Influence of permeation on air/water cleaning of spiral wound membrane NF/RO elements

Emile R. Cornelissen, Danny Harmsen, Erwin F. Beerendonk, Peter Wessels and Dick Van der Kooij

ABSTRACT

To improve hydraulic conditions, periodic air/water cleaning (AWC) is applied to a nanofiltration (NF) spiral wound membrane (SWM) element under permeation conditions to control biofouling and particulate fouling. A pilot study was carried out for 212 days with a vertically positioned SWM element fed by tap water enriched with a biodegradable compound (60 μg acetate-C l^{-1}). Operational parameters were daily recorded, rinse water was collected and analysed and a membrane autopsy was performed at the end of the experimental run. Normalized pressure drop (NPD) increased as a result of biofouling and particulate fouling, and could be controlled by periodic AWC, while the membrane transport coefficient (MTC) and retention based on the conductivity remained constant. Rinsing water from AWC contained biomass and particulate matter (iron), predominantly during the first few minutes of AWC. Membrane autopsy revealed active biomass and inorganic deposits (mainly iron and copper) at the inlet and outlet of the membrane element. The use of AWC in (vertically positioned) NF/RO SWM elements under permeation conditions improved the control of membrane fouling (especially biofouling and particulate fouling) and did not compromise the integrity of the membrane element (until day 60).

Key words | air/water cleaning, biofouling, membrane fouling, nanofiltration, spiral wound membrane

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INTRODUCTION

The use of membrane filtration in (drinking) water treatment is increasing (IDA 2006). One of the major problems in membrane filtration is membrane fouling, more specifically particulate fouling and biofouling, which negatively influences the performance of nanofiltration (NF) and reverse osmosis (RO) membrane processes (Khedr 2000). Membrane fouling can be controlled by periodic membrane cleaning to reverse the negative impact. The effectiveness of membrane cleaning depends strongly on the hydraulic conditions in spiral wound membrane (SWM) elements which are limited by the current design of NF/RO membrane installations (DOW 2008). To improve hydraulic conditions, periodic air/water cleaning (AWC) is introduced in vertically positioned SWM elements. AWC dramatically improved the control of membrane fouling (Cornelissen *et al.* 2007). All previous studies were performed under no permeation conditions with AWC. The effect of permeation on the AWC of SWM elements is the topic of this research.

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MATERIAL AND METHODS

Feed water and membrane material

A vertically positioned NF 2540-type SWM element (ESNA2 Hydranautics) was operated for 212 days at a transmembrane pressure of 700 kPa and a recovery of 14%.

Feed water was the locally available drinking water supplied over the period 12 April until 9 November 2007 (212 days) and was pretreated by 10 μm and 1 μm cartridge filters (Amafilter) to prevent high loads of particles entering the membrane element. A sodium acetate ($CH_3COONa{\cdot}3H_2O$ from Baker Analyzed) solution was added to the feed water entering the storage tank ($V = 1{,}000$ l; $t = 1$ h) to enhance microbial growth to obtain biofouling. The sodium acetate solution in the feed water was 60 μg acetate-C l^{-1}.

Method of operation

A vertically positioned SWM was fed (350 l h^{-1}) in up-flow by sodium acetate-enriched tap water to create biofouling. The actual sodium acetate dosing started at day 5. AWC started at day 30 and was carried out by adding pressurized air (600 kPa; 700 Nl h^{-1}) in co-current flow to the feed water. The AWC routine was carried out twice per week, and consisted of: (i) 5 minutes AWC; (ii) 1 hour copper sulfate sanitation (1 g l^{-1}); and (iii) 30 minutes AWC for day 30 to 160. After day 160 the copper sulfate sanitation duration was prolonged to 2 hours until the end of the run. At day 149 an extensive cleaning of the feed vessel and piping took place as a result of biofouling problems in front of the membrane element.

Sampling method

The pressure drop, permeate flow rate and permeate conductivity (only until day 60 due to technical problems) of the membrane element were daily recorded (except during weekends) to determine the normalized pressure drop (NPD), membrane transport coefficient (MTC) and retention in time (ASTM 2000). The rinsing water created during AWC was collected and analysed on day 30, 57, 58 and 61 by adenosinetriphosphate (ATP), non-purgeable organic compounds (NPOC), suspended solids, turbidity and inorganic compounds measurements. Furthermore, biomass organic and inorganic deposits were characterized (ATP, NPOC and ICP-MS) on the membrane surface during membrane autopsy after the experimental run. The membrane autopsy procedure was similar to that described by Vrouwenvelder *et al.* (1989).

RESULTS AND DISCUSSION

Feed water quality

The feed water was slightly alkaline and constant in temperature during the experimental run (Table 1). The NPOC and UVA_{254} levels were relatively low at the test location. The SUVA value was moderately high indicating a mixture of hydrophobic and hydrophilic organic fractions with both high and low molecular weights. The average turbidity value (after cartridge filtration) was relatively high, while the maximum turbidity value was found to be very high (0.58 NTU), indicating small particles entering the membrane element. For the inorganic parameters, only the calcium and bicarbonate concentrations were relatively high.

AWC under permeation conditions

The NPD of the membrane element doubled during the first 30 days, despite copper sulfate sanitation (Figure 1). When applying AWC at day 30, the NPD reverted back to the initial NPD-value (20 kPa) as a result of enhanced hydraulic cleaning of the membrane element. The NPD kept more or less constant until day 90, after which the NPD started to increase exponentially towards 68 kPa at day 149, which was ascribed to severe fouling of the membrane element due to visually observed biomass accumulation in the feed vessel and piping in front of the

Table 1 | Average feed water composition (12 April to 9 November 2007)

Parameter	Feed water
pH	7.9
T (°C)	15.6
NPOC (mg l^{-1})	1.9
UVA_{254} (m^{-1})	5.8
SUVA (l/mg m)	3.0
Turbidity (NTU)	0.23
Bicarbonate (mg l^{-1})	265
Calcium (mg l^{-1})	75.7
Chloride (mg l^{-1})	10.4
Iron (mg l^{-1})	0.10
Magnesium (mg l^{-1})	7.1
Silicate (mg l^{-1})	8.8
Sodium (mg l^{-1})	13.1

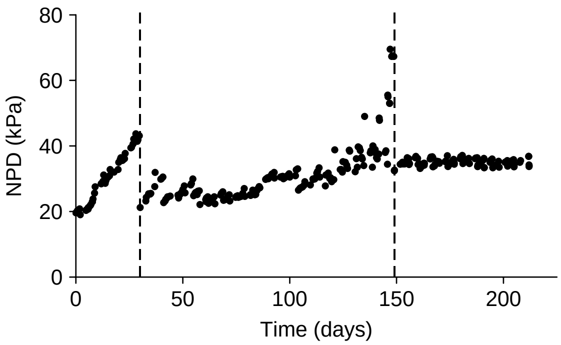

Figure 1 | Normalized pressure drop (NPD) with time for the membrane element operated with periodic air/water cleaning (twice per week) under permeation conditions. The dashed lines indicate (left) the start of the periodic air/water cleaning and (right) the extensive cleaning of the feed vessel.

membrane element. After cleaning of the feed vessel and replacement of the piping the NPD-value reverted back to 35 kPa. After doubling the copper sulfate sanitation time at day 149, the NPD remained constant until the end of the experiment.

AWC was found to be effective in controlling the NPD under permeation conditions, similarly to the results obtained from experiments under non-permeation conditions (Cornelissen *et al.* 2007). Furthermore, AWC is not a complete solution for controlling the NPD under permeation conditions and additional copper sulfate sanitation is necessary. A solid control of pretreatment processes of the membrane element is necessary to prevent excessive biomass production in the feed water.

During the experiment the MTC started at $0.8 \times 10^{11}\,\mathrm{m\,s^{-1}\,Pa}$ and slightly increased after starting AWC (Figure 2). During the experimental run after day 30 the MTC remained constant at $0.9 \pm 0.4 \times 10^{11}\,\mathrm{m\,s^{-1}\,Pa}$, indicating no effect of AWC on the MTC. Despite considerable biofouling and possibly particulate fouling as indicated by a substantial increase in NPD (Figure 1), the MTC remained constant. No flux decline phenomena were observed as a result of an extra resistance layer caused by particulate fouling as reported (Zhu & Elimelech 1997).

The retention based on conductivity remained constant at 98.5% during the first 60 days of the experiment, indicating no effect of the AWC on the membrane integrity (Figure 3). Despite considerable biofouling indicated by a doubling in NPD (before AWC at day 30), the retention remained constant.

The integrity of SWM elements is determined by changes in MTC and retention in time (DOW 2008). The MTC and retention (until day 60) remained stable during the experimental run using a SWM element under permeation conditions. The integrity of the SWM element remained uncompromised during 212 days of twice per week AWC (on the basis of MTC).

Analysis of the AWC rinsing water

Rinsing water from AWC mainly consisted of biomass (ATP) and particulate matter (iron and manganese) (Table 2) indicating that NPD increased as a result of a combination of biofouling and particulate fouling. The concentration of dissolved organic matter in the rinsing water was similar to the blank sample. At day 30, after 30 seconds a considerable amount of biomass and particulate matter was rinsed out, while after 2 minutes mainly

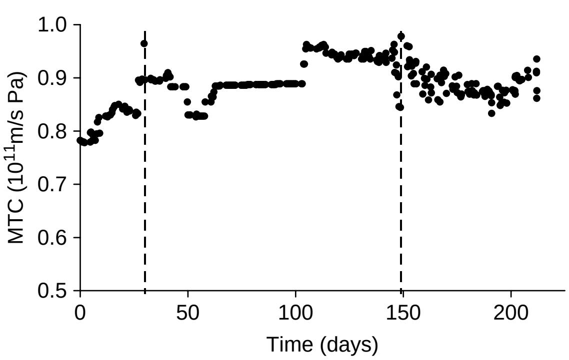

Figure 2 | Membrane transport coefficient (MTC) with time for the membrane element operated with periodic air/water cleaning (twice per week) under permea tion conditions. The dashed lines indicate (left) the start of the periodic air/water cleaning and (right) the extensive cleaning of the feed vessel.

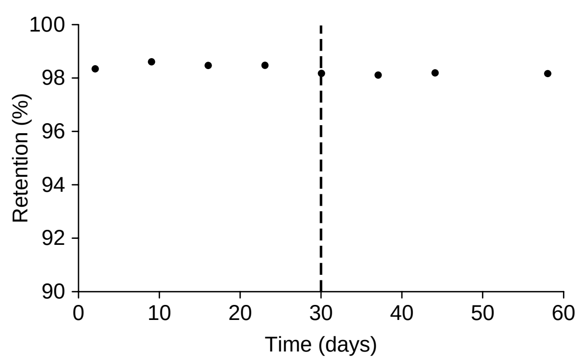

Figure 3 | Retention based on conductivity with time for the membrane element operated with periodic air/water cleaning (twice per week) under permeation conditions. The dashed line indicates the start of the periodic air/water cleaning.

Table 2 | Analysis of 1 l of collected flush water after air/water cleaning at day 30

Parameter	After 30 seconds	After 2 minutes	Blank
ATP (ng l^{-1})	2,170	238	0.7
Iron (mg l^{-1})	16	0.99	<0.1
Manganese (mg l^{-1})	0.28	<0.05	<0.05
NPOC (mg l^{-1})	2.3	2.1	2
Suspended solids (mg l^{-1})	47	2.9	1.5

biomass was rinsed out, albeit at much lower values. Similar results for only particulate matter were found at day 57, 58 and 61 as determined by both suspended solids and turbidity measurements (Table 3). Particulate matter was mainly removed in the first few seconds compared with after 2.5 minutes. Removal of particulate fouling and biofouling occurred in the first few minutes of AWC under permeation conditions, similarly to the results obtained under non-permeation conditions (Cornelissen *et al.* 2007).

Membrane autopsy study

The membrane element was taken from the pilot set-up at day 212 for autopsy. The analysis of the accumulated materials in the element was performed 11 days after removal of the elements from the set-up. A brownish deposit was evenly distributed over the length of the membrane element (Figure 4). High iron densities were found on the membrane surfaces, followed by lower copper, calcium and silicate densities (Table 4), possibly due to: (i) iron and silicate particulates/colloids present in the feed water (Gwon *et al.* 2003); and (ii) accumulation of iron and calcium in the biofilm (Ridgeway & Flemming 1996). High copper densities can be attributed to the copper sanitation during an AWC action. The levels of NPD increase (75%) found in this research indicated severe biofouling, which was, however, not confirmed by the ATP-values in membrane elements (1,400–3,000 pg ATP cm^{-2}). In comparison, feeding a pilot NF installation with 100 μg C l^{-1} sodium acetate-enriched tap water resulted in a NPD increase of 40% due to severe biofouling, measured as high ATP-values (45,000 pg ATP cm^{-2}) in a membrane element under non-permeation conditions (Cornelissen *et al.* 2007). The low ATP-values found in this research were attributed to the long gap between sampling and analysis (11 days cooled at 4°C). On the basis of the membrane autopsy study, the NPD increase during the experimental run was ascribed to biofouling and particulate fouling according to the results of the analysis of the rinsing water from AWC (see 'Analysis of the AWC rinsing water').

Figure 4 | Fouled membrane sheet from the membrane element operated under permeation conditions with AWC after 212 days. The flow direction was from right to left.

Table 3 | Analysis of 1 l of collected flush water after air/water cleaning at day 57, 58 and 61

		Suspended solids (mg l^{-1})	Turbidity (NTU)
Day 57	Directly ($t = 0$)	10.8	15.4
	After 2.5 minutes	3	1.7
Day 58	Directly ($t = 0$)	2.9	3.1
	After 2.5 minutes	1.1	1.0
Day 61	Directly ($t = 0$)	9.9	7.7
	After 2.5 minutes	0.4	0.76

Table 4 | Inorganic matter and biomass concentration at the inlet, middle and outlet position relative to the flow direction of the autopsied membrane sheet ($n = 3$)

Parameter	Inlet position	Middle position	Outlet position
ATP (pg cm^{-2})	1,400 ± 0.1	1,600 ± 0.1	3,000 ± 0.3
Aluminium (μg cm^{-2})	4.5	0.5	5.7
Calcium (μg cm^{-2})	18.0	2.0	11.6
Copper (μg cm^{-2})	32.5	2.8	44.6
Iron (μg cm^{-2})	170	17.5	226
Magnesium (μg cm^{-2})	0.4	<0.1	0.4
Silicate (μg cm^{-2})	13.5	1.2	13.8

Gradients in biomass concentration over membrane elements are commonly observed (Vrouwenvelder *et al.* 1989). When using AWC under non-permeating conditions, it was observed that the biomass concentration over the membrane elements was more constant over the length (Cornelissen *et al.* 2007). Similar results were obtained for biomass and inorganic deposits in this research for AWC under permeation conditions (Table 4). Biomass and inorganic matter concentrations were similar at the inlet and outlet position of the membrane element, while the middle part displayed lower concentrations. The gradient in biomass and inorganic deposits was not observed visually (Figure 4) and no solid explanation was found.

DISCUSSION

AWC was found to be effective under non-permeation conditions, although not 100% effective to control particulate fouling and biofouling. Similar results were found in this research for AWC on a membrane element under permeation conditions, from both operational parameters (NPD, MTC and retention) during a long filtration experiment and autopsy results. The results from this research indicate for the first time that permeation does not influence the effectiveness of AWC and vice versa, implying that AWC can be applied during normal operation of the (vertically positioned) NF/RO SWM elements.

AWC results in high shear forces in the membrane element, which might result in damage of the membrane element such as rupture of glue lines. During the experimental run the integrity of the 2,540-type SWM element remained uncompromised, measured by MTC and retention behaviour (until day 60), implying that AWC is a promising technique to control particulate fouling and biofouling in practical NF/RO SWM elements (under permeation conditions) without the risk of membrane damage.

The high substrate dosing applied in this work can be regarded as a worst case scenario for biofouling. Stable operation for 212 days under these extreme biofouling conditions can therefore be considered as a very long period. These results need to be validated under practical conditions for even longer periods to investigate: (i) the long-term effectiveness of AWC; and (ii) the long-term integrity of membrane elements operated under AWC conditions. Furthermore, in this research co-current feed flow and AWC was investigated. Detached biomass and inorganic deposits by AWC at the inlet of the membrane element will be transported through the entire length of the membrane element. Counter-current feed flow and AWC under permeation conditions of the membrane element is therefore preferred. Finally, a more fundamental basis needs to be provided in order to understand and improve AWC in SWM elements under permeation conditions, which is part of further research.

CONCLUSIONS

- Permeation does not influence the effectiveness of AWC in SWM elements.
- AWC in combination with copper sulfate sanitation is very effective in controlling biofouling and particulate fouling of SWM elements under permeation conditions.
- AWC is not 100% effective to control biofouling and particulate fouling, and a combination with a sanitation agent (copper sulfate) is necessary for better control.
- Most of the biofouling and particulate fouling is removed within the first minutes of AWC for SWM elements under permeation conditions.
- AWC does not compromise the integrity of the SWM element during 212 days with AWC twice per week.
- AWC is a promising technique to control membrane fouling during normal operation of (vertically positioned) NF/RO SWM elements.

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