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Evaluating impact of fouling on reverse osmosis membranes performance

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ABSTRACT

Membrane fouling is commonly defined as an undesirable formation of deposits on membrane surfaces and it is considered a major problem in most brackish, seawater, and waste water reclamation applications. Depending on nature of the scale and foulant, fouling processes will lead to decreased membrane fluxes, increased pressures needs, and/or increased permeate conductivity. In some cases, irreversible damage on membrane rejection properties may also occur. Membrane fouling will adversely affect reverse osmosis systems efficiency and result in increased operational costs and energy consumption. Although extensive research has been done in this field in the past, membrane fouling is an extremely complex process and still not fully understood. Historically, an important part of research conducted in membrane technology has been dedicated to understand fouling mechanisms in experiments designed to compare the effects of different variables in the behavior of one specific foulant (natural organic matter, colloids, etc.). There are fewer published works on fouling studies on membranes from actual operating plants which take into consideration the role of multiple composite fouling. In this article, results from over 500 membrane elements autopsied in Genesys membrane products laboratories in the last decade will be presented and reviewed. Statistical analysis will be used in order to establish relations between different types of foulants and factors affecting the fouling processes notably: feed water type/quality, pretreatment, operational conditions, and membrane position. The authors report the effects on membrane properties and performance derived from results of the autopsy procedure. They advocate the use of this technique to gain important data to improve the efficient operation of membrane plants.

Keywords: Reverse osmosis; Fouling; Autopsy

1. Introduction

Reverse osmosis (RO) is nowadays the most extended technology for desalination globally. Besides, it has become a viable technology for wastewater reclamation [1]. Commercial interest in RO technology is increasing globally due to continuous process improvements which lead to significant costs reductions. These advances mainly include developments in membrane properties and module design, process design, feed pretreatment, energy recovery devices,

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and operational strategies focused in energy consumption reductions. Nevertheless, fouling is still considered one of the major challenges for an efficient operation of RO facilities, including sea water desalination plants, brackish water desalination plants, industrial plants, and waste water reuse/tertiary facilities [1–3]. Main consequence of fouling phenomena on membrane systems is an increase in operational costs, mainly related to increased energy demand, additional labor for maintenance, chemical cleaning, and reductions in membrane life expectancy [4]. For that reason, new efforts continue on the development of membrane elements with enhanced properties, underlying major resistances to fouling processes, as greatest efficiency gains in these processes have arisen from this kind of improvements [5].

Fouling can be defined as the undesirable formation of deposits on a surface. From a practical point of view, fouling can be defined as the accumulation of foreign materials from feed water on the active membrane surface and/or on the feed spacer to the point of causing operational problems [6]. Even if this description is not very orthodox, it clearly describes how important it is to know how all the components that coexist in water may affect RO membranes performance.

Different classifications have been defined by researchers but most common are based in foulant chemicals nature. From simplest ones that just establish two main categories, biotic and abiotic fouling, [7] to more complex classifications that may also consider other processes rather than fouling [8]. Most common one is based in four main categories including biological, organic, inorganic, and colloidal/particulate [9], although depending on the scope of the work some subcategories can be underlayed.

The different fouling types that can affect RO systems are already well known [10]:

- *Biological fouling*. A biofilm is described as bacterial aggregates attached to a surface; the biofilm structure includes a matrix of bacterially produced extracellular polymeric substances (EPS). The EPS are composed of polysaccharides, proteins, and nucleic acids and have been proven to play a major role in biofouling formation and its behavior; effectively altering the porosity, density, water content, charge, and sorption properties of the biofilm [11].
- *Particulate/colloidal matter*. Colloidal matter deposition on membrane surfaces is a consequence from a poor pretreatment. Most common nature for this colloids is aluminosilicates (clays), as subproduct of chemically weathered rocks and is ubiquitous in all waters around the world [12], but also other constituents can be found (colloidal silica).
- *Inorganic fouling/scaling*. The primary cause of inorganic scaling is supersaturation. When the solubility of a salt is exceeded, the salt precipitates and forms scale. Surface crystallization produces solid crystals directly on membrane surface. As active sites are present on the membrane surface, nucleation originates on the membrane active sites and grows further [13].
- *Organic fouling*. Higher molecular weight straight-chained organics, such as humic and fulvic acids, are common foulants found in surface waters. These organics typically "blind" off sections of membrane so that water cannot properly permeate. Furthermore, organics provide nutrients that sustain microbial populations [14].
- *Metals*. Elemental metals such as iron and manganese can oxidize from soluble to insoluble forms within an RO membrane and precipitate on the membrane. Although they could be included in inorganic fouling category, their origin can be related to common operational practices. Iron and aluminum can be a problem when coagulants

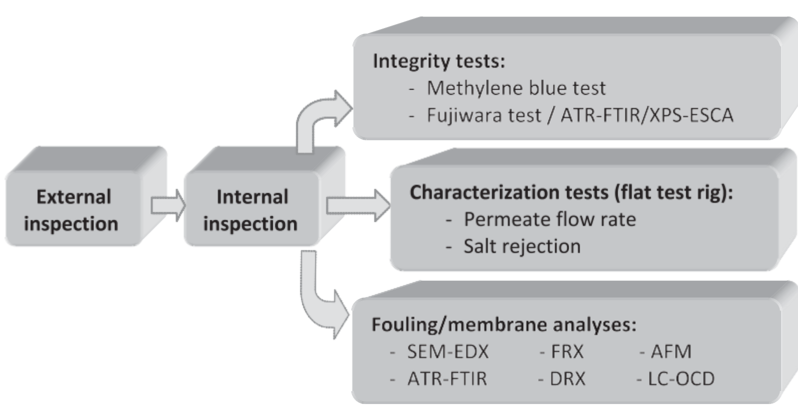


Diagram 1. Schematic description of main analysis carried out in membrane autopsies.

based in these metals are used to pretreat RO water. Both ferric chloride and alum are typically overfed and can carry over to post-precipitation and foul a membrane as a suspended solid [14].

Membrane autopsy is considered as the main tool for identifying fouling nature and establishing membrane failures causes. Next diagram shows schematically different stages and analytical techniques commonly involved during autopsy procedure, which were already explained in previous papers [12] (see Diagram 1).

Membrane autopsy is a destructive technique that implies high costs related to both analytical work and membrane replacement. In most cases, membrane autopsies are used as the last option troubleshooting tool. Commonly, autopsy techniques are used when severe problems related with decreases in flux, poor salt rejection, or increases in differential pressure values are detected in plant and no results with conventional remedial operational practices are achieved. Regardless, nowadays, some autopsies are carried out just for plant performance monitoring, cleaning practices optimization, or pilot tests evaluations, especially for big scale desalination projects as it offers a really objective evaluation of operational practices applied and membrane condition.

This work includes data from 500 autopsies, so it is a very valuable source of information from objective and real data. Although there are already publications compiling autopsy results [12,15], this article includes data from such a high number of studies, than it can be considered as a real data base of the most important fouling types and failures of RO plants around the world.

2. Results and discussion

Main causes of membrane failures can be classified into three categories: fouling, physical damage/abrasion, and chemical damage, which include oxidation processes. As explained previously, fouling is considered by both plant operators and researchers as one of the main reasons for membrane failure and deviations on expected performance. Data achieved in over 500 membrane autopsies recognize that severe fouling processes are considered as the main reason for membrane fouling in almost 60% of studied membranes, as shown in Fig. 1. However, chemical and physical damages appear as mild or slight failures. Although these data demonstrate that fouling is the main problem detected on membranes, this does not mean that it is the worst situation facing RO systems failures. Meanwhile in most of the cases, fouling can be

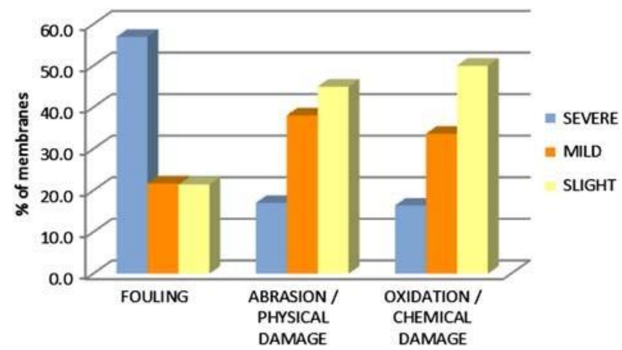


Fig. 1. Main membrane failures detected from membrane autopsies.

removed from membranes surface with chemical cleanings, both abrasion and oxidation are irreversible damages that make impossible to recover reference performance parameters. In any case, an effective fouling removal mainly depends on an accurate identification and on a fast application of cleaning procedures.

2.1. Types of fouling

Water treatment using RO membranes is a well known and broadly developed process. According to foulant classification discussed in previous chapter establishing five main categories (biofilm, particulate/colloidal matter, scales, metals, and organics), main foulant identified during autopsy process for membrane elements included in this study was reported and analyzed. Fig. 2 shows a summary of the main fouling relevance for each of these categories.

Biological and particulate/colloidal fouling state for 60% of the cases was reported. This fact shows the importance of pretreatment optimization in order to minimize membrane performance issues. Scaling/inorganic fouling processes have been identified as main foulant in 22% of the cases. Most commonly

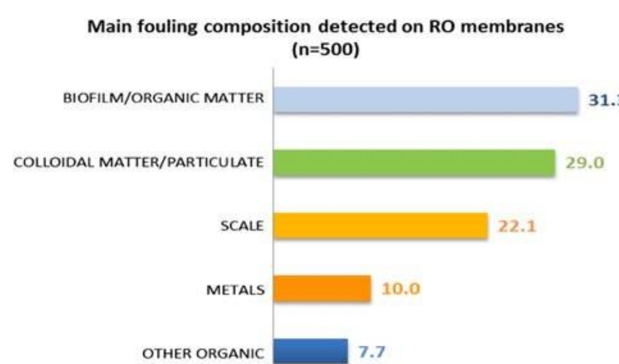


Fig. 2. Main fouling composition detected on membranes autopsies.

detected scales are silica and calcium carbonate (summarized data in Table 1). The most common metal detected on membranes is iron, which appears as oxide when it is the main fouling, and that it is commonly found as corrosion drag when it is found as a trace. Besides as main fouling components, it is very common to detect aluminosilicates (clays) and iron on most of the analyzed membranes.

Table 1
Main fouling compositions detected on membrane autopsies

Fouling group	Chemical composition	Characteristics	Expected membrane failures
Biofilm/ organic matter 31%	Protein derivatives/polysaccharides High level of micro-organisms	Jelly and sticky deposit on membrane surface On SEM-EDX, it is common to find particles and micro-organisms in it (Photographs 1 and 2)	″DP, #Flux, Salt passage Both foulings will gradually affect to membrane elements in first positions Micro-organisms presence can be difficult to remove
Other organic 8%	Some detected organic components: mineral oil, cellulose, chlorine compounds, polyacrilamide/ polyacrilates, dioxolane, and silicones	Thin and smooth covering of membrane surface (Photograph 3)	Organic matter is easy to remove and membrane condition can be recovered In extreme cases of biofilm, membrane degradation and failure can occur [16] Preventive action: Control of the population of micro-organisms in RO feed water and deficiencies in pretreatment
Colloidal matter 29%	>90% cases: Aluminosilicates	Conglomerate of very small particles (Photographs 4–6)	″DP, #Flux It will gradually affect all membranes, but first effects detected on lead elements The principal consequence of membrane fouling by clay minerals is an increase in hydraulic resistance resulting in a greater energy requirement to operate the process [17] Main cause: Deficiencies in pretreatment
Scales 22%	Calcium carbonate 35% Calcium phosphate 15% Calcium sulfate 10% Barium sulfate 4% Silica[a] 36%	Crystalline shapes, except for silica, which is also detected as amorphous (Photographs 7–12)	#Salt rejection, #Flux Scales mainly affect membrane elements in last position and they will negative affect membrane properties (rejection) if no action is taken immediately Preventive action: Accurate dosage of antiscalant
Metals 10%	Iron 67.7% Manganese 12.9% Aluminum 19.4%	Amorphous deposit (Photographs 13–15)	#Flux Main cause: Deficiencies in pretreatment

[a]Silica has been included in this scaling category, because from an autopsy analysis it is very difficult to distinguish between colloidal silica (deposit) and precipitated silica.

In order to review membrane position effect on fouling tendencies, data were analyzed separately for those membranes in the first and last positions. Data achieved are shown in Fig. 3 and verifies that most of the fouling groups appear at first position, except inorganic/scaling foulants, which are most frequent on last position. Nevertheless, it is also clear from data collected in this review that in some cases certain types of foulants can be detected in different positions than expected.

For identifying fouling nature different analytical techniques may be used, as it has been already specified in previous sections. Surface analyses techniques have played a decisive role in characterizing fouling on membrane elements. From all these, most widely used technique is Scanning electron microscopy with energy dispersive X-ray analysis (SEM-EDX). This technique is commonly used to study the membrane surface and to identify the elemental composition of its fouling. Elemental determination with the SEM-EDX system is based on analysis of X-rays produced via electron beam excitation of a sample area. This technique allows analysis of a membrane sample in selective areas. The limited depth of analysis (typically a few microns) and the possibility to select the area of interest under the electron beam allow for local analysis to reveal differences in composition. Table 1 includes a brief description of the main characteristics of the detected foulants, expected membrane failures related according to plant operation experience and related information provided with autopsied membrane elements. Additionally, characteristic images of each type of fouling obtained by SEM-EDX have been included. Additional data regarding membrane foulants incidence are also included.

The autopsied membranes included in this study come from many different plant types and applications. Regarding to water source type, most membrane elements have been applied for treatment of both sea and brackish water, considering other feed water sources (surface, effluents, etc.) as negligible. Fig. 4 shows a distribution of the different foulings detected on these two types of membranes. In order to provide valuable and objective conclusions, data included in this figure only considered those elements where fouling was considered as the main issue detected on the membrane and the main foulant was characterized. As it can be observed, the main two groups of fouling (biofilm and particulate/colloidal matter) appear on both kind of water as main groups of fouling. On the other side, scaling is mostly detected as main problem in brackish water and fouling seems to be more related to organic deposits and metals on sea water membranes. These results are consistent with the nature of the treated water and the historical problems found in plants.

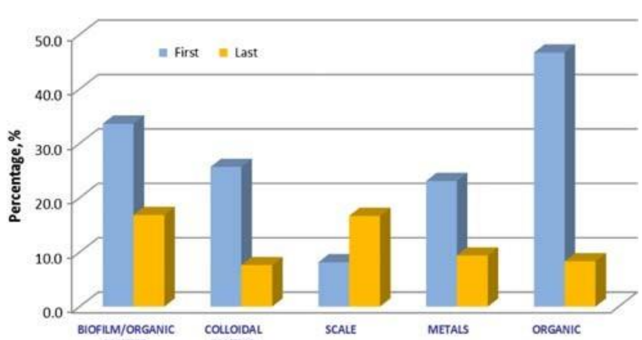


Fig. 3. Percentage of membranes detected on different plant positions vs. fouling group.

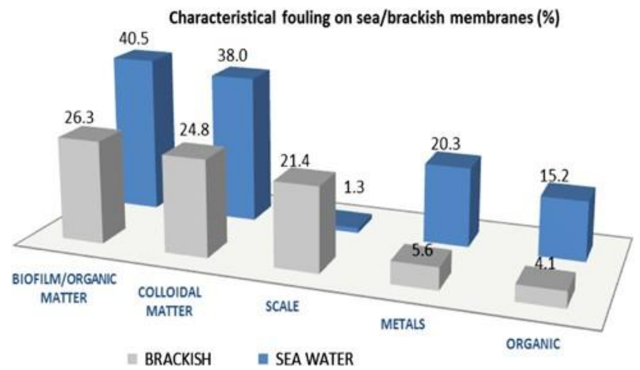
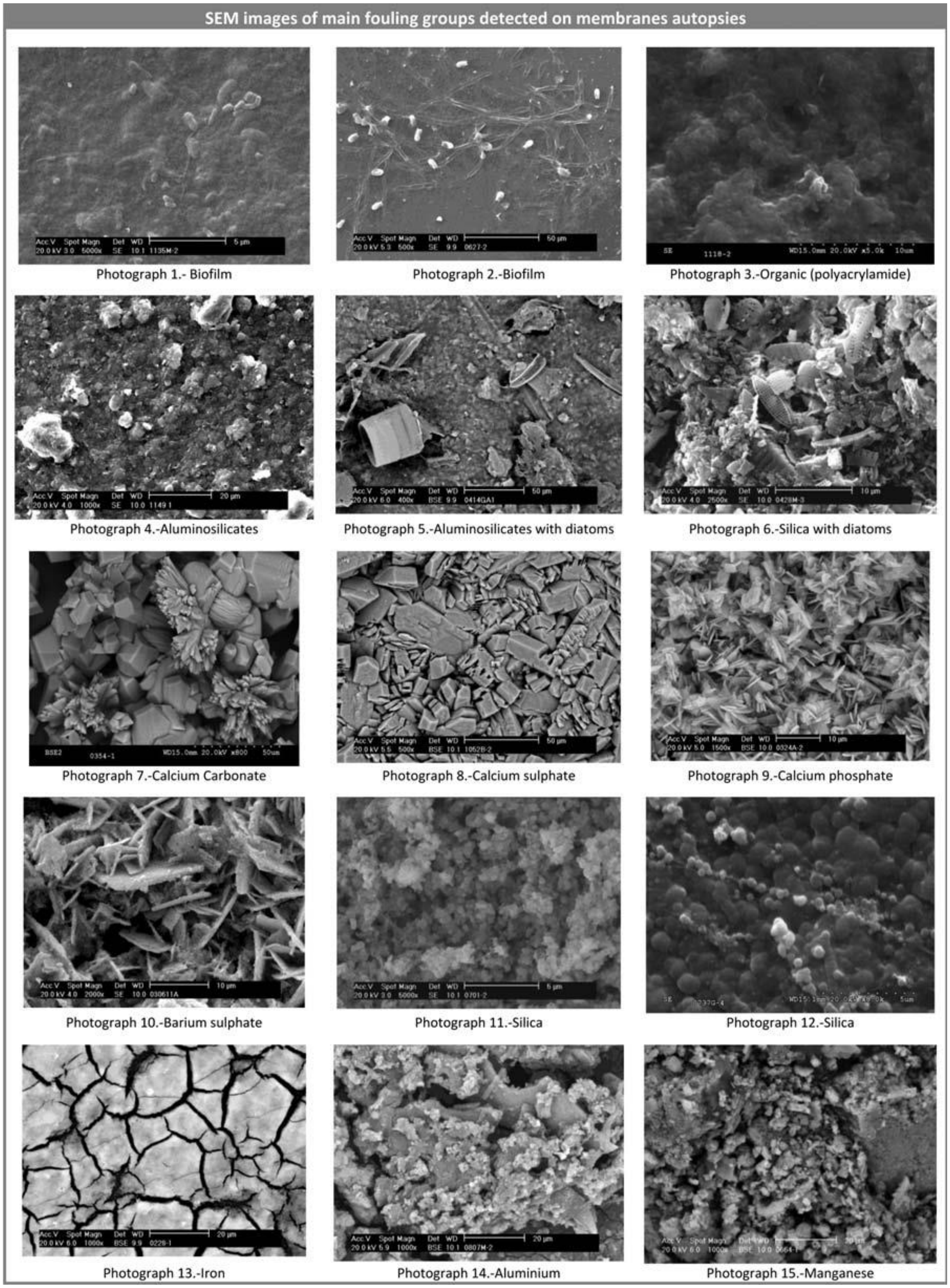


Fig. 4. Main fouling composition detected on membranes autopsies (%).

RO membranes in a typical operation are exposed to different types of foulants. Because of the complex nature of fouling, many mechanistic studies on RO membrane fouling have focused on one foulant type for the purpose of simplicity. However, it is very important to understand the effects of interactions between various foulant types on the fouling mechanisms [15]. Most of the published research papers on membrane fouling have focused only on a single, well characterized foulant, but the role of composite fouling needs to be considered and evaluated.

On membranes surfaces, fouling processes are very complicated and they are influenced by many factors. Besides, the presence of one preliminary fouling commonly causes that other components from water settle in it. Therefore, a deep study of membranes fouling must consider the "composite nature" of it, so it is necessary to take in account secondary components. Most of the studied autopsies for this article showed traces of different components and relevant presence of a secondary component for comparison were



detected on the 35% of the studied elements. These secondary components of fouling, considering the type of membrane, are graphically represented in Fig. 5. This figure verifies that, although some charac-

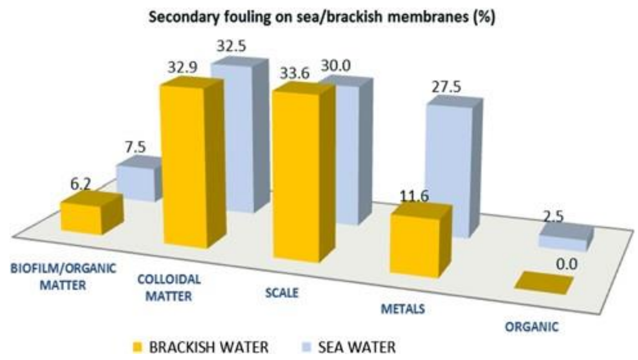


Fig. 5. Secondary fouling composition detected on membranes autopsies (%).

teristic components are not present on membranes as main foulants (e.g. scaling on sea water membranes), they may be present as secondary components. As figures show, secondary components are mainly inorganic for both kinds of water/membranes. In sea water, special attention should be paid to the presence of metals, since it is a clear consequence of corrosion processes.

Then, for a good performance of RO plants, we should consider all the skills that concern fouling and scaling independently of the water source [4].

As explained before, main fouling components are well known and characterized, but it is more difficult to find information about secondary components in order to anticipate and prevent their presence. Then, experimental data from autopsies are one of the most reliable sources for this information.

In Fig. 6, we have included the data of the secondary fouling components detected for each main group of fouling.

According to this figure, both biofilm and organic fouling are commonly accompanied by particulate/colloidal matter (aluminosilicates). However, when colloidal matter is the main component of fouling, it is more common to find scaling as secondary. On the other side, in scaling and metals fouling, the secondary component is more inorganic than organic.

Finally, Fig. 6 demonstrates that it is more usual to find a secondary component in organic foulings, probably due to their ability of retaining other components from water when they deposit on membranes surface. This last figure verifies also that colloidal matter is the most common secondary fouling found on membranes. Considering that, aluminosilicates are the second group of fouling observed as main problem in membranes, so pretreatment appears again as one of the main failures in water plant treatments.

2.2. Impact of fouling on membranes

In Table 1, we have reviewed the most common membrane failures expected from each fouling group and, on the other hand, we have demonstrated that it is more common to find a composite fouling than a pure one. Once again, the best way to test the influence of these factors on membranes performance is from experimental data.

In the following sections, we will try to face the main plant failures found for each type of fouling detected in membrane autopsies.

2.2.1. Increase in differential pressure (DP)

Most test rig devices have no instrumentation required for measuring DP, as they use membrane

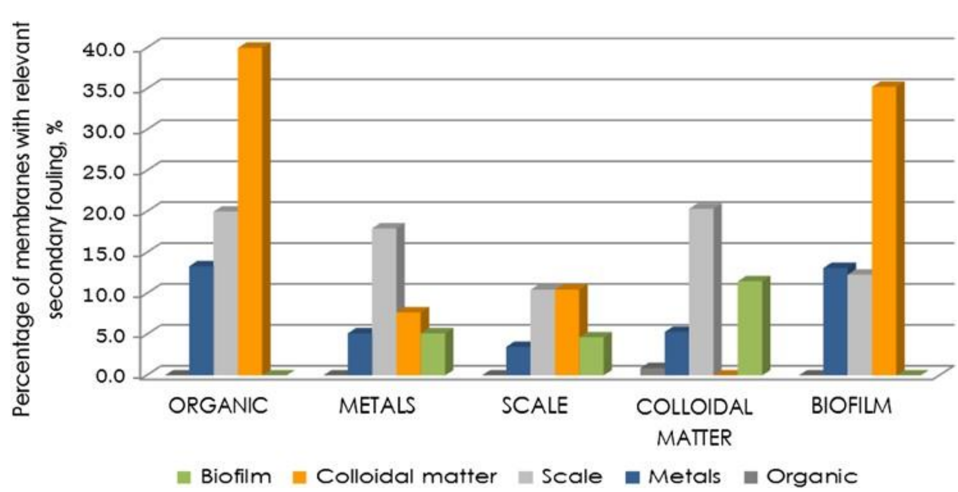


Fig. 6. Different components detected as secondary fouling vs. main fouling.

Table 2
Spacer protrusion on different fouling groups

Fouling	% of membranes with spacer protrusion
Biofilm	30.3
Organic	13.3
Colloidal matter	19.5
Scale	10.5
Metals	17.9

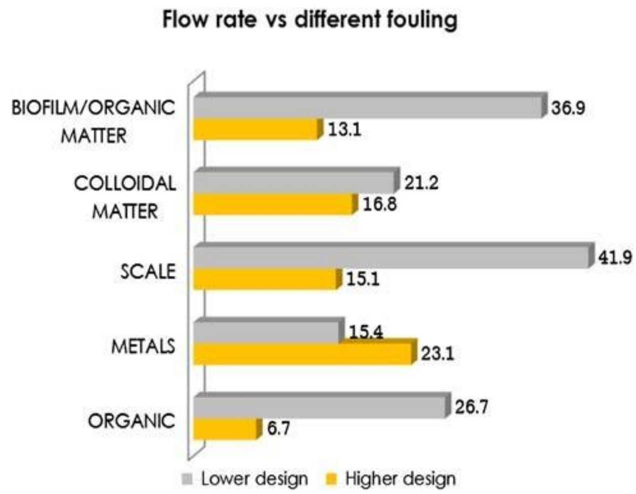


Fig. 7. Flow rate vs. fouling.

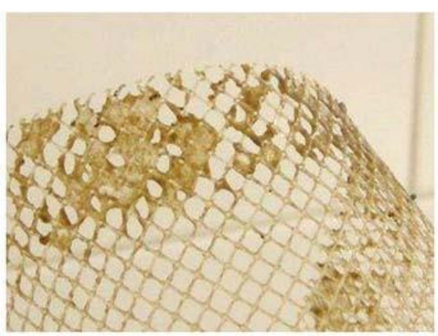
coupons with limited surface (commonly 5–20 cm^2), and it is not possible to check this parameter during autopsies. By the way, an increase in pressure differential is commonly associated to spacer protrusion phenomena and damages/fissures caused by the spacer on the membrane surface. Since these marks are difficult to quantify and verify, because the presence of fouling on the membrane surface may hide them, we will focus on spacer protrusion for evaluating its relation to fouling nature.

Table 2 includes the percentage of membranes with spacer protrusion for each group of fouling detected. As these results demonstrates, spacer protrusion was detected on every type of fouling, although the highest percentage of membranes with this problem was for biofilm. In membranes with biofilm, it is very common to find a relevant presence of the fouling on spacer, which contributes to material displacement (see photographs bellow) and increase in D*P*. The presence of fouling on spacer material can be found also in scaling processes (as shown in photograph 18).

group of fouling. Again, in order to evaluate just fouling influence on this operational parameter, only autopsies with fouling as main problem were used for flow rate and salt rejection evaluation.

Scaling is the group of fouling with a higher percentage of membranes with low flow (40%) and, on the otherside, it is also the group that shows the highest percentage of membranes with effect on rejection values. On the rest of fouling groups, only biological and organic fouling categories show a higher percentage of membranes where lower flow values were detected rather than higher, as particulate/colloidal matter and metals have a higher percentage with higher flux.

Photograph 16.- Spacer protrusion



Photograph 17.- Biofouling on spacer

Photograph 18.- Calcium Sulphate scale on spacer

2.2.2. Flow rate and salt rejection changes

Fig. 7 represents the variation of flow rate depending on the fouling detected on the membrane surface and Fig. 8 corresponds to the percentage of membranes with poor rejection values detected for each

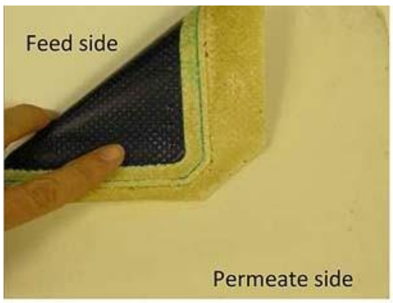
When severe fouling is detected, low flow rate would be expected, but between the autopsied elements showing fouling as main failure problem, a 22.7% showed a flow rate higher than design, so results are not accorded to membrane transport and fouling theories. Higher flux values in the presence of fouling imply damages on polyamide layer.

Concerning salt rejection, the 59% of the analyzed membranes with a severe problem of fouling showed poor rejection values. Fig. 8 shows that all the fouling categories can be related to decreased rejection properties. A bad rejection performance can be due to both the presence of fouling and the polyamide damages. If the reason is fouling, salt rejection could be recovered after effective cleaning procedures, but when the cause is polyamide damages, it would not be possible to recover original performance.

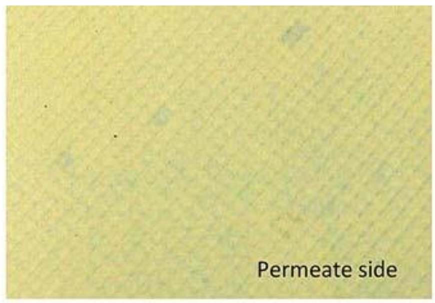
The lack of integrity on polyamide layer can be due to chemical damages (oxidation processes) and physical or mechanical damages (abrasion processes). Since we are just evaluating the impact of fouling on membrane surfaces, we will focus on the physical integrity of the polyamide and not on oxidation processes.

When we find sharp crystalline structures on the membrane surface, the cause of polyamide layer damage was showed obviously. But for other fouling groups, these damages can be produced on different ways. Fouled membranes produce less permeate flux, so plant operators tend to increase feed pressure to gain productivity. Operation under these conditions can result in spacer impact of the membrane surface. Concerning biofilm and other foulants that could adhere to spacer material, as already explained, increases in differential pressure are expected which produce telescoping and spacer displacement and the consequent membrane damage. Particulate/colloidal matter is commonly involved in membrane abrasion phenomena during regular cleaning procedures.

A fast experimental way to detect physical integrity failures is dye methylene blue test. In this test, a section of membrane is placed in a flat sheet test rig and a blue methylene solution is recirculated through it. Blue methylene is a high molecular weight molecule that should be easily retained by polyamide, unless it is damaged. Thus, if the membrane is damaged, dye passage will be observed on the permeate side of the membrane. Next photographs 19–21 show some examples of membrane coupons after this test.



Photograph 19.- Methylene blue test negative result



Photograph 20.- Methylene blue test positive result

Photograph 21.- Methylene blue test massive positive result

Table 3 includes the percentage of membranes with positive results in methylene blue tests. As it can be observed, except for membranes with organic fouling, most of the membranes show damages in some way on polyamide layer. As expected, scales are the fouling category with majority of damaged elements. By the way, how these damages affect membrane performance will depend on how relevant those damages are. So, it is important to take in account when positive results were slight or massive. Thus, Fig. 9 dem-

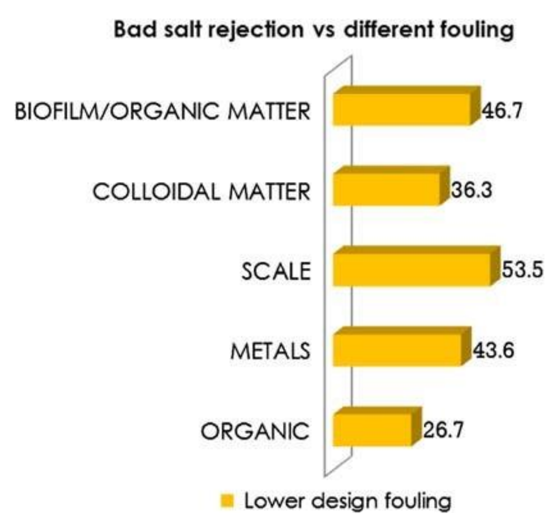


Fig. 8. Salt rejection vs. fouling.

Table 3
Percentage of membranes with polyamide damages

Fouling	% of membranes with positive result
Biofilm	87.1
Organic	8.3
Colloidal matter	83.7
Scale	95.8
Metals	78.1

onstrates that on membranes with biofilm and organic fouling, it is more frequent that damages are slight, meanwhile for scaling and metals, there is a higher percentage of membranes were this damage is massive.

Considering the level of damage, membranes with biofilms have the highest percentage of samples with slight damages and so it would be possible to recover their performance with cleanings. On the other hand, scaled membranes commonly show massive damages, so these results verify that this type of fouling processes produce irreversible consequences and membrane rejection properties could not be recovered after an effective deposit removal by chemical cleaning practices. Then, these results indicate that the main impact of fouling on RO membranes is the damage that can be produced on polyamide layer and on their rejection capabilities. Photographs 22–24 illustrate these phenomena (SEM micrographs).

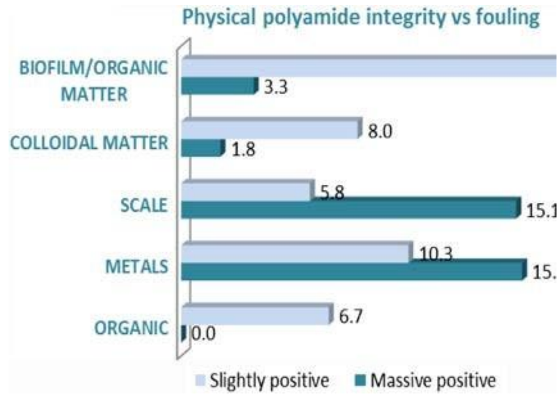
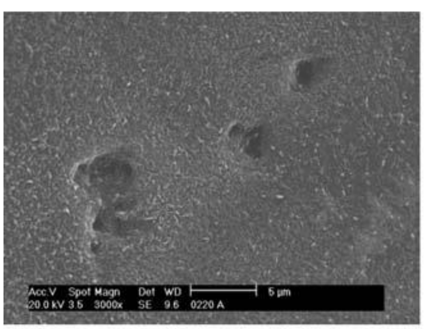


Fig. 9. Percentage of membranes with slight and severe polyamide damages vs. fouling.

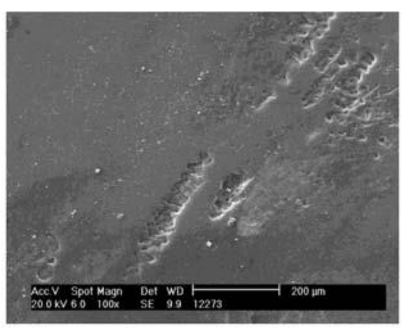
formance does not always correspond to the nature of the suspected fouling. This situation makes essential to carry out autopsies, in order to get the most accurate fouling identification for a reliable cleaning procedure.

On the other side, the information obtained from the 500 autopsies included in this study ends in some findings:

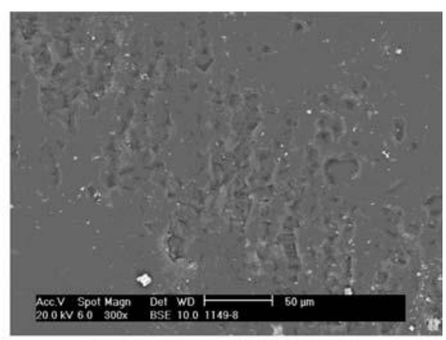
– Biofilm is the most common fouling found on membranes surfaces and it is the fouling that less damage may produce on polyamide layer, unless biofilm gets too well developed. In that case, a very high



Photograph 22.- Abrasion marks



Photograph 23.- Abrasion marks



Photograph 24.- Abrasion marks

3. Conclusions

The results of the membrane autopsies studied in this article verify most of the main items established in RO water treatment, concerning fouling and its impact on membranes performance: low flux, poor rejections, damages on polyamide layer, etc. Besides, all the results presented here indicate that it is almost impossible to find pure foulants and that their composite nature implies that membrane per-

presence of micro-organisms and the presence of secondary foulings make very difficult that cleaning procedures are effective and it is very probable to get high differential pressure and the corresponding damage.

– The other important problem found on membranes surface is particulate/colloidal matter, which was found also as one of the main secondary foulings. Aluminosilicates are more likely damaging polyamide layer by both abrasion marks and increases D*P* values, so pretreatment of RO plants is nearly the main problem to face in order to preserve membranes integrity and good performance.

– Scaling processes are probably the easiest problem to control with suitable antiscalants, but once the scale starts on the membrane surface, membrane performance failures are almost irreversible. Scales can be detected both on brackish and sea water membranes, so same prevention must be considered concerning antiscalant dosage for both kinds of water.

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