Air/water cleaning for the control of particulate fouling

E. R. Cornelissen, X. D. Viallefont, E. F. Beerendonk and L. P. Wessels

ABSTRACT

Fouling of feed spacers in spiral wound membrane elements due to particulate fouling and biofouling is one of the main operational problems of nanofiltration or reverse osmosis membrane plants. Removal of particulate fouling from spiral wound membrane elements is investigated using frequent air/water cleaning (AWC). In a pilot setup two spiral wound elements were operated in parallel and were fed by tap water containing suspended solids. The reference membrane (REF) was fed with tap water pre-filtered with a 1.0-μm cartridge filter and fouled within 50 days indicated by a 55% increase in the pressure drop. The second membrane element (AWC) was fed with unfiltered tap water (with an average turbidity of 0.3 NTU) resulting in a 73% increase in the pressure drop within a few days of operation. By using air/water cleaning, the pressure drop decreased to initial pressure drop values, indicating complete removal of particulate fouling. It was concluded that periodical air/water cleaning proved to be effective in controlling membrane spacer channel fouling as a result of particles in the feed water.

Key words | air/water cleaning, hydraulic cleaning, particulate fouling

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INTRODUCTION

The use of membrane filtration in water treatment is in many cases hampered by membrane fouling. Particulate fouling is a serious problem when using spiral wound nanofiltration (NF) and reverse osmosis (RO) membrane elements and can occur when membranes are applied for the production of drinking water, process water and for desalination of seawater (Li *et al.* 2006; Kwee & Song 2007). Pre-treatment processes, such as rapid sand filtration, physico chemical methods and ultrafiltration, are essential to control or prevent particles entering the NF/RO spiral wound membranes (Ebrahim & Malik 1987). Despite these measures, particulate fouling still can occur as a result of inefficient pre-treatment processes, for example due to integrity problems with ultrafiltration (UF) membranes (Gijsbertsen-Abrahamse *et al.* 2006). As a result of particulate fouling, operational problems occur, such as increase of pressure drop along the membrane channel and a decrease of the permeate flux. The negative effects of (particulate) fouling can be reversed by the use of a periodical membrane cleaning.

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A wide range of membrane cleaning procedures is applied in practice with variable success in removing fouling material (Whittaker *et al.* 1984). Usually a chemical cleaning alone is not sufficient to control particulate fouling, and additional hydraulic measures should be taken (Vrouwenvelder & Van Der Kooij 2001). UF membranes, which are commonly used for the removal of particulate matter, are for that reason frequently hydraulically cleaned. A combination of air and water is commonly used for hydraulic cleaning of capillary UF membranes (Verberk *et al.* 2001). The use of air/water cleaning (AWC) to control particulate fouling in spiral wound membrane elements is investigated in this work.

THEORY

The hydraulic conditions in the membrane channel, in combination with the composition of the feed water, define the particulate load to the system. Furthermore, the hydraulic conditions within the membrane elements define the processes at the liquid-solid interfaces. Membrane elements are available in different diameters and a commonly used type for small-scale applications has a length of 1 m, a diameter of 2.5 inch and includes an envelope wound around a central pipe collecting the product water. With one envelope, with a width of 1.3 m and a length of 1 m, the total feed-water exposed surface area in one membrane element is 2.5 m^2 (DOW 2008). A typical feed water flow rate for a 2.5 inch membrane element ranges from 0.11 to 1.4 $m^3 h^{-1}$. The average flow rate in the empty feed channel ranges from 0.1 to 0.4 $m s^{-1}$. The actual flow rate in the feed channel is higher due to the presence of the feed spacer which reduces the porosity (ε) of the feed channel and also causes turbulence. The flow of water in the feed channel results in a pressure drop (PD) which is given by:

$$\mathrm{PD} = \frac{1}{2}\frac{\xi \rho v^2 L}{d_h} \quad (1)$$

where ξ is the friction factor, ρ is the density of water (g cm^{-3}), v is the average flow rate ($m s^{-1}$) in the feed channel with feed spacer, L is the length (m) of the feed channel and d_h is the hydraulic diameter, which depends on the feed spacer dimensions in the feed channel. The hydraulic diameter is given by (Schock & Miquel 1987; Da Costa *et al.* 1994):

$$d_h = \frac{4\varepsilon}{\frac{2}{h} + (1-\varepsilon)\frac{4}{d_f}} \quad (2)$$

where h is the channel height and d_f is the filament thickness of the feed spacer. For a standard 26-mil (0.66 mm) feed spacer ($h = 0.55$ mm; $d_f = 0.66$ mm and $\varepsilon = 0.86$) the hydraulic diameter is 0.77 mm (Neal *et al.* 2003). The average flow velocity in the membrane channel is given by the feed flow rate and the dimensions of the flow channel and is given by:

$$v = \frac{Q_f}{\varepsilon \cdot w \cdot h} \quad (3)$$

where Q_f is the feed flow rate and w the width of the membrane envelope. The friction factor ξ depends on the flow rate. The relationship between ξ and the flow conditions in spiral wound membrane elements is given by (4):

$$\xi = 6.23 \cdot \mathrm{Re}^{-0.3} \quad (4)$$

where Re is the Reynolds number, a dimensionless characteristic for flow which is defined by:

$$\mathrm{Re} = \frac{\rho v d_h}{\eta} \quad (5)$$

where η (g $cm^{-1} s^{-1}$) represents the dynamic viscosity of water. For a typical membrane element the flow conditions in the feed channel are turbulent as a result of the feed spacer, where flow would be laminar in its absence. Turbulent flow conditions are essential for the mass transport away from the membrane surface into the bulk. From Equations (1)–(5) it is apparent that the pressure drop depends on a number of membrane system characteristics, the viscosity of the solution and the flow velocity.

Particulate or colloidal fouling is usually described in relation to a loss of productivity at a given pressure in the feed channel (Kim *et al.* 2006; Chong *et al.* 2008; Park *et al.* 2008). Productivity is expressed as Mass Transfer Coefficient (MTC), the specific flux, and is an operational performance characteristic of the membrane which is defined as the amount of product produced per unit of time, per unit of surface area and pressure. Particulate fouling depends on the hydraulic conditions (e.g. feed flow rate, transmembrane pressure, feed channel characteristics), feed water characteristics (e.g. particle size, charge and concentration, pH and ionic strength) and membrane characteristics (e.g. roughness, surface charge, hydrophobicity) (Park *et al.* 2008). In this work, the increase in pressure drop due to particulate fouling is important and the effect of air/water cleaning on this pressure drop is studied. Accumulation of particles in the feed channel causes a reduction of the free volume in the spacer filled feed channel. This leads to an increase in average flow velocity combined with a decrease in the hydraulic diameter of the membrane channel (see Equation (1)). Both will cause an increase in hydraulic pressure drop (Figure 1). For a clean 2540-type membrane element ($h = 0.55$ mm and $v = 0.16$ $m s^{-1}$) the feed spacer porosity is 0.86, which corresponds to a pressure drop of 22 kPa (220 mbar). When the feed spacer porosity decreases to 0.75, maybe as result of particulate fouling, the pressure drop increases to 41 kPa (410 mbar) (an 85% increase in pressure drop).

An increase of pressure drop and a decrease in MTC can occur separately or simultaneously. These operational

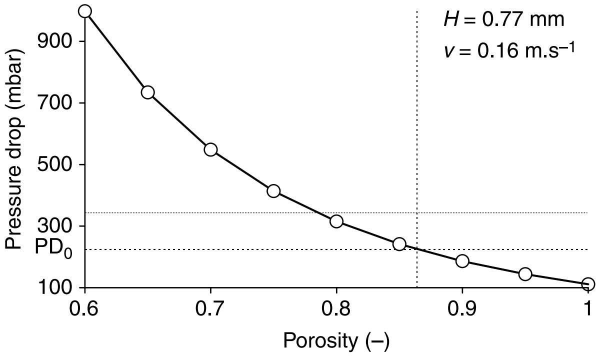

Figure 1 | Feed channel pressure drop in relation to the increase of the porosity of the feed spacer of a 2540-type membrane element. H = feed channel height; v = feed flow velocity; PD_0 = feed channel pressure drop for a clean membrane element.

problems require corrective measures to ensure productivity and reduce the loss of energy. For this purpose cleaning processes are applied and/or the feed water quality is improved by adapting pre-treatment. Usually cleaning is applied when the pressure drop increase over the entire membrane installation, which includes several stages with series of usually 4040-type or 8040-type membrane elements, exceeds 15% and/or the MTC decrease exceeds 15% (Graham *et al.* 1989). A pressure drop increase of 15% observed for the entire installation generally is mainly caused by a pressure drop increase in the front element of the first stage, in which the relative pressure drop may be close to 100% (Vrouwenvelder *et al.* 2008).

METHODS

Materials

Two 2540-type ESNA2 (Hydranautics) spiral wound nano-filtration membrane elements were used during the experiments. The membranes consisted of a composite polyamide layer, supported by a polysulfone substructure with a membrane area of 2.5 m^2, the feed spacer had a diamond pattern with a thickness of 0.66 mm. Both membrane elements were installed vertically for air/water cleaning and no permeation took place during the experiments.

The feed water was tap water supplied by pumping station Tull en't Waal and was produced from groundwater, treated by aeration and rapid sand filtration. Untreated feed water was characterized using on-line turbidity measurements (Sigrist KT65 white light turbidity meters) for 24 hours (4th Feb. 2009). Measuring accuracy is 0.02 NTU and the measuring interval is 2 minutes and 30 seconds (Vreeburg 2007). Furthermore, untreated feed water was characterized using particle count measurements (MetOne PCX) expressed as particle volume concentration and calculated assuming that the particles are spherical. Each measurement gives the number of particles per mL in 32 ranges with a width of 1 μm, ranging between 1 and 15 μm. Particle count measurements were carried out with a frequency of 2 minutes during one week (10th–18th Feb. 2009) (Vreeburg 2007).

Pilot set-up

Tap water entered a feed vessel equipped with a level switch for the protection of the feed pump (Grundfos CRN2). The total feed flow towards the pilot setup was 700 L/h and was distributed in down flow over two vertically mounted membrane elements (Figure 2). The flow per membrane element was adjustable and was kept constant at 350 L/h for each membrane element. The permeate outlets of both membranes were closed during the experiments, therefore no permeation occurred. The pressure difference over the feed spacer of both membrane elements was measured daily. During the weekends, operation of the membrane elements was stopped. The feed water for one membrane element (REF, reference element) was filtered by a 10.0 μm cartridge filter followed by a 1.0 μm cartridge filter. The feed water for the second membrane element (AWC, element frequently air/water cleaned) was not pre-treated.

Hydraulic cleaning with a mixture of air and water took place in up-flow direction (in counter current mode). Air/water cleaning was carried out when the pressure drop was increased up to 35 kPa (350 mbar) and at the end of the experiments.

Method and measurement scheme

Periodical air/water cleaning was performed by stopping the feed water flow to both elements and reversing the flow direction of the element to be cleaned while adding air. Pressurized air (6 bar) was used in an air to water ratio of 2:1, which is reported as a common air/water ratio

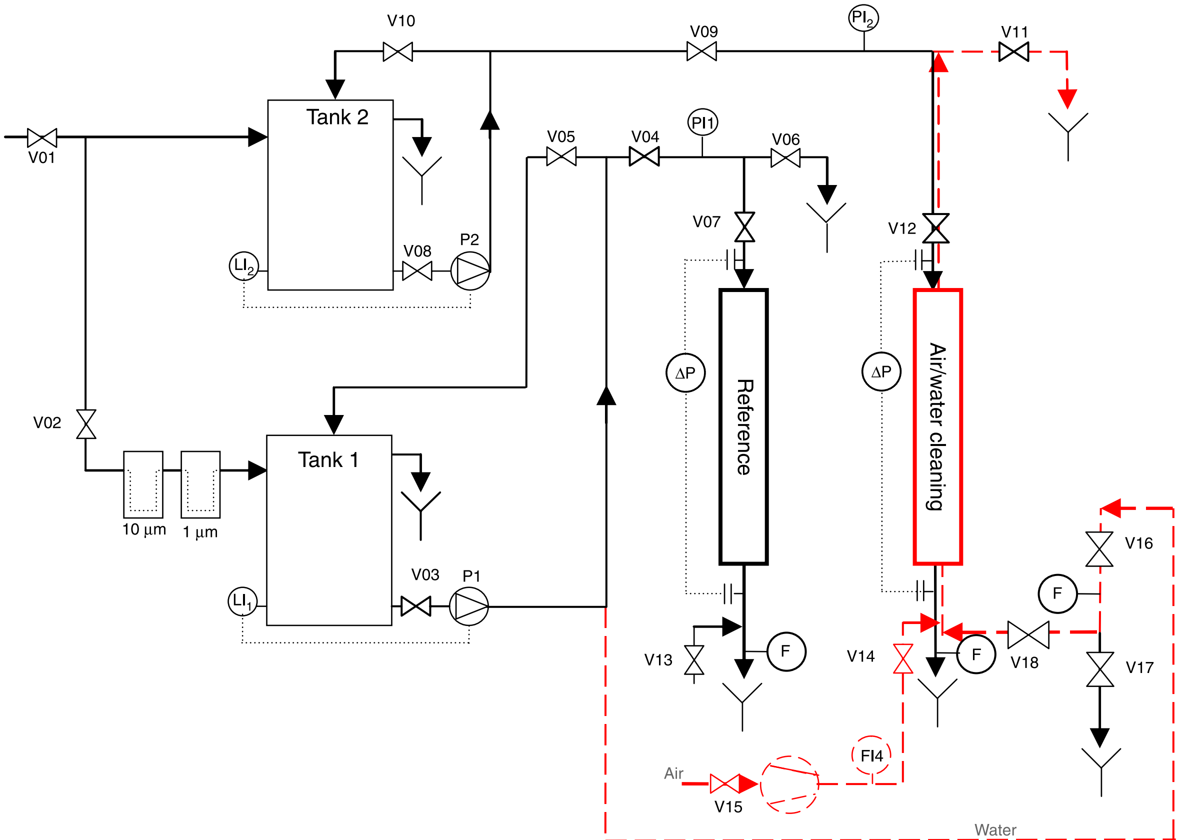

Figure 2 | Schematic diagram of pilot setup for air/water cleaning.

used for cleaning of ultrafiltration elements (Verberk *et al.* 2001). The water flow during cleaning was 350 L/h, and air flow was approximately 700 NL/h. When the pressure drop exceeded 35 kPa (350 mbar), the membrane elements were cleaned with air and water for 5 minutes. After the air/water cleaning process the pressure drop was measured again.

RESULTS

Feed water quality

The particle concentration in the feed water did fluctuate strongly as was found by turbidity and particle count measurements. The turbidity varied between 0.2 and 0.4 NTU and was 0.3 NTU on average (Figure 3). Maximum turbidity values were found around 11:00, 13:00 and 19:00, which was repeatedly found on other days (results not shown here). The total particle volume also considerably fluctuated in time within one week (Figure 4). The highest total particle volume peak values were found between 11:00 and 13:00 on each day, while lower total particle peak values were found between 18:00 and 19:00, which corresponds to the maximum turbidity values. Differences

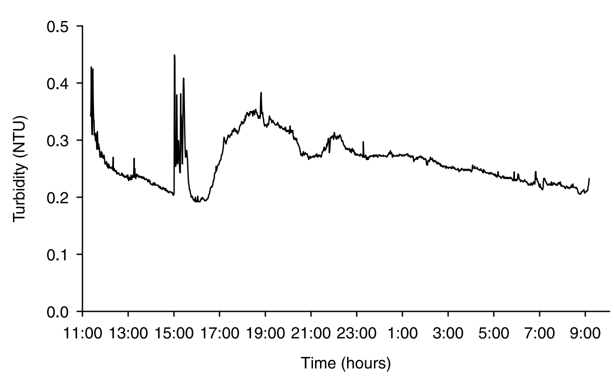

Figure 3 | Turbidity in time of feed water during one day (4th Feb. 2009).

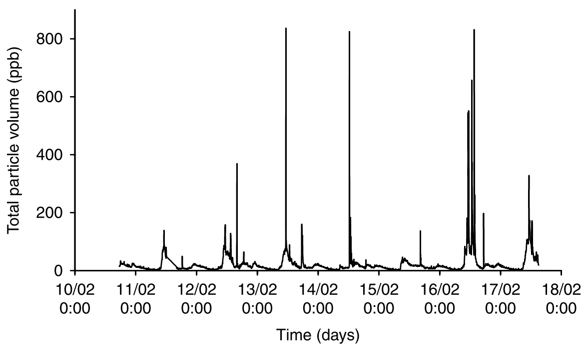

Figure 4 | Total particle volume in time of feed water during one week (10th to 18th Feb. 2009).

in total particle volume peak values were found between the different days; the highest total particle volume peak values were found on Friday, Saturday and Monday, while the lowest total particle volume peak values were found on Wednesday and Sunday. The maximum total particle volume value was 840 vol-ppb and the average total particle volume value was 13 vol-ppb (for particles between 1 and 15 μm). The average volume load of particles towards the membrane element without pre-treatment is therefore 4.6 μL particles/h. Temperature was approximately constant at 20°C, pH was 7.9, DOC was 1.9 mg/L and the iron content was 0.29 mg/L.

Reference membrane element (REF)

The pressure drop over the feed spacer of the reference element (REF) increased linearly in time from 22 kPa (220 mbar) to around 34 kPa (340 mbar) in 50 days' time, corresponding to a 55% increase (Figure 5). The membrane element was only operated for 36 days in this period, since installation was stopped seven times during the weekends. During the experiment, the cartridge filters needed to be replaced daily (except during the weekends). At the end of the experiment on day 50 the only air/water cleaning was performed resulting in a decrease to 23 kPa (230 mbar), corresponding to a 5% increase of the pressure drop compared to the initial pressure drop.

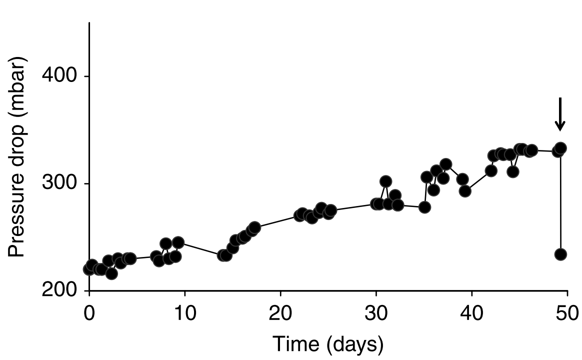

Figure 5 | Pressure drop development along the membrane channel of the reference element (REF) without air/water cleaning. At day 50 a 5-minute air/water cleaning was carried out (indicated by the arrow).

Air/water cleaned membrane element (AWC)

Air/water cleaning was necessary more frequently for the membrane element operated without pre-treatment to maintain the pressure drop below 35 kPa (350 mbar). The pressure drop increased within 2 days from 22 to 38 kPa (220–380 mbar), corresponding to a 73% increase (Figure 6). The first air/water cleaning procedures were performed after 2 and 4 days and could completely recover the original pressure drop. After the third air/water cleaning on day 10, the pressure drop after cleaning was 23 kPa (230 mbar), a 5% increase. The following five air/water cleaning procedures could recover the pressure drop to 23 kPa (230 mbar). After 50 days of operation, the pressure drop after air/water cleaning was 23 kPa (230 mbar).

Furthermore, it was found that the cleaning frequencies decreased as time progressed (Figure 6). There were three air/water cleanings required in the first 10 days of operation (or equivalent to an average fouling rate of 12 kPa/2 day

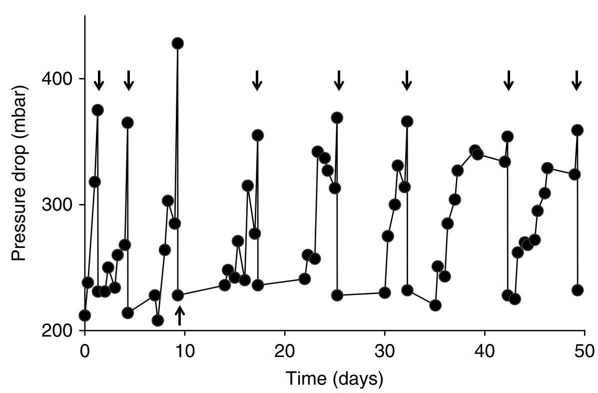

Figure 6 | Pressure drop development along the membrane channel of air/water cleaned membrane element (AWC). A total of eight air/water cleanings, each lasting for 5 minutes, were carried out in the 50-day experiment (as indicated by the arrows).

based on individual cycle), three air/water cleanings in the subsequent 25 days of operation (or equivalent to an average fouling rate of 12 kPa/3 day based on individual cycle) whereas only two cleanings were carried out from day 35 to day 50 of operation (or equivalent to an average fouling rate of 12 kPa/7.5 day based on individual cycle). The membrane operation performance appeared to have improved over time, which was unexpected especially when the pressure drop was restored to the original value after air/water cleaning.

DISCUSSION

A more or less linear pressure drop development over the membrane elements was observed, which was different from an exponential pressure drop increase caused by biofouling in similar membrane elements (Cornelissen *et al.* 2007). An exponential pressure drop development is expected on the basis of theoretical pressure drop considerations (theory) assuming a homogeneous fouling development. The linear increase of pressure drop over the feed spacer of the reference membrane element (REF) was ascribed to inhomogeneous particulate fouling due to incomplete removal of particles by the 10.0 and 1.0 μm cartridge filters. The mechanism of particulate fouling in spacer filled channels using AWC will be investigated in further research.

The membrane element fed with untreated water (AWC) displayed a rapid increase in pressure drop from 22 kPa to 38 kPa within 48 hours corresponding to a decrease in feed spacer porosity from 0.86 to 0.76 (assuming a uniform distribution of fouling). The available empty channel volume (V_{free}) in a membrane element is given by:

$$V_{\text{free}} = \varepsilon whL \tag{6}$$

The decrease in free volume according to Equation (6) due to feed spacer clogging over the whole membrane element is 72 mL. The volume of particles entering the feed spacer channel within 48 hours of operation is 0.2 mL, assuming a constant average total particle volume of 13 vol-ppb (based on particles > 1 μm and < 15 μm). The particle volume towards the membrane channel as determined by particle count measurement is only 0.3% of the volume loss calculated from the decrease in spacer porosity (Equation (6)). This percentage will be higher when particles < 1 μm are accounted for, which are not determined by particle counting. These smaller particles do contribute to membrane fouling as was clear from experiments with pre-filtered tap water (Figure 5). Apparently the feed flow resistance as a result of feed spacer clogging leading to pressure drop increase is located at a small part of the total length of the membrane element. By membrane autopsy it was visually observed that the dominant part of membrane fouling was located at the entrance of the membrane element (results not shown here).

Air/water cleaning proved to be very effective in reducing fouling in spiral wound elements due to particulate matter. The air/water cleaned membrane element (AWC) fed with untreated feed water, with an average feed water quality of 0.3 NTU, 0.29 mg/L iron and 13 vol-ppb total particle volume (for particles between 1 and 15 μm), performed well and experienced no shutdowns. Over a 50-day period of operation (36 days effectively), only a 5% pressure drop increase was observed with eight air/water cleaning cycles without using chemical cleaning. During the same period in the parallel reference membrane element (REF) the cartridge filters in the pre-treatment had to be replaced daily, indicating high levels of suspended solids in the feed water during the whole experiment, confirmed by turbidity and particle count measurements. Furthermore, we observed a distinct increase in the pressure drop to 34 kPa (340 mbar) over the reference element during 50 days of operation before air/water cleaning, corresponding to a decrease in feed spacer porosity of 0.86 to 0.78 (assuming homogeneous fouling) (Figure 1).

Fluctuations in air/water cleaning frequencies were explained by variations in feed water quality during the experiment with unfiltered tap water (Figure 6). Variations in feed water quality occurred in time and varied between different hours (Figure 3) and between different days (Figure 4). Variations in feed water quality were not monitored during the actual filtration experiment, but it is assumed that the feed water quality also varied between the different weeks of the filtration experiment.

A stable operation of a spiral wound membrane element was achieved with feed water containing more suspended solids compared to pre-treated feed water after normal sand filtration when applying frequent air/water cleaning. From practical experience with seawater desalination, it is known that (only) rapid sand filtration is used as a pre-treatment for spiral wound membranes (Kumar *et al.* 2006). Particulate fouling of spiral wound membrane elements, however, is still a concern in NF/RO applications where spiral wound membranes are horizontally housed in series in pressure vessels and air/water cleaning can not be applied (Van der Bruggen & Vandecasteele 2002). Based on this work it is possible to control particulate fouling in RO installations with rapid sand filtration pre-treatment by periodical air/water cleaning of the membranes.

The parallel experiments showed that after 50 days, there was no significant difference between the pressure drop values of both (REF and AWC) elements at the end of the experiment. Allowing more particles to enter a spiral wound membrane element (AWC) resulted in an increase of fouling rate compared to the reference membrane element (REF) with 1.0 μm and 10.0 μm pre-filtration. A more frequent air/water cleaning for the AWC element was needed to remove particulate fouling from the membrane element; however, this did not lead to more irreversible particulate fouling. A 5% increase in pressure drop values for both membrane elements after 50 days indicated that part of the particulate fouling could not be reversed by air/water cleaning. This phenomenon should be investigated in more detail in further research.

The membrane elements in this research were operated without permeation, which probably affected the feed spacer blocking behaviour. Drag forces during permeation will bring particles closer to the membrane surface and cause (additional) particle deposition and subsequent flux decline. Future research is therefore needed to investigate the effect of permeation on the particulate fouling rate and the control of particulate fouling using air/water cleaning. Also the ionic background of the feed water is of importance influencing electrostatic interactions between particles and the membrane surface. This can be investigated by different feed water qualities such as brackish water and seawater. Furthermore, the combination of biofouling and particulate fouling needs to be investigated further, since biofouling and particulate fouling are expected to influence one another.

CONCLUSIONS

- Periodical air/water cleaning of spiral wound membrane elements is effective in controlling particulate fouling caused by feed water containing a high load of particles;
- Increased particle loads lead to an increase in fouling rate of the spacer channel of the membrane element, which can be controlled by more frequent air/water cleanings;
- The remaining pressure drop after 50 days (effectively 36 days), probably caused by irreversible particulate fouling, was comparable for the two membrane elements fed with different loads of particles and operated with different air/water cleaning frequencies;
- Pre-treatment of NF/RO installations might be less critical when these installations are operated by periodical air/water cleaning of the membranes;
- The feed flow resistance as a result of feed spacer clogging leading to pressure drop increase is located at a small part of the total length of the membrane element.

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