Contents lists available at ScienceDirect

## Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# Fouling control in reverse osmosis for water desalination & reuse: Current practices & emerging environment-friendly technologies

Asif Matin <sup>a,b,\*</sup>, Tahar Laoui <sup>c,d,\*\*</sup>, Wail Falath <sup>a,b,e,\*</sup>, Mohammed Farooque <sup>f</sup>

<sup>a</sup> Center of Research Excellence in Desalination & Water Treatment, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia

<sup>b</sup> Center for Environment & Water, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia

<sup>c</sup> Dept. of Mechanical & Nuclear Engineering, University of Sharjah, Sharjah 27272, United Arab Emirates

<sup>d</sup> Desalination Research Group, University of Sharjah, Sharjah 27272, United Arab Emirates

<sup>e</sup> Dept. of Mechanical Engineering, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia

<sup>f</sup> Desalination Technologies Research Institute. Saline Water Conversion Corporation. Jubail. Saudi Arabia

#### HIGHLIGHTS

## GRAPHICAL ABSTRACT

- RO membrane fouling is a major obstacle in sustainable desalination.
- Current control strategies are inadequate and risk to the environment.
- Ecofriendly technologies e.g. osmotic backwashing and enzyme cleaning are promising.
- Feed spacer surface and geometry modification is effective for biofouling control.
- Application of EMFs and ultrasonic waves need further investigation.

ARTICLE INFO

Accepted 22 September 2020

Available online 6 October 2020

Received in revised form 22 September 2020

Article history:

Keywords:

Reverse osmosis Membrane fouling

Fouling control

Received 2 July 2020

Editor: Paola Verlicchi

Eco-friendly techniques

## ABSTRACT

Reverse Osmosis (RO) is becoming increasingly popular for seawater desalination and wastewater reclamation. However, fouling of the membranes adversely impacts the overall process efficiency and economics. To date, several strategies and approaches have been used in RO plants and investigated at the laboratory-scale for their effectiveness in the control of different fouling types. Amid growing concerns and stringent regulations for the conservation of environment, there is an increasing trend to identify technologies that are effective in fouling mitigation as well as friendly to the environment. The present review elaborates on the different types of environment-friendly technologies for membrane fouling control that are currently being used or under investigation. It commences with a brief introduction to the global water crisis and the potential of membrane-based processes in overcoming this problem. This is followed by a section on membrane fouling that briefly describes the major fouling types and their impact on the membrane performance. Section 3 discusses the predominant fouling control/prevention strategies including feedwater pretreatment, membrane and spacer surface modification and membrane cleaning. The currently employed techniques are discussed together with their drawbacks,

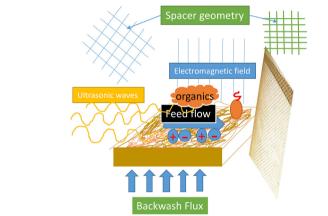
*Abbreviations*: BAC, biologically activated carbon; BSA, bovine serum albumin; DAF, dissolved air flotation; DOC, dissolved organic carbon; EPS, extracellular polymeric substances; HEMA, hydroxyethylmethacrylate; HMDP, hexamethylenediaminetetra (methylene phosphonic acid) – tetra phosphonate; NF, nanofiltration; NOM, natural organic matter; PFA, perfluorodecylacrylate; RO, reverse osmosis; RSF, rapid sand filtration; SDI, silt density index; SSF, slow sand filtration; SWRO, seawater reverse osmosis; TDS, total dissolved solids; TEP, transparent exopolymer particles; TSS, total suspended solids.

\* Corresponding authors at: Center of Research Excellence in Desalination & Water Treatment, King Fahd University of Petroleum & Minerals, Dhahran 31261, Saudi Arabia.

\*\* Correspondence to: Dept. of Mechanical & Nuclear Engineering, University of Sharjah, Sharjah 27272, United Arab Emirates.

E-mail addresses: amatin@kfupm.edu.sa (A. Matin), tlaoui@sharjah.ac.ae (T. Laoui), wfallata@kfupm.edu.sa (W. Falath).

Review









with some light being shed on the emerging technologies that have the ability to overcome the current limitations. The penultimate section provides a detailed discussion on a variety of eco-friendly/chemical free techniques investigated to control different fouling types. These include both control and prevention strategies, for example, bioflocculation and electromagnetic fields, as well as remediation techniques such as osmotic backwashing and gas purging. In addition, quorum sensing has been specifically discussed for biofouling remediation. The promising findings from different studies are presented followed by a discussion on their drawbacks and limitations. The review concludes with a need for carrying out fundamental studies to develop better understanding of the eco-friendly processes discussed in the penultimate section and their optimization for possible integration into the RO plants.

© 2020 Elsevier B.V. All rights reserved.

#### Contents

1.	Introduction	1				
1. 2.						
Ζ.	Membrane fouling					
	2.1. Inorganic					
	2.2. Organic					
	2.3. Biofouling					
	2.4. Colloidal					
3.	Current practices in fouling control					
	3.1. Pretreatment of feedwater					
	3.2. Membrane surface & spacer modification	8				
	3.2.1. Membrane surface	8				
	3.2.2. Spacer modification	8				
	3.3. Membrane cleaning	10				
	3.3.1. Chemical cleaning	10				
	3.3.2. Physical cleaning	10				
4.	Emerging eco-friendly fouling control technologies					
	4.1. Pretreatment	11				
	4.1.1. Microbial based	11				
	4.1.2. Gas purging					
	4.1.3. Electromagnetic fields & ultrasonic waves					
	4.1.4. Membrane-based					
	4.2. In situ methods					
	4.3. Membrane cleaning					
	4.3.1. Osmotic backwashing					
	4.3.2. Enzymatic cleaning					
	4.3.3. Ultrasonic waves					
5						
Declaration of competing interest.						
	Acknowledgments					
Refe	References					

## 1. Introduction

Water is an essential component for different sectors of economy such as agriculture, health, industry, tourism, and domestic consumption (Green et al., 2015). Continuously increasing population, rapid industrialization in many developing countries (Baten and Stummeyer, 2012), as well as water pollution from agricultural residues, sewage and industrial waste (Yao et al., 2016), has resulted in a disproportionately large imbalance between fresh water demand and supply (Oloukoi et al., 2013; Avrin et al., 2015; Cuerva et al., 2016). Currently, around one-third of the world's population is living in water-stressed regions with >1.2 billion people affected by clean water scarcity. According to reliable sources, these figures are expected to increase to around one-half (World Health Organization, 2014) and 3 billion (FAO, 2015), respectively, by the year 2025. In light of the above, the cost-effective and sustainable provision of clean and potable water has been identified by economists and researchers alike as the challenge for this century (Chen et al., 2018).

Desalination of seawater and inland water resources is a promising strategy to mitigate the current global water crisis and meet the rising demand for fresh water in a sustainable manner (Gu et al., 2013). Accounting for >95% of the planet's water resources, the water of the oceans represents a huge untapped reservoir that, if utilized in an efficient manner, can go a long way in overcoming the water shortage. According to recent statistics, there exists a total of around 20,000 desalination plants in >150 countries resulting in the daily production of ~100 million m<sup>3</sup> fresh water (Goh et al., 2018a, 2018b).

Furthermore, wastewater reclamation and reuse present a decent alternative in resolving the issue of water scarcity (Voulvoulis, 2018). Especially in regions with lack of natural water resources, the use of membranes for wastewater reclamation has proven to be a cost-effective route (Tang et al., 2016; Melián, 2020). For example, in Singapore, the municipal authorities installed two wastewater treatment plants with fairly large capacities, 32,000 m<sup>3</sup>/day and 40,000 m<sup>3</sup>/day, using Hydranautics' low fouling RO membranes (Lee and Tan, 2016; Timm and Deal, 2018). Several urban areas in different regions of the planet have also implemented large-scale projects on wastewater reuse (Hamoda et al., 2015; Kim et al., 2018a, 2018b).

The use of membranes in the above technologies is a very costeffective route to produce high quality water and has become widespread (Lin et al., 2016; Ochando-Pulido et al., 2016; Tang et al., 2016). The most popular membrane-based purification processes are Reverse Osmosis (Yang et al., 2017) (RO), Nanofiltration (Ribera et al., 2014) (NF), and Membrane Distillation (Bush et al., 2016) (MD). Among these, RO has the most widespread use in brackish & seawater desalination, drinking water production, and wastewater treatment (Gao et al., 2016), accounting for ~70% of the total installed capacity (Bashitialshaaer, 2020). The main advantage of this technique is that it is an energy-efficient technology for desalination, with energy costs now down to ~1.8 kWh/m<sup>3</sup>, much lower than that of other technologies (Kim et al., 2019a, 2019b). In the wake of more stringent regulations concerning public health and environmental protection, RO membranes possess the dual advantage of maintaining good water permeability and very high rejection rates for nearly all organic, inorganic and pathogenic micropollutants (Bieber, 2017).

Another important advantage of RO and similar processes, is the leverage to improve output parameters further by the incorporation of nanomaterials with exceptional transport characteristics. For example, novel materials such as zeolites, carbon nanotubes, and graphene, have the potential of high flux membranes for water desalination (Humplik et al., 2011). The ability to control the pore size at the Angstrom level (~ 0.5 nm) is important for the efficient working of such membranes. The recent increase in popularity of RO compared to contemporary desalination techniques can also be gauged from the number of research papers published in relevant scientific journals in the previous decade (Fig. 1).

However, membrane fouling is a major impediment to the use of membrane technology for such applications, because fouling is inevitable. Membrane fouling causes multiple adverse effects on the performance of RO systems (e.g., decreased production, water quality deterioration and decreased lifetime (Matin et al., 2011). The major types of fouling in water purification technologies include precipitation of inorganic salts, accumulation of suspended particulate matter, and formation of a biofilm that is a mixture of organic matter and microorganisms (She et al., 2016; Goh et al., 2018a, 2018b). The annual estimated costs for control and preventive measures undertaken to alleviate biofouling in the desalination industry worldwide is estimated to be ~ US\$ 15 billion (Flemming, 2011).

## 2. Membrane fouling

Fouling is defined as the undesired accumulation of deposits on the membrane surface that results in a decrease in water passage and a concomitant increase in solute passage across the membrane (Malaeb and Ayoub, 2011). Based on the foulant type, fouling is classified as follows (Fig. 2): (i) inorganic (scaling) (ii) organic, (iii) biofouling, and (iv) colloidal. In actual plants, fouling is a result of a combination of these different types and can be quite complicated.

Due to the relatively compact and nonporous nature of the RO membrane, surface fouling is comparatively more frequent than internal fouling. In contrast to internal fouling, surface fouling is easier to control by manipulation of feed water hydrodynamics and/or chemical cleaning (Du et al., 2017). Surface fouling development may be classified into three key mechanisms (Lei et al., 2016): cake, scale, and biofilm formation. Cake formation is caused by deposition of colloidal matter (inorganic, organic, or biological) onto RO membrane surface (Uppu et al., 2019). Scale formation results from heterogeneous crystallization of sparingly soluble mineral salts directly on RO membrane surface. Biofilm formation occurs when previously deposited microorganisms proliferate and colonize a membrane module (Vitzilaiou et al., 2019). The significance of RO fouling and its impact on global economics can be appraised by the recent surge of research activities in this field directed at a better understanding of the fouling mechanisms and design and development of more effective control and prevention strategies (Fig. 3).

Presented below is a brief yet concise description of the different fouling types and their impact on the overall process:

#### 2.1. Inorganic

Inorganic fouling or mineral scaling is the deposition of sparingly soluble salts e.g.  $CaCO_3$  (Mitrouli et al., 2016),  $CaSO_4$  (Benecke et al., 2018),  $SiO_2$ , etc. on the membrane surface to form hard scales (Antony et al., 2011). Fig. 4 depicts typical salt crystals deposited on a membrane surface. This type of fouling is more widespread in brackish water desalination (lakes, estuaries, wells etc.) where high water recovery ratios (> 70%) elevates the concentration of the above salts by a factor of 4–10 resulting in their spontaneous precipitation from the solution (Matin et al., 2019). However, it is also observed in seawater

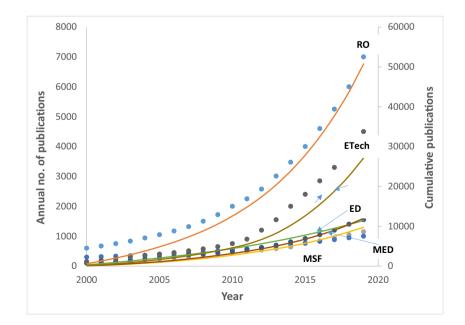


Fig. 1. Statistical data showing annual no. of publications (individual points) and cumulative publications (solid lines) for the major desalination technologies during the last two decades. (Reverse Osmosis [RO], Multi-Effect Distillation [MED], Multi-Stage Flash [MSF], Electrodialysis [ED]), Emerging technologies (ETech) include Nanofiltration [NF], Forward Osmosis [FO] and Membrane Distillation [MD]; Note the recent surge of interest in membrane-based processes compared to thermal counterparts.

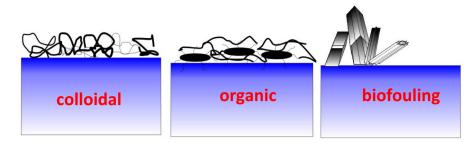
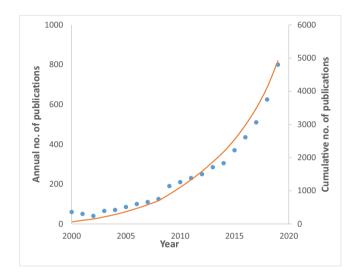


Fig. 2. Schematic sketch of the common types of fouling on RO membranes. Note the differences in the nature and morphology of the foulants.



**Fig. 3.** Statistics for annual no. of publications (blue dots) and cumulative publications (solid line) on RO fouling and control during the previous two decades. Note the exponential increase in the last 3–4 years.

desalination (Fortunato et al., 2020), in particular silica fouling that is usually present as silicates of Fe and Al that have a lower solubility as compared to SiO<sub>2</sub>.

The precipitated salt crystals deposit on the membrane surface and grow in the lateral direction resulting in complete surface coverage that is hard to remove (Ruiz et al., 2020). The presence of an extra barrier layer increases the hydraulic resistance of the membrane and therefore, yields a decline in permeate water flux as well as salt rejection. Another factor contributing to the deterioration of filtration characteristics is the formation of a cake layer that hinders the back-diffusion of salts away from the membrane surface (Shirazi et al., 2010). This results

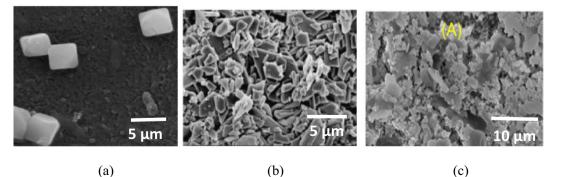
in an accumulation of salts near the membrane surface that increases the local osmotic pressure and reduces the driving force  $(\Delta P - \Delta \pi)$  for reverse osmosis (Ibrar et al., 2019).

Although a variety of mineral salts have been identified in membrane autopsy studies, the two most commonly encountered salts in the desalination of surface and ground waters are  $CaCO_3$  and  $CaSO_4$ (Hoang, 2015). The two proposed mechanisms for scale formation are homogeneous crystallization (nuclei formation in bulk solution), and heterogeneous precipitation (nuclei formation on membrane surface) (Shmulevsky et al., 2017). In the majority of scenarios, the latter mechanism prevails due to more favorable energetics and thermodynamics. However, it is widely believed that in actual situations, mineral scale formation typically occurs by a combination of the two contrasting mechanisms. In addition to the two major scale-forming salts mentioned above,  $Ca_3(PO_4)_2$  is a cause of concern in desalination of wastewater for domestic use (Rathinam et al., 2018).

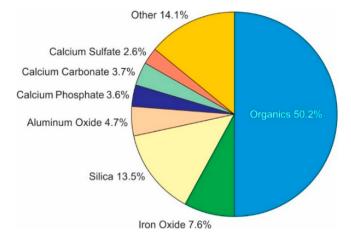
#### 2.2. Organic

Organic fouling is caused by the deposition and attachment of organic macromolecules on the membrane surface. The presence of a wide variety of organic matter, e.g. proteins, polysaccharides and humic substances, is ubiquitous in any water system, and depending upon the type of feedwater, their concentrations may vary. The two major categories of organic matter are: (i) Natural organic matter (NOM) produced by the degradation & decomposition of living organisms, and (ii) Transparent exopolymer particles (TEPs) derived from organic substrates released by aquatic organisms (Meng and Liu, 2015). Organic fouling is widespread and occurs in seawater desalination as well as wastewater reclamation; it is the most common fouling type after biofouling (Fig. 5).

It is well known that upon immersion in water, a conditioning film forms on the surface of most substrates that is composed of the organic substances mentioned above. The adsorbed organic compounds are capable of recruiting other types of foulants, e.g. microorganisms, to the surface (Dobretsov, 2009). The consequences of organic fouling are



**Fig. 4.** FESEM images depicting salt crystals deposition on membranes surfaces (a) Square-shaped CaCO<sub>3</sub> crystals after 10 h of operation with pretreated seawater (b) CaSO<sub>4</sub> grains of rhombohedral structure after cross-flow filtration without a scale inhibitor (Rahman, 2013) (c) Silica scaling after filtration studies with a laboratory setup at moderate pressure and cross-flow velocity (Tong et al., 2017).



**Fig. 5.** The incidence of different fouling types excluding biofouling from the autopsies of 150 different membranes. Note the dominant presence of organic fouling in exactly half of the cases (Kucera, 2019).

similar to its inorganic counterpart, a permanent decline in water flux as well as an increase of salt passage through the membrane. The organic compounds bind themselves to the polyamide layer of the membrane due to forces of adhesion that result from electrostatic and van der Waals interactions between the functional moieties from both sides (Yang et al., 2010). Further fouling development occurs as a result of foulant-foulant interactions that are facilitated by divalent cations (e.g. Ca<sup>2+</sup>). Fig. 6 shows some images and pictures of RO membranes fouled by organic matter during cross-flow filtration.

Natural Organic Matter (NOM) is the main organic foulant in the membrane treatment of surface waters, brackish waters and seawater and is made up of hydrophilic and hydrophobic fractions (Silva et al., 2019). The hydrophobic components are the major contributors to the permeate flux decline due to their greater adsorption on the membrane surface (Lee et al., 2020a, 2020b).

#### 2.3. Biofouling

Biofouling refers to the attachment of different bacterial species to the membrane surface. Microorganisms are almost always present in the feed water and their attachment to any immersed substrate is facilitated by the formation of a conditioning film comprised of organic macromolecules described in the previous section on organic fouling. The attached bacteria grow, multiply and excrete extracellular polymeric substances (EPS) to form a biofilm that is slimy in nature and difficult to remove. This type of fouling has been referred to as the "Achilles heel of membrane processes" and is the most problematic among the different types (Flemming et al., 1997). Biofouling is of the highest concern in wastewater treatment and reuse, followed by seawater desalination; statistics reveal that almost half of the cases of membrane fouling are caused by excessive growth of biomass (Komlenic, 2010). Fig. 7 is a collection of some pictures and images of biofouled RO membranes showing presence of bacterial cells and the biofilm on the membrane surface.

Biofouling occurs because the feed water for RO operation contains a variety of microorganisms that grow, multiply and form colonies in the presence of nutrients. In actual practice, multiple pretreatment procedures such as pre-filtration and disinfection are applied (Jamaly et al., 2014; Kim et al., 2015), however, even if 99.999% of the bacteria are eliminated, the remaining few will attach to the membranes surface and ultimately form a biofilm (Chinu et al., 2009). Membrane surface characteristics such as wettability, charge and roughness constitute an important parameter that has a strong influence on microbial attachment and hence, on biofilm formation, and as such membrane surface properties, such as wettability; charge; and topography, seem to be crucial (Habimana et al., 2014). Typically, hydrophobic negatively charged and rough surfaces are more prone to initial bacterial adhesion and biofouling.

A variety of microbial species/strains has been observed to contribute to biofilm formation in water desalination systems e.g. *Acinetobacter, Flavobacterium, Fusobacteria, Sphingobacteria, Arthrobacter, Cyanobacteria,* and *Aeromonas* (Vitzilaiou et al., 2019). In addition to the above species, studies have shown that microalgae produce EPS that play a crucial role in the formation of biofilms (Xiao and Zheng, 2016). One of the main precursors of membrane biofouling in desalination processes has been identified as transparent exopolymer particles (TEPs) (Nguyen et al., 2012). These comprise of sticky organic microgels that also include EPS, polysaccharides, proteins, acids, sulfates, etc. (Villacorte et al., 2009).

The consequences of biofouling on membrane processes are similar to other fouling types but more pronounced and can be summarized as follows (Kochkodan and Hilal, 2015):

- (a) Permanent decline in permeate flux due to presence of gel-like barrier in the form of biofilm.
- (b) Increase in salt passage across the membrane due to enhanced solute concentration polarization (back-diffusion of solutes hindered (Herzberg and Elimelech, 2007)
- (c) Increase in the differential pressure across the membrane module.
- (d) Degradation of the membrane and/or similar material in the RO module.

#### 2.4. Colloidal

Colloidal fouling is a result of the accumulation of particles intermediate in size between suspended solids and true dissolved solids

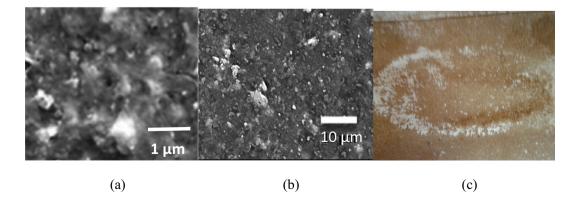


Fig. 6. Images of RO membranes fouled by organic matter during cross-flow filtration with a salt solution at pressures of ~800 psi (a & b) FESEM images showing sodium alginate deposition (Matin et al., 2016) (Shafi et al., 2017) (c) Camera pictures of humic acid deposition.

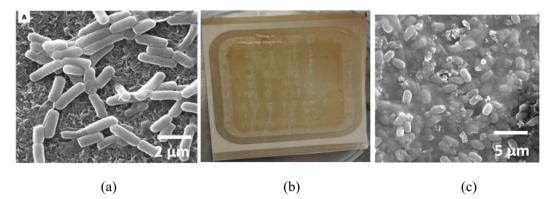


Fig. 7. Images depicting the presence of microorganisms on RO membranes (Abdulazeez et al., 2019) (a) *B. subtilis* cells in a colony formed after a few hour exposure to a suspension under no-flow (b) Presence of a biofilm (brown colour) after 8 h of cross-flow filtration at ~800 psi (c) *P. aeruginosa* cells embedded in an EPS matrix after filtration studies.

(1–1000 nm) (Xia et al., 2019) on RO membranes. Colloidal particles are difficult to deal with because they are small enough to pass through most pretreatment systems (dual media filtration, sand filtration, etc.), yet large enough to be rejected and concentrated at the membrane surface (Aimar and Bacchin, 2010). In practice, most of these particles are aluminosilicates with sizes ranging from 0.3 to 1.0 µm in diameter. Colloidal fouling is more pronounced in nanofiltration (NF) membranes as compared to reverse osmosis (RO) due to lower level of pretreatment carried out for the former (Yaun and Kilduff, 2010). However, inspite of the extensive pretreatment that removes a majority of particulate matter, this type of fouling still occurs in seawater reverse osmosis (Ruiz et al., 2020), in addition to brackish water desalination.

The colloids present in natural waters can be of different types (Chang, 2016); inorganic (SiO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub>/Fe (OH)<sub>3</sub>, AlSiO<sub>4</sub>, etc.), organic (polysaccharides, proteins, NOM, etc.), or biological (microorganisms, viruses, etc.) An important aspect that governs this type of fouling is the physicochemical interaction between the suspended particles and with the membrane surface (Yiantsios et al., 2005). The reversibility or irreversibility of the particle attachment to the surface is dictated by these interactions. Fig. 8 shows some images of membrane surfaces fouled by colloidal silica under different types of feed waters.

The main mechanism by which colloidal fouling interferes with the membrane performance is thought to be cake-enhanced concentration polarization (Ju and Hong, 2014). In this phenomenon, a porous cake layer enhances the concentration polarization near the membrane surface that increases the osmotic pressure and hence the driving force for permeate water passage, resulting in flux decline (Gutman and Herzberg, 2013). The other mechanism usually associated with biocolloids is the increase in hydraulic resistance due to the presence of an additional foulant layer (Dreszer et al., 2013). The increase in overall thickness through which the water has to pass reduces the membrane

permeability necessitating a higher operational pressure to maintain a constant flux.

## 3. Current practices in fouling control

## 3.1. Pretreatment of feedwater

An effective route for the control of different types of fouling is the use of feed water pretreatment system to remove foulants and their precursors. The pretreatment of seawater, or for that matter any other feed water, is performed to make it suitable for the main purification process (RO in this case) by lowering the levels/concentrations of relevant parameters namely total dissolved solids (TDS), turbidity, silt density index (SDI), bacterial species, and colloidal particles, to acceptable levels (Kavitha et al., 2019).

A reliable and adequate pretreatment system will ensure provision of good quality RO feedwater resulting in stable, long-term performance of RO membrane elements (Voutchkov, 2017). Moreover, slower fouling rates of the membrane and spacer will increase membrane lifetime, decrease cleaning frequency and hence, lower the overall maintenance costs associated with the plant. On the contrary, an ineffective or unreliable pretreatment may adversely affect plant productivity and operational costs due to one or more of the following: (i) high rates of membrane fouling resulting in more frequent membrane cleaning as well as reduced membrane life, (ii) low recovery rates, (iii) high operating pressure, and (iv) poor product quality (Anis et al., 2019).

The major pretreatment techniques and the configurations used in industrial RO plants and their utility are presented in Fig. 9. The usual sequence is as follows: after the initial screening to remove relatively large particles (> 1 mm), the seawater undergoes coagulation or a combination of coagulation and flocculation to facilitate the removal of algae and

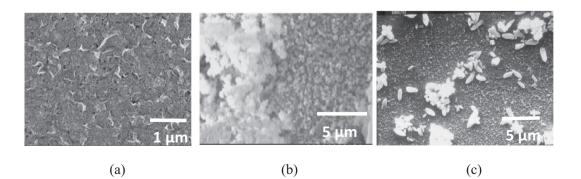


Fig. 8. FESEM images of different types of colloidal fouling on RO membranes (a) feed solution with 4200 ppm silica (Ho et al., 2016) (b) silica fouling with paper mill effluent (Ang et al., 2015) (c) fouling due to silica after aluminum addition (Gabelich et al., 2005).

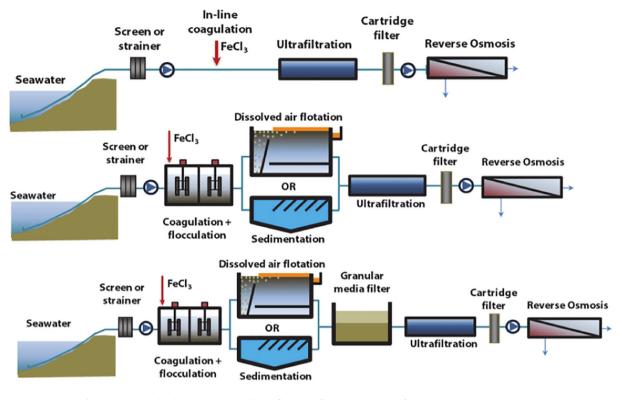


Fig. 9. A schematic sketch showing the possible configurations for RO pretreatment of seawater (Abushaban et al., 2017).

organic biopolymers such as humic substances. The most popular choice for the coagulant in RO plants is FeCl<sub>3</sub> due to the high-charge density of the cation and its relatively low solubility in seawater (Tabatabai et al., 2014). This is usually followed by dissolved air flotation (DAF) or sedimentation for the removal of suspended matter and algae. It is worth mentioning that DAF plants are also being used to remove oil slicks and reduce turbidity in addition to the foulants mentioned earlier. The feed is then flowed across an Ultrafiltration (UF) membrane either directly or via a granulated media filter (GMF).

With an average pore size in the range 20-50 nm, UF membranes are able to effectively block the passage of suspended particulates, colloidal materials, algae and pathogens and protect the RO module by physically separating the solids (Bonnélye et al., 2008). This results in excellent RO feedwater quality with very low levels of turbidity (< 0.1 NTU), SDI (1.0–2.5), and TSS (< 2 mg/L) (Valavala et al., 2011). In addition, compared to conventional pretreatment, the use of these membranes all but eliminates the need for RO disinfection/cleaning and can bring ~30% cost saving due to less space requirements.

Inspite of the above-mentioned advantages, there does exist a limitation with using UF for RO feed pretreatment. For example, UF membranes are very effective in the removal of microorganisms and suspended solids as confirmed by the findings from several pilot plant and commercial studies, yet they allow the passage of much smaller organic molecules that ultimately cause biofouling.

In addition to the above-mentioned techniques, chemicals for reducing the likelihood for biofouling and mineral scaling are added to the feed seawater. These include biocides, e.g. chlorine and sodium hypochlorite that mainly target the different bacterial species present in the RO feed water (Ayache et al., 2013; Sanchez, 2018). Similarly, scale inhibitors that are usually polyelectrolytes are added to interfere with the precipitation of sparingly soluble mineral salts such as CaCO<sub>3</sub>, CaSO<sub>4</sub>, etc., (Lee et al., 2020a, 2020b).

Some of the pretreatment technologies result in the control of multiple fouling types. For example, coagulation not only destabilizes suspended solids in water (Harif et al., 2012) but is also effective in reducing the content of dissolved organic carbon (DOC) (Hakizimana et al., 2015), which contributes to both organic and biofouling. Similarly, both suspended solids and microorganisms can be removed by media filtration (Monnot et al., 2016) using granular activated carbon that has the ability to remove a variety of toxic substances from water (Gamage and Sathasivan, 2017).

However, the conventional pretreatment techniques discussed above are not very efficient and produce feedwater of unsteady quality. The following major disadvantages of these techniques contribute to higher rates of RO membrane fouling and shorter RO membrane life: (i) Particles smaller than 10 µm in size are removed in less proportions, (ii) During filter backwash, there is a likelihood of breakthrough, and (iii) Significant fluctuations in the RO feed quality due to changing raw water conditions. Similarly, the use of biocides no matter how effective they may be, does not guarantee effective prevention of biofouling. Even if few microorganisms remain, they will still attach to the membrane surface, grow, multiply and eventually form a biofilm that will interfere with permeate water flux. Likewise, a particular antiscalant might be very effective in suppressing the precipitation of a certain mineral salt, but virtually ineffective against other salts. Also, findings of recent studies have shown common scale inhibitors to accelerate biofouling by acting as nutrients for the microorganisms (Ashfaq et al., 2019).

No matter how effective a certain pretreatment strategy is, it has always some limitations or drawbacks. For instance, dissolved organic carbon (DOC), identified as the dominant foulant in most RO membrane plants (Sioutopoulos and Karabelas, 2012), cannot be removed effectively by MF/UF membranes alone. The ideal strategy is to use a combination of different techniques in a manner that they complement each other. (Zhang et al., 2015) investigated the efficacy of different pretreatment combinations in mitigating organic fouling from a wastewater feed. They observed that a combination of ozonation, microfiltration and Biological Activated Carbon (BAC) resulted in minimal fouling as well as easy foulant removal from the membranes.

In some instances, the combination of a conventional technique with a membrane-based one did not significantly affect the proportion of foulant removal, but was helpful in reducing membrane fouling. For example, Ghaffour and co-workers (Li et al., 2016) investigated the removal efficiency of transparent exopolymer particles (TEP) by a combination of coagulation and ultrafiltration for minimizing SWRO fouling. They observed that coagulation with FeCl<sub>3</sub> at a pH of 5 resulted in a significant increase in the average size of TEPs; from colloidal range (0.1–0.4) to particulate size >0.4  $\mu$ m. Although the UF membranes removed most of the colloidal and particulate TEP when used alone, membrane fouling was significantly reduced after the coagulation step.

#### 3.2. Membrane surface & spacer modification

#### 3.2.1. Membrane surface

A novel strategy for fouling control and prevention in the spotlight for the last couple of decades is surface modification of the membranes (Rana and Matsuura, 2010). The main theme of this route is to alter the affinity of one or more foulant types to the membrane surface in a manner that its attachment and/or subsequent activity on the surface will be discouraged (Shahkaramipour et al., 2017). For example, it is wellknown that a majority of bacterial species readily adhere to hydrophobic and non-polar surfaces (Yuan et al., 2017) due to initial interactions that are hydrophobic in nature. Therefore, a feasible option explored by many researchers is to make the surface more hydrophilic. However, it should be kept in mind that such a surface will be more susceptible to fouling by hydrophilic organic matter that are usually present in feed waters (Mustafa et al., 2016).

Typically, for a surface modification technique to be effective for fouling control, one or more of the following surface properties need be modified: (i) wettability (Yin et al., 2017), (ii) roughness (Jiang et al., 2020), and (iii) electric charge (Wang et al., 2020). It is wellknown that the adhesion of different types of foulants e.g. bacteria, organics and to some extent salt crystals is influenced by the combination of these factors.

Researchers have explored several approaches to modify the surfaces of commercial RO membranes. These include:

- 1. Thin film deposition or surface coating (Jee et al., 2016).
- 2. Grafting of molecules (Liu et al., 2019) with desired functional groups.
- 3. Physical adsorption on the surface (Asadollahi et al., 2017).

Thin film deposition is perhaps the simplest of the above approaches and has been utilized by many researchers for membrane surface modification. A wide variety of techniques have been used under this category including, sol-gel (Kim et al., 2018a, 2018b), *initiated* Chemical Vapor Deposition (*i*CVD) (Ince et al., 2013; Yang et al., 2011), spin coating (Fadhillah et al., 2012), layer by layer assembly (Rahman et al., 2014) (Lbl). The materials used for surface modification are quite diverse and include polymeric molecules (PEG (Louie et al., 2011), (Sagle et al., 2009), PVA (Hachisuka and Ikeda, 2001), (Wu et al., 2006), QA compounds (Hibbs et al., 2016), organic compounds (silanes (Saffarimiandoab et al., 2019), zwitterionic (Azari and Zou, 2012; Shafi et al., 2015), polyelectrolytes, etc.), metals (Ag, Cu, etc.) and their oxides (TiO<sub>2</sub>,) as well as oxides of non-metals (e.g. graphene oxide). A summary of the different fouling control/prevention mechanisms associated with the different materials used for surface modification of RO membranes is provided in Table 1.

Gleason and co-workers (Matin et al., 2014a, 2014b) used an *initiated* CVD technique to deposit random amphiphilic copolymer films on commercial RO membranes. Two monomers with contrasting wetting properties, the hydrophilic HEMA and the hydrophobic PFA were polymerized and simultaneously deposited on flat sheet membranes. They observed that the presence of these coatings at near equal ratios of the two monomers significantly reduced the adsorption of a model protein (Baxamusa and Gleason, 2009), BSA, as well as attachment of *E. coli* cells in static flow conditions. Furthermore, in cross-flow filtration testing, the deposition of model organic foulants e.g. sodium alginate and humic acid was greatly reduced (Matin et al., 2016) and it was reversible (Shafi et al., 2017) compared to the commercial membrane.

Matin and co-workers recently fabricated antifouling and biocidal membranes by the deposition of a thin silane film and quaternization of its N atoms using a facile technique. The modified membranes not only discouraged the irreversible attachment of microorganisms but also inactivated the majority of bacteria that were able to attach. More importantly, the silane coating was able to maintain its efficacy in cross-flow conditions reminiscent of actual RO plants with negligible flux decline after prolonged operation with bacteria containing water.

An interesting approach to improve the fouling resistance of filtration membranes is to nano-structure their surface and tailor the surface topography (Bernards et al., 2008; Kang et al., 2008). In some instances, such a manipulation resulted in increased resistance to multiple and diverse types of fouling. For example, Cohen and co-workers grafted brush layers of two different polymers, the hydrophilic poly (methacrylic acid) (PMAA) and poly(acrylamide) (PAA), onto polyamide RO membranes. On comparison with low fouling commercial RO membranes, the modified membranes exhibited improved resistance to inorganic (Kim et al., 2010) and biofouling (Varin et al., 2013) and comparable resistance to organic compounds (BSA and SA) (Lin et al., 2010). The proposed antifouling mechanism is the screening of the polyamide layer from the different foulants types by the Brownian motion of the grafted polymeric chains that have been anchored terminally (Cohen et al., 2013).

#### 3.2.2. Spacer modification

Spacers, particularly the ones on the feed side, are important components of RO membrane modules with two main functions (i) separation

Table 1

A summary of the surface modification techniques used for fouling control and prevention in RO membranes.

Mechanism	Materials used	Findings	Limitations
Antifouling (Hydration layer formation)	Hydrophilic polymers (PEG, PDA (Baek et al., 2017) Zwitterions (PSBMA (Markovic et al., 2015), PCBMA)	Reduction in organics adsorption (BSA, SA, HA) & cell attachment	Fouling occurs in Long-term testing
Antimicrobial	Nanoparticles of Ag (Yang et al., 2009), TiO <sub>2</sub> (Khorshidi et al., 2018), GO (Huang et al., 2016), CNTs (Ihsanullah et al., 2016)	Good killing of different bacterial strains in static testing	Unstable in high-pressure flow conditons
Fouling release	Low surface energy polymers (PSVBP (Meng et al., 2014)	Good resistance to organics adsorption and easy release	Organic fouling occurs in long-term filtration
Antifouling & Antimicrobial	Silanes with Quaternizable Nitrogen	High bacterial killing Minimal flux decline in long-term testing Low bacterial adhesion in static	
Antiadhesive & Fouling Release	Amphiphilic copolymers (HEMA-PFDA (Matin et al., 2014a, 2014b) PAN-g-PEO (Asatekin et al., 2007)	conditions Good resistance to organic fouling (BSA, SA, HA)	Biofouling occurs in long-term tests
Antifouling Antimicrobial Fouling release	$\rm HFBM$ $+$ TOB (Wang et al., 2018 (hydrophilic OH, low surface energy CF and biocidal QA	Reduced foulant adhesion & high bacterial killing Good stability in long-term filtration	Tedious surface preparation

of the adjacent membranes layers, and (ii) reducing concentration polarization by the proper mixing of feed water (Haidari et al., 2018). They are typically made of polymers and optimized to maintain stable performance of membrane elements in feed waters of different compositions and varying process parameters. The hydraulic conditions of the feed channel; i.e. pressure drop and cross-flow velocity, are very much dependent on the geometry and configuration of the spacers.

Recent experimental investigations with NF/RO spiral wound membranes have shown the feed spacers to be the main contributors to biofouling by providing support to the microbes to attach and grow (Vrouwenvelder et al., 2009). It is the biomass accumulation on the spacer rather than on the membrane itself that causes most of the pressure drop increase in the feed channel and changes the flow velocity profile affecting the performance of membrane systems. Furthermore, the net-like structure of feed spacers, hinders the complete removal of the biofoulants (Creber et al., 2010) (inactivated by chemical cleaning agents) by trapping them (Bereschenko et al., 2011), resulting in rapid regrowth of the biofilm.

There are several characteristics of the spacer material that may influence bacterial adhesion and ultimate biofilm formation. These include the surface condition, geometry and thickness of the spacers used. Recent attempts by researchers on spacer modification for improved fouling control/prevention have been directed at one or more of these attributes. The surface modification of membranes using thin film deposition, molecular grafting, etc. has been extensively investigated by researchers in the last couple of decades and was discussed in detail in the previous section. In this section, we will begin by reviewing efforts made at altering the surface chemistry or morphology of feed spacers.

Vrouwenvelder and colleagues (Araújo et al., 2012) modified the surfaces of both membranes and spacers with hydrophilic, antimicrobial and biocidal coatings. Although very effective in short-term static bacterial adhesion and protein adsorption tests, none of the approaches were able to prevent or limit biofouling in long-term cross-flow conditions. In experiments carried out using a membrane fouling simulator, there was no significant reduction in the two major indicators of biofouling: (i) feed channel pressure drop, and (ii) biofilm accumulation as measured by ATP and TOC content. The failure of spacer surface modification with biocidal materials, e.g. Cu, is attributed to the coating agent toxicity being rendered ineffective by EPS secreted by the microorganisms. The bacterial strains possessing a higher antibiotic resistance, attach to and form colonies on the coating metal ultimately covering it with their extracellular polymeric substances, thus making it favorable for other micro-organisms to accumulate and join the EPS matrix

(Ronen et al., 2016) experimented with feed spacers coated with different types of coatings deposited by different techniques that exhibited distinct antifouling mechanisms. One of the spacers was modified by covalently binding polymeric quaternary ammonium groups (pQAs) by atom transfer radical polymerization (ATRP), while the other was embedded with silver nanoparticles (nAg) by sonochemical deposition. Cross-flow experiments were then conducted with a feed containing different microbial species at high concentrations and hydrodynamics simulating spiral wound modules. Permeate flux decline was considerably lower in both cases with a more steadier profile for the nano-Ag coated spacer probably due to the release of antibacterial species into the feed; the QA spacer showed a more localized killing upon contact strategy.

Kemperman and co-workers (Wibisono et al., 2015) experimented with a unique two-pronged approach in that they modified the feed spacer for reduced microbial attachment and then used the coated spacer for more effective cleaning. The antifouling properties of the spacers were enhanced by the deposition of hydrogels with different surface charge (positive, negative and neutral) using a plasmainduced polymerization. The negatively charged spacer showed reduced attachment of *E. coli* and significantly delayed biofilm formation. Furthermore, when this spacer was employed for biofilm removal in 2phase flow cleaning, the cleaning efficiency was enhanced as witnessed by a good flux recovery.

Other studies have focused on the influence of spacer geometry on fouling. The geometrical parameters include the spacer thickness, the internal strand angle, and the distance between the strands (meshsize). For example, (Park et al., 2016a, 2016b) investigated the effect of feed spacer thickness on membrane fouling behavior and the subsequent cleaning efficiency in a pilot plant operated almost a month. In addition, they also studied the fouling load distribution among the individual elements in pressure vessels by measuring normalized differential pressure. They observed that the use of spacers with more thickness not only reduced the fouling rate but also resulted in the elements being fouled uniformly.

(Radu et al., 2014) investigated deposition of microspheres mimicking bacterial cells in feed spacer channels for various spacer orientations under cross-flow conditions. They carried out both in situ microscopic observations as well as predictions of numerical models for particle trajectory calculations. The spacer orientations examined were the diamond orientation (D,  $\alpha = 45^{\circ}$ ) and the ladder orientation (L,  $\alpha =$ 90°). They observed particle deposition patterns specific to the type of spacer orientation and also influenced by cross-flow velocity.

(Siddiqui et al., 2016) performed a comprehensive and detailed study that included numerical modeling, 3-D printing and experimental fouling investigations with modified feed spacers. The following geometrical modifications with respect to the commercial ones were carried out: (i) reduction in filament angle (ii) increase in mesh size, and (iii) reduction in angle combined with mesh size increase. Based on mathematical modeling, they found that the pressure drop as a function of linear flow velocity was lowest for the spacers with a higher mesh size and the reduction in filament angle alone did not affect the pressure drop. These results were confirmed by experimental findings that revealed significantly lower pressure drop increase for the modified spacers (~ 500 mbar) as compared to the commercial ones (800 mbar) during the biofouling tests with the same amount of accumulated biomass as verified by the ATP measurements.

Since hydrodynamic conditions in the RO module are known to influence fouling, an interesting strategy is to combine spacer modification with manipulations in the flow conditions. Vrouwenvelder and colleagues used this approach (Vrouwenvelder et al., 2011) when they reduced the linear flow velocity in lead modules, changed the design for feed spacer and used an advanced cleaning strategy. They observed that this unique combination resulted in formation of biofilms that were fluffy in nature and can be easily removed from the lead membrane modules by the application of a reversed enhanced flush.

It is worth mentioning that a majority of surface modification endeavors discussed in the literature have shown to be effective only in short-term static testing for adhesion of microorganisms and/or biopolymer adsorption. The dilemma is that lab-scale static experiments use concentrated cultures of a particular species e.g. *E. coli*, whereas, in actual feedwaters, the fouling community is very diverse and highly complex (Bereschenko et al., 2008). The situation is very different in actual RO plants due to the complex nature of foulants, hydrodynamic conditions and the presence of other components such as feed and permeate spacers.

For instance, polymer brushes are considered for antibiofouling applications because of their proven ability to strongly discourage protein adsorption and hence, the attachment of microorganisms (Aubin et al., 2011; Bog et al., 2017). However, it has been shown that large shear forces during high-pressure cross-flow filtration may readily disrupt the structure of adsorbed polymer brush layers (Brzozowska et al., 2011), destroying the complex coacervate-brush structure. In a similar manner, biocidal nanoparticles of Ag, TiO<sub>2</sub>, GO, etc. deposited or incorporated in the membrane surface, gradually leach out in filtration studies. Another aspect worth considering is that a majority of surface modification strategies mentioned in literature have been directed against a certain type of fouling e.g. biofouling or organic fouling. This is a major handicap from a practical point of view since in actual feedwaters the chemistry is far too complex and synergistic effects of different fouling types play an important role. Therefore, there is a need to develop and design materials and techniques that will be simultaneously effective against multiple fouling types.

There have been some recent efforts in this direction. For example, Emadzadeh and co-workers (Emadzadeh et al., 2017) modified the surfaces of TFC membranes by incorporating nanoporous titanate nanoparticles (synthesized by solvothermal method) into the selective polyamide layer. Long-term filtration tests were performed with feedwater containing different types of foulants: organic, inorganic and multicomponent synthesized water, brackish water or seawater. In all scenarios, the flux decline of the modified membranes was significantly lower than the virgin one.

#### 3.3. Membrane cleaning

Cleaning of membranes after a certain time interval is essential to remove the mass of foulants irreversibly deposited on the membrane surface and to restore the permeate water flux and salt rejection to their pre-fouled levels during water and wastewater treatment processes (Yang et al., 2013). The major types of cleaning methods are (i) physical, and (ii) chemical (Madaeni and Samieirad, 2010); although there exist some biological techniques (Erkan et al., 2018), as well. The main indicators of the efficiency or effectiveness of any cleaning method are determined by hydraulic resistance removal and flux recovery (Sohrabi et al., 2011). Periodic cleaning results in significant flux recovery even after considerable flux decline. It is worth emphasizing that cleaning should be initiated when flux decline is around 10% because greater decline increases the irreversible proportion of the fouling that may not be overcome by cleaning.

## 3.3.1. Chemical cleaning

Of all the membrane remediation strategies after fouling has occurred to cause significant flux decline, chemical cleaning is indispensable (Vrouwenvelder et al., 2010). Based on their nature and mechanism of action, the commonly used chemical agents may be divided into the following categories: (i) acids, (ii) alkalis, (iii) surfactants, (iv) chelating agents, and (v) enzymes. Typically, a certain type of chemical is more effective against a particular fouling type (Table 2). For example, acids, such as HCl, HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub> are effective in removing mineral scaling formed by the deposition of sparingly soluble inorganic salts e.g. CaCO<sub>3</sub>, CaSO<sub>4</sub> (Filloux et al., 2015). On the other hand, alkaline solutions such as NaOH are more effective in removing organic and biofouling (Al-Amoudi and Lovitt, 2007).

The mechanism by which alkaline solutions (NaOH) work is to increase the solution pH that increases the negative charge on the organic foulant layer due to deprotonation (e.g. COO<sup>-</sup>) and hence it solubility. On the other hand, acids react with sparingly soluble salts e.g. CaCO<sub>3</sub>, CaSO<sub>4</sub> to convert them into their more soluble counterparts such as CaCl<sub>2</sub>. Chelating/sequestration agents form an integral part of any chemical cleaning procedure for RO membranes. For example, it is

#### Table 2

Typical combinations of chemical cleaning agents for the different fouling types.

Colloidal/Organic Alkali, surfactants & chelating agents   Metal oxides Citric acid (pH ~ 2) or sodium hydrosulfite   Silica NaOH at pH ~ 12   Carbonate scales (CaCO <sub>3</sub> ) Citric/HCl acid at pH ~ 2   Sulphate scales (CaSO <sub>4</sub> , BaSO <sub>4</sub> ) HCl solutions or sequestration agents (EDTA)   NaOH solutions, chelating or sequestration agents, surfactants and disinfectants	Fouling type	Chemicals used
Surfactants and uisinfectants	Metal oxides Silica Carbonate scales (CaCO <sub>3</sub> ) Sulphate scales (CaSO <sub>4</sub> , BaSO <sub>4</sub> )	Citric acid (pH ~ 2) or sodium hydrosulfite NaOH at pH ~ 12 Citric/HCl acid at pH ~ 2 HCl solutions or sequestration agents (EDTA)

well-known that divalent ions esp. Ca<sup>2+</sup> form a bridge between organic macromolecules such as alginate and humic acid that bound the foulant irreversibly to the membrane surface. Metal chelating agents remove such metal ions that weaken the structural integrity of the organic foulants (Fig. 10) and facilitate their removal (Hong and Elimelech, 1997). The commonly used chelating agent is ethylene diamine tetra acetic acid (EDTA) whose cleaning efficiency is very sensitive to solution pH (Chong et al., 2019). Similarly, surfactants increase the solubility of organic macromolecules by forming micelles around them (Cui et al., 2008).

The cleaning efficiency of any chemical used is thought to depend on two factors: (i) chemical reaction between the cleaning agent and the foulant(s) present, and (ii) mass transfer of the cleaning molecules from bulk phase to the foulant layer. In the case of biofouling, it has been observed that some cleaning chemicals acted more effectively on moderately fouled membranes as compared to heavily-fouled ones. One possible explanation is a lower mass transfer due to the more compact nature of the biofilm for the latter that may have reduced the permeability of the chemical into the fouling layer (Ang et al., 2011).

However, there are several inherent disadvantages associated with the use of chemicals in cleaning of fouled membranes. The first and foremost being the degradation of membranes as the polyamide layer is vulnerable to chemical attack by the different reagents used e.g. acids, alkalis and surfactants. In addition, since chemical cleaning requires frequent stoppage of the RO operation, this will result in lower production. Finally, the environmental issues related to the waste chemical disposal is also an important aspect that cannot be ignored (Qin et al., 2010). It should be noted that even with aggressive chemicals, a complete (100% effective) performance recovery of the RO membrane is rarely achieved.

#### 3.3.2. Physical cleaning

Physical cleaning methods use mechanical forces to dislodge and remove foulants from the membrane surface. This type of cleaning usually detaches loosely bound cake layers from the membrane surface, but is limited in its ability to eliminate all fouling layers. The effectiveness of physical cleaning tends to decrease over time, so that some portion of the fouling layer becomes irreversible. Based upon their configuration and the mode of force application, the conventional techniques in this category are as follows: (i) Flushing, (ii) Backwashing, (iii) Sponge Ball cleaning, and (iv)  $CO_2$  back permeation.

Flushing is done by passing the permeate water at high cross-flow velocities along the feed channel of the membrane module. Due to the shear forces caused by increased turbulence in the flow, many particulate matter adsorbed to the surface are removed and washed away. In forward flushing, the flow direction is from the feed to the brine and is particularly useful in the removal of colloidal matter (Ebrahim, 1994). On the other hand, in reverse flushing the flow direction is opposite and is beneficial to remove organic matter (Fig. 11).

Backwashing is a technique in which the original direction of permeate flow is reversed, i.e. from the permeate to the feed side. Regular backwashing is accomplished by applying a larger pressure on the permeate side of the membrane. This forces the permeate to flow across the membrane with some velocity and dislodge the contaminants present on the active layer. A special type of backwashing is osmotic backwashing, wherein, the flow reversal is achieved by replacing the regular feed with a highly concentrated salt solution. This is discussed in more detail in the next section.

Sponge ball cleaning involves the insertion of sponge balls made from polyurethane or a similar material into the membrane module for a very short duration and scrubbing the foulant deposited on the surface. It is usually used for cleaning membranes that have been heavily fouled by wastewater or industrial process water. Experiments conducted with secondary effluent from the terminal plant at Osaka, Japan, showed that scrubbing the foulant by a rubber sponge alone was sufficient to restore the permeate flux to the original level without

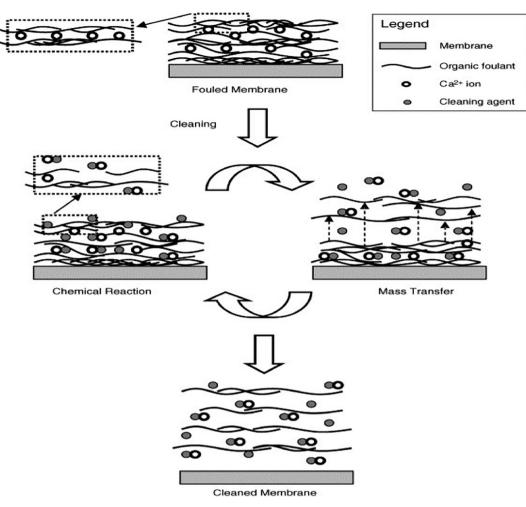


Fig. 10. Schematic sketch showing the mechanism of organic foulant removal from membrane surface. Successive foulant layers are linked to each other by Ca<sup>2+</sup> ions that form a bridge. Introduction of a cleaning agent disrupts the link between different layers that are then easily removed by hydrodynamics and mass transfer.

a need for sand filtration or cleaning chemicals (Yanagi and More, 1980).

## 4. Emerging eco-friendly fouling control technologies

Although many of the fouling control and remediation techniques mentioned in the previous section are effective and some are implemented on an industrial scale, yet there are some major disadvantages or handicaps associated with them. For example, none of the feedwater pretreatment routes have proved to significantly improve biofouling control primarily due to the fact that complete elimination of microorganisms present in the feedwater is quite impossible. Similarly, no antifouling/biocidal coating has shown the ability to completely resist bacterial adhesion or inactivate all the microorganisms upon contact and hence, cannot be implemented for full-scale plant operation.

Chemical cleaning of the fouled membranes, although very effective in removing most of the foulants and restoring the membrane performance to near initial levels, degrades the membranes, resulting in a considerable shortening of the expected lifetime, thereby necessitating frequent change and replacement, leading to higher operating costs. Furthermore, safe disposal of the used chemicals is a major challenge and a threat to the surrounding environment. Therefore, intense efforts are indispensable and underway to develop non-conventional technologies that will be both effective and implementable in real situations as well as friendly to the environment.

Here we present a brief description of some selective emerging technologies that have been under the research spotlight during the previous decade and possess the potential of overcoming the current obstacles and hurdles to cost-effective and sustainable desalination. Each of these techniques falls under one of the main categories discussed in the previous section: feedwater pretreatment, RO module surface modification, and membrane cleaning.

#### 4.1. Pretreatment

#### 4.1.1. Microbial based

Rapid sand filtration (RSF) or filtration through granular media is currently the most popular pretreatment technique for the removal of suspended solids (>0.35 mm) and reduction of silt density index (SDI) levels to ~ 4 in raw seawater. Recent investigations have also proven its capability of reducing particulate and dissolved organic carbon, chlorophyll and transparent exopolymeric particles (TEP). In comparison, slow sand filtration (SSF) with much higher residence times (12–24 h) and lower flow velocities is the obvious choice for wastewater treatment. The main advantage of SSF is the occurrence of biofiltration i.e. the decomposition of different types of organic pollutants present in the wastewater by microorganism that have the ability to form biofilm on the granular filter bed medium surface. It is thought that combining RSF with biofiltration will eliminate the need for using coagulants that represent an environmental hazard.

Zeev et al. (2013) investigated the effectiveness of a chemical-free, microbial-based pretreatment methodology based on bioflocculation. They utilized a novel RSF configuration with a highly porous filter media that provided a large surface area for bacterial growth and

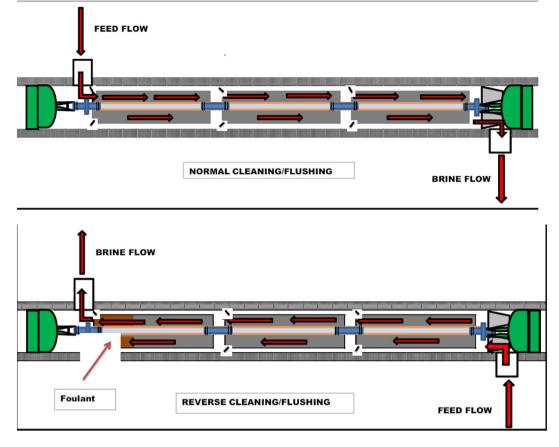


Fig. 11. Schematic sketch of DI water flushing of RO membrane modules (a) normal flushing (b) reverse flushing. The only difference is the reversing in the direction of water flow.

biofilm development. The decline in key parameters related to fouling such as silt density index (SDI), turbidity and transparent exopolymer particles (TEP) was monitored to assess the efficacy of this technology in reducing fouling potential. The findings after 1 year of continuous operation showed that the use of this 2-stage rapid bioflocculation filter (RBF) without any chemical additive was as effective as a regular RSF used with a coagulant  $Fe_2(SO_4)_2$ .

Another innovative and environment-friendly route for fouling control is the removal of organic nutrients present in feed water that support biofilm formation by using biological reactors, as proposed by Researchers at the Center for Biofilm Engineering at the University of Montana (Wend et al., 2003). Deliberate encouragement of biological growth within a component of the pretreatment train to substantially reduce the undesirable growth on reverse osmosis membranes was also a part of the strategy. The biological pretreatment columns used 2 types of packing media; (i) sand coated with Fe<sub>2</sub>O<sub>3</sub> for adsorption of natural organic matter (NOM), and (ii) biologically activated carbon (BAC) to promote biological growth. The findings showed that the biological treatment was capable of reducing downstream membrane fouling compared to untreated control water: reduction of fouling layer thickness by half and cell counts by four- to five-fold.

## 4.1.2. Gas purging

(Shahid et al.) utilized  $CO_2$  purging to control scale formation on RO membranes used for wastewater reclamation. The introduction of  $CO_2$  lowered the pH of the feed solution and consequently increased the solubility of scale-forming salts that were discharged in the concentrate stream rather than depositing on the membrane surface. In fact, several characterization findings such as membrane surface morphology (SEM/EDS) and surface chemistry (FTIR) indicated that gas purging alone was superior compared to the use of antiscalant in suppressing scale formation as well as better operational stability. In addition,  $CO_2$  is more

environment friendly in comparison to scale inhibitors that result in discharge of by-products in the concentrate stream.

In other instances, gas purging has been found to be effective in fouling control when used in combination with another strategy. Horseman and co-researchers (Horseman et al., 2019) combined periodic gas purging with the use of superhydrophobic membranes when using Membrane Distillation (MD) to concentrate a highly saline feed by 5fold. They observed that there was almost no crystal deposition and consequently no flux decline for the superwetting membranes as compared to a commercial hydrophobic membrane that had the presence of many crystal anchors and significant flux decline.

#### 4.1.3. Electromagnetic fields & ultrasonic waves

Electromagnetic fields (EMFs) have been used for many years to control deposition/scaling of inorganic salts on pipe walls of public and industrial water system (Lipus et al., 2011). Several researchers have investigated the efficacy of using electromagnetic fields for the control and prevention of mineral scale formation in RO membranes. The findings have been contrasting with ~15% of the studies showing the application of EMF to be virtually ineffective for fouling mitigation (Piyadasa et al., 2017). This is attributed mainly to the complex configuration in membrane modules including the presence of feed and permeate spacers and the hydrodynamic conditions prevailing near the membrane surface (Taherinejad et al., 2017). In addition, other parameters such as permeate water recovery are also thought to influence the efficacy of this eco-friendly technique for fouling control.

The suggested mechanism of fouling control/prevention is that EMFs enhance the coagulation of salt crystals such as CaCO<sub>3</sub>, which in turn decreases the likelihood of scale deposition on membrane surface. To confirm this hypothesis, (Piyadasa et al., 2017) examined the precipitation characteristics of CaCO<sub>3</sub> under the influence of pulsed electromagnetic fields (PEMFs) from two commercially available devices under controlled conditions. Ca(NO<sub>3</sub>)<sub>2</sub> and NaCO<sub>3</sub> were used as the parent solutions to precipitate CaCO<sub>3</sub>, the rate and profile of which was tracked, in parallel, by UV absorption at 350 nm and by turbidity measurements. The findings from the experiments coupled with the crystal morphology images (from SEM analysis) indicated that exposure to PEMFs from one of the devices enhanced the coagulation of the crystals.

(Jiang et al., 2019) investigated the effectiveness of EMF in fouling control during desalination of brackish groundwater using a pilot plant. The devices generating EMF were installed at two different locations: (i) before the cartridge filter, and (ii) in the RO feed inlet. The decline in permeate water flux was observed to reduce by 38.3% and 14.3% after >6 days and 15 days operation, respectively. Moreover, the foulant layer formed during the EMF application was less dense and easily removed by hydraulic flushing.

In addition to inorganic fouling, the application of EMFs has also been investigated for the control of biofouling. Similar to the abovementioned study on mineral scaling, Orbell and colleagues (Piyadasa et al., 2018) investigated the effect of two commercial pulsed EMF devices on the culturability of bacteria under controlled flow conditions. The findings showed the presence of both inhibitory as well as stimulatory effects on the viability of bacterial cells depending upon several factors such as device type, degree of flow and exposure time. An important observation from the experimental data was that generally, static conditions encouraged stimulatory effects i.e. enhanced the microbial activity, whereas flow conditions favored inhibitory effects implying reduced cell viability. This was very much consistent with findings from other studies (Faraj and Muhamad, 2012), (Bayir et al. 2015) reporting better microbial inactivation at a higher flow rate attributed to better mixing, allowing uniform treatment.

In addition to EMFs, ultrasound technology has also been investigated as a pretreatment option for fouling control. (Al-Juboori et al., 2012) studied the efficacy of using ultrasonic waves for controlling biofouling in an RO system. The ultrasound treatment was able to deactivate around~1000 colony forming units (CFUs) of *E. coli* per milliliter of the broth-based suspension and injuring >10% of the log survival of bacterial cells. This translated into a permeate flux recovery >0.1 L/ $m^2$ .h of the treated feed during the entire duration of the filtration test that lasted for two and a half days.

#### 4.1.4. Membrane-based

Membrane-based pretreatment has seen a recent increase in popularity as the conventional methods have not been very successful against deteriorating quality of feedwater. Micro- and ultrafiltration (UF) are mainly used for this purpose with some recent focus on nanofiltration (NF) (Kaya et al., 2015). (Herzberg et al. 2015) found that the salt rejection of RO membranes increased from ~94% to ~98.5% when microfiltration was used for pretreating secondary wastewater effluent. Similarly, Bae and co-researchers observed a 3-fold decrease in biofilm-forming bacteria with MF pretreatment as compared to the conventional media filtration (Bae et al., 2011). In addition to biofouling, membrane pretreatment was also found to be effective in the mitigation of particulate and colloidal fouling resulting in a lower permeate flux decline and an increase in the oxygen uptake rate.

Due to the vulnerability of polymeric MF membranes to fouling by biological and organic matter (Di Profio et al., 2011), a new and emerging concept is the use of low pressure ceramic membranes (Shang et al., 2014). Compared to their polymeric counterparts, these membranes are more oxidation-resistant, can tolerate backwashing at higher pressures (Hamad et al., 2013), and do not experience fiber breakage. In addition, ceramic membranes have a much lower fouling potential (Hofs et al., 2011) and are robust to chemical cleaning.

A promising alternative is to use NF membranes that are often termed as loose RO. With an average pore size of around few nanometers, these membranes are able to reject almost all particulate including colloidal and pathogenic matter and multivalent ions except for monovalent ions. The occurrence of inorganic fouling due to mineral scale formation is a pervasive problem in RO membrane desalination of inland water sources due to very high recovery ratios (70–90%). Operating at pressures around ~200 psi, NF membranes are able to reject between 75% and 90% of scale-forming divalent ions such as  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $SO_4^{2-}$ , and  $HCO_3^-$  (AbdelKader et al., 2018).

The high rejection of divalent salt ions as well as partial rejection of monovalent ions by the NF membrane have important practical applications as the osmotic pressure of the RO feed is lowered considerably. This implies possible operation at lower pressures resulting in a 25-30% decrease in energy consumption as well as the possibility of achieving a recovery rate > 70% for SWRO plants (Zhou et al., 2015). In addition, compared to actual RO membranes, these possess the inherent advantage of much higher water permeability that may be beneficial for the overall process economics. In a recent study, Tang and coresearchers (Yao et al., 2019) synthesized an NF-like FO (forward osmosis) membrane and used it for pretreatment of seawater. They observed that the fabricated membranes exhibited much higher water permeability and divalent ( $Ca^{2+}$ ,  $SO_4^{2-}$ , etc.) to monovalent ( $Na^+$ ,  $Cl^-$ , etc.) salt selectivity compared to a commercial reverse-osmotic-like (ROlike) FO membrane. Furthermore, the percentage rejection of both alginate and sulfate for the NF membranes was comparable to RO counterparts.

Another emerging concept is the application of hybrid organicinorganic membranes for pretreatment of feed waters. Antar and coresearchers (AbdelKader et al., 2019) synthesized such a membrane by depositing a layer of graphene oxide (GO) on the surface of PolyEtherSulfone (PES) using Polyacryl Amide (PAM) as an adhesive layer. The deposited GO was then reduced by using hydrogen iodide (HI) as a reducing agent and its attachment to the PES surface improved by PAM. Filtration tests with seawater revealed that the synthesized membrane gave a higher rejection for two important divalent cations, Mg<sup>2+</sup> and Ca<sup>2+</sup>, compared with commercial nanofiltration membrane (NF270).

#### 4.2. In situ methods

The previous section discussed techniques that target the feed reaching the RO membrane with the objective of minimizing the presence of one or more foulant types that are potential troublemakers. The following section focuses on membrane performance remediation after fouling has occurred to an extent that the membrane output, and hence, the process economics, have been significantly affected and need to be compensated. As discussed earlier, both of these strategies have their own limitations and therefore are not very efficient in fouling mitigation when used alone. This section will talk about the intermediate stage, that is, the techniques possessing the ability to interfere with the activities of the individual foulants on the membrane surface that lead to fouling aggravation.

Quorum sensing is a mechanism by which bacteria regulate gene expression in accordance with population density through the use of signal molecules. This allows microbial populations to regulate phenotype expressions, which in turn result in communication between them and coordination of group behavior. Formation of biofilms, production of EPS, and cell aggregation is a common phenotype associated with species such as *P. aeruginosa* (Sauer et al., 2002) and *Acinetobacter* (Bhargava et al., 2010), with the former also possessing the ability of rapid adaptation to changes in the surroundings.

Interfering with the QS regulatory system to disrupt the bacterial metabolism leading to EPS production and biofilm formation is a nondisinfectant biological approach to control membrane biofouling. A possible disruption pathway is to inhibit/suppress the production of molecules involved in QS signaling. (Ham et al., 2019) used structural derivatives of ginger, 6-gingerol analogs, to mitigate biofouling in RO membranes. They observed that these natural compounds were very effective in suppressing the formation of *P. aeruginosa* biofilms when used at low concentrations without affecting the growth of bacterial species. The resulting increase in permeate flux was in the range 35–50%, and moreover, there was no physical or chemical damage to the membrane surface.

Another strategy is to incorporate QS inhibitors into the membranes as deployed by Katebian and co-researchers (Katebian et al., 2018) who physically attached *vanillin* and *cinnamaldehyde* on RO membrane surfaces. Unlike the above study, in this case the membrane surface characteristics most notably the hydrophilicity was altered to reflect the QSI property. Biofilm formation was significantly disrupted as witnessed by >50% reduction in live & dead bacterial cells counts as well as polysaccharide production.

In some instances, the use of QSIs did not prevent biofilm formation, but the formed biofilm did not interfere with the RO performance. (Kim et al. 2019) used a fatty acid from a plant source, linoleic acid, to suppress *P. aeruginosa* biofilm formation. The film thickness was reduced by one third after application of small volumes of this natural product. Likewise, when used in a lab-scale RO system from the beginning, a highly porous biofilm formed that resulted in negligible flux decline when compared to control systems.

An important class of signaling molecules in quorum sensing are *N*-acyl homoserine lactones (AHLs) (Lade et al., 2014) produced by Gramnegative bacteria. Therefore, an effective biofouling control strategy is to target the activities of these molecules. (Siddiqui et al., 2012) investigated the efficacy of *Piper betle* extract (PBE) in suppressing the activities of AHLs for mitigation of membrane biofouling. They observed that this natural compound significantly reduced biofouling by inhibiting the production of AHL-related autoinducers (AIs). This was manifested by the decrease in EPS content of the biocake layer as shown by the reduced amounts of proteins and polysaccharides, two major components of the EPS. SEM images, showed the microbial count on the PBE-included surface to be much lower than the control.

#### 4.3. Membrane cleaning

#### 4.3.1. Osmotic backwashing

In micro- and ultrafiltration membrane systems, periodically reversing the permeate flow direction has proven to be effective in reducing fouling-related flux decline at both the laboratory and commercial scale (Raffin et al., 2012). This is accomplished by applying a larger pressure on the permeate side, causing the reverse flow of permeate into the feed side, and in the process removing deposited foulants as well as flushing the membrane pores. The same concept can be utilized in RO with the major difference being that the regular feedwater (brackish or seawater) is replaced by a salt solution of much higher concentration e.g. 1.5 M NaCl. The osmotic pressure causes the permeate water to flow in the opposite direction with some force and to dislodge the foulants attached to the membrane surface (Fig. 12).

Recently, researchers have also investigated this idea for the cleaning of RO membranes. For example, Elimelech and co-workers (Zeev and Elimelech, 2014) experimented this novel technique for a

commercial membrane fouled by an artificial wastewater. After heavy fouling of the membrane and formation of a dense and thick biofilm, the feed was replaced by a 1.5 M NaCl solution while maintaining an applied pressure of ~200 psi. The resulting backwashing by the permeate water resulted in the perforation and detachment of the biofilm accompanied by significant reduction in biofouling parameters, e.g. 70–90% in biovolume, > 70% removal of TOC and proteins. Approximately, 63% of the original flux was recovered.

Other studies investigated the efficacy of this potential technique for the removal of organic foulants from RO membranes. In a recent study, Hoek and co-researchers (Ramon et al., 2013) studied the effect of salt concentration and chemistries of the cleaning solution on the removal of a model organic foulant, alginic acid. They observed that the flux recovery and permeation rates of the membranes after cleaning increased with cleaning solution salinity (32, 64 and 96 g/L NaCl). Also, the best results for osmotic backwashing alone were comparable with a chemical cleaning combination of an alkali (NaOH) and a chelating agent (EDTA).

In addition to foulant layer perforation and detachment by the pressurized permeate flowing in the reverse direction, another mechanism is proposed for the removal of organic fouling from the membrane surface. Elimelech and co-workers<sup>206</sup> carried out a detailed and systematic study on cleaning of organic-fouled RO membranes using solutions of different monovalent cations (Na<sup>+</sup>, K<sup>+</sup>, Cs<sup>+</sup>, NH<sub>4</sub><sup>+</sup>) at different concentrations (25 mM, 100 mM). They observed the highest cleaning efficiency (75%) for NaCl at 25 mM and similar efficiencies (~ 90%) for all the salts at 100 mM. It was hypothesized that the foulant layer removal occurred in the following sequence (Fig. 13): (i) swelling in the presence of water, (ii) depletion of Ca<sup>2+</sup> in the foulant due to ion exchange with monovalent ions in cleaning solution, (iii) washing away of the loosened foulant by mass transfer.

Similarly, Semiat and colleagues experimented this emerging technology for membranes fouled by an inorganic salt,  $CaCO_3$ . They observed that an osmotic backwash cycle of only 20 s was sufficient to remove the salt crystals that had clogged the membrane and restore the permeate flux to its initial value. However, it should be noted that this was possible only when the cleaning was carried out immediately after the salt precipitation. It is well-known and observed that with the passage of time, the mineral scale hardens and becomes difficult to be removed<sup>46</sup>.

However, it is important to highlight that the majority of studies performed at laboratory-scale, including the studies reported above, deal with a single fouling type, e.g. biofouling alone, or a certain type of organic foulant. In actual RO plants, the fouling chemistry is very complex with the different types of fouling occurring simultaneously and influencing each other. Moreover, the hydrodynamics in the spiralwound RO modules is altogether different when compared to a simple lab setup that utilizes flat membrane sheets. Lastly, since backwashing involves pressurized flow in the opposite direction, the membranes being used must have pressure durability in both direction, which is generally not the case for spiral-wound membranes.

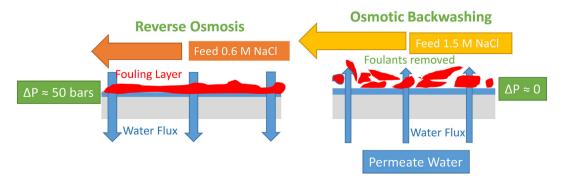


Fig. 12. Schematic sketch showing the comparison of reverse osmosis and osmotic backwashing. Note that in the latter, the permeate water is forced to flow in the reverse direction due to the high osmotic pressure and in the process dislodges the foulants that are transported away by high cross-flow velocity.

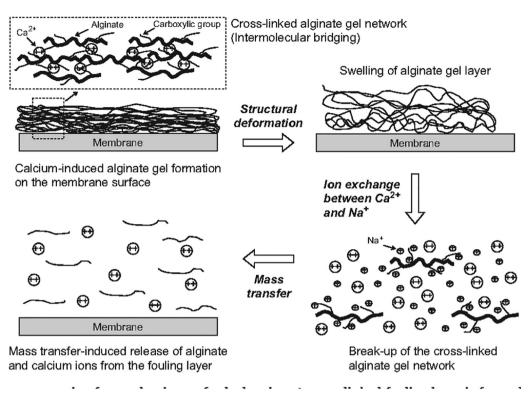


Fig. 13. Schematic sketch showing the stages of foulant deposition and its ultimate removal from the membrane surface using concentrated salt solution as the feed. The passage of saline water after fouling results in swelling of the foulant layer and its loosening due to replacement of the Ca with Na. The individual foulant layers are then swept away by the high cross-flow velocity (taken from Lee and Elimelech, 2007).

A group of scientists at the Saline Water Conversion Corporation (Farooque et al., 2014) in Jubail, Saudi Arabia, made an attempt for performance restoration of actual commercial RO membrane module using the OBW technique. They carried out both online and offline cleaning using a salt solution of very high concentration ~ 25% NaCl. However, the backwashing flux generated was only around half of the permeate flux during regular RO and not sufficient to cause significant foulant detachment and removal from the membrane surface. In fact, online cleaning with the hypersaline solution accelerated membrane fouling instead of mitigating it.

In spite of the uncertainty surrounding the implementation of this promising technique at the industrial scale, there exist some success stories in real plants. Desalination plants at the Gold Coast (Bartels et al., 2009), Australia, utilize the osmotic flushing offline i.e. when the high pressure pumps for RO plant are shut down. The permeate water is accumulated in elevated tanks in each train and is forced into the feed channels when the system is shut down for a very short duration ~30 s. This procedure facilitates in the removal of different foulant types e.g. biological, organic and colloidal present in the channel and/ or deposited on the membrane surface.

#### 4.3.2. Enzymatic cleaning

An alternate route for membrane cleaning is the use of environment-friendly enzymes to remove the biofoulants from RO membranes. In particular, enzymes have been found to be very effective in the removal of EPS protein foulants from the membrane surface by breaking the protein into small fragments (Puspitasari et al., 2010). The cleaning mechanism is hypothesized to be the hydrolysis of the proteinaceous and glycoprotein exopolymers surrounding the bacteria embedded in the biofilm matrix (Allie et al., 2003).

Although, the focus of recent attention, enzymes have been investigated for this application long before. (Whittaker et al., 1984) evaluated the effectiveness of different types of chemicals including surfactants, biocides, enzymes, etc. for the removal of biofilms from RO membranes. They found that a combination of enzyme-dispersant and antiprecipitant was one of the most effective way in cleaning as it reduced the original bacterial count by >90%.

The advantage of using enzymes for membrane cleaning is that they can function in mild conditions of pH, temperature and concentration that are not harmful to the RO membranes (Argüello et al., 2005). In addition, the use of eco-friendly enzymes significantly reduces the cost associated with chemicals and energy (Chen et al., 2006). Furthermore, unlike chemicals, enzymes have the ability to remove foulants from the membrane surface without altering the physico-chemical properties of the membrane.

(Khan et al., 2014) investigated the efficacy of several different enzymes in cleaning RO membranes operated continuously for almost 2 months in a rotating disk reactor. The initial screening shortlisted 3 of the most effective enzymes which were then utilized for removal of biofoulants accumulated on the membrane surface in neutral pH conditions and low dosage (50–150 ppm). The findings from the biofilm analysis and bacterial quantification indicated that protease and lipasebased enzymes had the highest cleaning efficiency and were able to restore the surface properties (wettability, roughness, etc.) to their original values. Even at lower dosing of 50 ppm, the enzymes were able to reduce the number of culturable cells in the biofilm by five logs.

In some instances, the use of enzymes alone for cleaning fouled membranes became more of a liability than improvement. (Rudolph et al., 2018) investigated the cleaning efficiency of polysaccharidedegrading and extractive-degrading enzymes on UF membranes used in the pulp and paper industry. They observed that the membrane permeability was lower after enzyme cleaning alone when compared to rinsing with DI water. Careful examination with SEM showed that the enzymes became additional foulants on the membrane surface. However, when the enzyme-cleaned membrane was cleaned with an alkaline chemical, the permeate flux was restored to the original level.

In general, investigations on the use of enzymes for cleaning fouled RO membranes are very limited. Further detailed studies need to be performed to gain further insights into the underlying mechanisms. It should also be kept in mind that enzymatic cleaning targets bio and organic fouling only; therefore, for effective cleaning of RO modules used in seawater desalination, it should be combined with other cleaning procedures that address the other fouling types i.e. inorganic scales and colloidal fouling.

## 4.3.3. Ultrasonic waves

The application of ultrasonic waves for enhancing permeation through the membrane has been extensively studied (Zhang et al., 2019). The underlying mechanism associated with this technique is cavitation that refers to the formation of bubbles in liquid, their growth and implosive collapse. Cavitation results in formation of hot spots in the liquid with extremely high local temperatures and pressures as well as high velocity microjet streams near the solid surface. The resulting combination of acoustic streaming and shear forces reduces the thickness of the boundary layer and hence its resistance to diffusion of water, resulting in an increased mass transfer coefficient and water permeability.

In addition to permeate flux enhancement, ultrasonic techniques have also been identified for membrane cleaning (Qasim et al., 2018). The effectiveness of an energy-efficient method was investigated using a focused ultrasound beam to produce cavitations for membrane cleaning. They observed that the membrane performance can be significantly restored with lower power.

(Feng et al., 2006) conducted a study to investigate the impact of online ultrasonic cleaning for a commercial RO membrane that was utilized for the filtration of synthetic solutions representing both organic (cellulose) and inorganic (CaSO<sub>4</sub>, FeCl<sub>3</sub>) foulants present in wastewater effluents. The findings showed significant increase (50–250%) in permeate flux after the cleaning procedure with no compromise on solute rejection for the different solutions. SEM images confirmed the near complete removal of the foulants by ultrasound with a surface morphology near identical to the original membrane.

The downside with ultrasound cleaning is the vulnerability of membranes to be damaged due to the intense cavitational collapse contingent on the power density, frequency, and the irradiation time of ultrasound. Zhang et al. conducted a detailed investigation on the main factors of ultrasonic technology influencing the filtration characteristics of a membrane as well as the influence of ultrasonic waves on membrane structure. They observed that although the use of ultrasound improved the membrane permeation yet in some cases membranes were damaged.

However, the membrane damage depended to a large extent on the ultrasonic power used. Generally, at low powers, no damage was reported, yet the cleaning efficiency and permeation enhancement were significant. For instance, Juboori and colleagues (Naji et al., 2020) investigated the direct application of low-power ultrasound (8–23 W), as an in-line cleaning and performance boosting technique for Air Gap Membrane Distillation used for treating RO reject streams. They observed a 100% increase in flux for the fouled membranes without any damage to the membranes.

Another important handicap is from the economy point of view. Weavers and co-workers (Lamminen et al., 2006) estimated the power requirement for ultrasound cleaning of DOW Chemical Filmtec NF270–4040 spiral wound NF membrane module used for processing ~4.5 million gallons of drinking water daily. It was calculated to be approx. 8.5 MW, that implied an additional cost of 0.45 cents per gallon of water produced. The operational cost will be further aggravated by the use of ultrasound transducers such as lead zirconate titanate (PZT) ceramics that could handle a high power to produce cavitations (Lu et al., 2009).

Table 3 summarizes the status of the above-mentioned eco-friendly fouling control/prevention technologies in terms of the level of advancement. As can be seen from the table, most of the techniques are still under investigation at the laboratory-scale and/or pilot plant level.

#### Table 3

Status of emerging eco-friendly fouling control/prevention strategies with regards to level
of advancement.

Technology & category	Current Status
Pretreatment	
Microbial-based	Pilot-plant testing
Gas purging	Laboratory-scale experiments
Electromagnetic field application	Pilot plant level
Membrane-based	Pilot-plant (Park et al. 2016)
In situ	
Quorum sensing	Laboratory scale experiments
Membrane cleaning	
Osmotic backwashing	Lab-scale investigations, pilot plant studies & field application
Enzymatic cleaning	Laboratory studies (membrane bioreactor <sup>214</sup> )
Ultrasonic waves	Experimental stage (cross-flow testing with flat sheet membranes)

This is because further investigations and findings are required for their full-scale integration into an RO plant. Of all these techniques, osmotic backwashing (OBW) is the only one that has been implemented in actual plants.

#### 5. Conclusions

Membrane fouling is a major impediment in sustainable desalination and water reuse by reverse osmosis. Fouling in actual plants is a very complex phenomenon that involves the synergistic effects of different fouling types on each other. Typical lab-scale fouling studies are carried out using a specific kind of foulant and are not representative of the actual conditions in RO plants. Among the fouling control strategies, feed water pretreatment using a sequence of different techniques, is perhaps the most effective on an industrial scale. However, a combination of conventional techniques alone is not sufficient to obtain a feed of good quality and delay fouling. More attention need to be focused on the integration of emerging and promising technologies such as nanofiltration, novel biocides and eco-friendly techniques e.g. electromagnetic fields with the conventional ones. In addition, combinations of individual processes should be selected in such a manner that they complement each other. For instance, the use of MF/UF membranes for the effective removal of suspended solids should be preceded by ozonation to increase the content of biodegradable organics and hence the fouling propensity of the membranes, and followed by BAC filters to remove the biodegradable organics.

Another strategy, membrane surface modification, has been investigated by many researchers and still remains a promising technique. However, there remains a big question mark over the scalability of most of the techniques and their effectiveness and consistency if scaled-up to the plant level. Another concern is their long-term stability under conditions of high pressure, variable temperature and continuous exposure to highly saline water with a complex foulant chemistry. As far as the antifouling performance is concerned, one can find many success stories in short-term static bacterial/protein adhesion tests; however, the surface-modified membranes have generally been found ineffective in long-term biofouling tests reminiscent of actual plant conditions.

Conventional cleaning with chemicals such as acids, alkalis, chelating agents and surfactants, is the current practice in industrial RO plants and no doubt is very effective in performance restoration of the fouled membranes. Given the inability of the current available physical cleaning methods in removing irreversibly attached foulants, it is considered absolutely essential when the permeate flux decline becomes significant resulting in operational cost increase. However, it has some major disadvantages, most notably lifetime shortening of membranes, high costs and environmental hazards. Furthermore, as discussed in this review, usually the chemicals are unable to completely remove the inactivated biomass due to obstruction by feed spacers. The remaining biomass results in rapid reformation of biofilms necessitating more frequent cleaning cycles.

Several innovative fouling control and remediation technologies are discussed in the penultimate section. Findings of studies at the laboratory scale are promising; however, none of them are in a ready state for implementation in actual plants. Further detailed investigations need to be carried out under varying conditions and with different types of foulants to develop a sound understanding of the fouling control mechanisms. Moreover, the feasibility at the pilot-plant level needs to be explored as the conditions are entirely different from the lab-scale.

Among the chemical-free cleaning methods, osmotic backwashing (OBW) appears to be promising as it has shown to be effective against different fouling types: biofouling, mineral scaling and organic fouling. In contrast, enzymatic cleaning mainly targets biofouling and has the inherent disadvantage of fouling aggravation instead of its mitigation. The other advantage with OBW is the easiness in scaling-up as it does not require any extra equipment as is the case with ultrasonic cleaning. However, once again, the efficacy of this technique needs to be proven at the pilot scale as one study showed otherwise.

To summarize, the global water situation is bleak and expected to worsen with the passage of time due to growing freshwater scarcity. Already, around a billion people do not have access to clean and potable water and this figure is expected to increase rapidly in the coming years. Due to their highly-efficient purification capabilities as well as energy efficiency, membrane processes such as reverse osmosis are expected to become more popular compared to other technologies for water treatment applications. However, there is a pressing need to improve the economics of these techniques to provide sustainable supplies of clean water at affordable cost throughout the planet, one of the two greatest challenges of this century (Geise et al., 2010).

#### **Declaration of competing interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

#### Acknowledgements

The authors would like to thank the Centers of Environment & Water and Desalination & Water Treatment, KFUPM and the Desalination Technologies Research Institute, Saline Water Conversion Corporation (SWCC), Al-Jubail for supporting this review via project no. CRDW2428.

#### References

- Abdelkader, B.A., Antar, M.A., Khan, Z., 2018. Nanofiltration as a pretreatment step in seawater desalination: a review. Arab. J. Sci. Eng. 43, 4413–4432.
- Abdelkader, B.A., Antar, M.A., Laoui, T., Khan, Z., 2019. Development of graphene oxidebased membrane as a pretreatment for thermal seawater desalination. Desalination 465, 13–24.
- Abdulazeez, I., Matin, A., Khan, M., Khaled, M.M., Ansari, M.A., Akhtar, S., Rehman, S., 2019. Facile preparation of antiadhesive and biocidal reverse osmosis membranes using a single coating for efficient water purification. J. Membr. Sci. 591, 17299.
- Abushaban, A., Rodriguez, S.G.S., Mondal, S., Goueli, S.A., Schippers, J.C., Kennedy, M.D., 2017. A new method of assessing bacterial growth in swro systems: method development and applications, The International Desalination Association World. Congress – São Paulo, Brazil.
- Aimar, P., Bacchin, P., 2010. Slow colloidal aggregation and membrane fouling. J. Membr. Sci. 360, 70–76.
- Al-Amoudi, A., Lovitt, W.L., 2007. Fouling strategies and the cleaning system of NF membranes and factors affecting cleaning efficiency. J. Membr. Sci. 303, 4–28.
- Al-Juboori, R.A., Yusaf, T., Aravinthan, V., 2012. Investigating the efficiency of thermosonication for controlling biofouling in batch membrane systems. Desalination 286, 349–357.
- Allie, Z., Jacobs, E.P., Maartens, A., Swart, P., 2003. Enzymatic cleaning of ultrafiltration membranes fouled by abattoir effluent. J. Membr. Sci. 218, 107–116.
- Ang, W.L., Mohammad, A.W., Hilal, N., Leo, C.P., 2015. A review on the applicability of integrated/hybrid membrane processes in water treatment and desalination plants. Desalination 363, 2–18.

- Ang, W.S., Yip, N.Y., Tiraferri, A., Elimelech, M., 2011. Chemical cleaning of RO membranes fouled by wastewater effluent: achieving higher efficiency with dual-step cleaning. J. Membr. Sci. 382, 100–106.
- Anis, S.F., Hashaikeh, R., Hilal, N., 2019. Reverse osmosis pretreatment technologies and future trends: a comprehensive review. Desalination 452, 159–195.
- Antony, A., Low, J.H., Gray, S., Childress, A.E., Le-Clech, P., Leslie, G., 2011. Scale formation and control in high pressure membrane water treatment systems: a review. J. Membr. Sci. 383, 1–16.
- Araújo, P.A., Miller, D.J., Correia, P.B., van Loosdrecht, M.C.M., Kruithof, J.C., Freeman, B.D., Paul, D.R., Vrouwenvelder, J.S., 2012. Impact of feed spacer and membrane modification by hydrophilic, bactericidal and biocidal coating on biofouling control. Desalination 295, 1–10.
- Argüello, M.A., Álvarez, S., Riera, F.A., Álvarez, R., 2005. Utilization of enzymatic detergents to clean inorganic membranes fouled by whey proteins. Sep. Purif. Technol. 41, 147–154.
- Asadollahi, M., Bastani, D., Musavi, S., 2017. Enhancement of surface properties and performance of reverse osmosis membranes after surface modification: a review. Desalination 420, 330–383.
- Asatekin, A., Kang, S., Elimelech, M., Mayes, A.M., 2007. Anti-fouling ultrafiltration membranes containing polyacrylonitrile-graft-poly(ethylene oxide) comb copolymer additives. J. Membr. Sci. 298, 136–146.
- Ashfaq, M.Y., Al-Ghouti, M.A., Qiblawey, H., Zouari, N., 2019. Evaluating the effect of antiscalants on membrane biofouling using FTIR and multivariate analysis. Biofouling 35, 1–14.
- Aubin, H.T., Chen, L., Ober, C.K., 2011. Fouling-resistant polymer brush coatings. Polymer 52, 5419–5425.
- Avrin, A.P., He, G., Kammen, D.M., 2015. Assessing the impacts of nuclear desalination and geoengineering to address China's water shortages. Desalination 360, 1–7.
- Ayache, C., Manes, C., Pidou, M., Croue, J.P., Gernjak, W., 2013. Microbial community analysis of fouled reverse osmosis membranes used in water recycling. Water Res. 47, 3291–3299.
- Azari, S., Zou, L., 2012, Using zwitterionic amino acid I-DOPA to modify the surface of thin film composite polyamide reverse osmosis membranes to increase their fouling resistance, J. Membr. Sci. 401–402:68–75.
- Bae, H., Kim, H., Jeong, S., Lee, S., 2011. Changes in the relative abundance of biofilmforming bacteria by conventional sand-filtration and microfiltration as pretreatments for seawater reverse osmosis desalination. Desalination 273, 258–266.
- Baek, Y., Freeman, B.D., Zydney, A.L., Yoon, J., 2017. A facile surface modification for antifouling reverse osmosis membranes using polydopamine under UV irradiation. Ind. Eng. Chem. Res. 56, 5756–5760.
- Bartels, C.R., Andes, K., Cannesson, N., Murphy, S., Cohen, D., 2009. "Energy Savings at the Gold Coast SWRO Plant", IDA 2009 – Perth (Poster Presentation).
- Bashitialshaaer, R., 2020. Solar-energy innovative and sustainable solution for freshwater and food production for Lake Titicaca Islands. European Journal of Engineering Research and Science 5, 436–442.
- Baten R., Stummeyer, K., How sustainable can desalination be?, 2012, Desalin. Water Treat. 1–9.
- Baxamusa, S.H., Gleason, K.K., 2009. Random copolymer films with molecular-scale compositional heterogeneities that interfere with protein adsorption. Adv. Funct. Mater. 19, 3489–3496.
- Benecke, J., Haas, M., Baur, F., Ernst, M., 2018. Investigating the development and reproducibility of heterogeneous gypsum scaling on reverse osmosis membranes using real-time membrane surface imaging. Desalination 428, 161–171.
- Bereschenko, L.A., Heilig, G.H.J., Nederlof, M.M., van Loosdrecht, M.C.M., Stams, A.J.M., Euverink, G.J.W., 2008. Molecular characterization of the bacterial communities in the different compartments of a full-scale reverse-osmosis water purification plant. *Appl.* Environ. Microbiol. 74, 5297–5304.
- Bereschenko, L.A., Prummel, H., Euverink, G.J.W., Stams, A.J.M., van Loosdrecht, M.C.M., 2011. Effect of conventional chemical treatment on the microbial population in a biofouling layer of reverse osmosis systems. Water Res. 45, 405–416.
- Bernards, M.T., Cheng, G., Zhang, Z., Chen, S.F., Jiang, S.Y., 2008. Nonfouling polymer brushes via surface-initiated, two-component atom transfer radical polymerization. Macromolecules 41, 4216–4219.
- Bhargava, N., Sharma, P., Capalash, N., 2010. Quorum sensing in Acinetobacter: an emerging pathogen. Crit. Rev. Microbiol. 36, 349–360.
- Bieber, S., 2017, Chapter 10 water treatment equipment for in-center hemodialysis, Handbook of Dialysis Therapy (Fifth Edition), pages 123-143.
- Bog, U., Pereira, A.S., Mueller, S.L., Havenridge, S., Parrillo, V., Bruns, M., Holmes, A.E., Emmenegger, C.R., Fuchs, H., Hirtz, M., 2017. Clickable antifouling polymer brushes for polymer pen lithography. ACS Appl. Mater. Interfaces 9, 2109–12117.
- Bonnélye, V., Guey, L., Castillo, J.D., 2008. UF/MF as RO pre-treatment: the real benefit. Desalination 222, 59–65.
- Brzozowska, A.M., Spruijt, E., deKeizer, A., Stuart, M.A.C., Norde, W., 2011. On the stability of the polymer brushes formed by adsorption of ionomer complexes on hydrophilic and hydrophobic surfaces. J. Colloid Interface Sci. 353, 380–391.
- Bush, J.A., Vanneste, J., Cath, T.Y., 2016. Membrane distillation for concentration of hypersaline brines from the Great Salt Lake: effects of scaling and fouling on performance, efficiency, and salt rejection. Sep. Purif. Technol. 170, 78–91.
- Chang, Q., 2016. Colloid and Interface Chemistry for Water Quality Control. Elsevier.
- Chen, B., Han, M.Y., Peng, K., Zhou, S.L., Shao, L., Wu, X.F., Wei, W.D., Liu, S.Y., Li, Z., Li, J.S., Chen, G.Q., 2018. Global land-water nexus: agricultural land and freshwater use embodied in worldwide supply chains. Sci. Total Environ. 613–614, 931–943.
- Chen, V., Li, H., Li, D., Tan, S., Petrus, H.B., 2006. Cleaning strategies for membrane fouled with protein mixtures. Desalination 200, 198–200.

- Chinu, K.J., Johir, A.H., Vigneswaran, S., Shon, H.K., Kandasamy, J., 2009. Biofilter as pretreatment to membrane based desalination: evaluation in terms of fouling index. Desalination 247, 77–84.
- Chong, V.Y.F., Koo, C.H., Thiam, H.S., Lai, S.O., 2019. Chemical cleaning of fouled polyethersulphone nanofiltration membranes with ethylenediaminetetraacetic acid. J. Appl. Membr. Sci. Technol. 23, 1–7.
- Cohen, Y., Lewis, G., Kim, M., Lin, N., 2013, "Fouling and scaling resistant surface nanostructured reverse osmosis membranes". U.S. Patent 8,445, 076, 5/21/2013.
- Creber, S.A., Vrouwenvelder, J.S., van Loosdrecht, M.C.M., Johns, M.L., 2010. Chemical cleaning of biofouling in reverse osmosis membranes evaluated using magnetic resonance imaging. J. Membr. Sci. 362, 202–210.
- Cuerva, L.G., Berglund, E.Z., Binder, A.R., 2016. Public perceptions of water shortages, conservation behaviors, and support for water reuse in the U.S. Resour. Conserv. Recycl. 113, 106–115.
- Cui, X., Mao, S., Liu, M., Yuan, H., Du, Y., 2008. Mechanism of surfactant micelle formation. Langmuir 24, 10771–10775.
- Di Profio, G., Ji, X., Curcio, E., Drioli, E., 2011. Submerged hollow fiber ultrafiltration as seawater pretreatment in the logic of integrated membrane desalination systems. Desalination 269, 128–135.
- Dobretsov, S., 2009, in Marine and Industrial Biofouling (Eds: H.-C. Flemming, P. S. Murthy, R. Venkatesan, K. Cooksey), Springer, Berlin.
- Dreszer, C., Vrouwenvelder, J.S., Paulitsch-Fuchs, A.H., Zwijnenburg, A., Kruithof, J.C., Flemming, H.-C., 2013. Hydraulic resistance of biofilms. J. Membr. Sci. 429, 436–447.
- Du, X., Wang, Y., Leslie, G., Lia, G., Liang, H., 2017. Shear stress in a pressure-driven membrane system and its impact on membrane fouling from a hydrodynamic condition perspective: a review. J. Chem. Technol. Biotechnol. 92, 463–478.
- Ebrahim, S., 1994. Cleaning and regeneration of membranes in desalination and wastewater applications: state-of-the-art. Desalination 96, 225–238.
- Emadzadeh, D., Ghanbari, M., Lau, W.J., Rahbari, S.M., Rana, D., Matsuura, T., Kruczek, B., Ismail, A.F., 2017. Surface modification of thin film composite membrane by nanoporous titanate nanoparticles for improving combined organic and inorganic antifouling properties. Mater. Sci. Eng. C 75, 463–470.
- Erkan, H.S., Turan, N.B., Engin, G.Ö., 2018. Chapter five membrane bioreactors for wastewater treatment. Compr. Anal. Chem. 81, 151–200.
- Fadhillah, F., Zaidi, S.M.J., Khan, Z., Khaled, M., Rahman, F., Hammond, P., 2012. Development of multilayer polyelectrolyte thin-film membranes fabricated by spin assisted layer-by-layer assembly. Appl. Polym. Sci. 126, 1468–1474.FAO Water Unit Water News: Water Scarcity 2015.
- Faraj, K.A., Muhamad, D.A., 2012. Effect of high magnetic field on gram negative bacteria. Eur. J. Sci. Res. 74, 240–243.
- Farooque, A.M., Al-Jeshi, S., Saeed, M.O., Alreweli, A., 2014. Inefficacy of osmotic backwash induced by sodium chloride salt solution in controlling SWRO membrane fouling. Appl Water Sci 4, 407–424.
- Feng, D., van Deventer, J.S.J., Aldrich, C., 2006. Ultrasonic defouling of reverse osmosis membranes used to treat wastewater effluents. Sep. Purif. Technol. 50, 318–323.
- Filloux, E., Wang, J., Pidou, M., Gernjak, W., Yuan, Z., 2015. Biofouling and scaling control of reverse osmosis membrane using one-step cleaning-potential of acidified nitrite solution as an agent, J. Membr. Sci. 495, 276–283.
- Flemming, H.-C., 2011. Microbial Biofouling: Unsolved Problems, Insufficient Approaches, and Possible Solutions. Springer, Berlin, Heidelberg, pp. 81–109.
- Flemming, H.-C., Schaule, G., Griebe, T., Schmitt, J., Tamachkiarowa, A., 1997. Biofouling the Achilles heel of membrane processes. Desalination 113, 215–225.
- Fortunato, L, Alshahri, A.H., Farinha, A.S.F., Zakzouk, I., Jeong, S., Leiknes, T.O., 2020. Fouling investigation of a full-scale seawater reverse osmosis desalination (SWRO) plant on the Red Sea: membrane autopsy and pretreatment efficiency. Desalination 114536.
- Gabelich, C.J., Chen, W.R., Yun, T.I., Coffey, B.M., Mel Suffet, I.H., 2005. The role of dissolved aluminum in silica chemistry for membrane processes. Desalination 180, 307–319.
- Gamage, S.M.K., Sathasivan, A., 2017. A review: potential and challenges of biologically activated carbon to remove natural organic matter in drinking water purification process. Chemosphere 167, 120–138.
- Gao, L.X., Rahardianto, A., Gu, H., Christofides, P.D., Cohen, Y., 2016. Novel design and operational control of integrated ultrafiltration – reverse osmosis system with RO concentrate backwash. Desalination 382, 43–52.
- Geise, G.M., Lee, H.-S., Miller, D.J., Freeman, B.D., Mcgrath, J.E., Paul, D.R., 2010. Water purification by membranes: the role of polymer science. J. Polym. Sci. B Polym. Phys. 48.
- Goh, P.S., Matsuura, T., Ismail, A.F., Ng, B.C., 2018a, The water-energy nexus: solutions towards energy-efficient desalination, Energ. Technol. 2 (2017); Global Water Intelligence IDA Water Security Handbook 2018–2019, ISBN: 978-1-907467-55-4, Oxford, UK.
- Goh, P.S., Lau, W.J., Othman, M.H.D., Ismail, A.F., 2018b. Membrane fouling in desalination and its mitigation strategies. Desalination 425, 130–155.
- Green, P.A., Vörösmarty, C.J., Harrison, I., Farrell, T., Sáenz, L., Fekete, B.M., 2015. Freshwater ecosystem services supporting humans: pivoting from water crisis to water solutions. Glob. Environ. Chang. 34, 108–118.
- Gu, H., Bartman, A.R., Uchymiak, M., Christofides, P.D., Cohen, Y., 2013. Self-adaptive feedflow reversal operation of reverse osmosis desalination. Desalination 308, 63–72.
- Gutman, J., Herzberg, M., 2013, Cake/biofilm enhanced concentration polarization, Encyclopedia of Membrane Science and Technology.
- Habimana, O., Semião, A.J.C., Casey, E., 2014. The role of cell-surface interactions in bacterial initial adhesion and consequent biofilm formation on nanofiltration/reverse osmosis membranes. J. Membr. Sci. 454, 82–96.
- Hachisuka, H., Ikeda, K., 2001, Composite reverse osmosis membrane having a separation layer with polyvinyl alcohol coating and method of reverse osmosis treatment of water using the same. US Patent 6,177,011 B1.

- Haidari, A.H., Heijman, S.G.J., van der Meer, W.G.J., 2018. Optimal design of spacers in reverse osmosis. Sep. Purif. Technol. 192, 441–456442.
- Hakizimana, J.N., Gourich, B., Vial, C., Drogui, P., Oumani, A., Naja, J., Hilali, L., 2015. Assessment of hardness, microorganism and organic matter removal from seawater by electrocoagulation as a pretreatment of desalination by reverse osmosis. Desalination 393, 90–101.
- Ham, S.-Y., Kim, H.-S., Jang, Y., Sun, P.-F., Park, J.-H., Lee, J.S., Byun, Y., Park, H.-D., 2019. Control of membrane biofouling by 6-gingerol analogs: quorum sensing inhibition. Fuel 250, 79–87.
- Hamad, J.Z., Ha, C., Kennedy, M.D., Amy, G.L., 2013. Application of ceramic membranes for seawater reverse osmosis (SWRO) pre-treatment. Desalin. Water Treat. 51, 4881–4891.
- Hamoda, M.F., Attia, N.F., Al-Ghusain, I.A., 2015. Performance evaluation of a wastewater reclamation plant using ultrafiltration and reverse osmosis. Desalin. Water Treat. 54, 2928–2938.
- Harif, T., Khai, M., Adin, A., 2012. Electrocoagulation versus chemical coagulation: coagulation/flocculation mechanisms and resulting floc characteristics. Water Res. 46, 3177–3188.
- Herzberg, M., Elimelech, M., 2007. Biofouling of reverse osmosis membranes: role of biofilm-enhanced osmotic pressure. J. Membr. Sci. 295, 11–20.
- Hibbs, M.R., McGrath, L.K., Kang, S., Adout, A., Altman, S.J., Elimelech, M., Cornelius, C.J., 2016. Designing a biocidal reverse osmosis membrane coating: synthesis and biofouling properties. Desalination 380, 52–59.
- Ho, J.S., Sim, L.N., Gu, J., Webster, R.D., Fane, A.G., Coster, H.G.L., 2016. A threshold flux phenomenon for colloidal fouling in reverse osmosis characterized by transmembrane pressure and electrical impedance spectroscopy. J. Membr. Sci. 500, 55–65.
- Hoang, T.A., 2015, Mechanisms of scale formation and inhibition. In Mineral Scales and Deposits, Scientific and Technological Approaches, 1st ed.; Z. Amjad, K. Demadis, Eds.; Elsevier: Amsterdam, The Netherlands, pp. 47–83.
- Hofs, B., Ogier, J., Vries, D., Beerendonk, E.F., Cornelissen, E.R., 2011. Comparison of ceramic and polymeric membrane permeability and fouling using surface water. Sep. Purif. Technol. 79, 365–374.
- Hong, S., Elimelech, M., 1997. Chemical and physical aspects of natural organic matter (NOM) fouling of nanofiltration membranes. J. Membr. Sci. 132, 159–181.
- Horseman, T., Su, C., Christie, K.S.S., Lin, S., 2019. Highly effective scaling mitigation in membrane distillation using a superhydrophobic membrane with gas purging. Environ. Sci. Technol. Lett. 6, 423–429.
- Huang, X., Marsh, K.L., McVerry, B.T., Hoek, E.M.V., Kaner, R.B., 2016. Low-fouling antibacterial reverse osmosis membranes via surface grafting of graphene oxide. ACS Appl. Mater. Interfaces 8, 14334–14338.
- Humplik, T., Lee, J., O'Hern, S., Fellman, B.T., Baig, M.A., Hassan, S.F., Atieh, M.A., Rahman, F., Laoui, T., Wang, E., Karnik, R., 2011. Nanostructured materials for water desalination. Nanotechnology 22, 1–19.
- Ibrar, I., Naji, O., Sharif, A., Malekizadeh, A., Alhawari, A., Alanezi, A.A., Altaee, A., 2019. A review of fouling mechanisms, control strategies and real-time fouling monitoring techniques in forward osmosis. Water 11, 695.
- Ihsanullah, Al Amer, A.M., Laoui, T., Abbas, A., Al-Aqeeli, N., Patel, F., Khraisheh, M., Atieh, M.A., Hilal, N., 2016. Fabrication and antifouling behaviour of a carbon nanotube membrane. Mater. Des. 89, 549–558.
- Ince, G.O., Matin, A., Khan, Z., Zaidi, S.M.J., Gleason, K.K., 2013. Surface modification of reverse osmosis desalination membranes by thin-film coatings deposited by initiated chemical vapor deposition. Thin Solid Films 539, 181–187.
- Jamaly, S., Darwish, N.N., Ahmed, I., Hasan, S.W., 2014. A short review on reverse osmosis pretreatment technologies. Desalination 354, 30–38.
- Jee, K.Y., Shin, D.H., Lee, Y.T., 2016. Surface modification of polyamide RO membrane for improved fouling resistance. Desalination 394, 131–137.
- Jiang, W., Xu, X., Lin, L., Wang, H., Shaw, R., Lucero, D., Xu, P., 2019. A pilot study of an electromagnetic field for control of reverse osmosis membrane fouling and scaling during brackish groundwater desalination. Water 11, 1015.
- Jiang, Z., Karan, S., Livingston, A.G., 2020. Membrane fouling: does microscale roughness matter? Ind. Eng. Chem. Res. 59, 5424–5431.
- Ju, Y., Hong, S., 2014. Nano-colloidal fouling mechanisms in seawater reverse osmosis process evaluated by cake resistance simulator-modified fouling index nanofiltration. Desalination 343, 88–96.
- Kang, G., Cao, Y., Zhao, H., Yuan, Q., 2008. Preparation and characterization of crosslinked poly(ethylene glyco) diacrylate membranes with excellent antifouling and solventresistant properties. J. Membr. Sci. 318, 227–232.
- Katebian, L., Hoffmann, M.R., Jiang, S.C., 2018. Incorporation of quorum sensing inhibitors onto reverse osmosis membranes for biofouling prevention in seawater desalination. Environ. Eng. Sci. 35, 261–269.
- Kavitha, J., Rajalakshmi, M., Phani, A.R., Padaki, M., 2019. Pretreatment processes for seawater reverse osmosis desalination systems—a review. Journal of Water Process Engineering 32, 100926.
- Kaya, C., Sert, G., Kabay, N., Arda, M., Yüksel, M., Egemen, Ö., 2015. Pre-treatment with nanofiltration (NF) in seawater desalination-preliminary integrated membrane tests in Urla. Turkey, Desalination 369, 10–17.
- Khan, M., Danielsen, S., Johansen, K., Lorenz, L., Nelson, S., Camper, A., 2014. Enzymatic cleaning of biofouled thin-film composite reverse osmosis (RO) membrane operated in a biofilm membrane reactor. Biofouling 30, 153–167.
- Khorshidi, B., Biswas, I., Ghosh, T., Thundat, T., Sadrzadeh, M., 2018. Robust fabrication of thin film polyamide-TiO<sub>2</sub> nanocomposite membranes with enhanced thermal stability and anti-biofouling propensity. Sci. Rep. 8, 784.
- Kim, D., Amy, G.L., Karanfil, T., 2015. Disinfection by-product formation during seawater desalination: a review. Water Res. 81, 343–355.

- Kim, D.-H., Park, S., Yoon, Y., Park, C.M., 2018a. Removal of total dissolved solids from reverse osmosis concentrates from a municipal wastewater reclamation plant by aerobic granular sludge. Water 10, 882.
- Kim, H.-S., Ham, S.-Y., Jang, Y., Sun, P.-F., Park, J.-H., Lee, J.H., Park, H.-D., 2019a. Linoleic acid, a plant fatty acid, controls membrane biofouling via inhibition of biofilm formation. Fuel 253, 754–761.
- Kim, J., Kim, C., Baek, Y., Hong, S.P., Kim, H.J., Lee, J.C., Yoon, J., 2018b. Facile surface modification of a polyamide reverse osmosis membrane using a TiO<sub>2</sub> sol-gel derived spray coating method to enhance the anti-fouling property. Desalin. Water Treat. 102, 9–15.
- Kim, J., Park, K., Yang, D.R., Hong, S., 2019b. A comprehensive review of energy consumption of seawater reverse osmosis desalination plants. Appl. Energy 254, 113652.
- Kim, M., Lin, N.H., Lewis, G.T., Cohen, Y., 2010. Surface nano-structuring of reverse osmosis membranes via atmospheric pressure plasma-induced graft polymerization for reduction of mineral scaling propensity. J. Membr. Sci. 354, 142–149.
- Kochkodan, V., Hilal, N., 2015. A comprehensive review on surface modified polymer membranes for biofouling mitigation. Desalination 356, 187–207.
- Komlenic, R., 2010. Biofouling: rethinking the causes of membrane biofouling. Filtration & Separation 47, 26–28.
- Kucera, J., 2019. Biofouling of polyamide membranes: fouling mechanisms, current mitigation and cleaning strategies, and future prospects. Membranes 9, 111.
- Lade, H., Paul, D., Kweon, J.H., 2014. Quorum quenching mediated approaches for control of membrane biofouling. Int. J. Biol. Sci. 10, 547–562.
- Lamminen, M.O., Walker, H.W., Weavers, L.K., 2006. Cleaning of particle fouled membranes during cross-flow filtration using an embedded ultrasonic transducer system. J. Membr. Sci. 283, 225–232.
- Lee, H., Tan, T.P., 2016. Singapore's experience with reclaimed water: NEWater. International Journal of Water Resources Development 32, 611–621.
- Lee, H.-J., Halali, M.A., Baker, T., Sarathy, S., Lannoy, C.-F., 2020a. A comparative study of RO membrane scale inhibitors in wastewater reclamation: Antiscalants versus pH adjustment. Sep. Purif. Technol. 240, 116549.
- Lee, S., Elimelech, M., 2007. Salt cleaning of organic-fouled reverse osmosis membranes. Water Res. 41, 1134–1142.
- Lee, Y.-G., Kim, S., Shin, J., Rho, H., Lee, Y., Kim, Y.M., Park, Y., Oh, S.-E., Cho, J., Chon, K., 2020b. Fouling behavior of marine organic matter in reverse osmosis membranes of a real-scale seawater desalination plant in South Korea. Desalination 485, 114305.
- Lei, Q., Zhang, M., Shen, L., Li, R., Liao, B.-Q., Lin, H., 2016. A novel insight into membrane fouling mechanism regarding gel layer filtration: Flory-Huggins based filtration mechanism. Sci. Rep. 6, 33343.
- Li, S., Lee, S.T., Sinha, S., Leiknes, T., Amy, G.L., Ghaffour, N., 2016. Transparent exopolymer particles (TEP) removal efficiency by a combination of coagulation and ultrafiltration to minimize SWRO membrane fouling. Water Res. 102, 485–493.
- Lin, L., Feng, C., Lopez, R., Coronell, O., 2016. Identifying facile and accurate methods to measure the thickness of the active layers of thin-film composite membranes - a comparison of seven characterization techniques. J. Membr. Sci. 498, 167–179.
- Lin, N.H., Kim, M.M., Lewis, G.T., Cohen, Y., 2010. Polymer surface nano-structuring of reverse osmosis membranes for fouling resistance and improved flux performance. J. Mater. Chem. 20, 4642–4652.
- Lipus, L.C., Ačko, B., Hamler, A., 2011. Electromagnets for high-flow water processing. Chem. Eng. Process. Process Intensif. 50, 952–958.
- Liu, T., Chen, D., Yang, F., Chen, J., Cao, Y., Xiang, M., Kang, J., Xu, R., 2019. Enhancing the permeability and anti-fouling properties of a polyamide thin-film composite reverse osmosis membrane via surface grafting of L-lysine. RSC Adv. 9, 20044–20052.
- Louie, J.S., Pinnau, I., Reinhard, M., 2011. Effects of surface coating process conditions on the water permeation and salt rejection properties of composite polyamide reverse osmosis membranes. J. Membr. Sci. 367, 249–255.
- Lu, J.-Y., Du, X., Lipscomb, G., 2009. Cleaning Membranes with Focused Ultrasound Beams for Drinking Water Treatment. Proceedings, IEEE International Ultrasonics Symposium.
- Madaeni, S.S., Samieirad, S., 2010. Chemical cleaning of reverse osmosis membrane fouled by wastewater. Desalination 257, 80–86.
- Malaeb, L., Ayoub, G.M., 2011. Reverse osmosis technology for water treatment: state of the art review. Desalination 267, 1–8.
- Markovic, M.G., Barclay, T.G., Constantopoulos, K.T., Markovic, E., Clarke, S.R., Matisons, J.G., 2015. Biofouling resistance of polysulfobetaine coated reverse osmosis membranes. Desalination 369, 37–45.
- Matin, A., Khan, Z., Zaidi, S.M.J., Boyce, M.C., 2011. Biofouling in reverse osmosis membranes for seawater desalination: phenomena and prevention. Desalination 281, 1–16.
- Matin, A., Khan, Z., Gleason, K.K., Khaled, M., Zaidi, S.M.J., Khalil, A., Moni, P., Yang, R., 2014a. Surface-modified reverse osmosis membranes applying a copolymer film to reduce adhesion of bacteria as a strategy for biofouling control. Sep. Purif. Technol. 124, 117–123.
- Matin, A., Shafi, H.Z., Khan, Z., Khaled, M., Yang, R., Gleason, K., Rehman, F., 2014b. Surface modification of seawater desalination reverse osmosis membranes: characterization studies & performance evaluation. Desalination 343, 128–139.
- Matin, A., Shafi, H., Wang, M., Khan, Z., Gleason, K., Rahman, F., 2016. Reverse osmosis membranes surface-modified using an initiated chemical vapor deposition technique show resistance to alginate fouling under cross-flow conditions: filtration & subsequent characterization. Desalination 379, 108–117.
- Matin, A., Rahman, F., Shafi, H.Z., Zubair, S.M., 2019. Scaling of reverse osmosis membranes used in water desalination: phenomena, impact, and control; future directions. Desalination Reviews 455, 135–157.
- Melián, J.A.H., 2020. Editorial sustainable wastewater treatment systems (2018–2019). Sustainability 12, 1940.

- Meng, J., Cao, Z., Ni, L., Zhang, Y., Wang, X., Zhang, X., Liu, E., 2014. A novel salt-responsive TFC RO membrane having superior antifouling and easy-cleaning properties. J. Membr. Sci. 461, 123–129.
- Meng, S., Liu, Y., 2015. New insights into transparent exopolymer particles (TEP) formation from precursor materials at various Na<sup>+</sup>/Ca<sup>2+</sup> ratios. Sci. Rep. 6, 19747. Mitrouli, S.T., Kostoglou, M., Karabelas, A.J., 2016. Calcium carbonate scaling of desalina-
- Mitrouli, S.T., Kostoglou, M., Karabelas, A.J., 2016. Calcium carbonate scaling of desalination membranes: assessment of scaling parameters from dead-end filtration experiments. J. Membr. Sci. 510, 293–305.
- Monnot, M., Laborie, S., Cabassud, C., 2016. Granular activated carbon filtration plus ultrafiltration as a pretreatment to seawater desalination lines: impact on water quality and UF fouling. Desalination 383, 1–11.
- Mustafa, G., Wyns, K., Buekenhoudt, A., Meynen, V., 2016. New insights into the fouling mechanism of dissolved organic matter applying nanofiltration membranes with a variety of surface chemistries. Water Res. 93, 195–204.
- Naji, O., Al-juboori, R.A., Bowtell, L., Alpatova, A., Ghaffour, N., 2020. Direct contact ultrasound for fouling control and flux enhancement in air-gap membrane distillation. Ultrason. Sonochem. 61, 104816.
- Nguyen, T., Roddick, F.A., Fan, L., 2012. Biofouling of water treatment membranes: a review of the underlying causes, monitoring techniques and control measures. Membranes 2, 804–840.
- Ochando-Pulido, J.M., Víctor-Ortega, M.D., Martínez-Ferez, A., 2016. Membrane fouling insight uring reverse osmosis purification of pretreated olive mill wastewater. Sep. Purif. Technol. 168, 177–187.
- Oloukoi, G.A., Urmilla, B., Vadi, M., 2013. Households' coping strategies for climate variability related water shortages in Oke-Ogun region. Nigeria. Environ. Dev. 5, 23–38.
- Park, H.-G., Cho, S.-G., Kim, K.-J., Kwon, Y.-N., 2016a. Effect of feed spacer thickness on the fouling behavior in reverse osmosis process — a pilot scale study. Desalination 379, 155–163.
- Park, M., Park, J., Lee, E., Khim, J., Cho, J., 2016b. Application of nanofiltration pretreatment to remove divalent ions for economical seawater reverse osmosis desalination. Desalin. Water Treatment 57.
- Piyadasa, C., Yeager, T., Gray, S.R., Stuart, M.B., Ridgway, H.F., Pelekani, C., Orbell, J.D., 2017. The influence of electromagnetic fields from two commercially available watertreatment devices on calcium carbonate precipitation, environ. Sci.: water res. Technol. 3, 566–572.
- Piyadasa, C., Yeager, T.R., Gray, S.R., Stewart, M.B., Ridgway, H.F., Pelekani, C., Orbell, J.D., 2018. Antimicrobial effects of pulsed electromagnetic fields from commercially available water treatment devices – controlled studies under static and flow conditions. J. Chem. Technol. Biotechnol. 93, 871–877.
- Puspitasari, V.L., Rattier, M., Le-Clech, P., Chen, V., 2010. Performances of protease and amylase cleaning for microporous membranes used in wastewater applications. Desalin. Water Treat. 13, 441–449.
- Qasim, M., Darwish, N.N., Mhiyo, S., Darwish, N.A., Hilal, N., 2018. The use of ultrasound to mitigate membrane fouling in desalination and water treatment. Desalination 443, 143–164.
- Qin, J.-J., Oo, M.H., Kekre, K.A., Liberman, B., 2010. Development of novel backwash cleaning technique for reverse osmosis in reclamation of secondary effluent. J. Membr. Sci. 346, 8–14.
- Radu, A.I., van Steen, M.S.H., Vrouwenvelder, J.S., van Loosdrecht, M.C.M., Picioreanu, C., 2014. Spacer geometry and particle deposition in spiral wound membrane feed channels. Water Res. 64, 160–176.
- Raffin, M., Germain, E., Judd, S.J., 2012. Influence of backwashing, flux and temperature on microfiltration for wastewater reuse. Sep. Purif. Technol. 96, 147–153.
- Rahman, F., 2013. Calcium sulfate precipitation studies with scale inhibitors for reverse osmosis desalination. Desalination 319, 79–84.
- Rahman, M.S., Aubin, H.T., Sasson, M.B., Ober, C.K., Nielsen, M., Elimelech, M., 2014. Control of biofouling on reverse osmosis polyamide membranes modified with biocidal nanoparticles and antifouling polymer brushes. J. Mater. Chem. B 2, 1724–1732.
- Ramon, G.Z., Nguyen, T.-V., Hoek, E.M.V., 2013. Osmosis-assisted cleaning of organicfouled seawater RO membranes. Chem. Eng. J. 218, 173–182.
- Rana, D., Matsuura, T., 2010. Surface modifications for antifouling membranes. Chem. Rev. 110, 2448–2471.
- Rathinam, K., Oren, Y., Petry, W., Schwahn, D., Kasher, R., 2018. Calcium phosphate scaling during wastewater desalination on oligoamide surfaces mimicking reverse osmosis and nanofiltration membranes. Water Res. 128, 217–225.
- Ribera, G., Clarens, F., Martínez-Lladó, X., Jubany, I., Martí, V., Rovira, M., 2014. Life cycle and human health risk assessments as tools for decision making in the design and implementation of nanofiltration in drinking water treatment plants. Sci. Total Environ. 466–467, 377–386.
- Ronen, A., Lerman, S., Ramon, G.Z., Dosoretz, C.G., 2016. Biofouling suppression of modified feed spacers: localized and long-distance antibacterial activity. Desalination 393, 159–165.
- Rudolph, G., Schagerlöf, H., Krogh, K.B.M., Jönsson, A.-S., Lipnizki, F., 2018. Investigations of alkaline and enzymatic membrane cleaning of ultrafiltration membranes fouled by thermomechanical pulping process water. Membranes 8, 91.
- Ruiz, S.G., Ramírez, J.A.L., Zerrouk, M.H., Lopera, A.E.-C., Alonso, J.M.Q., 2020. Study of reverse osmosis membranes fouling by inorganic salts and colloidal particles during seawater desalination. Chin. J. Chem. Eng. 28, 733–742.
- Saffarimiandoab, F., Gul, B.Y., Ilter, S.E., Guclu, S., Unal, S., Tunaboylu, B., Menceloglu, Y.Z., Koyuncu, I., 2019. Evaluation of biofouling behavior of zwitterionic silane coated reverse osmosis membranes fouled by marine bacteria. Prog. Org. Coat. 134, 303–311.
- Sagle, A.C., Van Wagner, E.M., Ju, H., McCloskey, B.D., Freeman, B.D., Sharma, M.M., 2009. PEG-coated reverse osmosis membranes: desalination properties and fouling resistance. J. Membr. Sci. 340, 92–108.
- Sanchez, O., 2018. Microbial diversity in biofilms from reverse osmosis membranes: a short review. J. Membr. Sci. 545, 240–249.

- Shafi, H.Z., Khan, Z., Yang, R., Gleason, K., 2015. Surface modification of reverse osmosis membranes with zwitterionic coating for improved resistance to fouling. Desalination 362, 93–103.
- Shafi, H.Z., Matin, A., Akhtar, S., Gleason, K.K., Zubair, S.M., Khan, Z., 2017. Organic fouling in surface modified reverse osmosis membranes: filtration studies and subsequent morphological and compositional characterization. J. Membr. Sci. 527, 152–163.
- Shahkaramipour, N., Tran, T.N., Ramanan, S., Lin, H., 2017. Membranes with surfaceenhanced antifouling properties for water purification. Membranes 7, 13.
- Shang, R., Verliefde, A.R.D., Hu, J., Zeng, Z., Lu, J., Kemperman, A.J.B., Deng, H., Nijmeijer, K., Heijman, S.G.J., Rietveld, L.C., 2014. Tight ceramic UF membrane as RO pre-treatment: the role of electrostatic interactions on phosphate rejection. Water Res. 48, 498–507.
- She, Q., Wang, R., Fane, A.G., Tang, C.Y., 2016. Membrane fouling in osmotically driven membrane processes: a review. J. Membr. Sci. 499, 201–233.
- Shirazi, S., Lin, C.-J., Chen, D., 2010. Inorganic fouling of pressure-driven membrane processes—a critical review. Desalination 250, 236–248.
- Shmulevsky, M., Li, X., Shemer, H., Hasson, D., Semiat, R., 2017. Analysis of the onset of calcium sulfate scaling on RO membranes. J. Membr. Sci. 524, 299–304.
- Siddiqui, A., Farhat, N., Bucs, S.S., Linares, R.V., Picioreanu, C., Kruithof, J.C., van Loosdrecht, M.C.M., Kidwell, J., Vrouwenvelder, J.S., 2016. Development and characterization of 3D-printed feed spacers for spiral wound membrane systems. Water Res. 91, 55–67.
- Siddiqui, M.F., Sakinah, M., Singh, L., Zularisam, A.W., 2012. Targeting N-acyl-homoserinelactones to mitigate membrane biofouling based on quorum sensing using a biofouling reducer. J. Biotechnol. 161, 190–197.
- Silva, R.C., Takizawa, Y., Nakaruk, A., Katouda, M., Yamanaka, A., Medina, J.O., Gomez, A.M., Tejima, S., Obata, M., Takeuchi, K., Noguchi, T., Hayashi, T., Terrones, M., Endo, M., 2019. New insights in the natural organic matter fouling mechanism of polyamide and nanocomposite multiwalled carbon nanotubes-polyamide membranes. Environ. Sci. Technol. 53, 6255–6263.
- Sioutopoulos, D.C., Karabelas, A.J., 2012. Correlation of organic fouling resistances in RO and UF membrane filtration under constant flux and constant pressure. J. Membr. Sci. 407–408, 34–46.
- Sohrabi, M.R., Madaeni, S.S., Khosravi, M., Ghaedi, A.M., 2011, Chemical cleaning of reverse osmosis and nanofiltration membranes fouled by licorice aqueous solutions, Desalination 267:93–100.
- Tabatabai, S.A.A., Schippers, J.C., Kennedy, M.D., 2014. Effect of coagulation on fouling potential and removal of algal organic matter in ultrafiltration pretreatment to seawater reverse osmosis. Water Res. 59, 283–294.
- Taherinejad, M., Derakhshan, S., Yavarinasab, A., 2017. Hydrodynamic analysis of spiral wound reverse osmosis membrane recovery fraction and permeate water flow rate. Desalination 411, 59–68.
- Tang, F., Hu, H.Y., Sun, L.J., Sun, Y.X., Shi, N., Crittenden, J.C., 2016, Fouling characteristics of reverse osmosis membranes at different positions of a full-scale plant for municipal wastewater reclamation, Water Res., 90 329-336.
- Timm, S.N., Deal, B.M., 2018. Understanding the behavioral influences behind Singapore's water management strategies. J. Environ. Plan. Manag. 61, 1654–1673.
- Tong, T., Zhao, S., Boo, C., Hashmi, S.M., Elimelech, M., 2017. Relating silica scaling in reverse osmosis to membrane surface properties. Environ. Sci. Technol. 51, 4396–4406.
- Uppu, A., Chaudhuri, A., Das, S.P., 2019. Numerical modeling of particulate fouling and cake-enhanced concentration polarization in roto-dynamic reverse osmosis filtration systems. Desalination 468, 114053.
- Valavala, R., Sohn, J., Han, J., Her, N., Yoon, Y., 2011. Pretreatment in reverse osmosis seawater desalination: a short review. Environ. Eng. Res. 16, 205–212.
- Varin, K.J., Lin, N.H., Cohen, Y., 2013. Biofouling and cleaning effectiveness of surface nanostructured reverse osmosis membranes. J. Membr. Sci. 446, 472–481.
- Villacorte, L.O., Kennedy, M.D., Amy, G.L., Schippers, J.C., 2009. Measuring transparent exopolymer particles (TEP) as indicator of the (bio)fouling potential of RO feed water. Desalin. Water Treat. 5, 207–212.
- Vitzilaiou, E., Stoica, I.M., Knøchel, S., 2019. Microbial biofilm communities on reverse osmosis membranes in whey water processing before and after cleaning. J. Membr. Sci. 587, 117174.
- Voulvoulis, N., 2018. Water reuse from a circular economy perspective and potential risks from an unregulated approach. Current Opinion in Environmental Science & Health 2, 32–45.
- Voutchkov, N., 31 May 2017, Pretreatment for Reverse Osmosis Desalination 1st Edition, Elsevier.
- Vrouwenvelder, J., Kruithof, J., Van Loosdrecht, M., 2010. Integrated approach for biofouling control. Water Sci. Technol. 62, 2477.
- Vrouwenvelder, J.S., von der Schulenburg, D.A.G., Kruithof, J.C., Johns, M.L., van Loosdrecht, M.C.M., 2009. Biofouling of spiral-wound nanofiltration and reverse osmosis membranes: a feed spacer problem. Water Res. 43, 583–594.

- Vrouwenvelder, J.S., Van Loosdrecht, M.C.M., Kruithof, J.C., 2011. A novel scenario for biofouling control of spiral wound membrane systems. Water Res. 45, 3890–3898.
- Wang, S., Mu, C., Xiao, K., Zhu, X., Huang, X., 2020. Surface charge regulation of reverse osmosis membrane for anti-silica and organic fouling. Sci. Total Environ, 715, 137013.
- Wang, Y., Wang, Z., Wang, J., Wang, S., 2018. Triple antifouling strategies for reverse osmosis membrane biofouling control. J. Membr. Sci. 549, 495–506.
- Wend, C.F., Stewart, P.S., Jones, W., Camper, A.K., 2003. Pretreatment for membrane water treatment systems: a laboratory study. Water Res. 37, 3367–3378.
- Whittaker, C., Ridgway, H., Olson, B.H., 1984. Evaluation of cleaning strategies for removal of biofilms from reverse-osmosis membranes. Appl. Environ. Microbiol. 48, 395–403.
- Wibisono, Y., Yandi, W., Golabi, M., Nugraha, R., Cornelissen, E.R., Kemperman, A.J.B., Ederth, T., Nijmeijer, K., 2015. Hydrogel-coated feed spacers in two-phase flow cleaning in spiral wound membrane elements: a novel platform for eco-friendly biofouling mitigation. Water Res. 71, 171–186.
- World Health Organization Water, 2014. Fact. Sheet 391.
- Wu, Z.C., Cai, Z.Q., Liu, X., Long, C.Y., Liu, F., Wang, S.L., 2006, Production method of low fouling composite reverse osmosis membranes, China Patent 200610051205.8.
- Xia, L., Vemuri, B., Saptoka, S., Shrestha, N., Chilkoor, G., Kilduff, J., Gadhamshetty, V., 2019. Chapter 1.8 - antifouling membranes for bioelectrochemistry applications, microbial electrochemical technology, sustainable platform for fuels, chemicals and remediation biomass. Biofuels and Biochemicals 195-224.
- Xiao, R., Zheng, Y., 2016. Overview of microalgal extracellular polymeric substances (EPS) and their applications. Biotechnol. Adv. 34, 1225–1244.
- Yanagi, C., More, K., 1980. ADVANCED REVERSE OSMOSIS PROCESS WITH DTOhfATIC 3PONGE BALL CLEANING FOR THE RBCLAMATION OF MUNICIPA.K. SEWAGE, Desalination 32, 391–398.
- Yang, H.-L, Lin, J. C.-T., Huang, C., 2009, Application of nanosilver surface modification to RO membrane and spacer for mitigating biofouling in seawater desalination, Water Res.43:3777–3786.
- Yang, J., Lee, S., Yu, Y., Kuk, J., Hong, S., Lee, S., Min, K., 2010. Role of foulant-membrane interactions in organic fouling of RO membranes with respect to membrane properties. Sep. Sci. Technol. 45, 948–955.
- Yang, J.Y., Li, Y.S., Huang, B., 2013. Research on refurbishing of the used RO membrane through chemical cleaning and repairing with a new system. Desalination 320, 49–55.
- Yang, R., Xu, J.J., Ince, G.O., Wong, S.Y., Gleason, K.K., 2011. Surface-tethered zwitterionic ultrathin antifouling coatings on reverse osmosis membranes by initiated chemical vapor deposition. Chem. Mater. 23, 1263–1272.
- Yang, Z., Sun, Y.X., Ye, T., Shi, N., Tang, F., Hu, H.Y., 2017. Characterization of trihalomethane, haloacetic acid, and haloacetonitrile precursors in a seawater reverse osmosis system. Sci. Total Environ. 576, 391–397.
- Yao, M., Yan, D., Kabat, P., Huang, H., Hutjes, R.W.A., Werners, S.E., 2016. Analysing monthly sectorial water use and its influence on salt intrusion induced water shortage in urbanized deltas. Sustain. Cities Soc. 26, 255–263.
- Yao, Z., Peng, L.E., Guo, H., Qing, W., Mei, Y., Tang, C.Y., 2019. Seawater pretreatment with an NF-like forward osmotic membrane: membrane preparation, characterization and performance comparison with RO-like membranes. Desalination 470, 114115.
- Yaun, Y., Kilduff, J.E., 2010. Effect of colloids on salt transport in cross-flow nanofiltration. J. Membr. Sci. 346, 240–249.
- Yiantsios, S.G., Sioutopoulos, D., Karabelas, A.J., 2005. Colloidal fouling of RO membranes: an overview of key issues and efforts to develop improved prediction techniques. Desalination 183, 257–272.
- Yin, Z., Yang, C., Long, C., Li, A., 2017. Influence of surface properties of RO membrane on membrane fouling for treating textile secondary effluent. Environ. Sci. Pollut. Res. 24, 16253–16262.
- Yuan, Y., Hays, M.P., Hardwidge, P.R., Kim, J., 2017. Surface characteristics influencing bacterial adhesion to polymeric substrates. RSC Adv. 7, 14254–14261.

Zeev, E.B., Elimelech, M., 2014. Reverse osmosis biofilm dispersal by osmotic backflushing: cleaning via substratum perforation. Environ. Sci. Technol. Lett. 1, 162–166.

- Zeev, E.B., Belkin, N., Liberman, B., Frank, I.B., Berman, T., 2013. Bioflocculation: chemical free, pre-treatment technology for the desalination industry. Water Res. 47, 3093–3102.
- Zhang, J., Northcott, K., Duke, M., Scales, P., Gray, S.R., 2015. Influence of pre-treatment combinations on RO membrane fouling. Desalination 393, 120–126.
- Zhang, R., Huang, Y., Sun, C., Xiaozhen, L., Bentian, X., Wang, Z., 2019. Study on ultrasonic techniques for enhancing the separation process of membrane. Ultrason. Sonochem. 55, 341–347.
- Zhou, D., Zhu, L., Fu, Y., Zhu, M., Xue, L., 2015. Development of lower cost seawater desalination processes using nano filtration technologies—a review. Desalination 376, 109–116.