REVERSE OSMOSIS

MEMBRANE FOULING - THE FINAL FRONTIER [1]

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Abstract

The phenomena of colloidal and bacteriological fouling of RO spiral wound brackish water membranes is reviewed. Generalizations from the literature about the mechanism of action of fouling are described. The monitoring methods used to measure colloidal and biofouling potential of feedwater is reviewed. The action of disinfectants and chemical cleaning agents on an established fouling layer is discussed. The need for extensive addition research is apparent.

Introduction

Premature failure of reverse osmosis (RO) membrane elements due to identified or unidentified membrane fouling substances costs thousands. to millions of dollars each year (l-4). Membrane fouling is becoming widely accepted as the single largest cause, if not the ONLY cause, of permeate flux decline at normal operating pressures and temperatures in brackish water systems. While RO fouling may not actually be the "final" frontier as the title asks, it is clearly a frontier, an incompletely known territory with many uncharted areas.

This article briefly reviews some of the oftentimes confusing, oftentimes conflicting research findings on fouling of spiral-wound, brackish-water RO membrane elements. This paper should not be regarded as strictly a literature review. Some of the generalizations and conclusions set forth are those of the authors, based upon direct experience and unpublished current information. These generalizations and conclusions may be modified or proven wrong with additional research.

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1 Published in Ultra Pure Water, vol 7 No 3, pp 25-36, 1990.

Membrane fouling

Fouling of RO membranes is defined operationally herein as the reduction in water transport per unit area of membrane (flux), caused by a substance or substances in the feedwater that accumulate either on or in the membrane. While there are several common causes of RO membrane fouling (5), this paper will focus primarily on biological and colloidal fouling, which are less well documented than other foulants such as inorganic scales.

Concentration polarization

Before discussing some of the fundamental aspects of colloidal and biofouling individually, it may be beneficial to first describe concentration polarization, a phenomenon that impacts all RO membrane fouling porocesses.

Figure la shows the three layers of material that are spirally wound about a perforated hollow tube to form a spiral- wound RO membrane element. The three layers are:

1. Feed channel spacer: Plastic cross-hatched screen material that separates the sheets of membrane, providing a space or channel for feed-water to enter the membrane element, contact the membrane sheets, then exit the membrane element as concentrate (becoming feedwater for the next membrane element in series, or becoming final waste).

2. RO Membrane: Sheets of semipermeable material (e.g., cellulose acetate or thin-film synthetic polymeric material) are permeable to water but relatively impermeable to dissolved and suspended solids.

3 Permeate water carrier: Fabric that separates sheets of membrane, providing a channel for permeate to flow to the central perforated tube of the membrane element.

Figure lb magnifies the feed channel, Figure lc magnifies the area just above the surface of the membrane, (which is where concentration polarization occurs) and illustrates the following discussion.

The action of the high-pressure pump forces feedwater with its dissolved and suspended solids are forced into the feed channel between two sheets of membrane. The bulk feedwater solution comes in contact with the membrane. Water diffuses through the membrane rapidly, but the dissolved and suspended particles in the water don't diffuse through the membrane readily, so they remain at the feedwater surface.

As additional feedwater enters the membrane element, additional water diffuses through the membrane, and additional dissolved and suspended solids accumulate at the membrane surface. These solids can accumulate to concentrations that exceed their concentrations in the bulk feedwater; and this phenomenon is referred to as concentration polarization.

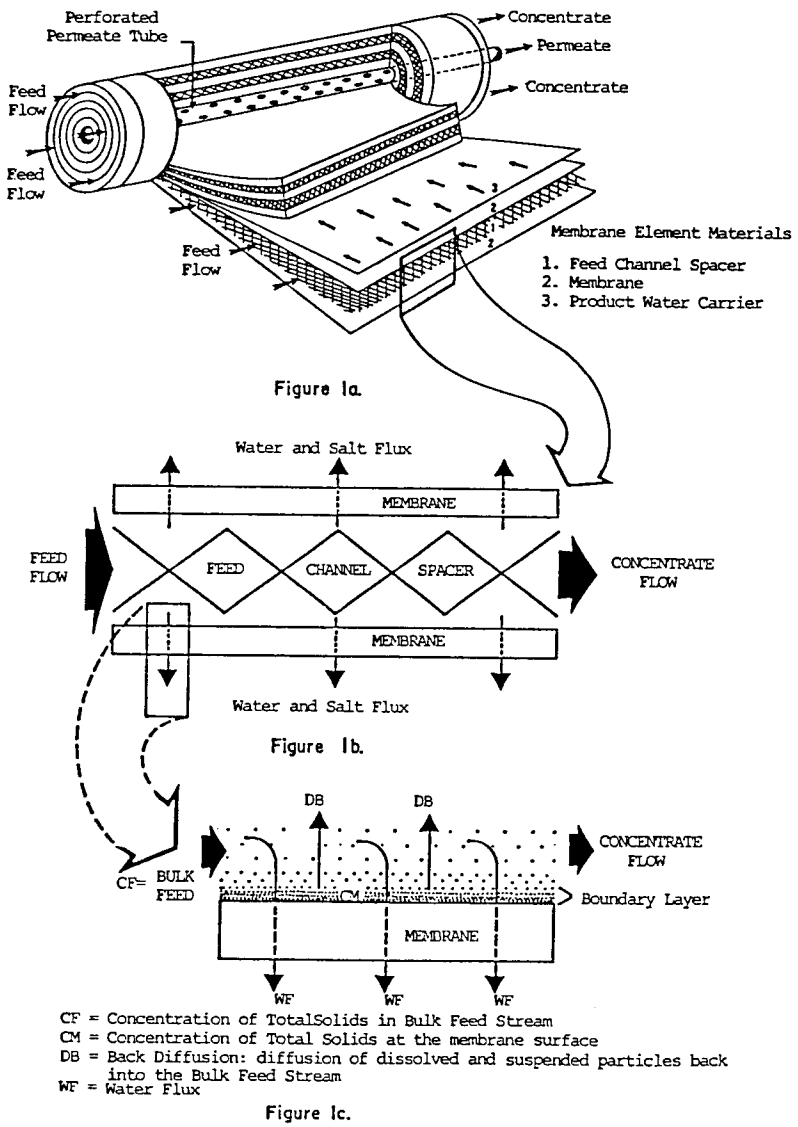


Figure 1. Concentration Polarization.

Concentration polarization occurs because there is a boundary layer of reduced turbulent mixing at the membrane surface (6). The thickness of the boundary layer depends upon the turbulence of the bulk feedwater flow. Increased velocity or turbulence (from the feed end of the membrane element to the concentrate end) will reduce the thickness of the boundary layer and will reduce concentration polarization.

Once dissolved or suspended solids become trapped in the boundary layer, their only means of escape is to diffuse back into the bulk feedwater stream. The larger the boundary layer, (due to lower bulk feed velocity), the slower the back diffusion of dissolved and suspended solids.

Concentration polarization plays a role in all RO membrane fouling phenomena. If concentrations of specific dissolved solids exceed their solubility in the boundary layer, they may precipitate and form a mineral scaling layer. Concentration polarization additionally provides concentration of nutrients needed by bacteria for their growth and metabolism. It also concentrates colloids, organics, and other fouling compounds.

Colloidal fouling

The mechanism of action for colloidal fouling of RO membranes is not entirely clear. Conflicting reports (3, 5-8) as to the ideal operating conditions required to reduce colloidal fouling suggest that several physicochemical factors may be involved. Two mechanisms, colloidal stability and concentration-polarization, appear to be fairly widely accepted.

Colloidal stability

Colloidal stability refers to the tendency of colloidal particles to settle out of solution. Stable colloids do not settle out readily; unstable colloids tend to agglomerate and settle out.

It is generally accepted that the stability of colloidal particles is due to their small size (typically less than two microns and many less than 1 micron) and the surface charges of the particles. The small size and density of the individual particles alone would permit them to stay in suspension indefinitely. Each particle generally has a net negative surface charge (7,9). The individual particles are prevented from coming into close contact with each other by the repulsive action of their negative surface charges (Figure 2a). This prevents agglomeration of smaller particles into larger, settleable particles that will settle out readily.

Stable colloidal particles are destabilized by partial or complete neutralization of their surface charge. This allows the particles to come into close enough contact to agglomerate (Figure 2b). Destabilization of particles by the addition of organic polymers and by inorganic salts such as alum and ferric chloride (coagulant aids) has been used for many years in clarifiers and filters to accomplish agglomeration of smaller particles into larger particles that can be settled or filtered out.

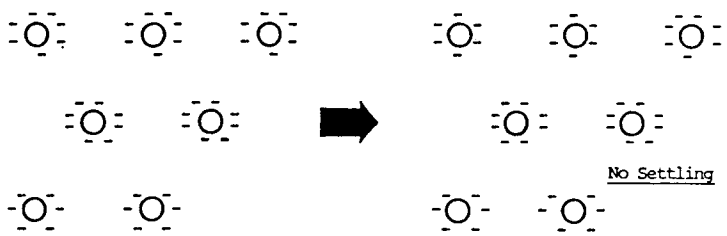


Figure 2a. Stable Colloids. Small particle size and negative surface charge does not permit particles to agglomerate and settle out.

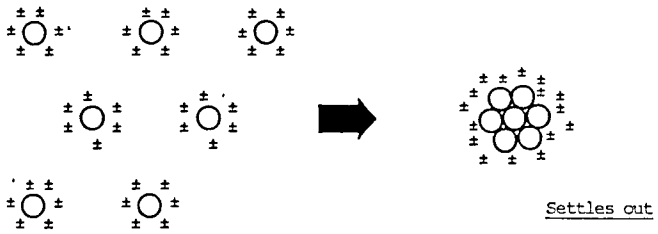


Figure 2b. Unstable colloids. Due to charge neutralization of the colloidal surface, particles tend to agglomerate and settle out.

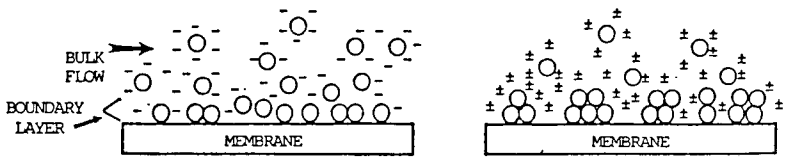


Figure 2c. Both stable and unstable colloids concentrate at the membrane surface. Stable colloids can more easily diffuse back into the bulk flow. Unstable colloids can agglomerate in the boundary layer which reduces diffusion back into the bulk flow.

Figure 2. Colloids Stability and Concentration.

In an RO system, three processes tend to destabilize colloids(7).

Colloid concentration: As the feedwater is passing through the membrane, colloids suspended in the feedwater are rejected by the membrane and are concentrated. For example: In a 75% recovery system, for every 100 gallons per minute (gpm) of feedwater, 75 gpm of permeate and 25 gpm of concentrate are produced. Since the colloids are rejected by the membrane, the number of particles in the original 100 gpm is now in 25 gpm. This is a 4x concentration.

Positive charge concentration: The addition of acid as an RO pretreatment adds positive hydronium ions that may neutralize the surface charge and therefore destablize colloidal particles. Additionally, the divalent cations (Ca^{++}, Mg^{++}, Fe^{++}, etc) are rejected well by the membrane. These cations are concentrated in the RO system as permeate is removed from the feedwater, and they provide a destabilizing environment for the colloids.

Concentration polarization: All particles will concentrate on the membrane surface. They are partially removed by diffusion of particles back into the bulk solution. Back diffusion is aided by turbulent flow. Larger agglomerated colloidal particles will diffuse less rapidly back into the bulk flow (Figure 2c). While these mechanisms may all be in operation during the fouling process, the physiochemical characteristics of the fouling material may play an important role in the extent of negative impacts caused by fouling and in our ability to remove the fouling material. Some colloids may actually bind to the membrane due to their chemical makeup and surface charge characteristics. Bound material probably creates more adverse effects and removal difficulties (10, 11).

Biofouling

What is known with certainty about bifouling? To be honest, not very much ! Some of the most enlightening information has been published by Ridgway et al. at Orange County Water District (10, 12-15). Those results, however, are specific to spiral-wound cellulose acetate membranes operated wrth a feedwater of secondary-treated sewage effluent. To quote Dr. Ridgway's (l0), "Fundamental research is still critically needed in nearly all aspects of RO membrane biofouling." Be that as it may, we will try to present a few biofouling mechanisms and observations that appear valuable and reasonable at this time. These generalizations come from Ridgway et al. as well as from other sources (9, 16-33). The following observations are typically made during a reload (subsequent set of new RO membrane elements). A new system starting up for the first. time goes through a debugging time period for the first few days and the initial events may not be noticed. However, many of the more typical events that may occur are summarized in Table A and Figure 3.

What's happening in or on the membrane to cause these changes shown in Table A? Is it compaction only.? Research findings appear not to favor this (7). Is it organics in the water that dissolve into the membrane and change its characteristic? Possibly (7)! Does colloidal fouling play a role? Probably. For this discussion we'll assume that 90% of the flux decline is due to biofouling. The research suggests that the following mechanism plays at least some role in the fouling process.

Table A

Typical events in the Life of an RO Membrane

Time	RO Performance	RO Membrane Surface
1-8 Hours	Highest Water Flux. Permeate TDS may be high during initial rinsing, then lowers, then rises slightly.	Clean membrane surface. Rapid sorption of dissolved organics, colloids and bacteria. Physical compaction of membrane.
1-14 days	Sharp Water flux decline (up to 10-15%). Permeate TDS may stay the same, go up or may even decrease.	Additional sorption of Colloids and bacteria. Microbial growth and multiplication. Biopolymer synthesis.
2 Weeks	Gradual Flux decline. When end of life water flux reduces by 10% to 15% or feed/concentrate differential pressure increases by 10- 15%, a chemical cleaning is required to bring water flux back to the expected flowrate based upon a standard gradual flux decline slope (Fig. 3).	Biofilm gradually develops in thickness. Concentration polarization enhanced. Membrane may deteriorate.

295

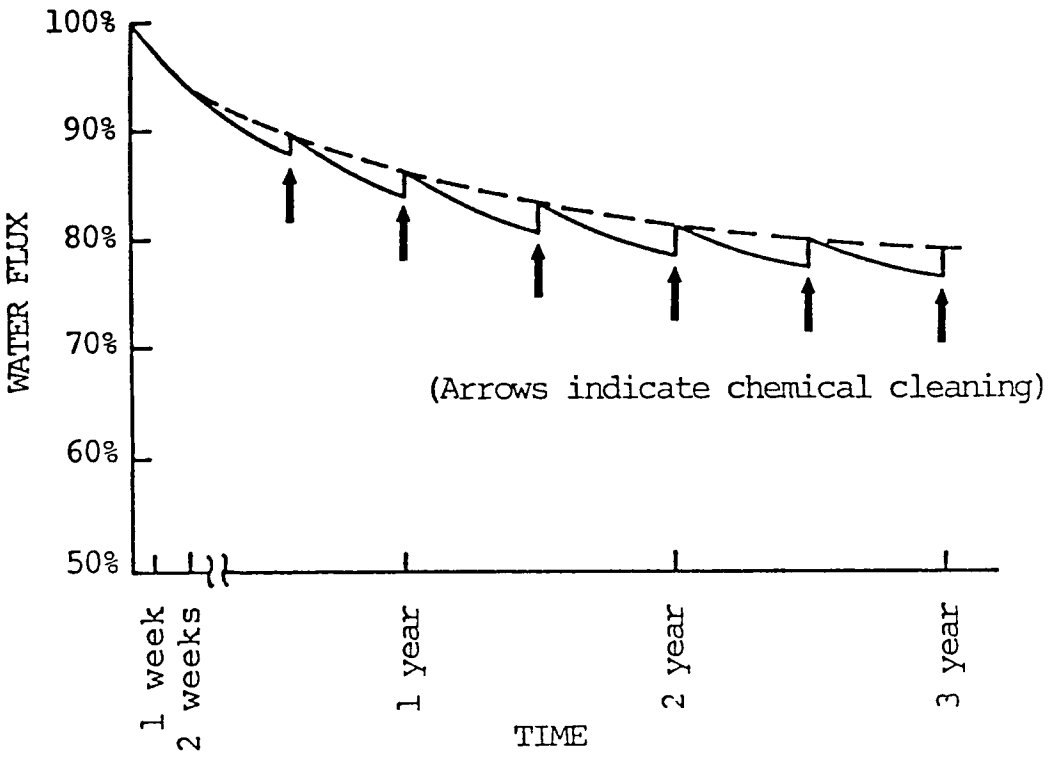


Figure 3. Water Flux as a Function of Membrane Life, Showing Effects of Cleaning.

296

The various stages of RO fouling (10, 18, 19) are shown in Figures 4a-d. During the first few hours of operation, concentration polarization increases the salt concentration at the surface of the membrane. This increases the osmotic pressure of the feedwater (wanting to pull water back from the permeate side) thereby reducing water flux. Concurrently, bacteria in the feedwater are attaching to the membrane. It appears that only certain bacteria can attach at this stage; and that there are a limited number of attachment sites, about 15% of the membrane area in one case (10, 13). During the next couple of weeks the bacteria either grow on the membrane, or else some other mechanism (like charge and/or hydrophobic changes on the membrane surface) allows the full membrane surface to be colonized by bacteria (12, 15, 19, 34). By day 16 (Figure 4c) the entire membrane is coated with bacteria several micrometers thick (15). Attached bacteria produce extracellular fibrils of polymeric substances such as hydrophobic mucopolysaccharides and glycoproteins (slime layer), which irreversibly attach the bacteria to the membrane. Additionally, it has been shown that these fibrils can concentrate organic and inorganic substances.

The combination of a continuous mircobial biofilm as well as the concentration of organics and inorganics may account for the rapid flux decline seen during the first couple of weeks. Another factor contributing to early flux decline may be the increase in concentration polarization due to the frictional resistance of the slime layer (10, 21, 25, 27) causing a greater boundary layer. It has been documented that a 42-inch water main with a biofilm of only 0.8 to 1.6 mm (l/32 to l/16 inch) had a flow reduction of 12% (27). Another incredible example of the fluid drag caused by biofilms was demonstrated when a biofilm with a thickness of 0.6 mm (25 mil) reduced flow in a 24-inch, 50 mile-long pipe by 55% (27).

As the biofilm matures, more bacterial growth and extracellular polymers are added to the biofilm. Biofilm growth eventually becomes limited by the shear force of the bulk feed stream (Figure 4d). It appears that the outermost layer of biofilm is the least compact (14). It may be that with time., the older biofilm layers compress, forming a more impermeable (hydrophobic) barrier; and/or perhaps salt concentration increases at the membrane surface due to the continuing hindrance to back diffusion produced by the growing and/or compressing biofilm. While the exact mechanism is not known, the reduction in water flux and increase in salt flux throughout the life of the membrane element is well known.

Monitoring

Monitoring RO systems for fouling potential is a standard operating procedure at most facilities. That's the good news! The bad news is that there aren't any good monitoring methods that accurately predict either colloidal or biological fouling. The following discussion considers the monitoring of colloidal fouling and of biofouling.

Colloidal fouling: The two commonly used analytical methods for monitoring suspended solids fouling potential in RO feedwater are measurement of turbidity and of the silt density index.

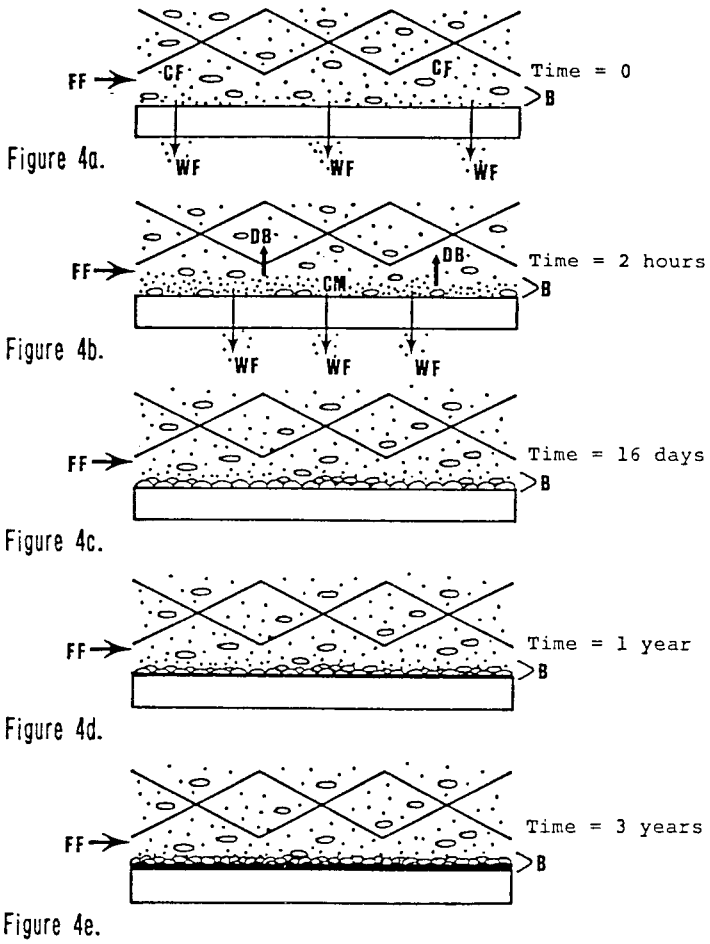
Turbidity measurement: Turbidity monitoring measures the presence of discrete particles in the feedwater by the ability of these particles to reflect light (Figure 5a). The assumption here is that there is a predictable correlation between the quantity of suspended particles in the feedwater and the amount of light that is reflected laterally from the surfaces of the particles. There are at least three limitations with using this method as an absolute indicator of fouling potential:

- Turbidity doesn't indicate the number of particles. Larger particles may reflect light in the same amount as hunderds or thousands of submicron particles depending upon sizes and shapes.
- Turbidity doesn't indicate the size of the particles. Particle size seems to be very important in the fouling process. Increased particle size at the membrane surface may reduce diffusion of the particles back into the bulk flow, thereby causing them to remain at the membrane surface. The size of particles on the membrane seems to be important. Relatively larger particles may not interfere as much with water transport as relatively smaller particles. This is presumably due to larger particles forming a looser compressible fouling layer; while smaller particles are able to form a tighter, incompressible, more impervious fouling layer (29, 35).
- In actuality, excellent feedwater turbidity values don't preclude fouling. Many facilities have experienced membrane failure due to fouling while maintaining less than the membrane manufacturers' usual turbidity limit of 1 NTU (1,7,26,28,36).

Silt density index measurement: Silt density index (SDI) is the standard method for determining the ability of feedwater suspended solids to plug a membrane and reduce water transport from feed side to permeate side. The procedure for SD1 is shown in Figure 5b.

There are at least three limitations of SD1 for predicting actual RO membrane fouling:

- SD1 filter membranes may not foul by colloids that foul RO membrane. The SD1 analytical membrane filter has a nominal (22, 35) pore-size rating of 0.45 micron. Many colloidal particles are less than 0.45 micron and readily pass through the analytical filter. These colloids may foul the RO system, but aren't measured by the SD1 test (8,29,35).
- SD1 filter membranes may foul by suspended solids that won't foul RO membranes. Recent research (35) has shown that fouling is most pronounced as the particle size is near the pore size of the analytical membrane. Particles approximately the same size as the pore size are able to completely plug the pore. Particles smaller than the pore size pass through the pore. Particles with diameters significantly larger than the pore size may not plug the pores (Figure 6).



CF = Concentration of dissolved and suspended solids in feed
CM = Concentration of dissolved and suspended solids at the membrane
B = Boundary layer
WF = Water flux
DB = Back diffusion
FF = Feed flux
○ = Bacteria
⁘ = Dissolved or suspended solids

Figure 4. Mechanisms of Membrane Fouling.

299

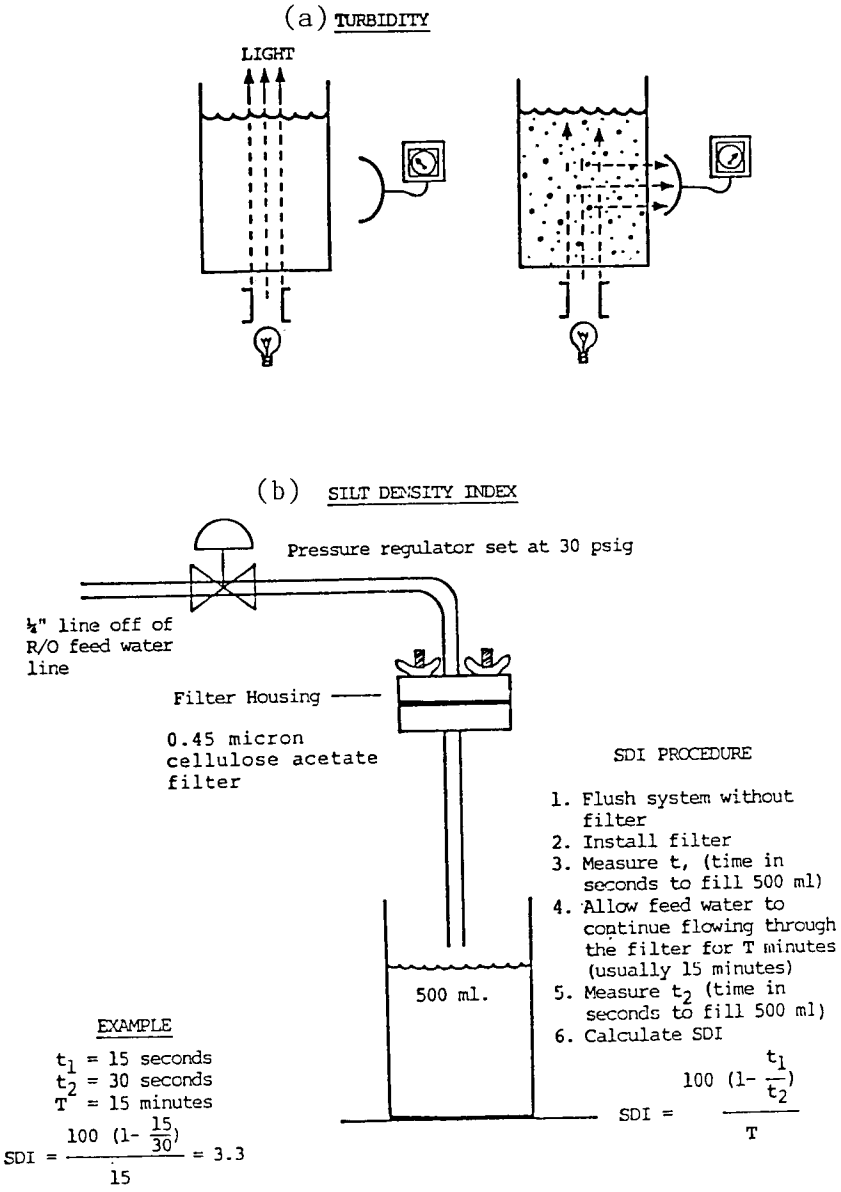


Figure 5. Turbidity and Silt Density Index.

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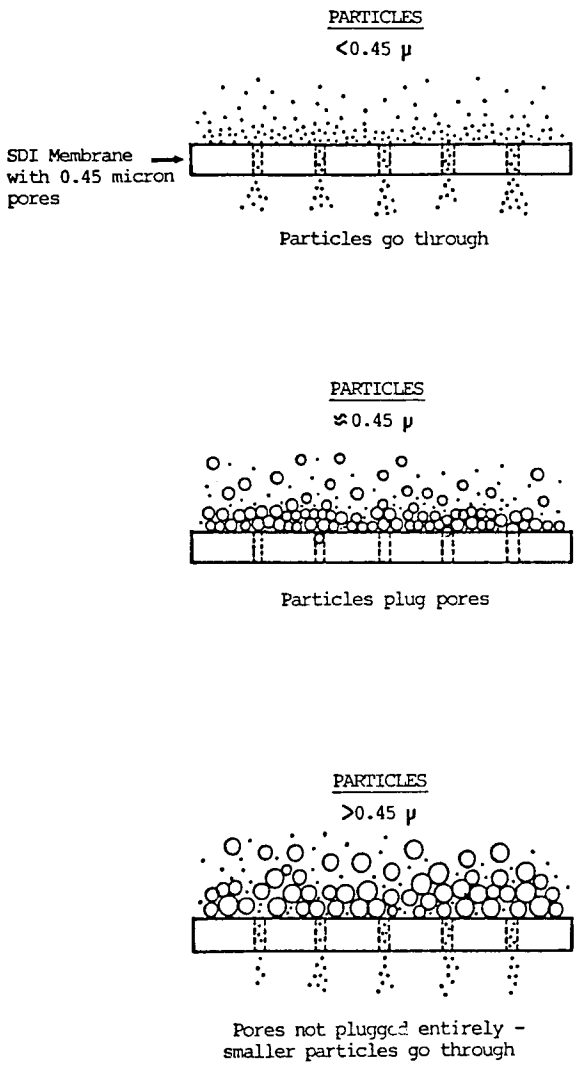

Figure 6. Fouling of Membranes by Particles.

301

- In actuality, excellent SD1 values don't preclude fouling. Several cases are reported where excellent SD1 values were measured from startup through RO system failure due to fouling (7, 28).

Even with their limitations, however, turbidity and especially SD1 are valuable qualitative predictors of RO fouling. We must use turbidity and SD1 to measure fouling potential, because they are all that we presently have.

Feedwater from surface water sources is oftentimes clarified and filtered as pretreatment to the RO. Turbidity measurement alone is insufficient to characterize such pretreated water. Both SD1 and turbidity should be monitored concurrently. Well water feeds usually don't require turbidity measurement. SD1 measurement, however, is still essential.

Biofouling: The fouling potential of bacteria is commonly monitored (when measured) by the standard plate count (SPC) or by field cassette monitoring (CM) (especially in high purity water systems). The theory is that the lower the number of viable bacteria in the feedwater and final water, the better. The number of bacteria is measured as colony-forming units (CFU) per milliliter, 100 milliliters, or liter of water sample. The population of bacteria that form colonies is referred to as the total plate count (TPC) bacteria. There are at least three limitations to these tests (37,38).

- The TPC bacteria represent only a portion of the total viable bacteria (alive and reproducing) that are actually presented because not all bacteria in the water can grow on the nutrients provided by the SPC or CM growth medium. Some of the water bacteria grow better on very low nutrient growth media.

- Only a portion of the total viable bacteria are measured because not all bacteria grow well at 35°C, the standard temperature for SPC and CM. This temperature is ideal for human pathogens but less suited for water bacteria accustomed to 15 to 25°C.

- Only a portion of total bacteria (total count) is measured because SPC and CM measure viable bacteria only. As we saw earlier in this article, even nonviable bacteria are capable of fouling the RO membrane.

Total counts may be more indicative of fouling potential. Total counts can be measured by fluorescent microscopy, scanning electron microscopy (SEM), and other visual counting methods. These are relatively expensive and time-consuming tests. Other tests are being sought as replacement.

Total organic carbon (TOC) has demonstrated a correlation with direct counts and provides valuable information (39). Limulus amebocyte lysate (LAL) analysis correlates well with direct counts and TOC (16, 39).

Disinfection

We might be inclined to think that killing bacteria prior to the RO should solve all our problems. Not so! Killing of bacteria by the use of chlorination may have little effect on the amount of biofilm formation (10, 34). While the chlorinated bacteria may not grow on plate count media, their bodies still attach, form a biofilm, and in one documented instance (34) either directly caused or at least were associated with higher flux decline, perhaps due to greater compactibility of dead bacteria over living bacteria.

Other disinfection methods may prove equally as troublesome. Ultraviolet light (UV) disinfection "kills" by damaging DNA (41) so that the bacteria can't reproduce. UV (like most, if not all, realworld disinfection schemes) only gives a percentage kill (41-43). Even if the kill is 99.99%, the living and dead bodies can still attach and form a biofilm as described above (20). The kills may be significantly less than 99.99% due to any substance, flow pattern, or lamp deficiency that prevents an effective, uniform dosage (41-46). The biofilm reduction potential of ozone is well documented. The rupturing of the bacteria due to oxidation of the cell wall liberates the cell contents (47-49). Since all of the organics will not be completely oxidized to carbon dioxide and water, the resulting total organic carbon (TOC) may increase and cell bodies may form a biofilm. Additionally, most noncellulosic membranes won't tolerate direct ozonation, requiring a UV or activated carbon deozonation step upstream of the RO. This elimination of biocide prior to the RO almost certainly ensures that viable bacteria will enter the RO membrane elements. Additionally, ozone may destabilize colloids (50) and increase their fouling potential.

At this point in time we can't be sure that any one or combination of disinfectants (51-52) is better than the others at reducing biofouling. We can be sure, however, of continued biofilm formation, flux decline, and other future challenges.

Cleaning

If we can't prevent fouling layers, can we remove them once they're formed? Maybe. Sometimes. Partially. That some of the foulant may be removed in many situations is well known. Practically ever facility has chemically cleaned its RO system with at least partial restoration of water flux and reduction of the feed/concentrate pressure differential.

There are indications that even most of the fouling layer can be removed. This was shown in perhaps the most scientifically conducted and documented research on RO membrane chemical cleaning, again from Ridgway et al. (18). The best cleaners in this case were ones that had one each of the following in the cleaning formulations:

Enzyme, to hydrolyze slime layer components

- Antiprecipitant, to solubilize inorganics
- Denaturing agent, to solubilize organics
- Bactericide, to kill living bacteria

The good news was that the formulations worked. The bad news was that they didn't work the same each time. It appears that the biofilm changes with time. The entire bacterial population may change during the life of the membrane element (10,28,34). If our old, reliable cleaner isn't working as well as it used to, it may be because the fouling layer has evolved. Also, the effectiveness of cleaning formulations may change over time.

So if we see that our old, reliable cleaner isn't working like it used to, it may be because the fouling layer has evolved. Effectiveness of current or new cleaners can be analyzed in-house

at the most sophisticated facilities, by chemical-cleaner vendors, or by other water treatment professionals.

Summary

Fouling occurs to some degree in all RO systems. There are several types of foulants, including inorganic foulants (colloids and precipitates) and organic foulants (dissolved organics and microorganisms). The mechanisms of action of the individual fouling processes have been minimally researched. The effect of combinations of foulmg processes is unclear.

Concentration polarization appears to be in operation with any fouling process or combination of processes. In general., increased feed/concentrate velocity and/or decreased water flux reduces foulmg.

Accurately monitoring the fouling potential of RO feedwater has many limitations at this time. Most existing measurements of colloidal and bacterial fouling potential are fairly good qualitative indicators but inaccurate quantitative indicators. Even with their limitations, they provide valuable information and must be used. Understanding their limitations, however, helps us to interpret the often confusing and conflicting results that we see.

Removal of an established fouling layer is attempted by exposing it to disinfectants and/or chemical cleaning agents. The effectiveness of removal varies from minimal to nearly l00%, depending upon the nature of the fouling layer, on the cleaning / sanitazing formulation used, and on the cleaning procedure. The fouling layer changes with time. Cleaning effectiveness of a single formulation may change accordingly.

It is obvious from the information presented in this article that much more research is needed.

Acknowledgment

Every attempt has been made to make this paper both accurate and readable. A special thanks for reviewing and editing this paper goes from the authors to Dr. Harry Ridgway, cheif research scientist, Orange County Water District, Fountain Valley, CA; and to Dr. Ata Hassan, Saline Water Conversion R&D Center, Kingdom of Saudi Arabia.

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