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# Three steps to control biofouling in reverse osmosis systems

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# ABSTRACT

Exposure to water containing micro-organisms causes biofouling on reverse osmosis (RO) membranes as they adhere, multiply and produce extracellular polymeric substances (ESP) which form biofilm on the surface of the membrane. As micro-organisms are present in virtually every water system, biofouling is one the most commonly encountered fouling types in large and small scale RO installations treating surface, waste- or seawater. Biofouling control is significantly improved when multiple methods are combined in an integrated approach and prevention methods employed in the RO stage itself are applied. In this study the impact of new membrane chemistry, feed spacer thickness and the use of non-oxidative biocide upon to the rate of biofouling in RO systems was investigated using a pilot-scale experiment involving small membrane elements subject to a high-fouling feed and autopsy-based analysis of membrane foulant loading and composition. The results were as follows: (1) The benefit of using the newest development in the family of fouling resistant (FR) membranes, DOW FILMTEC™ BW30XFR, was validated with side-by-side operation where lower rate of flux loss was observed when compared to the current industry standard membrane, BW30. (2) Thicker feed spacers provided reduced pressure drop and reduced rate of pressure drop increase during episodes of fouling. Overall organic foulant loading and bacterial counts were found to be reduced on membrane used in combination with thicker spacers. (3) The clear benefit of DBNPA dosing was observed with both shock and continuous dosing regimes. The benefit was most visible in the evolution of  $\Delta p$  as the treated elements operated at significantly lower  $\Delta p$ . Autopsy based results verified significantly lower organic fouling loading on the biocide treated element. These results point to the value of the studied factors – membrane chemistry, feed spacer configuration, and biocide dosing - for use with high-fouling feeds. The suggested route is to combine the components for use as an integrated strategy to solve biofouling. Combining a FR membrane with a thick feed spacer is preferred whenever a high potential for biofouling is seen. The use of targeted biocides in the pretreatment section will further result in improved fouling prevention and ensure long-term trouble free operation, maximizing the membrane lifetime and minimizing the operational expenses of the treatment system.

Keywords: Biofouling; Membrane and feed spacer modification; DBNPA

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# 1. Introduction

Exposure to water containing micro-organisms causes biofouling on reverse osmosis (RO) membranes. They adhere, multiply and produce extracellular polymeric substances (ESP) which form biofilm on the surface of the membrane [1]. As micro-organisms are present in virtually every water system, biofouling is one the most commonly encountered fouling types in large and small scale RO installations treating surface, waste- or seawater. If left uncontrolled, it can be a significant and persistent operational challenge with substantial economic consequences [1-3]. Biofouling research has been highly focused on various methods to control biofouling, but none of the previously proposed methods, when applied individually, have been able to solve the problem under every circumstances [4]. This work demonstrates that control is significantly improved when multiple methods are combined in an integrated approach [4].

The most common method to prevent fouling is to reduce or eliminate the contact of the foulant with the membrane by improving the pre-treatment section. Bacteria can be physically removed for example by low pressure membrane filtration (MF/UF) or inactivated by disinfection chemicals or UV treatment upstream of the RO membranes [3-5]. The drawback of these elimination strategies is that none can provide absolute elimination and the tendency of micro-organisms to reproduce results in situation where even a small amount of active bacteria surviving the pre-treatment can result in significant problems in RO section [6]. Additionally, a limiting factor for chemical disinfection is the incompatibility of the polyamide thin-film composite membrane with chlorine and other oxidizing chemicals commonly used for water disinfection [7]. A deactivation step with a reducing agent such as sodium metabisulfite is required before the feed water is put into contact with the RO membrane. Beyond the additional cost and complexity, the chlorination-dechlorination process has been reported to aggravate biofouling problems at some RO plants [3,7].

In view of these consideration, the need for biofouling prevention methods employed in the RO stage itself arises and this paper presents examples of three such techniques: (1) new fouling resistant FR membrane chemistry, (2) optimization of the feed spacer configuration for high-fouling or challenging feed water conditions by varying the spacer thickness and (3) the use of non-oxidative biocide (DBNPA), fully compatible with all materials of construction of the RO element. A range of pilot-scale experiments were carried out involving small membrane elements subject to a high fouling feed, followed by autopsy-based analysis of membrane foulant loading and composition. Results demonstrated the impact these techniques, alone and combined, upon permeate flux, feed-side pressure drop and response to fouling in RO systems.

# 1.1. Fouling resistant RO membrane chemistry

The surface chemistry of an active membrane can be modified such that it becomes FR, that is, more resistant to the adsorption and accumulation of the foulant. This is one of the recognized options to tackle fouling. As a leading RO membrane manufacturer, Dow Water & Process Solutions has long advanced this approach. The first generation of FR FILMTEC<sup>™</sup> membranes date back for more than a decade. Currently there are two FR chemistries available. The FILMTECTM FR membrane has a smooth, hydrophilic and strongly negative surface charge to make it less prone to the attachment and colonization of bacteria. It has an established track record against biofouling in surface and wastewater applications [8-11]. The most recent innovation is an extra FR membrane, BW30XFR, which in addition to biofouling control offers increased fouling resistance against particulate and organic fouling [12]. The improved biofouling resistance of the BW30XFR membrane compared to standard BW30 and extra low energy (XLE) membrane has been demonstrated using accelerated laboratory fouling tests. The results showed flux decline due to fouling with comparative magnitudes as follows: BW30XFR < BW30 < XLE. When run at constant feed pressure, the BW30XFR membrane lost less than 5% of its initial flux while the BW30 lost about 10% and XLE lost more than 20% of its initial flux [12]. In the same experiment, the BW30XFR also demonstrated an improvement in overall cleanability. The resistance to fouling and cleanability (degree of flux recovery upon cleaning) are not identical. Fouling resistance relates to the decrease of flux during operation and the rate at which this flux decrease occurs. Cleanability relates to the ability of the membrane to withstand cleaning protocols and the ability to recover the flux to its original level. Several cleanings will take place over the lifetime of the membrane element and harsh fouling will need aggressive chemical cleaning for removal. Biofouling is best removed with alkaline cleaners, with or without detergents. The unique feature of FILMTEC<sup>™</sup> elements is their ability to withstand repeated high pH cleanings, even up to pH 13, which is a key factor in cleaning biofouling. It has been previously reported that cleaning biofouling below pH 12 is not efficient [13].

# 1.2. Feed spacer configuration

Moving beyond membrane selection, RO element construction can be varied to reduce fouling and the effect of fouling upon system performance. The feed spacer has a critical role in biofouling development [14] and the spacer configuration can be optimized for high fouling potential or challenging feed conditions. One configurable parameter is the feed spacer thickness. It is often assumed that a thicker feed spacer will mitigate fouling and provide a reduction in pressure drop across the element [15]. The thicker spacer allows more void volume between the membrane leaves, providing more room for foulants and thereby increasing the time between cleanings in some systems, particularly systems where the cleaning criterion is based on a maximum allowed pressure drop. These would be plants with biofouling or high colloidal fouling. Unfortunately, high-quality comparative data on the effect of varying feed spacer thickness upon membrane element performance obtained under side-by-side operation in high-fouling feed water has been largely missing from the discussion [15].

# 1.3. Non-oxidizing biocide (DBNPA)

The use of a high efficiency, non-oxidizing, rapid kill biocide such as DBNPA (2,2-dibromo-3-nitrilopropionamide) is a more efficient biofouling control method than dosed-then-neutralized oxidizing disinfectants because the biocide can remain active throughout the RO membrane stage. DBNPA has been used for many years to prevent and remove biofouling in industrial RO systems [16,17]. It can be dosed either on continuous or in shock dosing mode and the dosing rate (concentration and frequency) can be optimized to account for varying plant conditions, such as seasonal changes. The largest benefit can be achieved when it is dosed further upstream of the RO section, such that biofouling control is achieved throughout the whole pre-treatment process (MF/UF, cartridge filters, etc.,). Most of the references related to the use of DBNPA at large scale installations describe successful operation over a period of years, but comparative data against a reference membrane demonstrating the effect has not been widely presented [18].

## 2. Materials and methods

In the present study three sets of experiments were carried out, each presenting one of the proposed steps for biofouling control: (1) new FR membrane chemistry, (2) optimization of the feed spacer configuration and (3) the use of non-oxidative biocide. In terms of experimental set up; pilot hard ware, data acquisition and control scheme the experiments were identical and are explained in Sections 2.1.1–2.1.3. Experiment specific details are explained individually in subsequent Sections 2.2–2.4.

## 2.1. Experimental setup

2.1.1. Pilot plant hardware, pretreatment and feed water properties

A small scale pilot apparatus, having up to eight single element RO pressure vessels in parallel positions was used in all experiments, allowing good quality side-by-side comparison (Fig. 1). The pilot unit received municipal effluent after secondary biological treatment and clarification. The process is shown schematically in Fig. 2. Minimal pre-treatment was used upstream of the RO membranes. This allowed the onset of the fouling to be fast and its effects easily observed. Details of the feed water composition and the size of used pre-filters during each experiment can be found from the Table 1.



Fig. 1. RO pilot plant with eight parallel vessels housing elements with outer dimensions of 2.5-in. in diameter and 14-in. in length.



Fig. 2. Process scheme of the pilot plant.

# 2.1.2. Data acquisition

Operational data was continuously logged for the following parameters: feed and permeate flow, feed pressure, pressure drop, feed and permeate conductivity, temperature, feed and permeate pH. The permeate flow and conductivity were normalized using the Film-Tec FTNorm program. The pressure drop was normalized according to Eq. (1), where  $Q_{avg}$  = average feed-side flow rate (equal to the arithmetic average of the feed and brine flow rates) and  $\mu$  = dynamic viscosity. Values marked ref correspond to the reference operating condition. The reference operating conditions to which the data were normalized included the same flux and recovery for all vessels:

$$d P_{\text{norm}} = d P \cdot \left(\frac{Q_{\text{avg,ref}}}{Q_{\text{avg}}}\right)^{1.4} \left(\frac{\mu_{\text{ref}}}{\mu}\right)^{0.6}$$
(1)

#### 2.1.3. Control scheme

The impact of fouling was observed in the flux decline and the increase of feed-side differential pressure. To the extent possible, the recovery and flux were

Table 1 Feed water analysis kept constant and equal for all elements. This provided a basis for direct comparison of the rate of fouling of the various membranes, element configurations and whether biocide treated or not.

# 2.1.4. Fouling analyses

Autopsy-based analysis of membrane foulant loading and composition were done after the experiments had finished, utilizing common techniques; physical and visual inspection, weighing and loss on ignition (LOI). Physical inspection is a non-destructive test to determine the physical integrity of the element and visual identification of potential foulants in comparison to the virgin product. Weighing a fouled element is a fast if somewhat approximate way to determine amount of accumulated foulant. LOI is a gravimetric method in which the foulant sample is dried at 110°C and then heated to 550°C. This procedure will determine the moisture content of the sample and will destroy the organic material present, indicating by gravimetric analysis whether the foulant is mainly inorganic or organic and whether the organic fouling is mainly due to biofouling.

### 2.2. Fouling resistant RO membrane chemistry

The effect of the membrane surface chemistry upon fouling was evaluated by comparing the industry standard BW30 membrane to extra FR BW30XFR membrane. In RO applications outside the laboratory, fouling is often a combination of many types of fouling and hence the fouling resistance of a membrane is best demonstrated with waters from real applications. Both RO elements had an active membrane area

Parameter	Unit	Experiment			
		Fouling resistant membrane average (min–max)	Feed spacer optimization average (min–max)	DBNPA	
				Shock dosing average (min–max)	Continuous dosing average (min–max)
Temperature	°C	25.6 (25.6–29.2)	24.8 (21.7–27.9)	17.7 (14.1–22.1)	25.6 (25.6–29.2)
pН	-	7.2 (7.1–7.3)	7.32 (7.05–7.57)	7.3 (7.1–7.6)	7.2 (7.1–7.3)
Feed conductivity	$\mu S \ cm^{-1}$	2,153 (1,823–2,300)	2,038 (1,522–2,553)	2,442 (1,469–3,453)	2,153 (1,823–2,300)
Suspended solids	mg l-1	12.2	11	7.3 (1–34)	12.2 (2–23)
Chemical oxygen demand	mg l-1	32.8	11	31.4 (16-67)	32.8 (19-45)
Biological oxygen demand	mg l <sup>-1</sup>	11	9	10.3 (5–31)	11 (5–19)
Pretreatment (bag/cartridge)	μm	50/5	NA/5	50/5	50/5

of 5.8 ft<sup>2</sup> ( $0.54 \text{ m}^2$ ) and a 34 mil feed spacer and both elements were continuously treated with a biocide (DBNPA), allowing a clear comparison between membrane chemistries.

#### 2.3. Feed spacer configuration

The effect of feed spacer thickness upon fouling was evaluated by comparing two different membrane types and for each membrane two different spacer thicknesses were tested. Comparisons were made between the spacers associated with like membrane to allow fair comparison. Within each like-membrane pair, the elements were run at similar flux and recovery. Details of the membranes and spacers are presented in the Table 2. The spacers were commercially available bi-planar extruded netting made from polypropylene. They were oriented such that the feed flow direction bisected the angle formed by the crossing sets of parallel strands. Caustic and acid cleanings were performed during the experiments according to the manufacturers' guidelines [7]. The experiment was conducted during summer time with warm water temperatures (Table 1), which provided optimal conditions for bio-growth.

Once the experiment was finished, the BW30 membranes were subjected to physical and visual inspection, weighing and LOI evaluations. In this experiment, the nature of biofouling was further analyzed with microbial analyses of the membrane surface using the two FR membranes. The membranes were autopsied and four coupons of 36 mm in diameter were removed and transferred into sterile 50 ml flasks. To each sample, 20 ml of a phosphate buffered saline pH 7.0–7.3 (PBS) containing 0.1% surface active agent (Tween 80) was added, along with glass beads to facilitate the removal of sessile microbes from the membrane surfaces during a 2 min vortexing step. Tryptic Soy Agar with Casein-Peptone-Soy meal-Peptone agar (TSA/CASO) was used, a standard growth media for aerobic bacteria. The plates were incubated for 2 d in 30°C. A standard viable count technique was used to determine the level of contamination.

## 2.4. Non-oxidizing biocide (DBNPA)

The effect of DBNPA was compared by operating membranes with and without biocide treatment sideby-side in the same pilot plant. Different membrane types, having the same element construction, were used. The membranes were FILMTEC LE, BW30, BW30FR and BW30XFR, each with 34 mil spacer and active membrane area of 5.8 ft<sup>2</sup> (0.54 m<sup>2</sup>). Periodic caustic cleaning-in-place (CIP) was carried out at pH 12.6 and 25°C during the experiments.

The experimentation used both shock and continuous dosing methods with different biocide concentrations and dosing frequencies. The dosing plan is presented in the Table 3. AQUCAR<sup>™</sup> RO-20 with 20% active DBNPA was used. During the continuous dosing experiment the 20% DBNPA solution was further diluted to 5% with polyethylene glycol (PEG).

Table 2 Details of 2.5-in. diameter elements

Name	Membrane type	Active area (ft <sup>2</sup> m <sup>-2</sup> )	Spacer thickness (in. mm <sup>-1</sup> )	Spacer strand count (strands per 10 cm)	Angle between strands (degree)
BW 22 mil	BW30	7.0/0.65	0.022/0.56	35	90
BW 34 mil	BW30	5.8/0.54	0.034/0.86	35	90
FR 34 mil	BW30FR	5.8/0.54	0.034/0.86	35	90
FR 46 mil	BW30FR	4.4/0.41	0.046/1.17	28	90

#### Table 3

Trial dosing methods

Dosing mode	Concentration (active) ppm	Frequency	Dosing time (min)
Online-shock	20	Five times per week	60
Online-shock	20	Three times per week	60
Online-shock	20	One time per week	60
Online-continuous	5	Continuous	NA
Online-continuous	2	Continuous	NA

The shock dosing trial was carried out during winter time with colder temperatures and the continuous dosing was carried out during summer with higher temperatures. Fluctuations were observed in the feed water quality during the experiments (Table 1). Elements from the shock dosing experiment were weighed and subjected to LOI analysis after the experiment and visual inspection was performed for the elements after the continuous dosing experiment.

# 3. Results

# 3.1. Fouling resistant RO membrane chemistry

Comparative operational data for permeate flux and salt passage obtained during 40 d of operation are presented in Fig. 3. The BW30 membrane showed a higher degree of fouling throughout the whole experiment when compared to BW30XFR membrane. At the end of the experiment, the BW30 had lost 22% and BW30XFR only 14% of the initial flux. Several additional pilot scale trials and full scale plant operation have since been conducted with the new BW30XFR membrane with different feedwaters and the results have demonstrated its capability to withstand high fouling conditions [19,20]. In the long run, the benefits of selecting FR membrane can be clearly seen in both decreased energy consumption and reduced cleaning frequency. Coupled with higher fouling resistance, the BW30XFR demonstrated lower salt passage, which is expected due the difference in the element specifications [21].

#### 3.2. Feed spacer configuration

Comparative operational data for permeate flux and differential pressure obtained during 65 d of operation are presented in Figs. 4 and 5 for BW30 and FR membranes respectively. Both pairs showed that a lower initial pressure drop was achieved with the thicker feed



Fig. 3. Permeate flux and salt passage evolution of BW30 and BW30XFR membranes.

spacer and the effect was magnified when the membrane became fouled. After approximately 50 d of operation, a very rapid pressure drop increase was observed with all membranes, which was concluded to be caused by rapid biofouling development.

For the BW30 membrane, the difference in pressure drop ( $\Delta p$ ) between the two spacer thicknesses (22 and 34 mil) was clear from the beginning, but grew significantly as the 22 mil element started to foul faster. The 22 mil element kept a stable pressure drop for 1 wk, after which it increased rapidly. The pressure drop with 34 mil spacer was more stable during the experiment. In terms of membrane flux, both membranes started at the same level and flux decrease was observed throughout the experiment. Caustic and acid cleanings restored the flux partly and decreased the pressure drop. The positive effect of the cleaning was much stronger with the thicker spacer element.

Significant difference in  $\Delta p$  evolution can be observed with the different spacer thicknesses associated with FR membranes. Both spacers showed stable performance for the first 10 d, after which the 34 mil pressure drop increased markedly. The 46 mil spacer showed some



Fig. 4. Flux and differential pressure evolution of the BW30 membrane.



Fig. 5. Flux and differential pressure evolution of the FR membrane.

increase due the fouling, but the performance was better restored upon chemical cleaning, coming closest to original values. During the highest fouling period, the  $\Delta p$  of the thinner spacer was as much as 10 times higher whereas that of the thicker spacer was only two times higher compared to initial values. No large difference between the different membranes was observed in terms of flux throughout the experiment; even the efficiency of cleaning was very similar.

# 3.2.1. Autopsy based analysis of membrane foulant loading and composition

The conclusions from operational data were verified with surface analysis from the fouled membranes. After the trial was finished the BW30 membranes were submitted for physical inspection, weighing and loss of ignition analysis. The FR membranes were submitted for microbiological analysis on the membrane surface.

The element construction of both BW30 elements was good, no visual damage such as telescoping of the element scroll was observed. The 22 mil membrane weighed 635 g and the 34 mil weighed 499 g. Compared to the weight of a new element (485 g), the weight increase is 31% and 3% for 22 and 34 mil elements, respectively, indicating that some degree of fouling had occurred with both membranes, although the fouling accumulation was significantly higher in the element with the thinner feed spacer.

Results of the LOI analysis are presented in the Table 4. The results show that almost all of the membrane foulant consisted of organic matter. The moisture content of both membrane samples was high, which indicated that most of the fouling present was biofouling. The amount of foulant was five to six times greater for the 22 mil sample than it was for the 34 mil sample.

The microbial analysis of FR membranes supported the finding of the LOI analysis. The primary contamination observed on the surface of the BWFR membrane was bacteria. The bacteria count on the surface of the thick 46 mil spacer was significantly lower than on the surface of the 34 mil spacer as can be seen in the Fig. 6.

Table 4 Results of the LOI analysis

Element type	Unit	BW 22 mil	BW 34 mil
Dry substance	%	21.01	4.77
Foulant distribution	g m <sup>-2</sup>	9.61	1.67
Inorganics	%	1.53	0.00
Inorganics distribution	g m <sup>-2</sup>	0.15	0.00
Organics	%	98.47	100
Organics distribution	g m <sup>-2</sup>	9.46	1.67



Fig. 6. Bacteria count found on the membrane surface.

This finding supported the results of the differential pressure evolution which indicated significantly lower level of fouling.

#### 3.3. Non-oxidizing biocide (DBNPA)

The effect of DBNPA online shock dosing was very similar with all tested membranes and followed the trending presented for the FR membrane (Fig. 7). The normalized permeate flux of the reference element was identical to the performance of the DBNPA treated membrane at the beginning of the experiment and the effect of increased fouling on the reference element can be seen after 3 wk of operation. At the end of the experiment, the DBNPA treated element had lost 23% of the initial permeate flux while the corresponding loss for the reference element was 38%. The performance decline of both elements can be justified by the fact that natural waste water is a combination of multiple fouling types and biocide is only expected to prevent biofouling. In terms of salt rejection, the impact of fouling was not clear as both DBNPA treated and reference membranes were showing similar evolution throughout the experiment (Fig. 8).



Fig. 7. Normalized permeate flux and  $\Delta p$  of BW30FR membrane.



Fig. 8. Normalized salt rejection of BW30FR membrane.

The performance difference between the biocide treated and reference elements was also well highlighted in the evolution of pressure drop over time (Fig. 7). As with permeate flux, the difference was obvious after 3 wk of operation. The  $\Delta p$  of the reference element continued to increase significantly, from 3% to 43%, inside the following 4 wk, while the treated membrane was more stable. A sudden differential pressure peak was observed for all membranes and the elements were immediately cleaned. The peak was caused by a sudden and rapid increase of feed water turbidity. A large amount of small particles (<5 µm) passed through the cartridge filters and blocked the feed spacers and caused an emergency shutdown which lasted over night. The performance of the BW30FR membranes was not fully restored, but the cleaning response was better with treated element. After 73 d of operation, the observed  $\Delta p$  increase was 32% (treated) and 192% (reference) when compared to initial values for the BW30FR.

During the first 3 wk of the continuous DBNPA dosing experiment, 5 ppm of active DBNPA was dosed and for the last 3 wk the concentration was lowered to 2 ppm. The fouling was strongest at the beginning of the experiment and the difference between the treated and non treated elements can be seen immediately, during the first day of operation. The phenomena differ from what was observed with shock dosing where the development of the fouling took longer. This can be partly related to the season. Continuous dosing was carried out during summer months and the increased ambient temperature was more favourable for rapid biogrowth. The seasonal phenomenon is often observed in larger RO plants.

The effect of DBNPA continuous dosing was very similar for all tested membranes and followed the trend presented for BW30XFR membrane (Fig. 9). The benefit of DBNPA was clearly visible both in the normalized flux and normalized  $\Delta p$ . For the first 25 d of operation, the flux evolution of dosed and reference membrane was the same; both membranes lost about 6% of their initial flux. For the remaining 15 d, the reference membrane continued to



Fig. 9. Normalized permeate flux and  $\Delta p$  of XFR membrane.



Fig. 10. Normalized salt rejection of XFR membrane.

decrease, ending with a flux loss of 21%. During the same time, the biocide treated membrane lost only 14% of its initial flux. In terms of salt rejection, the impact of fouling was not conclusive, but the DBNPA treated element did demonstrate moderately better salt rejection (Fig. 10).

The evolution of  $\Delta p$  was even more convincing. Significant differential pressure increases were observed after just 1 d of operation, measuring 39% and 105% for the dosed and reference membranes, respectively. After the first CIP the  $\Delta p$  of the treated membrane was restored to its original level whereas the reference membrane was still operating at around 88% higher  $\Delta p$  than initially. The third cleaning was most efficient in restoring the performance, but the difference between the elements remained clear; at the end of the experimentation, the observed  $\Delta p$  increase was 110% for the reference element while the treated element showed practically no  $\Delta p$  increase. Reducing the dosing concentration from 5 to 2 ppm did not show any a significant performance decline, which confirms the effectiveness of DBNPA at low use levels.

# 3.3.1. Autopsy based analysis of membrane foulant loading and composition

Elements from the shock dosing experiment were weighed after the experiment. The weight of both FR



Fig. 11. DBNPA treated BW30XFR.



Fig. 12. Reference BW30XFR.

elements was higher than initially, showing an increase of 13% and 24% for treated and reference element, respectively. LOI showed foulant distribution of 21.6 and 75.5 g m<sup>-2</sup> for the treated and reference element, respectively. The results are in line with the element performance presented above. Only visual inspection was performed for the elements after the continuous dosing experiment. The difference in fouling quantity was visible to the naked eye as seen in Figs. 11 and 12. Autopsy showed that all the membranes leaves were covered in brownish fouling and that the reference membrane showed a significantly higher degree of fouling.

### 4. Summary and conclusions

The impact of new membrane chemistry, feed spacer thickness, and the use of non-oxidative biocide upon feed-side pressure drop and membrane flux were investigated. A pilot-scale experiment was conducted using small membrane elements subject to a high-fouling feed, followed by autopsy-based analysis of membrane foulant loading and composition. The results were as follows:

• The benefit of using the newest development in the FILMTEC<sup>™</sup> family of FR membranes, BW30XFR, was

validated with side-by-side operation where lower rate of flux loss was observed when compared to the current industry standard membrane, BW30.

- Thicker feed spacers provided reduced pressure drop and reduced pressure drop increase during episodes of fouling. Overall organic foulant loading and bacterial counts were found to be reduced on membrane used in combination with thicker spacers.
- The clear benefit of DBNPA dosing was observed with both shock and continuous dosing regimes. The benefit was most visible in the evolution of  $\Delta p$  as the treated elements were operating at significantly lower  $\Delta p$ . Autopsy results verified significantly lower organic fouling loading on the biocide treated element.
- The shock dosing showed that frequent shocks were needed to keep the biofouling under control. Continuous biocide dosing resulted in more stable operation and no performance decline was seen when dosing was reduced from 5 to 2 ppm. This result shows how chemical use can be minimized without compromising efficacy.

These results point to the value of FR membranes, thicker feed spacers, and regular biocide dosing, especially for high-fouling feeds with significant potential to impact feed-side pressure drop. If a cleaning criterion based on maximum allowed pressure drop had been applied, rather than simultaneous cleaning of all elements, the data suggest that both the membrane treated with DBNPA and membrane elements with thicker feed spacers would have been cleaned less often.

A suggested path forward is to use an integrated approach for fouling control and not to rely on standalone fouling control strategies. A FR membrane combined with a thick feed spacer is recommended whenever a high potential for biofouling is seen. The introduction of targeted biocides upstream of the RO will result in improved fouling prevention and help to ensure long-term trouble free operation.

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#### References

- H.C. Flemming, Reverse osmosis membrane biofouling, experimental thermal and fluid, Science, 14(4) (1997) 382–391.
- [2] J.S. Vrouwenvelder and D. van der Kooij, Diagnosis, prediction and prevention of biofouling of NF and RO membranes, Desalination, 139(1–3) (2001) 65–71.
- [3] M. O. Saeed, A. T. Jamaluddin, I. A. Tisan, D. A. Lawrence, M. M. Al-Amri and K. Chida, Biofouling in a seawater reverse osmosis plant on the Red Sea coast, Saudi Arabia, Desalination, 128(2) (2000) 177–190.
- [4] J.S. Vrouwenvelder, Biofouling of spiral wound membrane systems, PhD thesis, chapter 13, (2009) pages 307–330.
- [5] D. Kim, S. Jung, J. Sohn, H. Kim and S. Lee, Biocide application for controlling biofouling of SWRO membranes – an overview, Desalination, 238(1–3) (2009) 43–52.
- [6] H.C. Flemming, G. Schaule, T. Griebe, J. Schmitt and A. Tamachkiarowa, Biofouling – the Achilles heel of membrane processes, Desalination, 113(2–3) (1997) 215–225.
- [7] FILMTEC<sup>™</sup> Reverse Osmosis Membranes Technical Manual, chapter 2.6.3 Chlorination/Dechlorination, page 58 (2009).
- [8] P. Sehn, A. Song, Y. Maeda and E. Zhao, Eliminating Biological Fouling by Combining Two Technologies: Fouling Resistant Membranes and Non-Oxiziding Biocide, Conference proceedings of International Forum on Water Industry conference, Shanghai (2006).
- [9] K. Majamaa, P. Aerts, C. Groot, L. Paping, W. Van den Broek and S. Van Agtmaal, Retrofit of an integrated membrane system to maximize water recycling at an industrial complex, Desalin. Water Treat., 18 (2010) 17–23.

- [10] S. Coker and P. Sehn, Four years field experience with fouling resistant reverse osmosis membranes, Desalination, 132 (2000) 211–215.
- [11] K. Majamaa, C. Connolly and K. Hoehn, Performance of Fouling Resistant Reverse Osmosis in Australia, Conference proceedings of Ozwater'10 conference, PB06.04, Brisbane (2010).
- [12] A. Roy, J. Jemenez and W.E. Mickols, A Critical Flux Loss Phenomena Found in Particulate and Oil/Surfactant Fouling in Seven Commercial TFC Reverse Osmosis Membranes, Conference proceedings of NAMS conference (2009).
- [13] FILMTEC<sup>™</sup> Reverse Osmosis Membranes Technical Manual, chapter 6.7 Effect of pH on Foulant Removal, page 126 (2009).
- [14] J.S. Vrouwenvelder, D.A. Graf von der Schulenburg, J.C. Kruithof, M.L. Johns and M.C.M. van Loosdrecht, Biofouling of spiral-wound nanofiltration and reverse osmosis membranes: a feed spacer problem, Water Res., 43(3) (2009) 583–594.
- [15] K. Majamaa, J.E. Johnson and P. Aerts, Improved Reverse Osmosis Element Construction Reducing Differential Pressure in High Fouling Conditions, proceedings of the IDA world conference, DB09-075, Dubai (2009).
- [16] U. Bertheas, K. Majamaa, A. Arzu and R. Pahnke, Use of DBNPA to control biofouling in RO systems, Desalin. Water Treat., 3 (2009) 175–178.
- [17] S. Najmy, J. Adnett and U. Bertheas, Application development for the on-line application of DBNPA to RO-systems, proceedings of the Aachener Membran-Kollouqium, Aachen (2007).
- [18] K. Majamaa, U. Bertheas, V. Gomez Cortes, J. Cugat Mata and A. Arzu, Field Trial to Optimize the Use of DBNPA in WRU Application, proceedings of the IDA world conference, DB09-076, Dubai (2009).
- [19] K. Majamaa, W.E. Mickols, D. Wang, Y. Yuan and S. Beardsley, Tackling Membrane Fouling with Novel Membrane Chemistry, proceedings of the IDA world conference, DB09-167, Dubai (2009).
- [20] K. Majamaa, S. Rosenberg, A. Shrivastava, I. Chen, Performance Review of Extra Fouling Resistant Membranes In Industrial Applications, proceedings of the IWA Regional Conference and Exhibition On Membrane Technology and Water Reuse (IWA-MTWR 2010), Turkey (2010).
- [21] FILMTEC<sup>TM</sup> Reverse Osmosis Membranes Technical Specifications (www.filmtec.com).