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REVERSE OSMOSIS MEMBRANE BIOFOULING:
Causes, Consequences and Countermeasures

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REVERSE OSMOSIS MEMBRANE BIOFOULING: CAUSES, CONSEQUENCES AND COUNTERMEASURES

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ABSTRACT

Over the past two decades, membrane filtration in the form of microfiltration (MF) and ultrafiltration (UF) has proliferated globally to become the gold standard for removing suspended and colloidal solids, pathogens and emulsions from virtually every type of water source – traditional and non-traditional, fresh and saline. Hence, everywhere and anywhere filtration or clarification is required membranes are now rapidly replacing conventional flotation, sedimentation, and filtration processes.

Generally, MF/UF membranes suffer from two key limitations – fouling and integrity. Membrane integrity issues are somewhat specific to hollow fiber filters in municipal applications, where fiber breakage can be a frequent occurrence and significant operating costs are incurred due to the labor and downtime associated with integrity testing and fiber pinning. This is exacerbated for high fouling feed waters where frequent backwashing and cleaning are required to combat flux decline, where the mechanical limits of hollow fibers are overextended during flux maintenance actions (backwashing and cleaning). Over time, MF/UF plant capacities can drop off significantly due to fouling and fiber pinning.

This paper reviews state-of-the-art and emerging MF/UF technologies including ceramic membranes, novel polymeric membranes and advanced control strategies all of which promise next-generation improvements for MF/UF membranes.

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1. INTRODUCTION

A number of general reviews have been written on bacterial adhesion, biofilm formation and biofouling[1-3]. Several reviews specifically focus on biofouling causes, consequences and countermeasures in reverse osmosis (RO) membrane systems[4-9]. Most historical research – both laboratory and field scale – has focused on seawater RO desalination membrane plants with some emphasis on wastewater RO reuse plants. Not much has been reported in the open literature on brackish groundwater RO systems, but biofouling can be a significant problem there as well.

Biofouling negatively affects both RO system operating cost and performance. Biofouling degrades product water quality through "biofilm-enhanced concentration polarization" (BECP), which increases salt passage and elevates trans-membrane osmotic pressure. Moreover, biofouling increases the overall cost of water treatment by demanding more pretreatment, increasing RO system operating pressure (i.e., energy demand), increasing membrane cleaning frequency and reducing membrane useful life[10-12].The keys to fending off biofouling in RO systems or recovering from biofouling once it takes root include:

1. Understanding site-specific processes governing biofilm formation,
2. Implementing effective biofouling pretreatment ahead of RO membranes, and
3. Monitoring biofouling to enable more proactive and effective membrane cleaning.

Herein, we briefly review the mechanisms of microbial adhesion, biogrowth and biofilm formation in RO systems, common biofouling control methods for reducing the formation of biofouling in RO systems and the most common strategies for biofilm monitoring.

2. MECHANISMS OF RO MEMBRANE BIO-FOULING

Ridgway, Flemming and co-workers conducted pioneering research on RO membrane biofouling 20 to 30 years ago[5, 6, 8, 13].Their work elucidated fundamental physical, chemical and biological factors governing biofouling in real RO membrane plants. Later, Hoek and co-workers studied bacterial deposition, adhesion and removal from polymeric filtration and desalination membranes[14-19], as well as physical-chemical characteristics and bacterial phylogeny of biofilms formed in several different RO membrane plants[16, 20].

Vrouwenvelder and co-workers have focused on bacterial biofilm formation, biogrowth, inhibition and monitoring in a combination of theoretical, laboratory and field studies[21-30]. Fane and co-workers have also studied RO membrane biofouling causes, consequences, inhibition and monitoring through a number of unique methods[31-36].

In general (Figure 1), bacteria present in RO feed waters undergo transport to the membrane surface where initial (reversible) adhesion occurs (1). Irreversible adhesion is associated with biosynthesis of extracellular polymeric substances (EPS) (2) and a biofilm results from subsequent growth and multiplication at the expense of feedwater nutrients (3). Complex biofilm formation ensues which involves communication (quorum sensing) and gradient formation of biological functions among cells located in different zones of the biofilm (4). Eventually a fully formed biofilm sloughs off viable bacteria cells and nonviable cell debris to downstream locations (5).

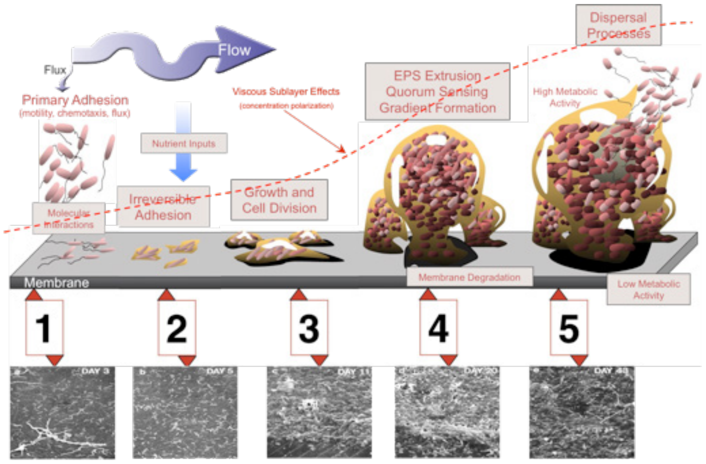

Figure 1a. Illustration and SEM images of biofilm formation mechanisms and evolution over time[37, 38]

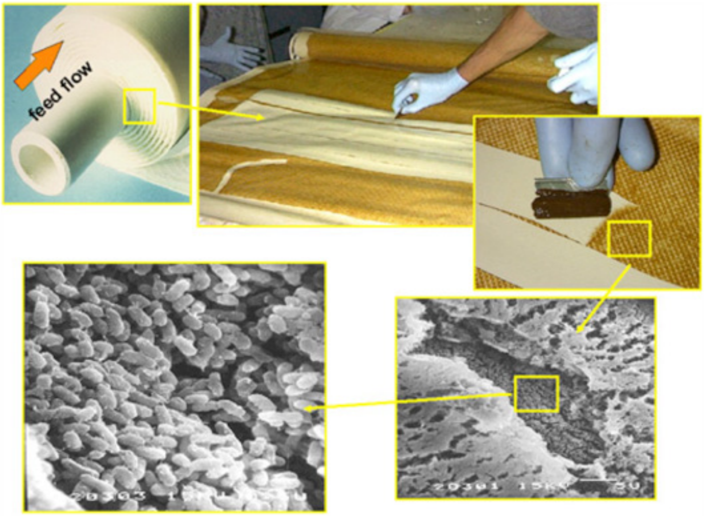

Figure 1b. pictures and SEM images of biofilms formed on RO membranes at the Orange County Water District[39].

Whereas the basic processes of bacterial adhesion and biofilm formation are similar in membrane systems and other natural and industrial systems, membranes are perhaps uniquely impacted by pressure-driven water and solute transport phenomena that influence biofouling kinetics.40 Somewhere between the biofilm formation Stages 4 and 5 shown above, symptoms of membrane fouling emerge which could include:

- a reduction in membrane water flux due to establishment of a gel-like diffusion barrier (i.e., the biofilm)
- biofilm enhanced concentration polarization accompanied by lowered solute rejection
- an increase in the net differential pressure (NDP) across the RO modules
- biodegradation and/or biodeterioration of the membrane polymer or other module construction materials (e.g., polyurethane-based glue lines)
- establishment of concentrated populations of primary or secondary human pathogens on membrane surfaces.

In general, the assimilable organic carbon (AOC) content of the influent natural organic matter (NOM) drives biogrowth throughout a water treatment plant[24]. Bacterial EPS is primarily responsible for hydraulic and osmotic losses from biofilms on RO membranes[10]. Starved cells do not produce much EPS and so RO membranes may be significantly colonized, but biofouling may not be noticeable until adequate AOC becomes available. During algae blooms the influent AOC content at seawater RO plants can be elevated even if algal biomass is removed by pre-filtration; at wastewater RO plants occasional upsets at the upstream wastewater treatment plant can send more AOC into the downstream RO and induce biofouling events. Alternatively, brackish groundwater plants usually have less than 1 ppm of influent total organic carbon (TOC) of which only a fraction comprises AOC, and rarely would TOC fluctuate dramatically such that introduction of AOC would be sufficient to drive a biofouling event. So why does it happen?

At RO plants with low influent AOC, pretreatment chemicals can introduce enough AOC into RO membrane systems - over and above the influent NOM-derived AOC - to stimulate significant biogrowth and biofouling[25, 41]. For example, Vrouwenvelder observed metabolically active biofilm bacteria, despite low AOC levels in an RO plant feed water. However, an increase in AOC was observed following the addition of acid (to prevent CaCO3 scaling) which coincided with a high biofilm formation rate observed through a significant increase in net differential pressure (NDP). More recently, a well-controlled laboratory study suggests a significant increase in biofilm formation on RO membranes in the presence of both polyacrylate-based (PAA) and polyphosphonate-based (PPP) antiscalants[42].The PAA increased biofilm formation by membrane surface modification which enhanced bacterial adhesion, while the PPP most likely accelerated biofilm formation by serving as an additional source of phosphorous which is often a limiting nutrient in water treatment systems[29, 43].Therefore, selection of acid and antiscalant products should take into account their potential contribution to site specific membrane biofouling.

According to Saeed, another important consideration for plants using chlorine for disinfection is the location of sodium metabisulfite (SBS) injection[44].Typically, SBS is used to remove chlorine ahead of RO membranes to prevent their degradation. Saeed observed reduced RO membrane biofouling potential when the SBS dosing point was placed between dual media filters and cartridge filters, but higher biofouling potential was observed as the SBS dosing point was moved forward along the pretreatment line, closer to the RO membranes. It is known that antiscalants can be oxidized by traditional water disinfectants and oxidants. Therefore, even when using antiscalant and acid products with quantifiably low AOC, one must consider the possibility that locating the antiscalant and/or acid injection point prior to SBS injection could lead to oxidation and enhanced biodegradability of either (1) antiscalant polymers or (2) the non-oxidative biocides used in acid and antiscalant formulations. This could introduce AOC formation just as the water enters the RO membrane system and over-time lead to a biofouling problem. Accordingly, the location of SBS dechlorination relative to acid and/or antiscalant injection points must be considered.

3. METHODS FOR MITIGATING RO BIOFOULING

Flemming states, "Countermeasures (of RO membrane biofouling) require a three step protocol: (1) detection, (2) sanitation, and (3) prevention[5, 6]." Some level of pre-filtration can reduce the load of colloidal and particulate matter that directly foul RO membranes through cake formation and enhanced concentration polarization[45],but also to reduce the influent load of viable biofilm forming microorganisms[46].

An effective chemical disinfection regime that inhibits biogrowth throughout the RO system greatly improves performance, reliability and economics by reducing the feed pressure, cleaning frequency, cleaning chemical costs, plant downtime and operator intervention[47]. In addition to pretreatment, each RO membrane plant must develop an optimized approach to RO membrane cleaning (both frequency and chemical regime) as well as RO element rotation and replacement[48]. However obvious these statements may appear, there is no universally successful combination of pretreatment and maintenance. Why? The principal reason is that modern polyamide composite RO membranes cannot be continuously exposed to free chlorine without rapid degradation and loss of rejection. Without the ability to continuously chlorinate a water system, biofouling control becomes complex and subject to a milieu of site-specific nuances.

Biocides such as chlorine, ozone, chlorine dioxide and UV have been used to inhibit biogrowth on RO membranes. Free chlorine is most frequently used, but is known to degrade RO membranes, produce disinfection by-products, and sometimes leads to enhanced biogrowth on RO membranes because it must be quenched using sodium bisulfite[49]. Ozonation of bromide-rich saline waters can produce bromate, which is classified as a carcinogen by USEPA and WHO, and which, may not be completely rejected by RO membranes; hence, may have limited application for drinking water production. UV treatment is gaining in popularity since it does not produce disinfection byproducts, it quenches chlorine without using sodium bisulfite, and may break down some organic matter in the RO feed water. However, it may not be effective where assimilable organic carbon (AOC) exists in RO feed water because it only provides up front disinfection, but no residual biogrowth inhibition throughout the RO membrane system.

The use of chloramines (combined chlorine) can be an effective means to control RO membrane biofouling in wastewater reclamation plants[61]; although, some reports suggest chloramines are not effective for biofouling control[62]. Variances may come from different qualities of tertiary wastewater being fed into the RO membranes. Moreover, not all RO membrane manufacturers offer warrantee coverage on their membranes when continuous chloramine dosing is part of the RO plant operational strategy. Combining membrane filtration and chloramination as pretreatment can be more effective at fouling control because it reduces influent colloidal matter while also inhibiting biogrowth.

At a full scale seawater RO demonstration plant, Hoek and co-workers evaluated different methods of disinfection following chlorination, microfiltration and dechlorination including: granular activated carbon (GAC), ultraviolet (UV) irradiation, and chlorine dioxide (ClO_2) to prevent biofouling on downstream RO membranes[63]. The UV pretreatment was not effective while GAC and ClO_2 performed well, although some membrane damage was observed for the ClO_2 fed membranes.

Table 1. Summary of biogrowth inhibitors, mechanisms, advantages and disadvantages

Inhibitor	Mechanism(s)	Advantage(s)	Disadvantage(s)
Chlorine	Oxidative inhibition	• Prevents biofilm at low doses • Familiar to operators • Well proven • Accepted practice	• Chlorination byproducts • Membrane degradation
Chloramines	Oxidative inhibition	• Prevents biofilm at moderate doses • Familiar to operators • Well proven • Accepted practice	• Chloramination byproducts • Membrane degradation
Chlorine dioxide	Oxidative inhibition	• Well proven biogrowth inhibitor • Prevents biofilm at very low doses (~1ppm) • Does not directly degrade RO membranes	• Chlorite/chlorate only known byproducts • Sometimes carries sufficient free chlorine residual that it degrades RO membranes
ICI	Oxidative shock	• Periodically retards bio-growth • Reduced byproducts • Reduced degradation	• Does not prevent biofilm formation • Lack of operator familiarity • Not well-proven for polyamide RO membranes
DBNPA	• Releases 2,2-dibromo-3-nitrilopropionamide • Degrades cell membrane protein leads to lysis	• Used widely in industrial systems • Can be combined with chlorine • Meets FDA requirements • EPA registered	• More expensive than chlorine • Lack of operator familiarity • Not well-proven in water treatment
NO donors	• DNA damage • Degradation of iron sulfur centers	• Non-oxidative, • So compatible with RO membranes • Inhibits biogrowth at lab scale	• No regulatory approval • More expensive than chlorine • Lack of operator familiarity • Not well-proven
DCC	• Releases HOCl and isocyanuric acid • Maintains low free available chlorine (FAC)	• Used widely in swimming pools, industrial cooling systems, & hospitals	• No regulatory approval • More expensive than chlorine • Lack of operator familiarity • Not well-proven
Phosphorus removal	Lack of essential nutrient	• Can be completely effective when phosphorous is the limiting nutrient	• May not work if phosphorous is not a limiting nutrient • Eliminates some highly effective anti-scalants

Intermittent chlorine injection (ICI) has been demonstrated to successfully reduce biofouling on cellulose triacetate (CTA) membranes in seawater containing heavy metals, but a systematic study was required to identify the optimal combination of dose, duration and frequency for the plant-specific feed water quality, pretreatment sequence and RO membrane array[64].

A non-oxidative biocide, 2,2-dibromo-3-nitrilopropioamide (DBNPA), can also be used to minimize and/or eliminate problems due to biofouling accumulation and to ensure long-term performance of a RO system[65]. DBNPA is compatible with RO membrane materials and is highly rejected by RO membranes, and has already been demonstrated successfully in field studies on full-scale RO systems[47]. The major drawback of this biocide is its cost, which is very high relative to conventional water treatment disinfectants but could be economic if used intermittently.

Most recently, nitric oxide (NO) donor compounds proved very effective at removing both biofilm bacteria cells and EPS with MAHMA NONOate [6-(2-Hydroxy-1-methyl-2-nitrosohydrazino)-N-methyl-1-hexanamine, NOC-9] being the optimal NO donor compound[31]. Nitric oxide is toxic to bacteria; the mechanism for this includes DNA damage and degradation of iron sulfur centers into iron ions and iron-nitrosyl compounds. However, many bacterial pathogens have evolved mechanisms for nitric oxide resistance suggesting the same could happen in a RO membrane plant[66]. In another recent study, biofilms grown on RO membrane were inactivated by dichloroisocyanurate (DCC) as much as chlorine; normalized flux and salt rejections of DCC-exposed membranes were stable, while rejection by chlorine-exposed membranes decreased to 80%[67].

4. RO MEMBRANE CLEANING AND BIOFILM REMOVAL

Membrane cleaning chemicals and protocols are prescribed by membrane manufacturers and chemical vendors, which typically are not involved until an RO plant is already experiencing major operational disruption. So, each scenario is addressed in an ad hoc manner. While there are generalizable approaches to cleaning, there are no known universally successful protocols for cleaning fouled RO membranes. An alkaline clean is often performed before or after an acid clean when targeting removal of both organic and inorganic foulants. Typical cleaning agents include acids (HCl, HNO_3, H_2SO_4), bases (NaOH, NH_3OH), complexing agents like EDTA, surfactants like SDS, and their combination[68]. In the case of organic and biological fouling, it is typical to apply alkaline cleaning solutions containing various combinations of surfactants, chelating agents, enzymes and chaotropic agents[69-71].

In one recent study at a wastewater RO plant, two stages of caustic and detergent cleaning (NaOH + SDS) followed by acid provided effective recovery of initial RO membrane flux and rejection. Sometimes chlorine and other biocides are also applied as part of a cleaning regimen or intermittently between cleaning intervals as in the ICI method described above[64, 72]. ***To achieve the highest cleaning efficiency, the cleaning solution(s) chemistry, sequence, flow velocity, temperature, duration, and frequency must be studied and optimized for a given RO installation – as they vary by water quality, type of fouling materials, type of RO membrane, RO pretreatment processes and RO system operating conditions. Moreover, cleaning regiments may need updating as feed water quality, pretreatment efficacy, and RO membranes change over the life of a plant.***

Biofouling Monitoring Strategies

As mentioned above, one key to mitigating biofouling in RO systems is choosing an adequate biofilm monitoring technique that (1) gives early warning indication of the onset of biofouling and (2) confirms the effectiveness of biofouling control measures employed. A recent trend in membrane research involves the use of ex situ, side-stream fouling detectors applied as early warning sensors at full-scale and pilot plants. For example, Vrouwenvelder and co-workers developed several ex situ fouling detectors they call a “membrane fouling simulator” (MFS) to monitor biological fouling in RO and NF filtration of surface and ground water[27, 78, 79]. The MFS provided (i) the same hydraulic behavior as spiral wound membrane modules, (ii) reproducible results, and (iii) effective early warning of biological fouling by monitoring tangential pressure drop through the system.

Subsequently, Vrouwenvelder and co-workers used the MFS to elucidate new fundamental insights into membrane biofouling mechanisms, particularly focusing on “fouling as a spacer problem” and methods of inhibiting biofilm formation and removing biofilms once formed[21-23, 29, 30, 80-86].

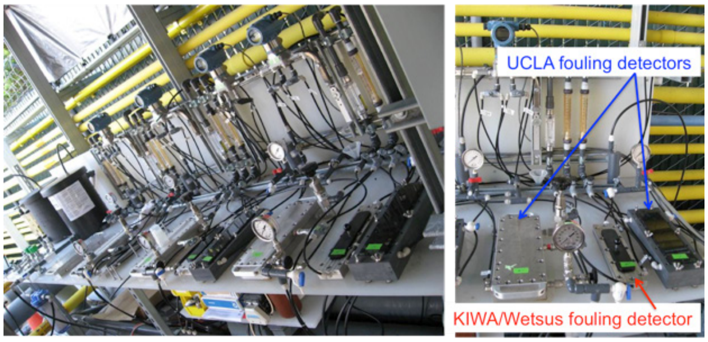

Figure 2. Research conducted at the Long Beach Water Department’s seawater RO/NF demonstration plant focusing on NF/RO membrane biofouling mechanisms, prevention and monitoring. Three different membrane fouling detectors were compared to understand potential bias due to different monitoring device configurations.

A major limitation of the MFS device is that it was designed for low pressure (<50 psi) operation such that there was no water permeation through NF/RO membranes during their studies. Over the past decade, Hoek and co-workers extended the range of working pressures for their direct microscopic observation system from microfiltration (<50 psi) up to seawater RO (<1200 psi)[15, 17, 19, 87-91]. In the course of this work, they developed new insights about attachment and removal of bacterial biofilms onto polymeric membranes, particularly focusing on detection of early stage biofilm formation and use of their fouling detectors to optimize operating conditions and cleaning methods. More recently, Hoek and Vrouwenvelder joined forces to evaluate the use of low-pressure (LP) and high-pressure (HP) membrane fouling detectors (MFDs) at the LBWD desalination demonstration plant[92]. Permeate flux, TDS rejection, and differential pressure drop data derived from two LP-MFDs did not correspond well with the full-scale plant, whereas the HP-MFD mimicked the full scale plant behavior well. Moreover, the HP-MFD was much more sensitive than the full scale plant. Hence, the HP-MFD gave early warning detection of fouling and membrane degradation and proved useful in optimizing membrane cleaning protocols.

Others researchers have employed similar fouling detector devices in lab and full-scale studies. For example, Cohen and co-workers extended a laboratory-scale optical membrane module into a novel, ex situ scaling observation detector (EXSOD)[93]. Subsequently, they used their EXSOD system to conduct a number of studies on the feasibility, scaling limits and optimized scale inhibition methods for RO desalination of brackish agricultural waters[94-97]. A more recent study by Duranceau and co-workers successfully employed sacrificial 4” by 40” spiral wound elements as “canary modules” to optimize chemical pretreatment at a brackish groundwater RO plant[98]. ***Membrane fouling detector technology is available for anyone to employ and these devices can be used not only for early warning of RO membrane biofouling, but also for other forms of fouling, mineral scaling, membrane damage and cleaning efficacy in any membrane process. Ex situ monitoring of RO membrane performance is now a reality and should be considered part of best practices at all full-scale installations.***

5. CONCLUSIONS & KEY TAKE-AWAYS

In conclusion, the lack of chlorine tolerance by commercially available NF/RO membranes makes it very difficult to prevent biogrowth and biofouling; hence, biofouling is the “tail that wags the dog” at membrane desalination plants. Biofouling drives the design, operation and cost of membrane desalination. Water Planet’s team of experts have helped a large number of municipal and industrial clients around the world through our MembranePRO® service offering. We have a number of related case studies available for download from our website at www.waterplanet.com/knowledgecenter.

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Dr. Eric M.V. Hoek, Ph.D. has over 20 years experience in water treatment membrane technology teaching, research, commercial product development and professional consulting as well as various entrepreneurial and philanthropic activities. Dr. Hoek is a founder of Water Planet and inventor of PolyCera membranes and IntelliFlux controls technologies. Previously, Dr. Hoek was a UCLA Engineering professor as well as the inventor of thin-film nanocomposite membrane technology and co-founder of NanoH$_2$O (now operating as LG Water Solutions).

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