Desalination and Water Treatment
www.deswater.com

doi: 10/5004/dwt.2011.2810

34 (2011) 112–116
October

The influence of particles on biofouling behavior in spiral wound membrane elements

Emile R. Cornelissen[a,*], Audrey Berteloot[b], Danny Harmsen[a], Erwin Beerendonk[a], Dick van der Kooij[a]

[a]*KWR Watercycle Research Institute, Nieuwegein, The Netherlands*
Tel. +31 306069538, +31 6 53508928 (mobile), email: emile.cornelissen@kwrwater.nl
[b]*Ecole Nationale Supeririure de Chime de Mulhouse (ENSCMu), Mulhouse, France*

Received 3 September 2010; Accepted 3 January 2011

ABSTRACT

Spiral-wound membrane (SWM) elements for nanofiltration (NF) and reverse osmosis (RO) need pre-treatment to prevent clogging of the feed spacer channel by particles and/or biomass. Usually rapid sand filtration or ultrafiltration is applied for this purpose. The effect of (the removal of) particles on biofouling processes in spiral-wound membrane (SWM) elements was investigated in this work by using a pilot set-up consisting of three parallel SWM elements. Local tap water supplemented with acetate to promote biofouling was fed to (i) a reference element, (ii) an element with one hour of daily dosing of copper sulfate to control biofouling and (iii) a third element with an additional pre-filtration to eliminate particles from the feed water of the element. The additional filter element behaved similarly to the reference element with respect to membrane transfer coefficient (MTC) decrease and pressure drop increase. Also results for membrane autopsy for biological and organic parameters were similar. The results demonstrated that the applied particle removal did not have a significant effect on the biofouling behavior in SWRO elements.

Keywords: Biofouling; Particulate fouling; Copper sulfate; Spiral wound membrane; Reverse osmosis; Membrane fouling

1. Introduction

Membrane fouling, in particular biofouling, is one of the main drawbacks in the application of spiral-wound nanofiltration (NF) and reverse osmosis (RO) membrane elements potentially leading to operational problems and deterioration of the water quality [1]. In order to control membrane fouling, two basic strategies are employed in practice, i.e., intensification of the pre-treatment before NF/RO and regular membrane cleaning of spiral wound membrane (SWM) elements. A common way to detect membrane fouling is the monitoring of operational parameters during membrane filtration specifically the increase in pressure drop which cannot differentiate particulate fouling and biofouling [2]. The aim of this study is to systematically investigate the effect of particles on biofouling.

2. Experimental part

A pilot study was carried out using three parallel 2.5-inch SWM elements (DOW Filmtec; TW30-2521) in cross-flow operation at a recovery of 10% per element, i.e., (i) a reference element, (ii) an element with one hour of daily copper sulfate dosing (1 g/l at pH 2) to inactivate biomass and (iii) a third element with an

*Corresponding author.

Presented at the AMS6/IMSTEC10, The 6th conference of the Aseanian Membrane Society in conjunction with the 7th International Membrane Science and Technology Conference, Sydney Australia, November 22–26, 2010

extra pre-filtration (using 1 µm cartridge filters) to eliminate particles entering the element (Fig. 1 and Fig. 2). These three elements were fed with pre-filtered (using 10 µm cartridge filters) tap water (without disinfectant residual) enriched with acetate (100 µg C/l) to promote biomass development while the operational parameters were investigated. Permeate production was evaluated using the mass transport coeffient (MTC) defined as the ratio of the flux (J) and the transmembrane pressure (TMP):

Fig. 1. Set-up using three parallel 2.5" SWM elements.

$$J = \frac{1}{A}\frac{\Delta V}{\Delta t} \tag{1}$$

$$\mathrm{TMP} = \frac{P_F - P_C}{2} - P_P \tag{2}$$

$$\mathrm{MTC} = \frac{J}{\mathrm{TMP}} \tag{3}$$

In which A is membrane area, ΔV is produced permeate volume, Δt is time, P_F is feed pressure, P_C is concentrate pressure and P_P is permeate pressure. Pressure drop (PD) increase is a measure for clogging of the feed spacer channel due to either biofouling or particulate fouling.

$$\mathrm{PD} = P_F - P_C \tag{4}$$

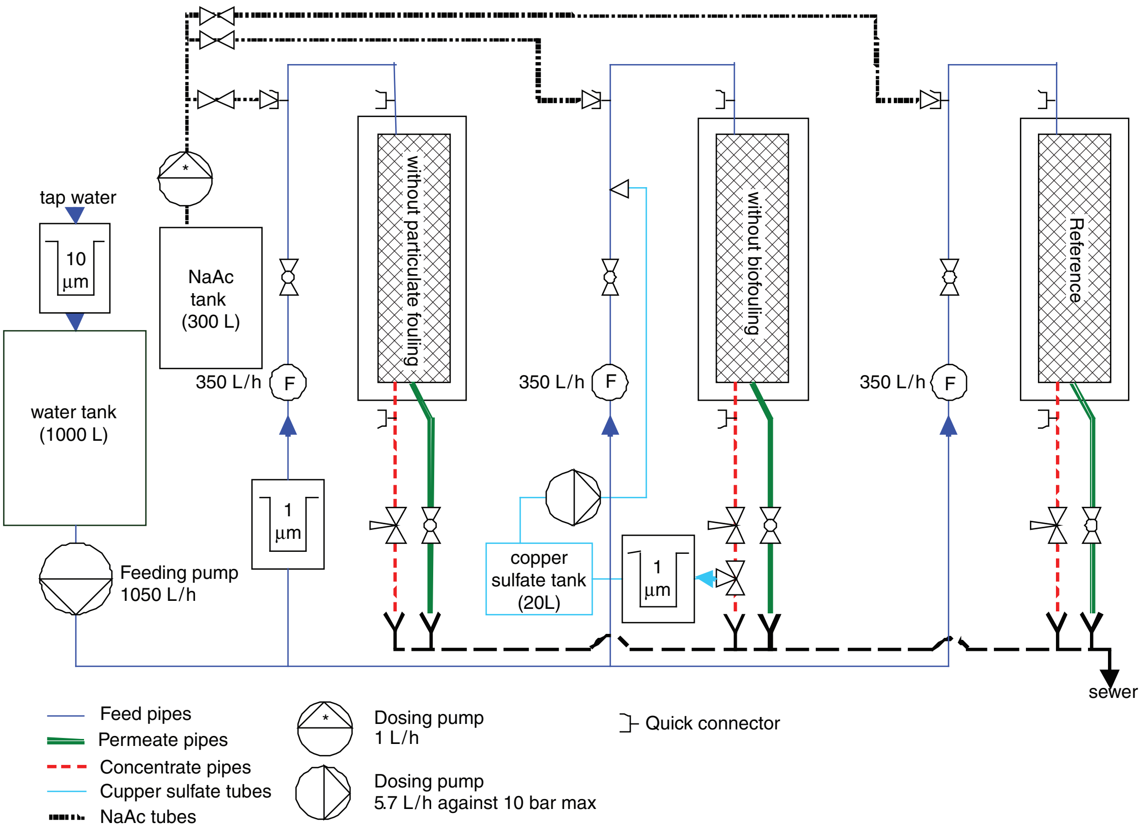

Fig. 2. Schematic depiction of the three element set-up used for the study of the influence of particles on biofouling.

The increase in pressure drop can be described as a first order process [3] by:

$$Ln\left(\frac{PD_t}{PD_0}\right) = R_f t \tag{5}$$

In which PD_t is the current pressure drop, PD_0 is the pressure drop at $t = 0$ and R_f is the fouling rate. The permeate quality is monitered by the permeate conductivity. The rejection (R) is defined as the ratio between the feed and permeate conductivity.

$$R = 1 - \frac{\sigma_P}{\sigma_F} \tag{6}$$

In which σ_F is the electrical conductivity of the feed and σ_P is the electrical conductivity of the permeate.

More details about the composition of the tap water was published by Cornelissen et al. [4]. At the end of the experiments membrane autopsies were performed to analyze the biological (ATP, TDC and carbohydrates), organic (TOC) and inorganic (ICP-MS) [5] deposition on the membrane surfaces. Average turbidity values of the feed water for the membrane element were respectively 0.25, 0.35 and 0.15 NTU for the reference, biocide and filter element.

3. Results

At day 0, the pressure drop values were respectively 17.5 mbar for element 1, 24.8 mbar for element 2 and 21.9 mbar for element 3. Calculated values of 26.5 mbar were obtained for a clean SWM element, indicating obstructions in the flow channel induced for example by the pressure vessels.

Fig. 3 show the evolution of the relative pressure drop (PD) between day 0 and day 21 (end of the run). Until day 7, the increase of pressure drop in the 3 elements was linear, the pressure drop increased from 17.5 to 37.8 mbar for element 1, from 24.8 to 27.5 mbar for element 2 and from 18.5 to 30.0 for element 3. The relative PD-increase percentages were respectively 116, 10.9 and 62%. After this, a different PD increase patterns were observed. The rate of PD increase in the 'biocide' element was negligible compared to the two other elements. The PD increase of the biocide element was linear and at the end of the run, after 21 d, the relative pressure drop reached 17.2 mbar, i.e., an increase of 69.4%.

The element 1 and 3 showed similar PD increases and were comparable. After day 7, the PD increase was exponential and reached levels of respectively 263 and 332 mbar (i.e., 1505 and 1797%). Because of this high fouling rate, the acetate dosing was stopped during the weekend due to practical considerations. Stopping the dosing of acetate induced a decrease of the relative pressure drop in the element 1 and 3 but not in the element 2. After this stage, the relative pressure drop was respectively 235, 18.2 and 255 mbar. Between day 17 and the end of the run, day 21, the acetate dosing was re-started and caused an increase of the relative pressure drop for element 1 and 3. At the end of the run, the relative pressure drop had reached 423 mbar for element 1 and 712 mbar for element 3. If we considered the part of the run between day 0 and day 14, the calculated fouling rates (R_f) were respectively 0.44, 0.16 and 0.54 ln PD_i day^{-1} [3].

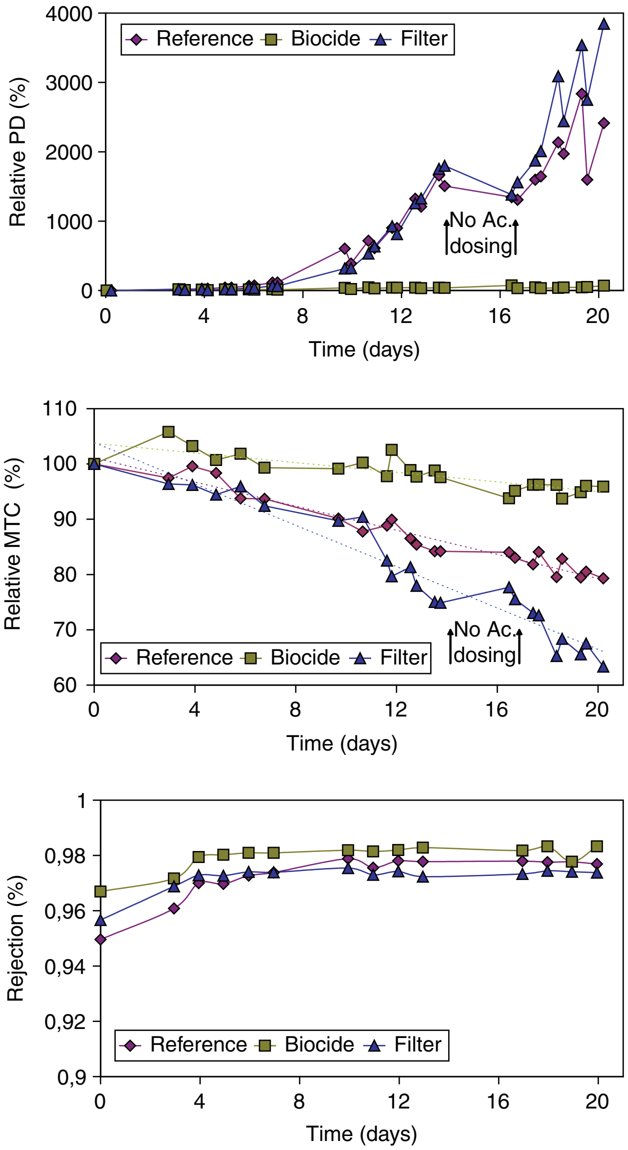

Fig. 3. Relative membrane transfer coefficient (MTC), relative pressure drop (PD) and rejection in function of time.

Fig. 3 illustrates the evolution of the Mass Transfer Coefficient (MTC) with time. The slope was lower for element 3 (–1.94) than for element 1 (–1.12) which was lower than for element 2 (–0.41). The evolution of the MTC correlates with the evolution of the relative pressure drop noticed before.

The rejection slightly increased during the biofouling experiment within the first 4 d to remain constant after (Fig. 3). This behavior did not seem to be related to the type of treatment of the membrane element, since it was similar for all three SWM elements.

Visual observation (Fig. 4 and Fig. 5), revealed that element 1 and element 3 were more fouled than element 2. This corresponds to the behavior of the operational parameters of the three elements as was described above. Visually, element 1 appeared to be more severely fouled than element 3, which is consistent with the results of the operational parameters of the membrane elements during the biofouling experiment.

Fig. 4. Visual inspection of the three membrane elements at the inlet side.

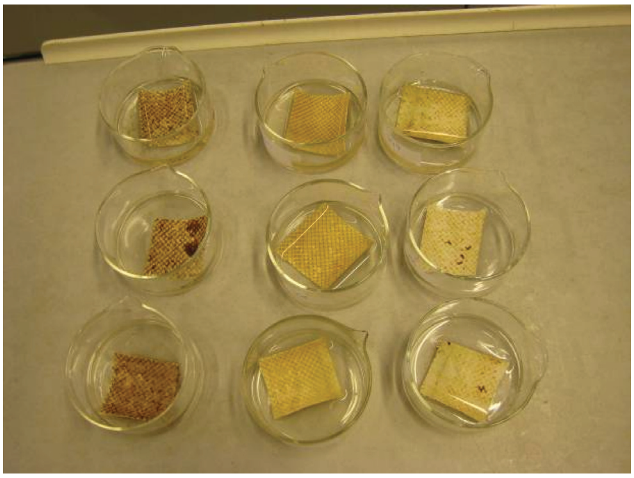

Fig. 5. Membrane coupons obtained from the three elements (left to right) at different positions of the membrane leaf from top to bottom.

Results of membrane autopsy at day 21 of the biofouling experiment indicated biomass development within the membrane elements. ATP, TDC and TOC analysis demonstrated that biomass accumulation had occurred (Fig. 6). ATP, TDC and TOC results showed lower values for element 2 than for element 1 and 3. Between element 1 and 3, ATP results and TOC and TDC results were respectively higher and lower. The ATP values followed the evolution of the pressure drop, contrary to the TDC and TOC results.

The membrane autopsy results at day 21 of the biofouling experiment revealed deposits of iron, manganese, copper, calcium and silicon, indicating particulate fouling (Fig. 7). Calcium and silicon deposit values were below the detection limit (<10 μg/cm²). The average iron deposit were low and was 43 μg/cm² for element 1; 14 μg/cm² for element 2 and 15 μg/cm² for element 3. Average manganese deposit values were

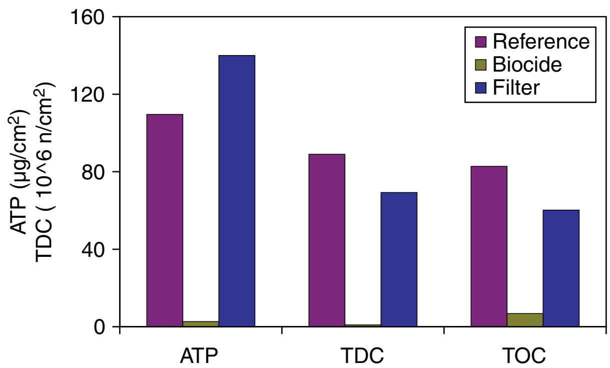

Fig. 6. Biological and organic deposits determined after membrane autopsy.

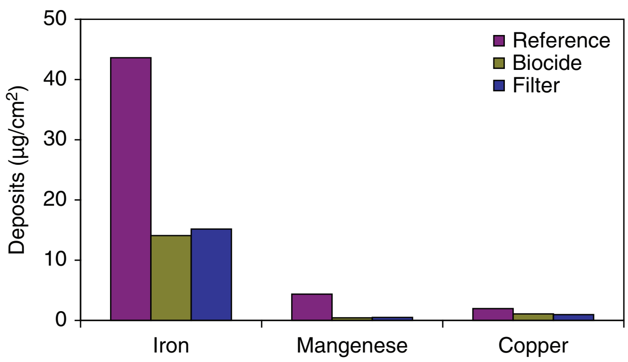

Fig. 7. Inorganic deposits determined after membrane autopsy.

low (maximum 4.4 µg/cm² for element 1) and below 0.5 µg/cm² for elements 2 and 3. The average copper deposit values were very low. The higher deposit values were found in element 1 (2.0 µg/cm²). In element 2 and 3 the deposit values were respectively 1.0 µg/cm² and 1.1 µg/cm². The overall trend for particulate fouling was that element 1 had more particulate fouling compared to element 2 and 3, the deposits of iron, manganese and copper in element 1 were respectively 9, 2 and 3 times higher than the deposits element 2 and 3. All the inorganic deposits were very low.

4. Discussion

Higher fouling rate (R_f) values corresponded to (i) a higher relative decrease in MTC, (ii) higher values of biological and organic deposits (ATP, TDC and TOC) and (iii) higher inorganic deposits (Cu, Fe, Mn). A low fouling rate (R_f) was found for the biocide treated element, while similar higher fouling rates were found for the reference and filtered element. An exception to this trend a high fouling rate for the filtered element combined with a lower deposition of Fe and Mn. This was attributed to the removal of Fe and Mn by the additional cartridge filters.

The general trend for the biofouling experiment was that the element supplied with filtered feed water was (slightly) more fouled than the reference element, on the basis of relative MTC decrease, relative pressure drop increase and biomass deposit measured as ATP. This was hypothesized by the addition of conservation liquids from the cartridge filters which acted as an additional nutrient to biomass growth. These cartridge filters were replaced every two days to avoid passage of particles of filters which were exhausted. Another general trend was the lower fouling behavior of the element which was regularly treated with a biocide (copper sulfate). Copper ions act as an enzyme toxicant which penetrates into bacterial cells very easy. Bacterial cells start to suffocate rapidly in the presence of copper [6]. The effect of biocide treatment was apparent from a lower MTC decrease, a lower pressure drop increase and lower deposition of biomass (measured as ATP and TDC), organic matter (measured as TOC) and inorganic matter (Fe, Mn and Cu).

5. Conclusions

A decrease in particles did not have a significant effect on the biofouling behavior in SWRO elements, monitored by (i) pressure drop, flux decrease and rejection and by (ii) membrane autopsy. Using daily copper sulfate dosing at low pH, on the other hand, did lead to complete control of biofouling. Temperature and nutrient dose did result in an increase of biofouling monitored by the same parameters.

The results of the experiments suggest that in practice the removal of inorganic particles (e.g., iron, manganese) in the feed of SWM RO elements will not significantly reduce biofouling behavior. The experiments were, however, carried out at a high concentration of acetate and a relative low concentration of particles.

Acknowledgements

The Dutch drinking water supply companies are acknowledged for their financial support, which made it possible to conduct this research. This work was performed in the TTIW-cooperation framework of Wetsus, centre of excellence for sustainable water technology (www.wetsus.nl). Wetsus is funded by the Dutch Ministery of Economic Affairs. The authors like to that the participants of the research theme “Schoon Water Technologie” for the fruitful discussions and their financial support. Furthermore, the authors wish to acknowledge Harry van Wegen (KWR), Sidney Meiering (KWR) for the construction of the three SWM element set-up. Wim Hijnen (KWR) for microbiological advice and Julien Ogier (previously KWR) for his assistance during the operation of the three SWM element set-up.

References

[1] J.C. Kruithof, J.C. Schippers, P.C. Kamp, H.C. Folmer and J.A.M.H. Hofman, (1998). Integrated multi-objective membrane systems for surface water treatment: prctrcatment of reverse osmosis by conventional treatment and ultrafiltration." Desalination, 117(1–3) 37–48.

[2] J.S. Vrouwenvelder, C. Hinrich, W.G.J. van der Meer, M.C.M. van Loosdrecht and J.C. Kruithof, (2009). "Pressure drop increase by biofilm accumulation in spiral wound RO and NF membrane systems: Role of substrate concentration, flow velocity, substrate load and flow direction." Biofouling, 25(6) 543–555.

[3] W.A.M. Hijnen, D. Biraud, E.R. Comelissen and D. Van Der Kooij, (2009). "Threshold concentration of easily assimilable organic carbon in feedwater for biofouling of spiral-wound membranes." Environ. Sci. Technol., 43(13) 4890–4895.

[4] E.R. Cornelissen, D. Chasseriaud, W.G. Siegers, E.F. Beeren donk and D. van der Kooij, (2010). Effect of anionic fluidized ion exchange (FLX) pre-treatment on nanofiltration (NF) membrane fouling." Water Res., 44(10) 3283–3293.

[5] F.H. Butt, F. Rahman and U. Baduruthamal. (1997). "Hollow fine fiber vs. spiral-wound reverse osmosis desalination membranes part 2: Membrane autopsy." Desalination, 109(1) 83–94.

[6] V.B. Rodin, S.K. Zhiglctsova, V.S. Kobelev, N.A. Akimova and V.P. Kholodenko, (2005). "Efficacy of Individual biocides and synergistic combinations." Int. Biodeter. Biodegrad., 55(4) 253–259.