Treatment of surface water with hydraulically cleaned reverse osmosis modules



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ABSTRACT

This thesis is part of AiRO-project which is performed in co-operation with Vitens, KWR Watercycle Research Institute, Hatenboer-Water and Evides. The commissioning organization of this thesis was Vitens. The objective of the AiRo -project is to study a method to clean vertically positioned spiral-wound reverse osmosis membranes hydraulically with a mixture of air and water. The objective of this thesis was to study if it is possible to treat surface water with hydraulically cleaned membranes and how much pretreatment is required for stable operation.

The first part of the thesis is theoretical framework about membrane techniques based on a literature survey. The second part of the thesis is research carried out with a reverse osmosis pilot installation. To study the performance of a hydraulically cleaned reverse osmosis module six shortterm experiments were performed with different cleaning frequencies and different pretreatment filter pore sizes. It was found out that treatment of surface water is possible with hydraulically cleaned reverse osmosis modules and hydraulic cleaning is an effective way to clean reverse osmosis modules. After air-water cleaning a reduction was observed in transmembrane pressure and an increase in permeability. Under a high particle load the feed channel pressure drop was reduced efficiently with hydraulic cleaning. According to this study extensive pretreatment is not needed but less pretreatment requires shorter hydraulic cleaning frequency. Also rinsing water quality was studied and it was found out that after 5 minutes of air-water cleaning the rinsing water quality did not compare to the feed water quality. Further research with long-term experiments is suggested to study the pretreatment requirements and performance of hydraulically cleaned reverse osmosis membrane treating surface water, and air-water cleaning time when surface water is used as rinsing water.

Keywords Reverse osmosis membranes, hydraulic cleaning, fouling.

Pages 60 p. + appendices 17 p.

TIIVISTELMÄ



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TIIVISTELMÄ

Tämä opinnäytetyö on osa AiRO-projektia, jonka yhteistyötahot ovat Vitens, Vitens, KWR Watercycle Research Institute, Hatenboer-Water ja Evides. Opinnäytetyön toimeksiantajana toimi Vitens. AiRO-projektin tavoitteena on tutkia pystysuoraan asennettujen spiraalikierteisten käänteisosmoosikalvojen puhdistusta hydraulisesti veden ja ilman avulla. Opinnäytetyön tavoitteena oli tutkia onko pintaveden käsittely mahdollista hydraulisesti puhdistetuilla käänteisosmoosikalvoilla ja kuinka laaja esikäsittely tarvitaan tasaiseen prosessiin.

Työn ensimmäisessä osassa on perehdytty yleisesti kalvotekniikkaan käyttäen lähteinä erilaisia julkaisuja. Työn toinen osa on tutkimus, joka suoritettiin käänteisosmoosipilottilaitteistolla. Hydraulisesti puhdistettavan käänteisosmoosimoduulin suorituskykyä arvioitiin kuudella lyhytkestoisella kokeella, joissa puhdistusväliä ja esikäsittelysuodattimien silmäkokoa muutettiin. Todettiin, että pintaveden käsittely on mahdollista hydraulisesti puhdistetuilla käänteisosmoosikalvoilla, ja että hydraulinen puhdistus on tehokas menetelmä käänteisosmoosimoduulin puhdistukseen. Ilma-vesi-puhdistuksen jälkeen havaittiin palautuminen paine-erossa kalvon läpi sekä läpäisevyyden kasvu. Suuren partikkeli kuorman aikana syöttökanavan painepudotus palautui tehokkaasti hydraulisen puhdistuksen ansiosta. Tämän tutkimuksen mukaan laajaa esikäsittelyä ei tarvita, mutta suppea esikäsittely vaatii lyhyemmän puhdistusvälin. Myös huuhteluvedenlaatua tutkittiin ja havaittiin, että viiden minuutin huuhtelun jälkeen huuhteluvedenlaatu ei vastannut syöttövedenlaatua. Pitkän aikavälin tutkimuksia ehdotetaan esikäsittely tarpeen määritykseen, pintavettä käsittelevän käänteisosmoosilaitteiston suorituskyvyn määrittämiseksi, sekä huuhteluajan määritykseen, kun huuhteluvetenä käytetään pintavettä.

Avainsanat Käänteisosmoosikalvot, hydraulinen puhdistus, tukkeutuminen.

Sivut 60 s. + liitteet 17 s.

NOMENCLATURE

AWC Abbreviation for air-water cleaning. Cleaning of membrane

with mixture of air and water.

Concentrate Waste stream with retained components. Also called reten-

tate or brine.

Feed Water to be treated which enters the membrane system. In

cross-flow filtration equal to concentrate + permeate.

Flow Volume of feed water entering the membrane system, L/h or

 m^3/h .

Flux Permeate flow through membrane, L/h m².

Fouling Clogging of membrane resulting in performance loss.

MF Abbreviation for microfiltration.

NF Abbreviation for nanofiltration.

Osmotic pressure Minimum pressure which prevents the movement of solvent

to the concentrated solution.

Permeate Water stream which passes through the membrane.

Pressure drop Pressure difference between feed channel and concentrate

channel of membrane.

Retentate Another word for concentrate.

Retention A percentage of material that is removed.

RO Abbreviation for reverse osmosis.

Specific flux Flux through membrane divided by transmembrane pressure,

L/m²*h*bar.

Transmembrane

pressure (TMP) The driving force for membrane filtration. The pressure dif-

ference between feed channel and permeate channel.

UF Abbreviation for ultrafiltration.

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1 INTRODUCTION

The application of membrane techniques in water treatment and wastewater treatment is growing. Membrane processes have become better functioning and more cost effective than they were a few years ago. Now they are an efficient and reliable way to treat water. (Liikanen 2007, 7.) Due to population growth and increasing demand for water, new methods to create clean water have to be found. Conventional sources for fresh water such as rivers, lakes, and groundwater are overused or misused. Desalination of salty water and water reuse offer one solution for water shortage. (Greenlee, Lawler, Freeman, Marrot & Moulin, 2009, 2318.) Reverse osmosis and nanofiltration are established processes for desalination of sea water and brackish water (Liikanen 2007, 7). Not only have the water resources become scarce but also the requirements for water quality have tightened. Reverse osmosis and nanofiltration are the finest membranes and they can separate substances from water until ion-level. These membranes are applied in water treatment, desalination of seawater and brackish water, in recovery of wastewater, and production of water for industry (Van der Kooij, Hijnen & Cornelissen 2009, 19).

Membrane processes have one disadvantage. Particles and biomass can accumulate on the membrane and spacer mesh and cause fouling leading to decrease of productivity or increase in pressure drop across the feed channel. Because of this extensive pretreatment is needed and membranes have to be cleaned frequently. Other filtration processes like sand filtration, microfiltration and ultrafiltration, can be cleaned hydraulically and backwashed. Reverse osmosis and nanofiltration processes have not been cleaned hydraulically. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 959, 964, 1435.) This thesis is part of AiRO -project which is performed in co-operation with Vitens (The Netherlands), KWR Watercycle Research Institute (The Netherlands), Hatenboer-Water (The Netherlands) and Evides (The Netherlands). The objective of the AiRo project is to study a method to clean vertically positioned spiral-wound reverse osmosis membranes with a mixture of air and water. The cleaning process is similar to the one that has been used for ultrafiltration membranes: membrane is cleaned by injecting air and water through the bottom section of the element.

The objective of this thesis is to study if it is possible to treat surface water with hydraulically cleaned membranes and how much pretreatment is required for stable operation if any is required at all. The first five chapters of the thesis deal with basic information of micro- and ultrafiltration and nanofiltration and reverse osmosis processes. The sixth chapter contains calculations applying to membranes. The materials, methods, results and conclusions of the research are in the chapters 7-9.

2 MEMBRANES

Membrane filtration is a separation technique of which the principle is shown in figure 1. A membrane is a thin layer of material which works as a barrier to certain particles, molecules or substances. It is semipermeable which means that some components can permeate through it and some components are retained by the membrane. As a result of a membrane filtration the feed water separates into two streams: the product stream, also called permeate, which contains the permeable components and the waste stream, called concentrate or retentate, which contains the impermeable components. The goal of membrane filtration is to produce a product stream from which the targeted compounds are removed. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 956-957.) This chapter presents the classification of membranes.

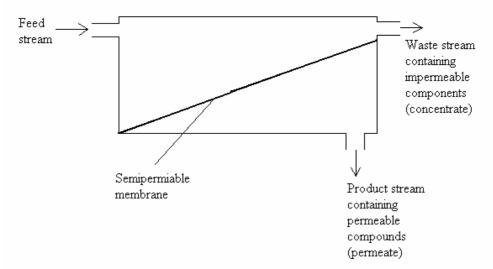


FIGURE 1 The principle of membrane process (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 956.)

2.1 Classification of membranes

The currently used membrane processes for water treatment are microfil-tration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). In all these processes the driving force is the pressure difference between the feed water channel and the permeate channel, also called as transmembrane pressure. The transmembrane pressure is effected by osmotic pressure. The basic difference between membranes is the presence and absence of pores. Membranes can be classified into two processes: micro- and ultrafiltration (MF and UF) and nanofiltration and reverse osmosis (NF and RO). (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 956-957, 964.) Figure 2 shows the classification of membranes.

The differences between MF/UF and NF/RO processes are noticeable. The basic difference is that MF/UF are porous membranes and NF/RO are considered not to have pores. MF/UF is mainly used for particle removal and NF/RO achieves much bigger variety of removed substances including dissolved solutes. Because the size of rejected particles is different the applications of MF/UF and NF/RO differ too. MF/UF is used for removal of particles and micro-organisms and NF/RO is used for seawater and brackish water desalination, softening, natural organic matter removal or removal of toxic compounds or specific contaminants. Consequently the typical source water for reverse osmosis and nanofiltration is seawater or brackish water and for micro- and ultrafiltration it is surface water but also NF/RO membranes are used for surface water. MF/UF are porous so the flux through membrane is higher and applied pressure is lower than in NF/RO. The main separation method for MF/UF is straining, in other words separation by particle size. In NF/RO separation is based on differences in solubility or diffusivity.

Membranes are used for many purposes in variety of fields and industries and the difference between membrane filtration and reverse osmosis as used in water treatment might not be correct for other industries. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 956-958, 1430.) In this thesis membranes are observed only from the water treatment sector's point of view.

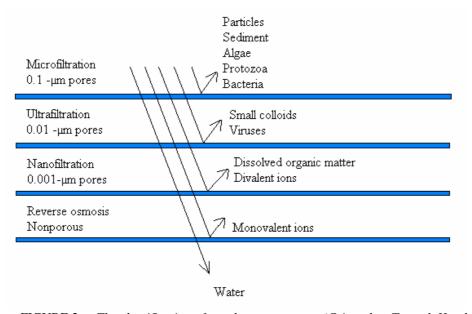


FIGURE 2 The classification of membrane processes (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 957).

3 MICROFILTRATION AND ULTRAFILTRATION

Microfiltration and ultrafiltration are used mainly for particle removal. In early days of membrane filtration the application of MF/UF were limited, consisting only of MF/UF membrane process and disinfection. This treatment was used for waters which needed only particle removal and disinfection. Now micro- and ultrafiltation is combined with other processes and it can be used for different source waters. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1011.) MF/UF is also used as pretreatment for nanofiltration and reverse osmosis processes (Liikanen 2007, 8). In this chapter the focus is on microfiltration and ultrafiltration and their structure and material, module configuration, process description, filtration mechanisms, and application.

3.1 Membrane structure and material

In water treatment membrane filtration is often performed through hollow fiber membranes. Hollow fibers are very thin tubes. Their outside diameter ranges from 0,5 to 2 mm and the wall thickness ranges from 0,07 to 0,6 mm. There are also other configurations such as flat sheet, tubular, or spiral-wound membranes. The configuration of membrane defines its packing density. The benefit of membranes is that they have a big surface area per volume of equipment so they are compact. They need less space than traditional water treatment processes. For example with hollow fibers 1 m² of footprint may contain as much as 100 m² of membrane area. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 964, 966.)

The type of material used for membrane is important. It has strong effect on membrane performance. An ideal membrane has many characteristics: it should be able to produce high flux through membrane without fouling, it should be physically durable, chemically durable and stable, non-degradable, and cheap. Many different materials are used for membranes. Most common materials used for water treatment are polypropylene (PP), polyvinylidene fluoride (PVDF), polysulfone (PS), polyethersulfone (PES), and cellulose acetate (CA). These materials are used in water treatment but different kinds of materials are used for other industries for example ceramic materials. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 980-981, 983-984.)

3.2 Module configuration

Membrane modules are made by putting thousands of hollow fibers into a module. One module can contain 8 to 700 m² of membrane area. These modules are set up either in pressure-vessel systems or submerged systems. In pressure-vessel systems membranes are put into a pressure-vessel shell and shells are arranged in skids or racks. One skid or rack can con-

tain from 2 to 300 pressure-vessel modules depending on production requirements. To generate driving force, transmembrane pressure, feed water is pumped with high pressure to the feed side of membrane while permeate side stays at atmospheric pressure. Due to the pressure difference between the feed and permeate, water is transported through the membrane. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 966-968.) Figure 3 shows a typical pressure-vessel configuration.



FIGURE 3 A pressure-vessel system mounted on rack (Shubham Inc.).

In submerged systems modules are placed in an open feed tank. Driving force is created by applying a negative pressure on the permeate side of the membrane. These systems are also called sometimes vacuum-based systems. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 966-969.) A submerged module is shown in picture 4.

Both systems have advantages. Submerged systems are in larger modules than pressure-vessel systems but they need less valves and piping. In pressure-vessels each module has to be piped separately for permeate and feed water. Submerged systems operate with individual tanks which can be operated separately so during cleaning or maintenance the whole production does not have to be stopped. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 967, 969, 971.)



FIGURE 4 Picture of submerged module for wastewater treatment (Koch membrane systems 2008).

3.3 Process description

The water that passes through membrane becomes the product water, permeate, and the retained water on the feed side is called concentrate or retentate. Since the membrane pores are too small to allow the solids in the feed to go through the membrane, the solids will accumulate on the membrane over time. This causes the transmembrane pressure to increase or the flux to decline. To prevent the pressure from becoming too high membrane plants operate with backwash and filtration cycles. During the backwash air and/or water is pumped from the permeate side to the feed side of the module. Backwash removes the accumulated material from the membrane surface. After backwashing the filtration process starts again. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 964.)

However, backwash does not remove all substances from the surface and so gradually more and more material is present on the membrane. This results in a loss in the membrane performance which is also called fouling. To remove fouling membranes are cleaned with chemicals. Cleaning is carried out by soaking the membranes into a washing solution containing surfactants, acids or bases. Membranes are cleaned regularly but the washing frequency depends on feed water quality, the process settings and membrane material. It ranges from few days to months. Even with chemical cleaning membranes have a limited life-time. Chemical cleaning and accumulation of material make the membrane fragile and it starts to degrade so finally membrane has to be replaced. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 964-966.)

3.4 Filtration mechanism

The main filtration mechanism for membrane filtration is sieving but separation occurs also through adsorption and cake formation. The principle of

sieving is that particles big enough do not pass the membrane pores and they are retained on the surface. Cake is formed on the surface when bigger particles are first strained on the surface and block the way from smaller ones that could pass the membrane pores. Adsorption occurs when small material enters the pores and is adsorbed on the pore walls. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 984.)

3.5 Applications

Micro- and ultrafiltration are mainly a particle and micro-organism removal processes. Even though for viruses the rejection is not complete, MF and UF provide good hygiene quality of water regarding bacteria, protozoa and algae. In drinking water treatment membrane filtration can replace coagulation-settling, sand filtration and disinfection processes and MF/UF can be used as pretreatment for nanofiltration and reverse osmosis. Possible source waters for MF/UF can be fresh surface water, waste water, or ground water. When no other demand for water quality is present than particulate removal, pretreatment for membrane filtration is minimal. To prevent clogging of fiber tubes and damaging fiber material, the feed water will be prefiltered with cartridge filters or microscreens. Because particles are removed by physically straining them, chemical addition like coagulation is not mandatory. This saves costs for treatment plants with respect to chemical handling because changes in feed water quality do not effect on membrane filtration as much as conventional treatment. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 975, 1020. Liikanen 2007, 8.)

Micro- and ultrafiltration are also used to remove more challenging components from the feed water for example organic material or taste and odour. For organic matter micro- and ultrafiltration by themselves are usually not sufficient. Removal of organics with micro- or ultrafiltration is limited to 10-30 % depending on membrane and source water. With tighter ultrafiltration membranes the removal of organic matter may be 60-70 %. Organic matter removal can be improved by using coagulant pretreatment or powdered activated carbon-membrane reactors. In submerged systems the coagulant or activated carbon can be added to the feed tank. In pressure-vessel systems coagulation and activated carbon can be added straight into the feed water before membrane modules. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1011-1013. Liikanen 2007, 8-9.) In post-treatment pH is adjusted and disinfectant is usually added to prevent microbial growth in the distribution system (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1021-1022. Liikanen 2007, 8).

4 REVERSE OSMOSIS

Reverse osmosis membranes are the finest membranes. They are applied in water treatment, in desalination of sea or brackish water, for waste water reclamation and for water production for industry (Van der Kooij, Hijnen & Cornelissen 2009, 19). This chapter deals with reverse osmosis including nanofiltration and reverse osmosis describing diffusion, membrane material, membrane configuration, process description, pretreatment, filtration and rejection mechanisms, and applications.

4.1 Diffusion

Reverse osmosis is based on diffusion. Osmosis is diffusion through a semi-permeable membrane. Figure 5 shows the differences between diffusion, osmosis and reverse osmosis. In figure 5a a wall separates two liquids. One liquid is pure water and the other one is a solution of a salt in water. When the wall is removed the water mass is in an unbalanced state and it will go back into an equilibrium state. Salt molecules start to move to pure water side on the right and water molecules will move to the left side. The movement of salt molecules from high concentration to low concentration is called diffusion.

If there is a semi-permeable membrane between solutions as in figure 5b, only the water molecules can move. The membrane prevents the movement of salt ions. The balance is regained by water flowing to the left side. As a result, the water level on the concentrated side rises. The flow of water from low concentration to high concentration through a semi-permeable membrane is called osmosis. In figure 5c the concentrated side is pressurized and the water molecules are forced to permeate through the membrane to the pure water side. The flow of water from a high concentration to a low concentration through semi-permeable membrane by applying an external force is called reverse osmosis.

Between two solutions with different concentrations a difference in osmotic pressure exists. The osmotic pressure is the minimum pressure which prevents the movement of water molecules to the concentrated solution. To make reverse osmosis possible the external pressure has to exceed osmotic pressure. When osmotic pressure and external pressure are equal, no water is flowing. When external pressure exceeds the osmotic pressure, the water starts to flow from left to right. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1444-1445, 1448, 1450.)

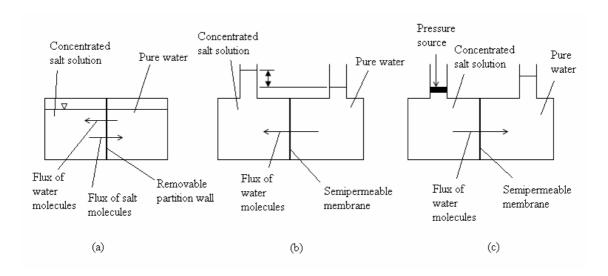


FIGURE 5 The mechanisms of diffusion, osmosis and reverse osmosis (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1444).

4.2 Membrane material

The most common materials used for RO/NF membranes are cellulose acetate (CA) and polyamide (PA). Thin-film composite membranes are made of two or more materials cast onto each other. The benefit of thin-film is that properties of membrane can be chosen independently according to materials. RO/NF membranes are extremely thin, only 0.1 to 2 μ m. The active layer allows for selective watertransport but rejects solutes that may have similar molecular size compared to water. Reverse osmosis membranes are dense which means they do not have pores. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1441-1442.) The transport mechanism is based on solution-diffusion (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1450).

4.3 Membrane configuration

Usually reverse osmosis membranes are produced in spiral-wound elements or hollow-fine-fiber elements. Hollow-fine-fiber elements are similar to membrane filtration except that reverse osmosis fibers are thinner, about the thickness of human hair. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1438, 1440.) In this thesis spiral-wound membranes were used.

4.3.1 Spiral-wound membrane element

Spiral-wound membrane elements are more complicated than hollow-fiber modules. In spiral-wound elements two membrane sheets are placed with their backs together and active layers on the outside. They are glued together from three sides to form an envelope. In the envelope a carrier spacer is placed which collects permeate and forms a permeate channel. The fourth side of envelope is glued to a perforated permeate tube in

which permeate is collected. Between envelopes are feed spacers that separate the envelopes from each other and create a feed channel and give rise to turbulent flow of the feed water. Envelopes are wound around permeate tube. During operation feed water enters the feed channel and part of the water penetrates through the membrane and spirals its way to the permeation tube. Part of the water exits on the other end of module as concentrate and continues to the next element. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1438-1440. Van der Kooij, Hijnen & Cornelissen 2009, 21.) Figure 6 shows the construction of spiral-wound membrane and figure 7 demonstrates the cross-cut of spiral-wound membrane.

Spiral-wound membranes operate with cross-flow filtration in which the feed flow is parallel to the membrane surface and permeate flow is perpendicular to membrane. Spiral-wound elements are available in different diameters. The most common diameter is about 20 cm (8'') with length of 1 m. This kind of element with 24 envelopes of 0.75 cm width and 1 m length has membrane area of 36 m². (Van der Kooij, Hijnen & Cornelissen 2009, 30.)

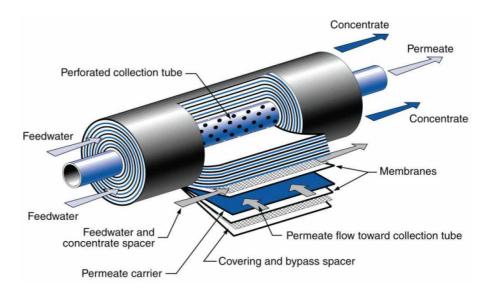


FIGURE 6 Spiral-wound reverse osmosis membrane element (Hallsby 2006).

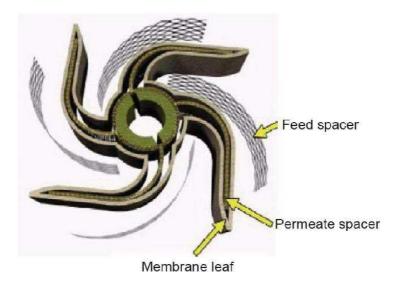


FIGURE 7 A cross-cut of spiral-wound membrane (Hallsby 2006).

4.3.2 Module configuration

Spiral-wound elements are usually arranged in pressure vessels containing six to eight element in series (Van der Kooij, Hijnen & Cornelissen 2009, 21). An example of these pressure vessels is shown in figure 8. Elements in a pressure vessel are connected together so that the permeate tubes are attached. From the first membrane element the concentrate continues to the feed channel of the next element and so on until the concentrate has flowed through all the elements and exits the vessel. When the water flows through the feed channel to the next element, it generates a pressure drop. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1440.) As fouling develops the pressure drop will increase (Van der Kooij, Hijnen & Cornelissen 2009, 27).



FIGURE 8 Reverse osmosis pressure-vessels in water treatment plant in USA (Compton engineering 2007).

4.4 Process description

Reverse osmosis process is similar to other membrane processes. Water enters the system from other end and the stream divides into permeate and concentrate streams. The driving force in reverse osmosis is the pressure difference between applied and osmotic pressure differentials as presented in formula 1 (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1454).

FORMULA 1 Reverse osmosis driving force for water flux through membrane (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1454).

$$\Delta P_{NET} = \Delta P - \Delta \pi = (P_F - P_P) - (\pi_F - \pi_P)$$
 in which,

 ΔP_{NET} = net transmembrane pressure, bars

 ΔP = pressure difference, bars

 $\Delta \pi$ = osmotic pressure difference, bars

The subscripts p and f refer to permeate and feed, respectively.

Reverse osmosis membranes are operated in pressure vessels. A group of pressure vessels operated parallel is called a stage. From a stage either the concentrate or permeate can be directed to next stage. If the concentrate is directed to the next stage it increases the recovery and it's called a multistage system. If the permeate is fed to next stage it's called a two-pass system. This will increase the permeate quality. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1435-1436.) For sea water desalination reverse osmosis plant operates with one- or two-pass systems. These days many plants are using only one-pass but the number of passes depends on production water quality standards. Some plants might even have to use four-passes. (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2331.) Figure 9 shows the flow diagram of Ashkelon reverse osmosis sea water plant in Israel which uses four-pass system. It's the largest reverse osmosis desalination plant in the world with 330000 m³ of drinking water per day. (Sauvet-Goichon 2007, 75.) The use of staging differs with brackish and sea water. Brackish water uses stage system in which the concentrate is feed water to the next stage but sea water uses permeate as feed to the next pass (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2331, 2335). The next section describes the air-water cleaning of NF/RO membranes.

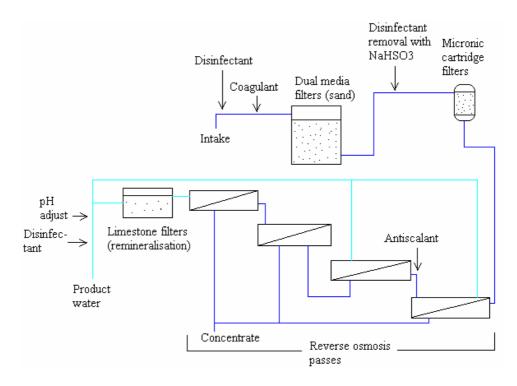


FIGURE 9 Simplified process flow of Ashkelon reverse osmosis sea water plant in Israel (Sauvet-Goichon 2007, 77).

4.4.1 Air-water cleaning

Reverse osmosis processes are continuous so there is no backwash cycle (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1435). Chemical cleaning which is performed with acids or bases is considered the only way to clean RO/NF membranes and restore flux and reduce salt passage (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2329). In this thesis backwash with mixture of air and water was studied. Using air in cleaning of membranes is quite new method and it has different names such as air sparging, air-water flushing, or air scouring. Application of air in membrane processes started in the 1990s. Methods of using air in the process can be divided into two groups, prevention of fouling by using air continuously during filtration, and cleaning with air between filtration cycles. (Cornelissen, Vrouwenvelder, Heijman, Viallefont, van der Kooij & Wessels 2007, 95.) In this thesis the application of air was used as a cleaning method.

Air-water cleaning or using air during filtration is widely research with micro- and ultrafiltration membranes. Use of air is reported to be efficient in removing particulate fouling and enhancing flux. (Cabassud, Laborie & Laine 1997, 97. Li, Ghosh, Bellara, Cui & Pepper 1998, 80-81.) Only few studies were performed on air-water cleaning in reverse osmosis processes (Cornelissen, Vrouwenvelder, Heijman, Viallefont, van der Kooij & Wessels 2007. Cornelissen, Harmsen, Beerendonk, Wessels, & Van der Kooij 2009. Cornelissen, Rebour, Van der Kooij & Wessels 2009.).

4.5 Pretreatment

Pretreatment is required basically in all reverse osmosis systems (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1436). The goal for pretreatment is to prevent fouling on the membrane (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2329). The next sections describe methods for pretreatment and post-treatment.

4.5.1 Conventional pretreatment

Conventional pretreatment for sea and brackish water usually consists of chemical addition, disinfection, media filtration and cartridge filtration. The extent of pretreatment depends on source water. Chemical addition depends on the source water quality. Coagulant dosing is added if water will be filtered with media filtration like sand filtration. Disinfection is used to prevent biofouling. Not all the membrane materials tolerate disinfection chemicals so the chemical has to be removed before membrane for example with activated carbon or sodium bisulfite. Scaling control is before or after cartridge filtration. Scaling control consists of pH adjustment and/or antiscalant dosing. This is done to prevent precipitation of salts on the membrane. During operation the concentrate side becomes more concentrated with salt and the salt concentration can become higher than the salt's solubility. Then the salts may start to precipitate and damage the membrane. With pH adjustment the goal is to change the solubility of salts and antiscalants prevent the precipitate formation or crystallization. pH adjustment is done with acid at the same time with coagulant dosing. Cartridge filtration is the last pretreatment step and it is applied to remove the larger particles that passed media filtration. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1436-1437. Van der Kooij, Hijnen & Cornelissen 2009, 22. Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2329-2330.) The flow diagram of Ashkelon reverse osmosis sea water plant in figure 9 shows a conventional pretreatment process.

When reverse osmosis is used for surface water, biofiltration (sand) with coagulation is commonly used as part of pretreatment. Variable combinations of ozonation, sedimentation and activated carbon filtration with biofiltration are used in pretreatment process. (Van der Kooij, Hijnen & Cornelissen 2009, 46-47.)

4.5.2 Membrane pretreatment

A new method for pretreatment is the use of micro- or ultrafiltration. They give defined protection against particles. Membrane filtration has the advantage of non-chemical treatment and it can replace the granular filtration of conventional pretreatment which needs chemical dosing. Micro- and ultrafiltration membranes have also backwash possibility. They are more flexible to changes in feed water quality than conventional pretreatment methods. Because of the good rejection of micro- and ultrafiltration, reverse osmosis membranes age slower. (Greenlee, Lawler, Freeman, Mar-

rot & Moulin 2009, 2330-2331.) Recent studies show that ultrafiltration has become the most tested and studied membrane filtration pretreatment (Van Hoof, Minnery, Mack 2001, 164-166. Halper, McArdle & Antrim 2005). The disadvantage of pretreatment with membrane filtration is fouling of the pretreatment membranes themselves. Fouling can be reduced by the use of inline coagulation. Coagulant can not be applied at the same time with antiscaling agent. Coagulant and antiscaling chemical form together a complex which is a very difficult foulant. (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2330-2331.) Ultrafiltration has yet another disadvantage. According to studies, ultrafiltration results in a very good rejection of particles but it does not remove material that causes biofouling. (Van der Kooij, Hijnen & Cornelissen 2009, 46, 49. Vrouwenvelder, van Paassen, van Agtmaal, van Loosdrecht, Kruithof 2009, 41-42).

4.5.3 Post-treatment

Post-treatment for product water is simple. It consists of pH adjustment, possible removal of gases, and remineralization. Depending on salt concentration the permeate might be blended with another water to increase or decrease the salinity. Hardness will be increased to achieve the typical taste of drinking water. Alkalinity and pH are also adjusted to prevent corrosion. Dissolved gases permeate well through membrane and if the source water contains hydrogen sulphide it will be stripped. Also disinfectant is added to prevent microbial growth in distribution network. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1437-1438. Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2336-2337.)

4.6 Filtration and rejection mechanisms

With nonporous reverse osmosis membranes the flux through membrane is different than in MF/UF. First the water is adsorbed on the membrane surface, then it diffuses through membrane and desorbs on the permeate side. This model is called solution-diffusion model. With nanofiltration membranes the flux is a combination of diffusion and the same pore flow-method as in micro-, and ultrafiltration. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1450-1451. Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2322-2323.) Although the newest studies present, that flux through nanofiltration membranes is more controlled by diffusion (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2322).

Rejection can occur in different ways. Most common mechanisms are electrostatic rejection at the membrane surface, solubility and diffusivity through membrane or straining. Nanofiltration and reverse osmosis membranes are often negatively charged so negatively charged ions may be rejected based on electrostatic repulsion and positively charged ions may be rejected to sustain electroneutrality in the feed and permeate side. Polar and hydrogen-bondable functional groups in membrane increase the solubility of polar compounds, such as water, and develop larger flux of water through the membrane. Large molecules have lower diffusivity through

membrane or they are not able to pass the membrane at all. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1453.)

4.7 Applications

Reverse osmosis processes are used for many different applications because of their tight membrane and ability to reject various compounds. Next sections deal with applications of reverse osmosis and nanofiltration membranes including desalination, natural organic material removal, specific contaminant removal and organic contaminant removal.

4.7.1 Desalination

The main application for reverse osmosis is desalination of sea water or removal of dissolved solids. Total dissolved solids means the sum of ions in the solution (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 18, 77). Different types of membranes are used for different product water. To produce demineralised water high rejection type reverse osmosis membrane is used. If the product water is normal drinking water then more loose type of reverse osmosis or nanofiltration membrane is used. (Van der Kooij, Hijnen & Cornelissen 2009, 24.) Even though other techniques for desalination exist reverse osmosis seems to be the main technique in the future (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2322).

4.7.2 Natural organic material

Other application is removal of natural organic material (NOM). When NOM gets contact with disinfectants, harmful disinfection by-products (DBP) are formed. Generally NOM is easier to remove from water than by-products. Colour removal has been very effective with nanofiltration and colour is usually caused by organic material. This makes nanofiltration effective process to control disinfection by-products. Nanofiltration has been also used for softening in USA for three decades. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1433. Liikanen 2007, 8. Van der Kooij, Hijnen & Cornelissen 2009, 24.)

4.7.3 Specific contaminants

Reverse osmosis can also remove specific contaminants. One common application is nitrate removal from ground water. With reverse osmosis membrane the rejection can be between 73-93 %. With nanofiltration the rejection is not so good so they are not usually used for this application. Reverse osmosis is also used for fluoride and arsenic removal. (Van der Kooij, Hijnen & Cornelissen 2009, 24.) In Finland at the water treatment plant in Laitila reverse osmosis is used for fluoride removal from ground water. Reverse osmosis reduces the fluoride concentration by more than 95 %. At the same time it reduces the aluminum concentration to approv-

able level. Because the rejection of fluoride is so high, the targeted level is reached easily by mixing the reverse osmosis water with pre-treated water. (Liikanen 2007, 8.)

4.7.4 Organic contaminants

Reverse osmosis is able to remove organic contaminants such as pesticides, pharmaceutically active compounds and personal care products. Retention of these contaminants depends on solute properties, membrane properties and operating conditions. Generally these compounds are removed very well with both nanofiltration and reverse osmosis. (Van der Kooij, Hijnen & Cornelissen 2009, 25.) However, use of reverse osmosis to remove specific contaminants is not very cost effective because other cheaper techniques exist and disposal of concentrate might be challenging (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1433).

5 MEMBRANE FOULING

Fouling is the most important issue for membrane applications. It causes flux decline and shortens the membrane life. Fouling can be categorized by different characters: mechanism, reversibility and foulants. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 993, 994.) Surface fouling and fouling in pores are the two fouling mechanisms that are commonly detected. Fouling causes water flux decline, increase of transmembrane pressure drop and feed channel pressure drop, and salt passage through NF/RO membranes. (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2327). Permanent loss of performance after cleaning is called irreversible fouling. Reversible fouling is fouling that could be removed by backwashing or cleaning. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 994.) The next sections describe four common types of fouling: particulate fouling, organic fouling, scaling, and biofouling.

5.1 Particulate fouling

Source water for reverse osmosis is often sea water or brackish water and compared to fresh surface waters sea water has less particle content. However sea water treatment plants that treat water from open water intake are typically fouled by particles and organic matter. Particle fouling is caused by sand, sludge, silicates, salt precipitates and remains of microorganisms. Particle fouling causes cake formation on the membrane and plugging in the feed channel or piping. From micro- and ultrafiltration membranes particle fouling is easy to remove with backwash but NF/RO processes do not have a backwash cycle. Big part of particles exits the membrane in the concentrate because of turbulence flow in the membrane elements. If the load of particles is too big or there is not enough turbulence, particles will start accumulating which results in salt passage through NF/RO membrane, pressure drop over membrane elements and a decrease in water flux. Ultrafiltration and microfiltration as a pre-

treatment for reverse osmosis give excellent particle removal. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1468. Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2327-2328. Van der Kooij, Hijnen & Cornelissen 2009, 26.)

5.2 Organic fouling

Natural organic matter is a term often used when describing organic material. Natural organic matter (NOM) is a term used to characterize a complex group of organic chemicals originating from biological activity in water bodies such as metabolic activity of algae or micro-organisms. It can also be washed from land into water. It is composed of biological matter, reaction products between NOM molecules or reaction products between NOM molecules and inorganic components. This makes it very complex mixture of different chemical features. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 81-82.) NOM consists of particles, biological material and dissolved organic compounds. It can be partly removed by backwashing from MF/UF membranes (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 998, 1000-1001). Also coagulation and activated carbon treatment as part of MF/UF can help to reduce organic content and thus decrease fouling.

In NF/RO processes NOM precipitates and adsorbs on the membrane surface and causes decrease in water flux (Van der Kooij, Hijnen & Cornelissen 2009, 27). Organic fouling can be reduced by pretreatment with biofiltration or very tight ultrafiltration membranes are also able to reduce the organic load (Liikanen 2007, 8. Mosqueda-Jimenez, Huck 2009, 65).

5.3 Scaling

Scaling is fouling by inorganic substances. Scaling occurs when the concentration of salts exceeds the solubility and they start to precipitate. They crystallize on the membrane surface. Micro- and ultrafiltration membranes allow salts to permeate through the membrane so the salt concentration will not rise on the membrane surface. Scaling is mainly a problem of NF/RO membrane processes. Scaling on membrane is shown in figure 10. In sea and brackish water there are lots of inorganic ions. The main ions are calcium, magnesium and barium. Concentration polarization is a phenomenon which occurs when dissolved ions accumulate in a thin layer of the feed water. It is the ratio of salt concentration at the membrane surface and in the bulk solution. Concentration polarization decreases water flux through membrane and increases salt transport through membrane. It leads also to scaling. Water flux declines because higher concentration on the membrane surface causes higher osmotic pressure which leads to the overall pressure difference decrease. Salt transport increases due to increase in concentration and decrease in water flux. Scaling is prevented by using antiscalants which increase the threshold of concentration when the ions start to crystallize and disturb the formation of crystal structure. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1462, 1470. Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2323, 2327-2328. Van der Kooij, Hijnen & Cornelissen 2009, 26.)



FIGURE 10 Scaling of spiral-wound membrane (WaterForum Online 2005).

5.4 Biofouling

Biofouling is accumulation and attachment of micro-organisms on membrane surface where they form a biofilm. Biofouling is troublesome because it can not be controlled by reducing microbes in the feed water. If there is any microbe left, it will multiply as long as nutrients are available. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1001. Flemming 1997, 382.) Part of NOM can be used by micro-organisms as nutrient. Assimilable organic carbon (AOC) is ready to use energy source for microbes and if it's available in big concentrations that means that microbes have a lot of potential to grow. So the biofouling potential can be derived from nutrient concentration in the system. (Van der Kooij, Hijnen & Cornelissen 2009, 27, 45-46.)

A biofilm is formed always when micro-organisms have a surface to attach. Micro-organisms can attach to the membrane and they are difficult to remove during backwash. On the membrane they start to excrete gel-like extracellular material that protects them from cleaning and results in additional fouling. The possibilities to prevent biofouling are disinfection, biocide dosing and nutrient reduction by biofiltration. Disinfection kills micro-organisms but if the dead biomass is not removed a new biofilm will grow on it fast using the biodegradable compounds from the dead mass. According to studies limiting nutrient concentration is an effective way to control biofouling (Griebe, Flemming 1998, 156. Hu, Song, Ong, Phua, Ng 2005, 128, 132). Biofilm forms in phases. It occurs when the biofilm growth exceeds the threshold of interference. Because it's impossible to kill all the micro-organisms from the system, the other option is to live with biofilm formation as long as it does not lead to biofouling. The

threshold of interference is the limit below which the biofilm does not interfere with membrane performance. (Flemming 1997, 383, 387-388. Griebe, Flemming 1998, 156. Hu, Song, Ong, Phua, Ng 2005, 128, 132. Van der Kooij, Hijnen & Cornelissen 2009, 27.)

Biofouling results mainly in pressure drop increase but it can also decrease the permeate flux and salt rejection on NF/RO membrane. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 84. Flemming 1997, 383, 387-388. Hu, Song, Ong, Phua, Ng 2005, 128, 132. Van der Kooij, Hijnen & Cornelissen 2009, 27.) Figure 11 shows biofouling on the surface of autopsied reverse osmosis membrane. Even though biofouling has been noticed and studied a few decades, it still remains a difficult problem that is hard to control (Van der Kooij, Hijnen & Cornelissen 2009, 29).



FIGURE 11 Biofouling on spiral-wound reverse osmosis membrane (WaterForum Online 2008).

6 CALCULATIONS

This chapter presents some of the basic equations for calculating rejection, recovery and water flux, solute flux and silt density index for micro- and ultrafiltration and nanofiltration and reverse osmosis processes.

6.1 Rejection

The extent to which material that is retained by a membrane is called rejection. Rejection can be calculated for specific components or using an overall property, such as turbidity. For example salt rejection is one of the performance indicators for reverse osmosis. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 984. Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2323-2324.) Formula 2 shows the equation for rejection. The equation is used in all types of membrane filtration.

FORMULA 2 Equation for calculating rejection for membrane filtration (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 984, 1443).

$$R = 1 - \frac{C_P}{C_F}$$

in which,

R = rejection, dimensionless

 C_P = permeate concentration, mole/L or mg/L

 C_F = feed concentration, mole/L or mg/L

6.2 Recovery

Recovery is the fraction of the feed water that becomes permeate. The equation shown in formula 3 applies only for nanofiltration/reverse osmosis membranes. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1455.)

FORMULA 3 Equation for recovery for reverse osmosis and nanofiltration membranes (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1455).

$$R_{w} = \frac{Q_{P}}{Q_{F}}$$

in which,

 R_{w} = recovery, dimensionless

 $Q_P = \text{permeate flow, m}^3/\text{s}$

 $Q_F = \text{feed flow, m}^3/\text{s}$

For MF/UF recovery is typically very high, 95-98 % and it is the ratio of net water production to gross water production as shown in formula 4. The equation is only valid for dead-end filtration (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1011.)

FORMULA 4 Equation for recovery for micro- and ultrafiltration (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1011).

$$R_{_W} = \frac{V_{_F} - V_{_{BW}}}{V_{_E}}$$

in which,

 R_{w} = recovery, dimensionless

 V_F = volume of water fed to the membrane, m³

 V_{BW} = volume of water used during backwash, m³

6.3 Water flux

Water flux calculation differs for the different types of membrane filtration because with nanofiltration and reverse osmosis the osmotic pressure affects the flux. The next sections show water flux calculations for NF/RO and MF/UF.

6.3.1 Water flux through reverse osmosis membranes

The water flux through reverse osmosis or nanofiltration membrane is driving force times mass transfer coefficient. The water flux through reverse osmosis membrane is described in formula 5.

FORMULA 5 Equation for water flux through reverse osmosis and nanofiltration membranes (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1454).

 $J_{\scriptscriptstyle W} = k_{\scriptscriptstyle W} (\Delta P - \Delta \pi)$

in which.

 J_W = volumetric flux of water, L/m²*h

 $k_w = \text{mass transfer coefficient for water flux (MTC), L/m}^2 *h*bar$

 ΔP = transmembrane pressure difference between feed and permeate, bars

 $\Delta \pi$ = osmotic pressure difference between feed and permeate, bars

Mass transfer coefficients are given by the membrane manufacturers or calculated from clean water flux experiments. Osmotic pressure is calculated by the van't Hoff equation which is similar to ideal gas law (pV=nRT). It's derived from ideally diluted solution which is not usually the case in reverse osmosis systems. So a coefficient has to be added to account for diluteness and behaviour of solutions. Osmotic pressure depends on the concentration of water in the system and dissociation of solutes in the water. When these factors are taken into account and included in the van't Hoff equation, we can calculate the osmotic pressure equation, shown in formula 6. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1446, 1455.)

FORMULA 6 Equation of osmotic pressure (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1446).

 $\pi = i\phi CRT$

in which,

 π = osmotic pressure

i = number of ions when solute is dissociated

 ϕ = osmotic coefficient, unitless

C = concentration of all solutes, moles/L

R = universal gas constant, 0,083145 L * bar/moles * K

T = temperature, K

To be able to evaluate the performance loss caused by fouling or membrane aging, the flux has to be corrected for temperature. Temperature effects water viscosity and correction factors for temperature are given by membrane manufacturers. If these factors are not available the flux can be corrected with factor shown in formula 7.

FORMULA 7 Equation for temperature correction factor (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1456).

 $TCF = (1,03)^{T-25}$ in which, TCF= temperature correction factor T= feed water temperature, $^{\circ}C$

6.3.2 Water flux through micro- and ultrafiltration membrane

The water flux equation for micro- and ultrafiltration membranes is shown in formula 8, which is derived from Darcy's law.

FORMULA 8 Equation for water flux in micro- and ultrafiltration (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 990).

$$J = \frac{\Delta P}{\mu \kappa_{\scriptscriptstyle M}}$$

in which,

J = water flux through membrane, L/m² *h or m/s

 ΔP = differential pressure between feed and permeate side (transmembrane pressure), bar or Pa

 μ = viscosity of water, kg/m * s

 κ_{M} = membrane resistant coefficient, 1/m

In some climates the temperature difference between winter and summer can be more $20\,^{\circ}\text{C}$ which means that at summer flux can be much higher than at winter. Temperature variations can be adjusted by calculating equivalent flux at standard temperature. Formula 9 is often used for that.

FORMULA 9 Equation for correcting temperature for water flux in micro- and ultrafiltration (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 992).

$$J_S = J_M (1.03)^{T_S - T_M}$$

in which,

 J_s = water flux in standard temperature (20 °C), L/m² *h

 J_M = water flux at measured temperature, L/m² *h

 T_S = measured temperature, ${}^{\circ}C$

 T_M = standard temperature, ${}^{\circ}C$

With standard temperature 20°C formula 9 is accurate within 5 % for waters with temperature range 1-28°C. Because flux is also dependent on pressure the normalized pressure is calculated as shown in formula 10. It's called specific flux. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 992.)

FORMULA 10 Equation for specific flux for micro- and ultrafiltration (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 992).

$$J_{SP,20^{c}C} = \frac{J_{S}}{\Delta P}$$

in which.

 $J_{SP,20^{\circ}C}$ = specific flux at 20°C, L/m² *h*bar

 $J_{\rm S}$ = water flux at standard temperature (20 °C), L/m² *h

 ΔP = differential pressure between feed and permeate side (transmembrane pressure), bar

The effect of fouling on water flux can be defined by calculating the percent loss of specific flux.

6.4 Solute flux through membrane

The solute flux through membrane can also be presented as performance measure. The solute flow is only calculated for reverse osmosis and nanofiltration membranes. (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2323.) The equation is shown in formula 11.

FORMULA 11 Equation for solute flux through reverse osmosis or nanofiltration membranes (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1454).

$$J_S = k_S(\Delta C) = k_S(C_{feed} - C_{permeate})$$

in which,

 $J_s = \text{flux of solute, mg/m}^2 \text{ h}$

 k_s = mass transfer coefficient for solute flux, L/m² *h or m/h

 $C_{\it feed}$ = concentration in the feed solution, mole/L or mg/L

 $C_{\it permeate}$ = concentration in the permeate solution, mole/L or mg/L

The mass transfer coefficient is given by manufacturers (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1455).

6.5 Silt density index

The fouling potential of water can be described with SDI. Silt density index is a timed filtration test performed with 0,45 µm filter in constant 2,07 bar pressure. The standard time of the test is 15 minutes and the filtration continues all this time. First the duration time of collecting 500 ml of permeate is measured. After collecting 500 ml of permeate the filtration continues without measuring the volume. When 15 minutes have passed from the beginning of the test another 500 ml sample is filtered and the time to filter that sample is measured. From these filtering times the SDI is calculated as shown in formula 12. (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1469. Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2327.)

FORMULA 12 Equation for calculating SDI (Crittenden, Trussel, Hand, Howe & Tchobanoglous 2005, 1469).

$$SDI = \frac{100\% \cdot (1 - \frac{t_1}{t_2})}{t}$$

in which.

 t_1 = time required to filter the first 500 ml sample, min

 t_2 = time required to filter the final sample after 15 min filtration, min t = the time from the beginning of the test to the beginning of final sample filtration, (15) min

An SDI value of 3 or less is preferred for feed water for reverse osmosis. Values of 4-5 are also tolerated and they are usually achieved with conventional pretreatment. (Greenlee, Lawler, Freeman, Marrot & Moulin 2009, 2327.)

7 MATERIALS AND METHODS

This thesis discusses the treatment of surface water with reverse osmosis membranes that were cleaned with a mixture of air and water, and the minimum pretreatment that is required to achieve stable operation. The research objective was to find out what the optimum air-water cleaning frequency is for the installation and how much pretreatment is required to successfully operate the pilot. This chapter describes materials, the pilot set-up, methods and measured parameters.

7.1 Materials

This thesis was part of AiRO-project which is performed by Vitens (Leeuwarden, the Netherlands), KWR Watercycle Research Institute (Nieuwegein, the Netherlands), Evides (Rotterdam, the Netherlands), and Hatenboer-Water (Schiedam, the Netherlands). The locations of cooperation partners are shown in appendix 7. The research was performed with pilot installation and surface water from canal Potmarge. The pilot was provided and built by KWR. The pilot was equipped with a Hydranautics reverse osmosis element type ESPA2-4040. The technical data of the module is given in table 1 and a picture of the membrane is shown in figure 12.

TABLE 1 Technical data of the reverse osmosis membrane (Membranes.com. 2009).

Membrane element	Hydranautics ESPA2-4040
Membrane diameter	10 cm
Membrane length	1 m
Material	Thin-film composite polyamide
Membrane area	$7,90 \text{ m}^2$
Average salt rejection	99,6 %
Minimum salt rejection	99,4 %
Permeate flow	3000 L/h
Maximum feed flow	3600 L/h
Maximum operating temperature	45 °C
Maximum applied pressure	41,6 bar
Feed water pH range	3,0-10,0



FIGURE 12 Reverse osmosis membrane type used in the experiments.

7.2 The pilot set-up

The installation was situated in Leeuwarden in Greunsweg in one of Vitens' water distribution stations. The feed water was surface water pumped from Potmarge canal with a submerged pump. The pump was in a metal basket which sieved the biggest particles (leaves, fishes, and rocks) from the water. Photos of canal Potmarge and the intake point are in appendix 1. The water was pumped to the feed water tank of 1 m³ and was pretreated by a cartridge filter with a pore size of $100 \, \mu m$. The feed tank was covered with black plastic to prevent the algae blooming and equipped

with a level switch ASV-Stübbe NIS 1 to prevent the tank from overflowing. From the feed tank the water flew gravitationally to the installation. The installation was equipped with a low pressure pump (DP pumps DPVE 2-20) supplying the feed water to three cartridge filters placed in parallel. Only two of the cartridge filters were in use at the same time. Third one was only used when the filters had to be changed. After the cartridge filters there was a high pressure pump (Grundfos CRNE 3-23) that provided the flow to the module.

The installation was operated with 1500 L/h feed flow at 10 % recovery. The feed flow was kept constant at 1500 L/h with 150 L/h permeate flow resulting in a recovery of 10 %. Because there was no feed water flow meter in the installation the concentrate flow and permeate flow were adjusted to 1350 L/h and 150 L/h respectively. The membrane was vertically positioned and the flow of feed water was from the top of the module to the bottom. The permeate was collected from the top of the membrane. The concentrate exited from the bottom of the membrane. There were three sample points in the installation: after the cartridge filters, in the permeate line and in the concentrate line. There were two flow meters and both of them were manually read. The concentrate flow meter had a range of 300-3000 L/h and the permeate flow meter had a range of 25-250 L/h.

There were four pressure measuring points: before and after cartridge filters, after the second pump in feed line, and after membrane in concentrate line. Pressure drop between feed channel and concentrate channel was measured as an indicator of fouling. In all experiments this pressure drop was measured manually every 15 minutes by a single pressure sensor and an array of valves. The result was read from the display of pressure transmitter (Endress-Hauser Cerabar S, PMC71).

A compressor supplied the air for the air-water cleaning. The air pressure was measured with a manometer after the compressor. The air flow was measured with a flow meter after the compressor. When air-water cleaning was applied the flow of water was stopped and the flow direction changed to from bottom to top. Air was then added into the water stream. The air-water cleaning was applied for 5 minutes with air pressure of 6 bars and air flow of 3000 L/h. The water flow during the cleaning was 1500 L/h so the air/water ratio was 2:1. When stopping air-water cleaning the air flow was turned off but water was left to flow through membrane for 3 minutes. This was done to remove the remaining air from the system. A schematic picture of installation is shown in figure 13 and a picture of the pilot is shown in figure 14.

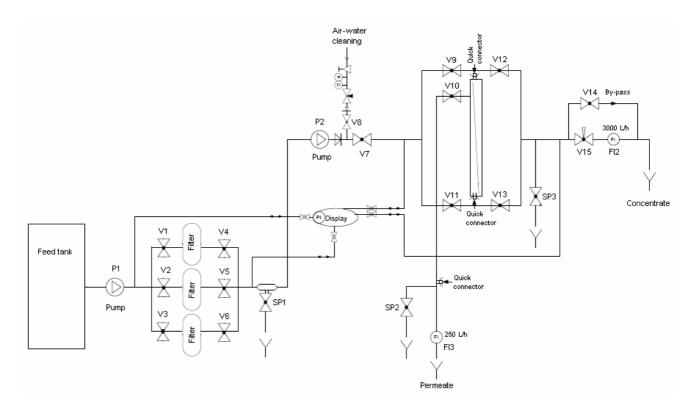


FIGURE 13 A schematic picture of the pilot installation.

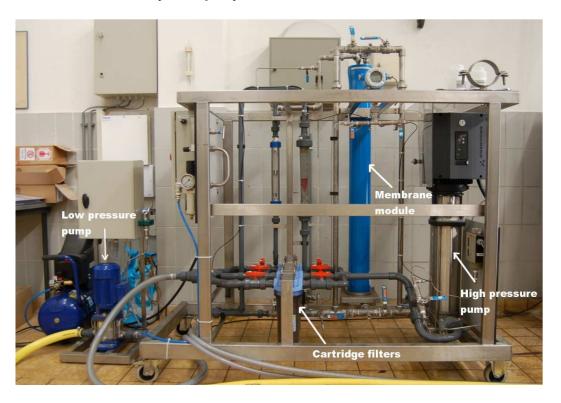


FIGURE 14 The pilot installation for the experiments.

7.3 Methods

Four experiments were conducted to determine the required air-water frequency for stable operation and to study the pretreatment requirements. In addition two reference experiments were performed. The next sections describe methods for air-water cleaning experiments and reference experiments.

7.3.1 Air-water cleaning frequency experiments

The pilot had not been used for surface water before so in the first four experiments the cleaning frequency was studied as well as the pretreatment requirements. Conditions during the experiments are given in table 2. In the first experiment air-water cleaning was applied after every seven hours of operation. The size of the cartridge filters before the module were 5 μm . Experiment 2 was also performed with air-water cleaning frequency of seven hours. The cartridge filter size was 20 μm . Experiment 3 was performed without cartridge filters before the module and with a cleaning frequency of 14,5 hours and experiment 4 also without cartridge filters with a cleaning frequency of 24 hours.

Temperature, conductivity, turbidity and particle amounts were measured every 30 minutes. Experiments consisted of four runs. Each run lasted 7,5 hours, since the pilot was running only during working hours so the total running time for each experiment was 30 hours.

TABLE 2	Conditions o	f air-water	cleaning	frequency	erneriments
	Conamons o	ı air-waier	Cleaning	meguency	experiments.

Experiment	Run	Run time	Filter size	Feed flow	Recovery	AWC frequency
1	1	7,5 h	5 μm	1500 L/h	10 %	7 h
	2	7,5 h	5 μm	1500 L/h	10 %	7 h
	3	7,5 h	5 μm	1500 L/h	10 %	7 h
	4	7,5 h	5 μm	1500 L/h	10 %	7 h
2	1	7,5 h	20 µm	1500 L/h	10 %	7 h
	2	7,5 h	20 µm	1500 L/h	10 %	7 h
	3	7,5 h	20 µm	1500 L/h	10 %	7 h
	4	7,5 h	20 µm	1500 L/h	10 %	7 h
3	1	7,5 h	No filters	1500 L/h	10 %	
	2	7,5 h	No filters	1500 L/h	10 %	14,5 h
	3	7,5 h	No filters	1500 L/h	10 %	
	4	7,5 h	No filters	1500 L/h	10 %	14,5 h
4	1	7,5 h	No filters	1500 L/h	10 %	
	2	7,5 h	No filters	1500 L/h	10 %	
	3	7,5 h	No filters	1500 L/h	10 %	
	4	7,5 h	No filters	1500 L/h	10 %	24 h

7.3.2 Reference experiments

Reference experiments were performed with a membrane of the same type as the one used in experiments 1-4. No air-water cleaning was applied. The objective was to see how fast a new membrane fouls without air-water cleaning. The conditions during reference experiments are in table 3. Two reference experiments were conducted. The first experiment was carried out with a cartridge filter of size 5 μ m and the second with a cartridge filter size of 20 μ m.

TABLE 3 Conditions of reference experiments.

Experiment	Run	Run time	Filter size	Feed flow	Recovery	AWC frequency
1	1	7,5 h	5 µm	1500 L/h	10 %	no AWC
	2	7,5 h	5 μm	1500 L/h	10 %	no AWC
	3	7,5 h	5 μm	1500 L/h	10 %	no AWC
	4	7,5 h	5 μm	1500 L/h	10 %	no AWC
2	1	7,5 h	20 µm	1500 L/h	10 %	no AWC
	2	7,5 h	20 µm	1500 L/h	10 %	no AWC
	3	7,5 h	20 µm	1500 L/h	10 %	no AWC
	4	7,5 h	20 µm	1500 L/h	10 %	no AWC

7.4 Parameters

To determine the performance of the membrane and the efficiency of airwater cleaning several parameters were measured during tests. Table 4 shows the parameters which were measured on-site and parameters which were analyzed in laboratory. Samples for on-site analyses were taken every 30 minutes from the feed water tank, after pretreatment filters and permeate. Samples for laboratory analyses were taken once during each experiment, except during the first air-water cleaning experiment no samples were taken and the results of the fourth experiment were lost in the laboratory.

 TABLE 4
 Measured parameters during the experiments.

Laboratory analyses	On-site analyses
pН	Turbidity
TOC and DOC	Temperature
Color	Conductivity
Suspended solids	Particle count
Colony forming units	
UV extinction	
Hardness	
Alkalinity	
Ions:	
Ca^{2+}	
Fe ²⁺	
Mg^{2+}	
Mn^{2+}	
Na ⁺	
K^+	
Cl	

Turbidity measurements were performed with a Hach 2100P Turbidimeter, temperature and conductivity were measured with a WTW Multi 340i, and particle counts were measured with a Met One laser particle counter. The particle counter measured particles of sizes 2, 5, 10, 15, 20 and 30 μm .

8 RESULTS AND DISCUSSION

8.1 Feed water quality

The feed water was analyzed in the laboratory and on-site. Samples were taken both from the feed tank water and from the sample point after the pretreatment steps just before membrane. The results of laboratory analysis in appendix 2 show that water from both of these sample points was low quality water. The lab results were not available of the first and fourth experiment but as can be seen the quality did not change a lot between the experiments. The colour remained high (50-70 mg Pt/Co/l) and TOC was also high (~15 mg/l). The UV extinction was very high which showed that the water contained a lot of organic matter. The water had a high biofouling potential since there were a lot of colony forming units during the first experiments.

In all experiments the feed water turbidity was high which could also be observed visually as can be seen in figure 15. The quality of feed tank water and water after pretreatment was almost the same during the experiments 1-4. During the first four experiments the feed water turbidity remained quite stable between 8 to 12 NTU as shown in appendix 3. Spikes in the feed tank turbidity are probably caused by the level switch which

turned the pump off and let the water stand in the pipe and the pump turned on right before the sample was taken.



FIGURE 15 Samples of feed tank, after pretreatment, permeate and concentrate water.

The turbidity depended on the particle amount of the smallest particles (2, 5 and $10~\mu m$) as can be seen in figure 16. The total particle amounts varied between 15000 and 30000 particles per ml during the first four experiments. Particle amounts are shown in appendix 4. On rainy days the particle count rose only a bit to almost 30000 particles per ml while it was otherwise around 25000 particles in ml. In the period of the experiments there were only three to four rainy days. In experiment 1 in runs 3 and 4 it was raining almost the whole day. During the first reference experiment the first three runs it was raining during the night and also in the first run of reference experiment 2. During experiment 2 in run 2 the weather was hot. This resulted in a decrease in particle count.

When the cartridge filter size was changed or taken completely away during the first four experiments the particle amounts increased only for about 2000 particles per ml and turbidity remained approximately the same in the first four experiments. Figure 17 shows the ratio of 2 µm particles compared to the total particle amount. Over 40 % of total particles were 2 μm so it could be that the filter size of 5 μm was too large pore size to affect the particle amount. About 98 % of the total particle amount was composed of small particles (2, 5 10 µm) as can be seen in figure 18. In experiments 1, 3 and 4 the ratio of small particles was close to each other. In experiments 1 and 2 the filter sizes were 5 and 20 µm respectively but the ratio of small particles in experiment 2 was smaller than in experiment 1. The reason for this is perhaps the hot weather during experiment 2 which reduced the total particle amount and because of that the ratio of small particles seems smaller. In reference experiments the small particle amounts are vice versa. This could be because the total particle load was higher during the reference experiments and the effect of 5 µm filter on the small particle amount showed more clearly which can also be seen in figure 17. During the second reference experiment and cartridge filters

were effectively filtering the large particles. This was detected when the filters had to be changed in the middle of the experiment.

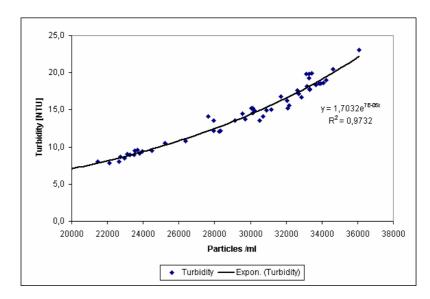


FIGURE 16 Turbidity compared to the total particle amount of 2, 5, 10 µm particles in the reference experiment 1 in the sample after pretreatment.

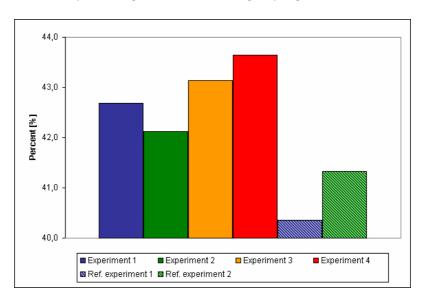


FIGURE 17 Amount of 2 μm particles from total particle amount in pretreated water in all experiments.

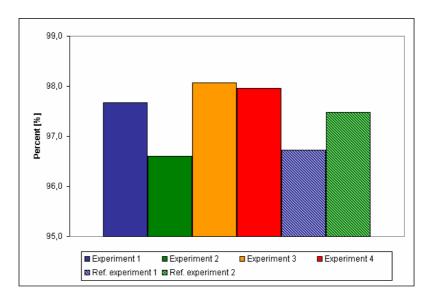


FIGURE 18 The amount of small (2, 5, 10 μ m) particles from total particle amount in pretreated water in all experiments.

During the reference experiments the turbidity and particle count of the pretreated water diverged from the feed tank turbidity and particle count measured in the feed tank. The turbidity and particles in the feed water after pretreatment began to rise. The turbidity reached almost 20 NTU while the water in the feed tank had turbidity of 10 NTU. The particle count in the pretreated water in the first reference experiment was over 35000 particles per ml while in the feed tank the count was below 30000 particles per ml.

At the end of the first reference experiment one of the large supply tanks at the distribution station was emptied for maintenance work and the outlet was next to the pump in the canal. This caused material on the bottom of the canal to rise and mix in the water which led to sudden fouling of the feed tank filter and also a lot of small pieces of plants and sand flushed to the cartridge filters in the installation. This can be seen as a big spike in the turbidity and particle counts. At this point the pilot was turned off for 45 minutes until the flow of supply tank decreased. Turbidity and the particle count in feed tank water and especially in the water after the pretreatment remained high during the second reference experiment but the values were lower than in the reference experiment 1. Appendix 2 table 11 shows that during the reference experiment 2 in the sample after the pretreatment had an extremely high colour value. The colour in the feed tank sample was normal. However, all the other parameters which could affect the colour, such as TOC/DOC, iron or magnesium, were normal levels. Only turbidity was high too. This indicates that between the feed tank and after pretreatment was a particle source that was polluting the water. Probably during the draining of the supply tank a lot of particles deposited in the bottom of the feed tank and they were flushed to the installation during the last reference experiment or deposit of particles were somewhere else in tubing between the feed tank and installation.

8.2 Permeate quality

Permeate quality was stable and high throughout the experiments. Retention based on conductivity was around 99,1 % (appendix 5) in all experiments and turbidity mainly between 0,15 and 0,20 NTU (appendix 3). Small changes in retention between experiments are caused by lower feed water conductivity and spikes are due to one unit change in permeate conductivity. In the first reference experiment the retention started high which is normal for a new membrane but towards the end of experiment it decreased to 98,9 %. At the beginning of the second reference experiment it was back to 99,1 %. Laboratory analysis in appendix 2 shows that permeate quality was very high during all the experiments. Salts were removed almost completely (over 98 %) and also other contaminants were removed with high retention (96-99 %). The results prove that during the experiments the membrane was not damaged by the air-water cleaning.

8.3 Air-water cleaning experiments

In these experiments the objective was to study the optimum air-water cleaning frequency and pretreatment requirements. The next sections describe the effect of AWC on pressure drop, transmembrane pressure and membrane permeability.

8.3.1 The effect of air-water cleaning on feed channel pressure drop

The air-water cleaning was applied on the membrane to observe its effect to control membrane fouling. Cornelissen, Vrouwenvelder, Heijman, Viallefont, Van der Kooij & Wessels (2007) found dramatic improvement in fouling control when AWC was used. The mixture of air and water removed fouling from the membrane and the feed spacer. The feed channel pressure drop increases due to fouling so pressure drop was observed to detect the effect of AWC.

The feed channel pressure drop, the pressure difference between feed and concentrate lines, was developing slowly during the first four experiments considering the low feed water quality. In the first test AWC was applied after every seven hours of operation. Figure 19 shows the feed channel pressure drop during first experiment. Arrows show the cleaning times. No decrease in feed channel pressure drop was observed after cleaning but this could be due to minimal fouling. The feed channel pressure drop increased only 0,02 bars (9,5 %) during the first test which showed that there was only slight fouling. During the experiment the pressure drop remained stable between 0,21 and 0,23 bar which could be due to AWC.

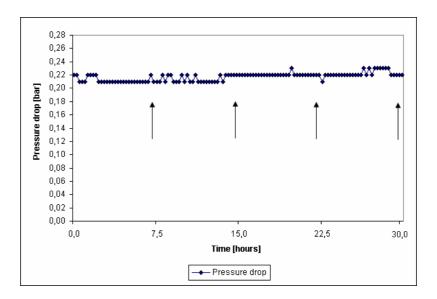


FIGURE 19 Feed channel pressure drop during experiment 1 (filter size 5 μ m). Arrows indicate AWC times when the cleaning was applied for 5 minutes.

Before the first experiment the pilot was operated for 26 hours with 5 μm cartridge filters and different feed flows and recoveries. During this period membrane was cleaned sporadically eight times. The feed channel pressure drop increased 10 % during that time. After the first experiment the increase in feed channel pressure drop was 15 % compared to the initial value of clean membrane. This indicated slight fouling. The total operating time after the first experiment was 56 hours. Considering the quality of feed water, fouling was developing slower than expected.

The second experiment was not showing faster fouling despite the larger pore size filters. The particle count during this experiment was also lower due to a very warm day in run 2. The feed channel pressure drop increased as much as in the first experiment, 0,02 bars (9 %). The total increase compared to initial pressure drop was 20 % after 86 hours (3,5 days) of operation. The pressure drop recovered 0,01 bars after the three first cleanings but the fourth cleaning did not show any effect on the pressure drop as can be seen in figure 20. The cleaning frequency was the same as in the first experiment which may have kept the pressure drop from increasing but at the same time the particle load was also smaller than in experiment 1 which could slow down the fouling. So low particle load and short cleaning frequency together might have caused the slower fouling.

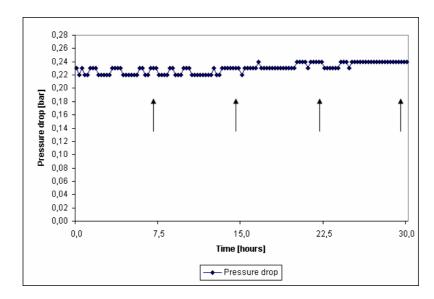


FIGURE 20 Feed channel pressure drop during experiment 2 (filter size 20 μ m). Arrows indicate the AWC times when the cleaning was applied for 5 minutes.

In the third experiment the cartridge filters were removed and cleaning frequency was longer. As can be seen in figure 21 the membrane was cleaned only twice during the third experiment. The feed channel pressure drop increased 0,03 bars (12,5 %) during this experiment. The total increase compared to the initial value was 35 % which showed that the membrane was getting more fouled. The particle count was the same as in the experiment 1 which indicated that longer cleaning frequency caused the faster fouling.

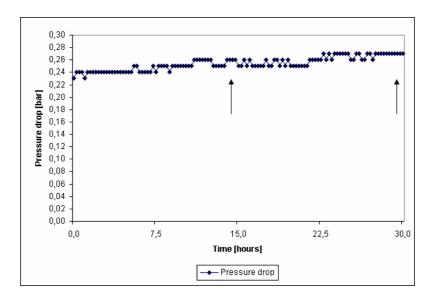


FIGURE 21 Feed channel pressure drop during experiment 3 (no filters). Arrows indicate the AWC times when the cleaning was applied for 5 minutes.

In the fourth experiment the objective was to study the decrease in feed channel pressure drop after air-water cleaning. In the experiment the membrane was cleaned only once after 24 hours of operation. As can be seen in figure 22 the pressure drop was reduced from by 0,01 bar to 0,29 bars. A complete recovery of the feed channel pressure drop was not

achieved. The membrane was cleaned three times and after each cleaning step the pressure drop was determined and when no recovery in pressure drop was detected the membrane was cleaned again. The details of each cleaning are in table 5. The third cleaning was applied with an air/water ratio of 3/1 with air flow of 3000 L/h and water flow of 1000 L/h. After this last AWC the pressure drop reduced 0,01 bars and because it seemed that a higher recovery could not be reached the cleaning was stopped. The initially used air/water ratio of 2/1 was chosen according to previous studies but further studies should be made with different air/water ratios. Cornelissen, Rebour, van der Kooij & Wessels (2009) found that turbulence increased in experiments performed with membrane fouling simulator (MFS) when the air/water ratio was higher. Turbulence prevents particles to accumulate on membrane and thereby prevents fouling so higher turbulence during AWC should be more effective (Van der Kooij, Hijnen & Cornelissen 2009, 26).

During experiment 4 the pressure drop increased 0,03 bars (11 %) which was as much as in experiment 3. Shorter cleaning frequency than in the experiment 3 did not lead to faster fouling. After the fourth experiment the total increase in pressure drop was 50 %. Fouling was attached strongly to the membrane which indicated that fouling might be mainly biofouling. Also the slow development suggests to biofouling. Vrouwenvelder, Graf von der Schulenburg, Kruithof, Johns & van Loosdrect (2009) found that biofouling is a feed spacer problem so the fouling was attached also on spacer. When the spacer gets fouled it affects mainly feed channel pressure drop because the resistance in the flow channel increases. Spacer fouling also causes development of flow channels when parts of the spacer mesh becomes clogged and the flow of water goes around the clogged parts. (Vrouwenvelder, Graf von der Schulenburg, Kruithof, Johns & van Loosdrect 2009, 589-590.) This might also affect the flow of AWC and interfere the cleaning flow distribution.

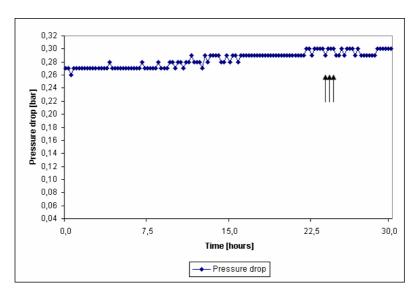


FIGURE 22 Feed channel pressure drop during experiment 4 (no filters). Arrows indicate AWC times when the duration were 5 min, 10 min, and 5 min respectively.

 TABLE 5
 Air-water cleanings in experiment 4.

Cleaning	Water flow	Air flow	Air-water cleaning time
1	1500 L/h	3000 L/h	5 min
2	1500 L/h	3000 L/h	10 min
3	1000 L/h	3000 L/h	5 min

Figure 23 shows the fouling rate of the air-water cleaning experiments. The fouling rate of two preliminary runs before the experiments is added in the figure. During those runs the feed flow and recovery were the same as in the air-water cleaning experiments. As can be seen in figure 23 the fouling rate increased significantly between preliminary runs and the first air-water cleaning experiment. During the preliminary runs the membrane was cleaned four times sporadically during 14 hours of operation. So the longer cleaning frequency in the experiment 1 compared to preliminary runs had a strong effect on the fouling rate which indicates that shorter cleaning frequency keeps the fouling rate lower. There was an increase between fouling rates of the experiments 1 and 2 which shows that the fouling was actually developing faster in experiment 2 than in experiment 1. The difference between fouling rates in experiments 2 and 3 was almost the same as between experiments 1 and 2. After 100 hours of operation when the filters were removed the fouling rate increased substantially. So without filters the cleaning frequency should be shorter than 14,5 hours which was used in experiment 3. According to the fouling rate it could be that during continuous run the module would have to be cleaned a few times a day.

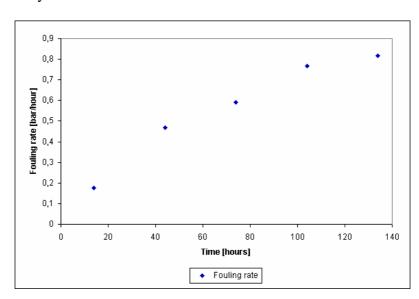


FIGURE 23 Fouling rate of air-water cleaning experiments.

The particle count during the experiment 3 was not differing much from the experiment 1 which indicates the same particle load in those two experiments. Cornelissen, Rebour, van der Kooij & Wessels (2009) found that daily AWC was more efficient than weekly AWC. According to previous studies, experiments 1, 2 and 3, and the fouling rate between preliminary runs and experiment 1, it seems that shorter AWC frequency

slowed the fouling down. The results from the experiment 4 unfortunately did not support this conclusion since the longer AWC frequency did not lead to faster fouling. But the particle counts were a bit lower than in the experiments 1 and 3. Laboratory results were not available from the experiment 4 but judging from other laboratory results it can be assumed that the feed water quality should have been quite similar during the fourth experiment. The small difference between fouling rates of the experiments 3 and 4 could be due to lower particle load during the experiment 4.

The effect of AWC was hard to detect from feed channel pressure drop measurements because a clear reduction in pressure drop could not be reached. The total net operating time at the end of the experiment 4 was 6 days. During this time the pressure drop increased 50 %. Figure 24 shows the increasing of pressure drop during the experiments. The two preliminary runs are included in the graph the same way as in the fouling rate graph. At the beginning from 15 to 75 hours of operation the pressure drop increase was steady since the fouling rate was low. Between 80 and 100 hours of operation the pressure drop increased substantially which was due to higher fouling rate. Also between 100 and 140 hours of operation the pressure drop made a steep increase. This also suggests significant difference between the experiments with cartridge filters and without cartridge filters combined with longer AWC frequency. The exponential increase reminds of biofilm development (Flemming 1997, 383). This could also suggest that the fouling was mainly biofouling. But it has to be taken into account that the membrane carried a historical fouling from one experiment to another and that could interfere the experiments.

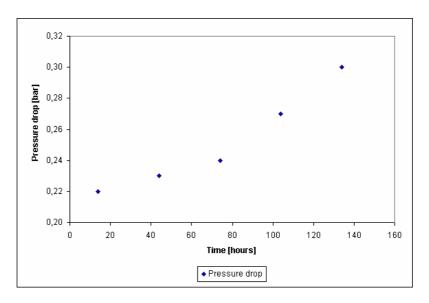


FIGURE 24 Feed channel pressure drop during air-water cleaning experiments.

In the previous study the effect of AWC to control biofouling was studied and an increase of 50 % in feed channel pressure drop was found after 16 weeks of operation on a reverse osmosis membrane which was cleaned daily by AWC (Cornelissen, Vrouwenvelder, Heijman, Viallefont, Van der Kooij & Wessels 2007, 98). In that study feed water was tap water filtered with 1 µm cartridge filters to prevent heavy particle load and dosed

with sodium acetate to increase the biofouling potential. The surface water used in this study had more fouling potential than the feed water used in the study of Cornelissen, Vrouwenvelder, Heijman, Viallefont, Van der Kooij & Wessels (2007) since the feed channel pressure drop increased in shorter period of time to the same value. Faster fouling in this study could also be due to the combination of particle and biofouling which is expected to influence each other (Cornelissen, Viallefont, Beerendonk & Wessels 2009, 9). Long-term experiments are necessary to study the performance of hydraulically cleaned RO membrane fed with surface water. Long-term experiments will also show what effect changes in the surface water quality will have on the performance of a hydraulically cleaned membrane.

8.3.2 The effect of air-water cleaning on transmembrane pressure

Transmembrane pressure is the pressure difference between feed and permeate side of the membrane. In reverse osmosis process permeate is near the atmospheric pressure so the TMP was calculated with feed pressure and assuming that the permeate side is in the atmospheric pressure of 100 kPa.

The transmembrane pressure increased during the experiments. It started from 650 kPa in experiment 1. Already in the first experiment it rose to 680 kPa as can be seen in figure 25. The average transmembrane pressure during experiment 1 was 660 kPa. The increase in transmembrane pressure showed that the membrane was fouling. A sudden increase happened after 15 hours at the beginning of third run and also at the beginning of fourth run. These increases could indicate a strong fouling. The feed channel pressure drop increased 0,01 bar at the beginning of the third run which correlates with transmembrane pressure but no increase in the feed channel pressure drop was observed at the beginning of fourth run. The pressure dropped suddenly during the third run after 20 hours which was due to the clogged feed tank filter which stopped the installation. When the pilot was started again the pressure was lower. So the pressure might also change due to start-ups.

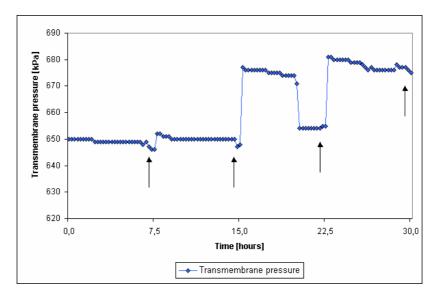


FIGURE 25 Transmembrane pressure during experiment 1. Arrows indicate the AWC times.

Only a slight effect of AWC was detected in the feed channel pressure drop but the transmembrane pressure decreased after AWC. As can be seen in figure 25 the transmembrane pressure during the experiment 1 decreased slightly after each AWC. This shows that the membrane was cleaned during AWC. During the experiment 2 it showed clearly that the transmembrane pressure dropped after each air-water cleaning as can be seen in figure 26. This much clearer drop also showed that in the experiment 1 fouling had not developed so much that the air-water cleaning could show a noticeable decrease in the TMP or feed channel pressure drop. In the second experiment the transmembrane pressure increased from 620 to 720 kPa which is a remarkable increase. The increasing compares to feed channel pressure drop which was also steadily increasing for 0,01 bar after 15 hours of operation. The average transmembrane pressure was 675 kPa which indicates that average TMP increased 15 kPa from experiment 1. The AWC applied at the end of the first run reduced the TMP for 60 kPa. The transmembrane pressure at the second run was lower than in the first run. Either the AWC at the end of the first run cleaned the membrane very well and the pressure remained low or the start-up affected the TMP. The next three AWCs reduced the TMP to value of 645 kPa which showed that AWC was effectively cleaning the membrane.

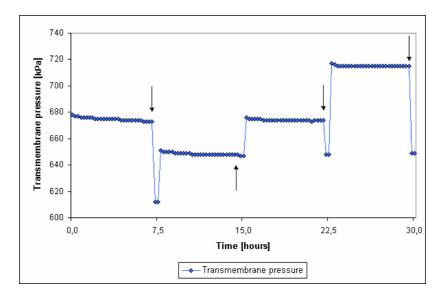


FIGURE 26 Transmembrane pressure during experiment 2. Arrows indicate AWC times.

Also during experiment 3 the transmembrane pressure increased. As can be seen in figure 27 it reached 750 kPa while the TMP at the beginning of the experiment 3 was 690 kPa. TMP increased 30 kPa between runs 1 and 2, and 3 and 4. The same steps were found on the feed channel pressure drop as the pressure increased 0,01 bar. The average transmembrane pressure was 720 kPa which indicates a 45 kPa increase of average TMP from the experiment 2. The average increase can be compared to faster fouling which could be seen also in the feed channel pressure drop.

It can be clearly seen that transmembrane pressure decreases after every AWC but when the next run is started it has risen again. When pilot was started in the morning, brown dirty water could be seen in the concentrate flow meter. The start up flushed away removable fouling that developed during the night when the pilot was standing still. The "stair" looking increase could also be irreversible fouling that developed during the night. Between the experiments was weekend while the pilot was not running. After each experiment clean tap water was fed to the pilot and left in the tubing to prevent extensive microbial growth during the weekend. At the end of the first experiment the transmembrane pressure was 680 kPa as can be seen in figure 25. The second experiment also started with a transmembrane pressure of 680 kPa as can be seen in figure 26. So no fouling effect was found after weekend. This could indicate that the "stair" effect was biofouling. The same end and start-up pressures were also found between experiments 3 and 4.

With continuous run the "stairs" could be avoided. It disturbed the analysis of the results because a definite reason for the effect can not be presented.

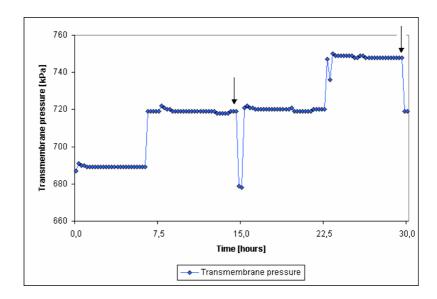


FIGURE 27 Transmembrane pressure during experiment 3. Arrows indicate AWC times.

In the experiment 4 the average TMP was 740 kPa which indicates a 20 kPa increase compared to the experiment 3. As was detected from the feed channel pressure drop the average transmembrane pressure also showed minor fouling compared to the experiment 3. During the fourth experiment TMP was more stable. The high point of almost 800 kPa could be due to the feed channel pressure drop increase of 0,01 bar. The same increase in the feed channel pressure drop happened after 28 hours which reduced the flux and the feed pressure had to be adjusted. This is shown in figure 28 as an increase in TMP. The air-water cleaning after 24 hours of operation reduced the transmembrane pressure. It can be concluded that the transmembrane pressure indicated the efficiency of AWC clearer than pressure drop in the feed channel and AWC reduced the transmembrane pressure.

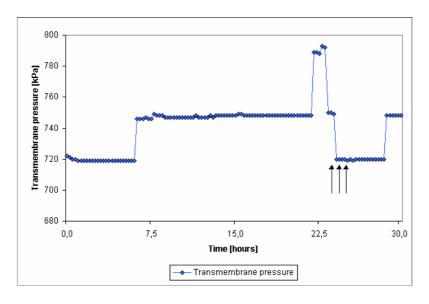


FIGURE 28 Transmembrane pressure during experiment 4. Arrows indicate AWC times.

Biofouling is mainly a feed spacer problem (Vrouwenvelder, Graf von der Schulenburg, Kruithof, Johns & van Loosdrect 2009, 589-591). The transmembrane pressure indicates the resistance through the membrane and therefore it indicates the fouling on the membrane surface as can be seen in figure 29. A reduction in transmembrane pressure was observed after air-water cleaning which indicated that AWC cleaned the membrane surface. Biofouling was attached mainly on the spacer and was not so easily removed. Particles might not be deposited on the spacer but on the membrane surface. Air-water cleaning has found to be effective also on particle removal (Cornelissen, Viallefont, Beerendonk & Wessels 2009, 8). So the reduction on transmembrane pressure could indicate the removal of particle fouling.

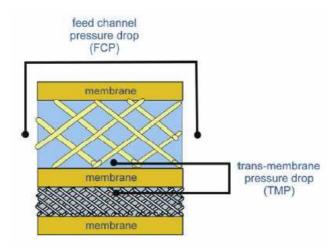


FIGURE 29 Pressure drops in membrane sheets: feed channel pressure drop and transmembrane pressure drop (Vrouwenvelder, Graf von der Schulenburg, Kruithof, Johns & van Loosdrect 2009, 584).

8.3.3 The effect of air-water cleaning on membrane permeability

Flux is related to transmembrane pressure. When fouling increases the transmembrane pressure, flux decreases. The normalized flux for temperature and pressure is called mass transfer coefficient or permeability. The flux was calculated by dividing the permeate flow with membrane area as can be seen in formula 13. The flux during all the experiments can be found on appendix 6.

FORMULA 13 Equation for flux.

$$J = \frac{Q_p}{A}$$
in which,
 $J = \text{flux}$, L/h m²
 $Q_p = \text{permeate flow}$, L/h
 $A = \text{membrane area}$, m²

The permeability (MTC) was calculated and normalized for temperature and pressure. The calculations for MTC are shown in formulas 14-16.

FORMULA 14 Equation for MTC.

$$MTC = \frac{J \cdot TCF_{MTC}}{\overline{P} \cdot 3600 \, \text{sec}}$$

in which,

MTC= mass transfer coefficient, m/s Pa

J= flux, L/h m²

 TCF_{MTC} = temperature correction factor for MTC, unitless

 \overline{P} = average feed channel pressure, Pa

FORMULA 15 Equation for temperature correction factor for MTC.

$$TCF_{MTC} = e^{(1/(T_M + 273)) - (1/(T_{Ref} + 273))}$$

in which,

 TCF_{MTC} = temperature correction factor for MTC, unitless

 T_M = measured temperature, °C

 T_{Ref} = reference temperature, °C (in this 20 °C were chosen for reference temperature)

FORMULA 16 Equation for average feed channel pressure.

$$\overline{P} = \frac{P_F - P_C}{2}$$

in which,

 \overline{P} = average feed channel pressure, Pa

 P_F = feed pressure, Pa

 P_C = concentrate pressure, Pa

After air-water cleanings the permeability rose which indicates that the membrane was cleaned. The same effect was not seen on flux. Flux and permeability were both stable during the experiments. During the first experiment the average flux was 19,3 L/h m². As can be seen in figure 30 the permeability was slightly decreasing which is due to increase in the transmembrane pressure during the first experiment. This indicated fouling. The average permeability was 7,2 *10⁻¹² m/s Pa. During the first experiment an increasing and decreasing can be seen during each run. An increase could indicate the flushing of fouling that developed between the runs and decrease a new fouling on the membrane.

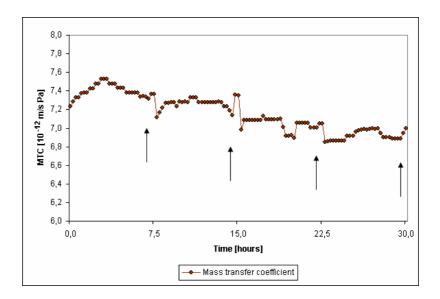


FIGURE 30 Permeability during experiment 1. Arrows indicate AWC times.

In the second experiment the average flux was 19,5 L/h m² which is slightly higher than in the experiment 1. Small changes can be seen between the runs but generally flux remained stable. The average MTC of 7,1*10⁻¹² m/s Pa showed a slight increase. The permeability declined during the runs 2-4. The increase on TMP was observed during the runs 2-4 which correlates on permeability so the membrane was fouling. After each AWC permeability increased which indicates a cleaning of the membrane.

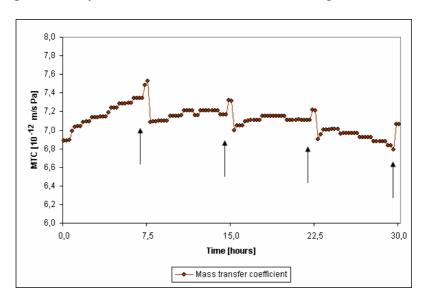


FIGURE 31 Permeability during experiment 2. Arrows indicate AWC times.

During experiment 3 flux remained about 19 L/h m². The average flux had decreased to 19,0 L/h m². The permeability correlated increases and decreases of transmembrane pressure as can be seen in figure 32. In the first run permeability was high and then in the second and third run it was quite stable and in the fourth run it declined. The changes indicate that the membrane was slowly fouling. The average MTC was 6,5*10⁻¹² m/s Pa which indicates a small decline compared to experiment 2. The average transmembrane pressure increased 45 kPa from the experiment 2 which

perhaps caused the slight decrease in MTC and flux. The effect of AWC can be clearly seen on permeability.

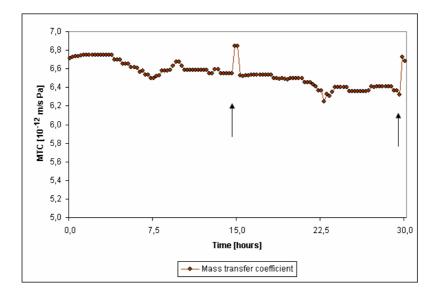


FIGURE 32 Permeability during experiment 3. Arrows indicate AWC times.

The flux was stable during the fourth experiment. It remained around 19,0 L/h m². The average flux of 19,0 L/h m² also proves that there was no change compared to the experiment 3. The permeability during the experiment 4 is shown in figure 33. The average permeability was $6.4*10^{-12}$ m/s Pa which was almost the same in the previous experiment. Compared to the transmembrane pressure which had the increase of 20 kPa from the experiment 3, the permeability showed that small increase in transmembrane pressure was not affecting the MTC. The MTC was declining during the first three runs which indicates fouling but after the AWC it increased indicating that AWC was enhancing the normalized flux.

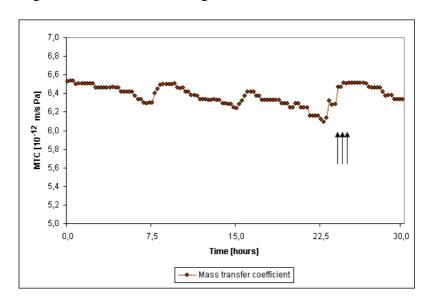


FIGURE 33 Permeability during experiment 4. Arrows indicate AWC times.

In previous studies the effect of particle or biofouling on flux has been controversial. Some studies report the flux decline and some report no ef-

fect on flux (Speth, Gusses & Summers 2000, 35. Zhu & Elimelech 1997, 3656, 3658. Cornelissen, Harmsen, Beerendonk, Wessels & Van der Kooij 2009, 4. Vrouwenvelder, van Paassen, van Agtmaal, van Loosdrecht, Kruithof 2009, 41-42). In this study the transmembrane pressure increased 170 kPa (27 %) from the initial value, the flux declined ~2 L/h m² (12 %) and permeability declined ~1,2*10⁻¹² m/s Pa (16 %) from their initial values. During experiments while the feed channel pressure drop was rising the transmembrane pressure increased too which shows a connection in fouling development. The fact that experiments were not performed on continuous runs makes the comparison difficult because the transmembrane pressure increased step by step. The average values of flux and permeability between experiments were not declining significantly. The decline of the flux and permeability from their initial values during the complete experiment series were 12 % and 16 % respectively, which indicates that there was a decline. It could be assumed that if the experiments had continued the flux and MTC would have continued decreasing. In transmembrane pressure the reduction after AWC can be clearly seen. Also the MTC increased after air-water cleaning. These results are in contradiction to the fact that flux is not affected by fouling. The results suggest that air-water cleaning reduces transmembrane pressure and enhances permeability.

8.4 Reference experiments

Two reference experiments were performed to see how fast fouling develops on membrane which is not cleaned by AWC. The results indicated that the fouling was developing at the same rate as in the air-water cleaning experiments. During the first reference experiment the pressure drop increased 0,02 bars (10 %) which is as much as in the first experiment. The pressure drop of the reference experiment 1 is in figure 34.

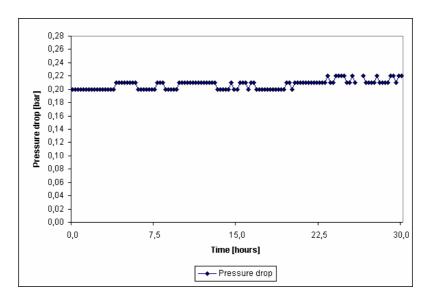


FIGURE 34 Feed channel pressure drop during reference experiment 1 (filter size 5 µm).

During the second reference experiment the fouling was not faster as can be seen on figure 35. The pressure drop increased less than in the first reference experiment, only 0,01 bar. The total increase compared to the initial value was 15 % after 59 hours of reference experiments. After 56 hours of operation with air-water cleaning the pressure drop increase was the same. During the reference experiments the particle count and turbidity were higher than in the experiments 1-4. The reason for slow fouling could be that the feed water had less biofouling potential. The water temperatures were lower which could have reduced the biofilm formation and make the fouling develop slower.

One cleaning was applied at the end of the second reference experiment and the pressure drop was recovered 0,01 bar almost to the initial value. In the experiments 1-4 the AWC was not recovering the pressure drop at all or only for short time but in the reference experiment after the AWC the feed channel pressure drop remained at 0,21 bar until the end of the experiment. This could indicate that most of the fouling was caused by particles because also the transmembrane pressure decreased remarkably after air-water cleaning. Also the huge particle count during the draining of supply tank might have caused particle fouling. This is also supported by the cartridge filters which had to be changed after two runs during the second reference experiment.

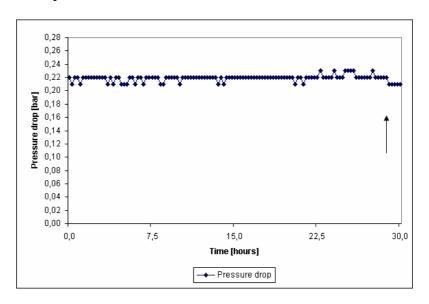


FIGURE 35 Pressure drop during reference experiment 2 (filter size 20 µm). Arrow indicates the AWC.

The transmembrane pressure was high during both of the reference experiments as can be seen in figures 36 and 37. The average pressure in the reference experiment 1 was 820 kPa and in the second reference experiment 845 kPa. So the increase of average transmembrane pressure between the experiments was 25 kPa. A substantial decline in TMP was detected after AWC which proves that huge part of fouling was flushed away during the AWC.

The transmembrane pressure was changing through the first reference experiment. After the first run it decreased which could be due to rinsing of material that is added in the new membranes for safe transport. The particle count was high during the third run which perhaps caused the TMP increase after 15 hours of operation. After the draining of the supply tank the pressure was unstable and went down. It is possible that the particles and other material that got flushed into the installation caused disturbance in the low pressure pump. During the second reference experiment the transmembrane pressure was also unstable. During the second reference experiment the TMP was decreasing but at the beginning of the fourth run it was high again. The increase could not be due to particles because the particle count was the same as during the previous run. The explanation could be the malfunction of the low pressure pump because right before the pilot was shut down after the last reference experiment the pump seemed to break down. The high transmembrane pressures could indicate that the low pressure pump was not operating properly during the reference experiments or during the reference experiment 2.

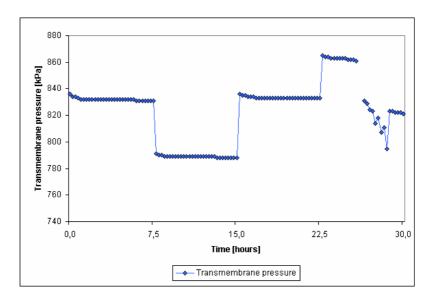


FIGURE 36 Transmembrane pressure during reference experiment 1.

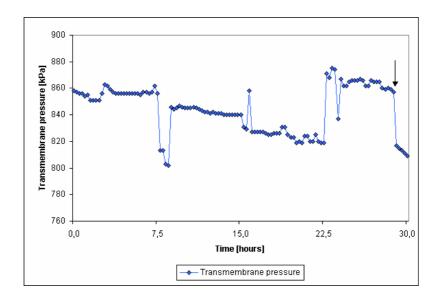


FIGURE 37 Transmembrane pressure during reference experiment 2.

The flux was stable in both of the reference experiments. The average flux was 19,3 L/h m² in the first reference experiment and 19,0 L/h m² in the second reference experiment. At the end of both experiments the flux declined a bit. The permeability was low which correlates to high TMP. The average MTC on first reference experiment was 5,9*10⁻¹² m/s Pa and 5,7*10⁻¹² m/s Pa on the second reference experiment. So the average flux and MTC did not decline during the reference experiments. The permeability was low in the fourth run of the first reference experiment and at the same time TMP was high which indicates strong fouling happened between runs 3 and 4. During the second reference experiment permeability had an increasing trend during three first runs which correlates the decrease of TMP. This indicates that fouling was decreasing. At the beginning of the second run the MTC started with a high value but it decreased during the run. Also transmembrane pressure started low which means that fouling had decreased between the runs 1 and 2. It could be that material which was added on the membrane at the factory was flushed from membrane and it caused the decreasing of fouling. Another reason could be the malfunction of the low pressure pump.

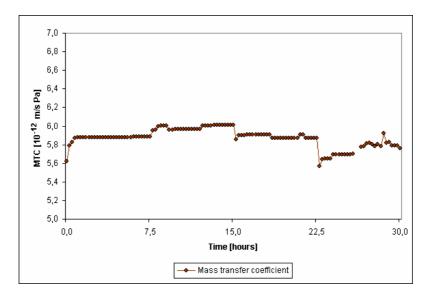


FIGURE 38 Permeability during reference experiment 1.

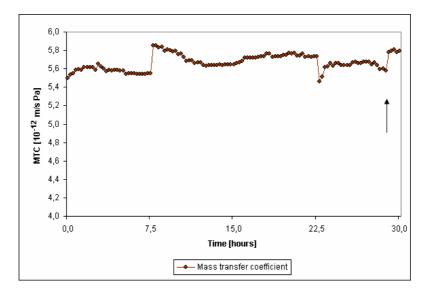


FIGURE 39 Permeability during reference experiment 2. Arrow indicates AWC.

The high TMP throughout the reference experiments indicates that the membrane was fouled but the feed channel pressure drop was not high. The fouling was not developing faster even though the feed water quality was worse in turbidity and particles than in the experiments 1-4. Only two reference experiments were operated and the same duration than experiments 1-4 would give better comparison. Also parallel modules with and without AWC would give better results. With surface water which changes according to seasons it is most effective to have parallel modules to observe the performance of air-water cleaned membrane. The reference experiments are not giving a definite comparison of air-water cleaning because the changes in feed water quality and possible troubles with the equipment. Parallel experiments are needed to fully compare the performance of air-water cleaning. The recovery of the pressure drop at the end of the reference experiment 2 proved that AWC is an effective way to clean reverse osmosis membranes treating surface water.

8.5 Rinsing water

Cleaning time was chosen according to earlier studies which revealed that the main part of the removable matter was removed during the first minutes of AWC and that no significant removal took place after five minutes of cleaning (Cornelissen, Vrouwenvelder, Heijman, Viallefont, Van der Kooij & Wessels 2007, 97. Cornelissen, Harmsen, Beerendonk, Wessels & Van der Kooij 2009, 5). Rinsing water was collected twice during experiments and analyzed on-site. Figure 40 shows the rinsing water during experiment 2 in run 2 after 15 seconds, 1 minute and 5 minutes rinsing. This sample was rinsing water of 7 hours AWC frequency. Visual observation showed that AWC is removing the impurities from the membrane. The on-site analysis results of rinsing water are shown in table 6. The sample after 15 seconds was too concentrated to be analyzed. In previous studies it was found that the quality of rinsing water after five minutes was almost the same as the feed water (Cornelissen, Vrouwenvelder, Heijman, Viallefont, Van der Kooij & Wessels 2007, 97. Cornelissen, Harmsen, Beerendonk, Wessels, & Van der Kooij 2009, 5). As can be seen in table 6 the turbidity was still high and the particle count was slightly higher than in the feed water. This indicates that AWC was still removing particles and biofilm from the membrane.

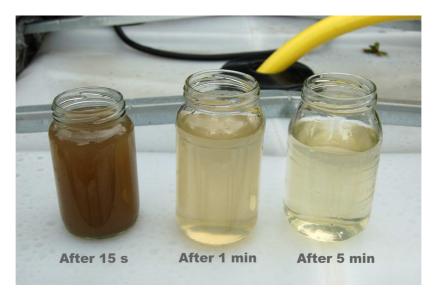


FIGURE 40 Rinsing water of experiment 2 run 2 (AWC frequency 7 hours) after 15 second, 1 minute and 5 minutes rinsing.

TABLE 6 Rinsing water quality on experiment 2 run 2 compared to feed water quality before air-water cleaning.

Time	Temperature	Turbidity	Conductivity	Total particles/ml
After 1 min rinsing	19,8 °C	29 NTU	749 μS/cm	29555
After 5 min rinsing	19,8 °C	9,51 NTU	749 μS/cm	20660
Feed water before AWC	19,9 °C	6,21 NTU	748 μS/cm	19735

The other rinsing water sample was taken during the experiment 4 when AWC frequency was 24 hours. Figure 41 and table 7 show that rinsing wa-

ter had deeper colour when the cleaning frequency was longer. Also "flakes" or small pieces of removed bacteria could be recognized in the 15 second rinsing water sample and also in the 1-minute sample. Comparison of feed water before AWC and the sample after 5 minutes rinsing show that particles were still being removed. These results differ from previous researches which concluded that particles are mainly removed during the first minute of AWC and mainly biomass was rinsed after 2 minutes (Cornelissen, Harmsen, Beerendonk, Wessels, & Van der Kooij 2009, 5). The rinsing water samples in the study of Cornelissen, Harmsen, Beerendonk, Wessels, & Van der Kooij (2009) were analyzed in a laboratory which made it possible to compare the results between many parameters. In this study rinsing water was not analyzed in a laboratory but only on-site and the comparison was based on turbidity, conductivity and particles. High turbidity after 5 minutes of rinsing showed that AWC was still removing particles. These results indicate that longer AWC frequency needs longer AWC time to wash away more impurities. Also the amount of fouling could have effect since during the second experiment the membrane was not fouled as much as at the end of the experiment 4. So the rinsing time might be derived from the quantity of fouling. The rinsing time should be studied for AWC when surface water is used for rinsing.

In reverse osmosis membrane autopsies main part of the biomass is found close to the inlet of the membrane (Vrouwenvelder, Graf von der Schulenburg, Kruithof, Johns & Van Loosdrecht 2009, 589. Cornelissen, Vrouwenvelder, Heijman, Viallefont, Van der Kooij & Wessels 2007, 99). This is because the nutrient concentration is also high near the inlet. Particles are probably spread more throughout the membrane. In this study the membrane was positioned vertically with the water flow from top to bottom. The air-water cleaning was applied from bottom to top. When the cleaning is applied the biomass near the top is first flushed away but it might take more time for the particles to be removed since they are distributed to a larger area. Also the spacer fouling creating flow channels causes the removal to become more difficult. In studies of Cornelissen, Harmsen, Beerendonk, Wessels, & Van der Kooij (2009) the feed water was filtered with 10 and 1 um cartridge filters and in the study of Cornelissen, Vrouwenvelder, Heijman, Viallefont, Van der Kooij & Wessels (2007) the feed water was filtered with 1 µm cartridge filters. This prevented the high load of particles to enter the membrane and thus does not include the removal of particles in the rinsing time.

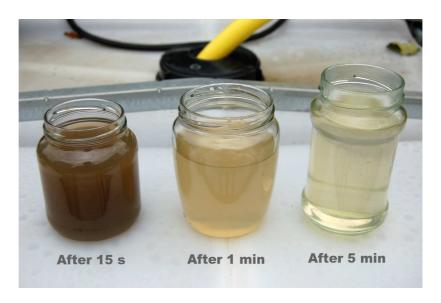


FIGURE 41 Rinsing water of experiment 4 run 4 (AWC frequency 24 hours) after 15 seconds, 1 minute and 5 minutes rinsing.

TABLE 7 Rinsing water quality on experiment 4 run 4 (AWC frequency 24 hours) compared to feed water quality before AWC.

Time	Temperature	Turbidity	Conductivity	Total particles/ml
After 1 min rinsing	17,8 °C	115 NTU	951 μS/cm	43925
After 5 min rinsing	17,8 °C	27,1 NTU	955 μS/cm	31819
Feed water before AWC	17,8 °C	7,56 NTU	956 μS/cm	21447

9 CONCLUSIONS

The results show that air-water cleaning is an effective way to clean reverse osmosis membranes. The performed experiments were short-term experiments when the pilot was running only during working hours which appeared to be not an ideal way for the study. But it is concluded that it is possible to treat surface water with hydraulically cleaned reverse osmosis membranes. Long-term experiments are needed for further studying of the performance of hydraulically cleaned RO membrane treating surface water. The experiments should be done with parallel modules when one of the modules is cleaned and the other one is a reference module. The results of this study indicate that shorter cleaning frequency controls the fouling more efficiently. It can be predicted that in continuous run in surface water treatment the reverse osmosis membranes have to be cleaned with AWC a few times a day. By long-term experiments the optimal AWC frequency can also be defined.

The results of this study showed enhancement of permeability when AWC was applied and also reduction in transmembrane pressure which indicates that the membrane was efficiently cleaned by AWC. The feed channel pressure drop was not recovered as effectively as transmembrane pressure which indicates that the spacer was not cleaned as well as the membrane.

Reference experiments proved that particles were effectively removed with AWC.

It was found that the air-water cleaning time should be chosen according to the fouling quantity or depending on the cleaning frequency. The fouling was both particle and biofouling which could have affected the rinsing time. Further study is required for rinsing time when rinsing is applied with surface water. Further studies are also suggested to study the optimal air/water ratio.

These experiments showed that it is possible to treat surface water with hydraulically cleaned reverse osmosis membranes and the pretreatment do not have to be extensive. The results showed that less pretreatment requires short cleaning frequency. The performance of pretreatment would also be better observed with long-term experiments and it would give a better overview how often filters need to be changed. In these experiments the filters were dirty and changed after 30 hours of operation which suggests that during long-term operation they would have to be changed every day. In short-term experiments the coarse cartridge filter of 100 μm was enough pretreatment to prevent the severe fouling of the membrane.

Based on these experiments modifications in the pilot are also suggested. Parallel modules are required and the pilot should be made more automatic. A data logger for pressure drop or a differential pressure transmitter and digital flow meters for feed and permeate would make the results more accurate and the pilot easier to operate.

Overall further studies are required but air-water cleaning seems to be an applicable technique for treatment of surface water with reverse osmosis membranes. It is possible to control fouling with hydraulic cleaning but it seems that only air-water cleaning is not enough to clean membranes. Hydraulic cleaning can not yet replace chemical cleaning but it is a cost-effective cleaning method and reduces the costs caused by fouling.

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APPENDIX 1/1

CANAL POTMARGE AND INTAKE POINT



FIGURE 42 Canal Potmarge in Leeuwarden. Arrow shows the intake point.



FIGURE 43 Canal Potmarge. Photo taken from the intake point.

APPENDIX 1/2

CANAL POTMARGE AND INTAKE POINT



FIGURE 44 Intake point.

APPENDIX 2/1

LABORATORY ANALYSIS

TABLE 8 Laboratory results on sample date 8.9.2009 (experiment 2).

Parameter	Unit	Feed tank	After pretreatment	Permeate
pН	-	7,56	7,61	5,69
Colour	mg Pt/CO/l	46	49	<3
TOC	mg/l	14	14	<0,5
DOC	mg/l	12	13	<0,5
Suspended solids	mg/l	7,8	9,3	<0,5
UV 254	1/m	37	37	<1,0
Colony forming units	/ml	>100000	>100000	22
Hardness	mmol/l	1,53	1,57	<0,02
	${ m O}$	8,5	8,8	<0,1
$\begin{array}{c} Ca^{2+} \\ Fe^{2+} \\ Mg^{2+} \\ Mn^{2+} \end{array}$	mg/l	43,0	44,3	<0,5
Fe ²⁺	mg/l	0,248	0,276	<0,01
Mg^{2+}	mg/l	11,0	11,2	<0,1
Mn^{2+}	mg/l	0,064	0,049	<0,005
Na ⁺	mg/l	110	111	1,30
K ⁺	mg/l	10	10	0,3
Cl	mg/l	140	140	<3

TABLE 9 Laboratory results on sample date 17.9.2009 (experiment 3).

Parameter	Unit	Feed tank	After pretreatment	Permeate
рН	-	7,61	7,75	5,86
Colour	mg Pt/CO/l	71	77	<3
TOC	mg/l	14	13	<0,5
DOC	mg/l	13	12	<0,5
Suspended solids	mg/l	6,8	7,4	<0,5
Total dissolved solids	mg/l	560	550	<5
UV 254	1/m	40	41	<0,2
Alkalinity	mg/l	206	206	<10
Hardness	mmol/l	1,72	1,78	<0,02
	$^{\circ}\mathrm{D}$	9,6	10,0	<0,1
Ca ²⁺ Fe ²⁺ Mg ²⁺	mg/l	47,6	49,3	<0,5
Fe ²⁺	mg/l	0,221	0,235	<0,01
Mg^{2+}	mg/l	12,9	13,3	<0,1
Mn^{2+}	mg/l	0,059	0,053	<0,005
Na ⁺	mg/l	121	124	1,50
\mathbf{K}^{+}	mg/l	11	11	<0,1
Cl	mg/l	160	160	<3

APPENDIX 2/2

LABORATORYANALYSIS

TABLE 10 Laboratory results on sample date 1.10.2009 (reference experiment 1).

Parameter	Unit	Canal	Feed tank	After pretreatment	Permeate
pН	-	7,73	7,65	7,74	5,74
Colour	mg Pt/CO/l	64	60	65	<3
TOC	mg/l	15	16	15	<0,5
DOC	mg/l	15	12	15	<0,5
Suspended solids	mg/l	10	6,6	7,6	<0,5
Total dissolved	mg/l	610	580	600	<5
solids					
UV ₂₅₄	1/m	43	43	43	<0,2
Alkalinity	mg/l	229	228	230	<10
Hardness	mmol/l	1,87	1,88	1,86	<0,02
	${ m O}$	10,5	10,5	10,4	<0,1
Ca ²⁺	mg/l	51	51,3	51	<0,5
Fe ²⁺	mg/l	0,24	0,22	0,21	<0,01
Mg^{2+}	mg/l	14,5	14,6	14,4	<0,1
Mn^{2+}	mg/l	0,056	0,048	0,048	<0,005
Na ⁺	mg/l	138	138	138	1,13
K ⁺	mg/l	13	13	13	<0,1
Cl	mg/l	190	190	190	<3

TABLE 11 Laboratory results on sample date 8.10.2009 (reference experiment 2).

Parameter	Unit	Feed tank	After pretreatment	Permeate
pН	-	7,62	7,47	5,66
Colour	mg Pt/CO/l	47	160	<3
TOC	mg/l	14	14	<0,5
DOC	mg/l	13	13	<0,5
Suspended solids	mg/l	7,0	8,2	<0,5
Total dissolved	mg/l	540	540	<5
solids				
UV ₂₅₄	1/m	40	40	<0,2
Alkalinity	mg/l	180	179	<10
Hardness	mmol/l	1,64	1,79	<0,02
	$^{\circ}\mathrm{D}$	9,2	10,0	<0,1
Ca ²⁺	mg/l	45,6	49,7	<0,5
Fe ²⁺	mg/l	0,218	0,228	<0,01
Mg^{2+}	mg/l	12,3	13,4	<0,1
Mn^{2+}	mg/l	0,031	0,023	<0,005
Na ⁺	mg/l	109	116	1,22
K^{+}	mg/l	10	11	0,2
Cl ⁻	mg/l	150	150	<3

APPENDIX 3/1

TURBIDITY DURING EXPERIMENTS 1- 4 AND REFERENCE EXPERIMENTS 1-2.

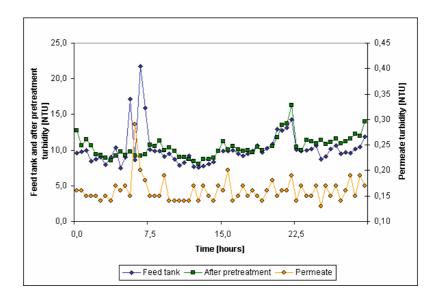


FIGURE 45 Turbidity during experiment 1 (filter size 5 µm).

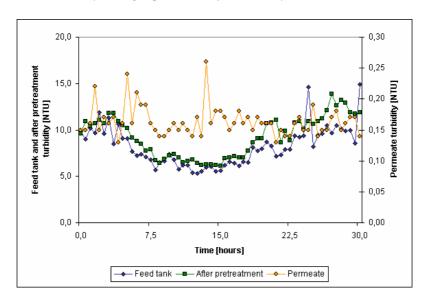


FIGURE 46 Turbidity during experiment 2 (filter size 20 µm).

APPENDIX 3/2

TURBIDITY DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

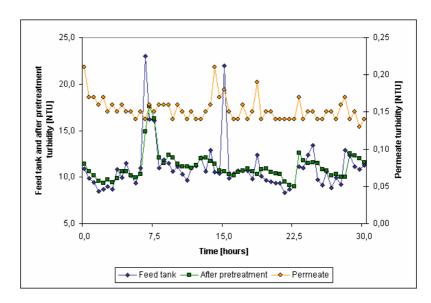


FIGURE 47 Turbidity during experiment 3 (no filters).

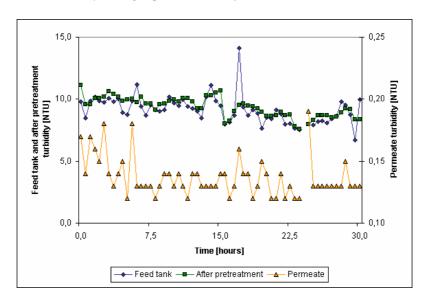


FIGURE 48 Turbidity during experiment 4 (no filters).

APPENDIX 3/3

TURBIDITY DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

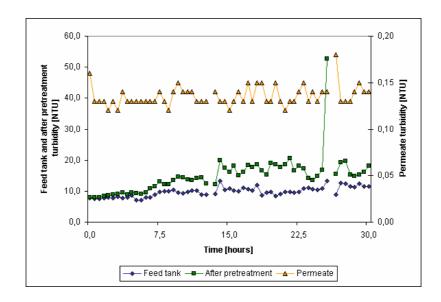


FIGURE 49 Turbidity during reference experiment 1 (filter size5 µm).

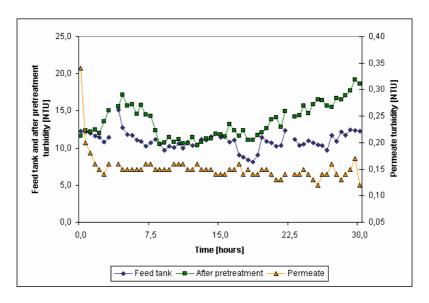


FIGURE 50 Turbidity during reference experiment 2 (filter size 20 µm).

APPENDIX 4/1

PARTICLES DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

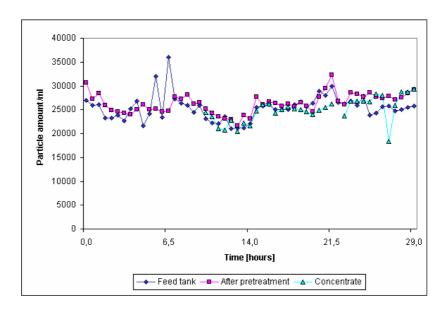


FIGURE 51 Particles during experiment 1 (filter size 5 µm).

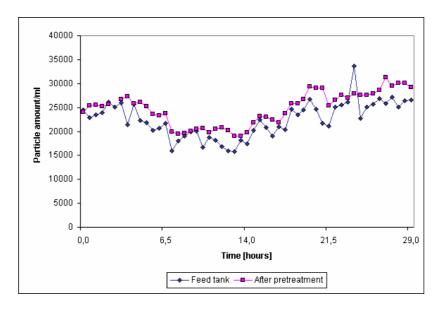


FIGURE 52 Particles during experiment 2 (filter size 20 μm).

APPENDIX 4/2

PARTICLES DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

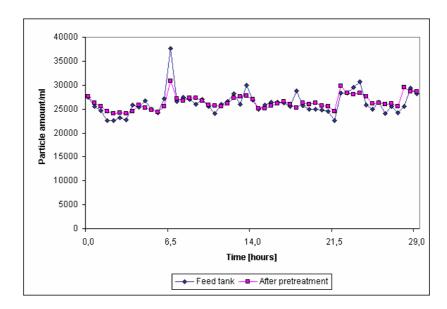


FIGURE 53 Particles during experiment 3 (no filters).

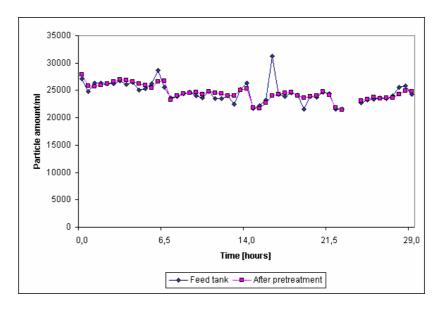


FIGURE 54 Particles during experiment 4 (no filters).

APPENDIX 4/3

PARTICLES DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

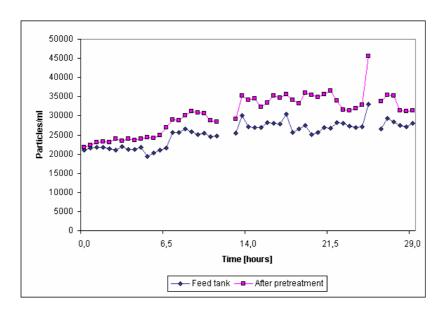


FIGURE 55 Particles during reference experiment 1 (filter size 5 µm).

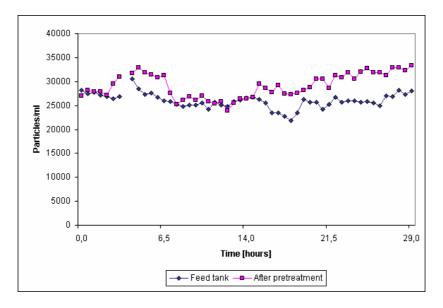


FIGURE 56 Particles during reference experiment 2 (filter size 20 µm).

APPENDIX 5/1

RETENTION DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

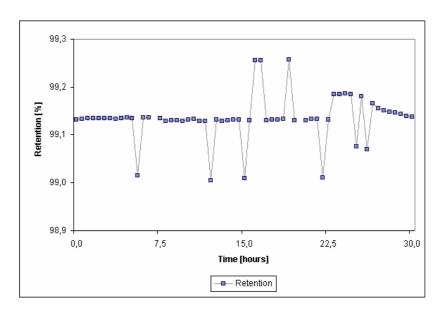


FIGURE 57 Retention during experiment 1.

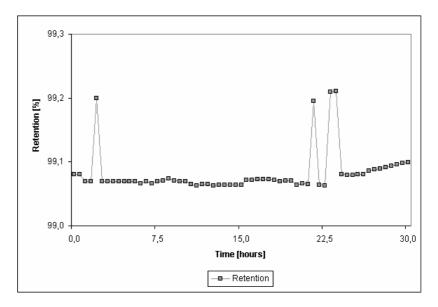


FIGURE 58 Retention during experiment 2.

APPENDIX 5/2

RETENTION DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

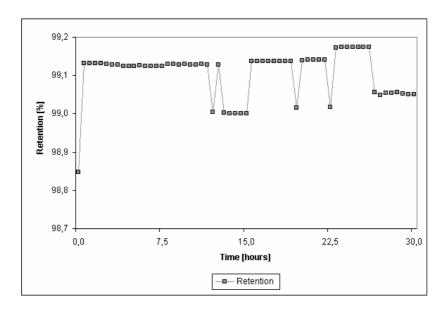


FIGURE 59 Retention during experiment 3.

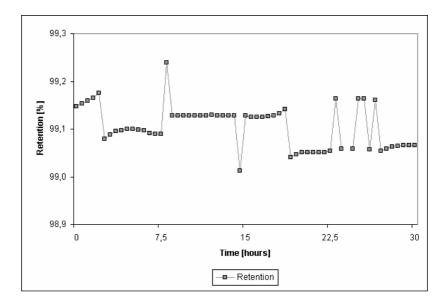


FIGURE 60 Retention during experiment 4.

APPENDIX 5/3

RETENTION DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

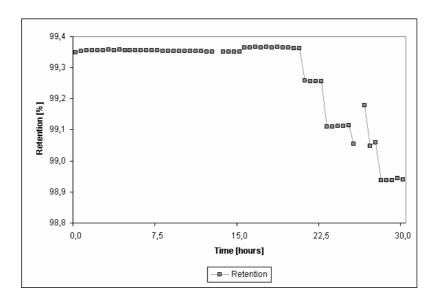


FIGURE 61 Retention during reference experiment 1.

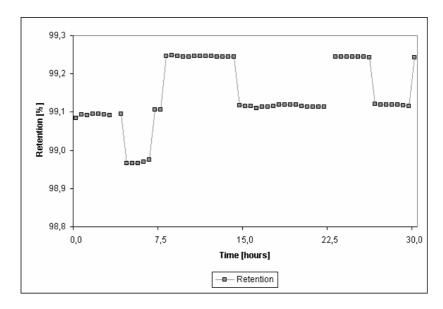


FIGURE 62 Retention during reference experiment 2.

APPENDIX 6/1

PERMEATE FLUX DURING EXPERIMENTS 1-4 AND REFERENCE EXPERIMENTS 1-2.

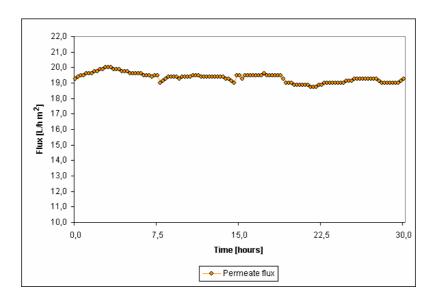


FIGURE 63 Permeate flux during experiment 1.

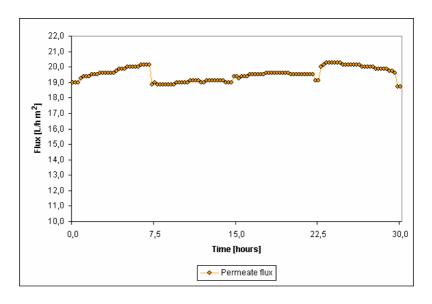


FIGURE 64 Permeate flux during experiment 2.

APPENDIX 6/2

PERMEATE FLUX DURING THE EXPERIMENTS 1-4 AND THE REFERENCE EXPERIMENTS 1-2.

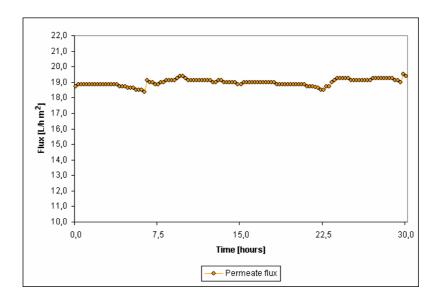


FIGURE 65 Permeate flux during experiment 3.

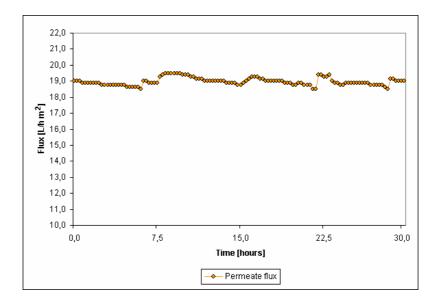


FIGURE 66 Permeate flux during experiment 4.

APPENDIX 6/3

PERMEATE FLUX DURING THE EXPERIMENTS 1-4 AND THE REFERENCE EXPERIMENTS 1-2.

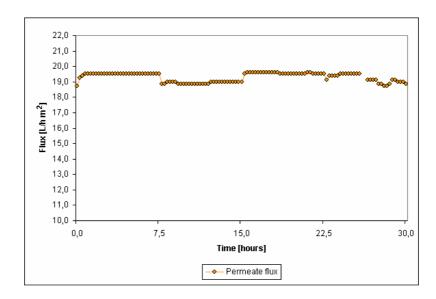


FIGURE 67 Permeate flux during reference experiment 1.

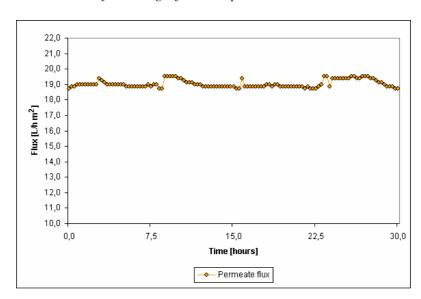


FIGURE 68 Permeate flux during reference experiment 2.

APPENDIX 7

LOCATIONS OF COOPERATION PARTNERS



FIGURE 69 Locations of cooperation partners.