Filtration characteristics in dead-end ultrafiltration of wwtp-effluent

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Filtration characteristics in dead-end ultrafiltration of wwtp-effluent

Proefschrift

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aan de Technische Universiteit Delft,
op gezag van de Rector Magnificus prof.dr.ir. J.T. Fokkema,
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geboren te Den Helder

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Jelle Roorda
's-Gravenhage, April 2004

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1 Ultrafiltration for advanced treatment of municipal wastewater

1.1 From wastewater treatment to water reclamation

1.1.1 Focus change in the treatment of wastewater

In nowadays-developed countries like the Netherlands, human wastewater was collected and used in agriculture, or disposed in streets and surface waters until the early 19th century. Around 1850 citizens became aware of the hygienic aspects of wastewater and initiated the collection and also the treatment of the wastewater (Asano and Levine, 1996). A new era started after World War II; due to the industrial growth and urbanisation wastewater caused serious environmental problems. In many countries the treatment of wastewater became compulsory by law around 1970. Initially, focused the treatment of the wastewater on the removal of oxygen consuming pollutants (ammonia and BOD) and was later on followed by removal of nutrients to decrease eutrophication of receiving water bodies. Nowadays, in most western countries the major part of the wastewater is treated in order to protect the water quality of the receiving rivers and lakes. In the near future the driving force may even shift towards the shortage of fresh water resources. The focus will change towards resource management. In the coming decennia the treatment of wastewater will develop into the reuse of wastewater constituents and the reclamation of the water.

Shortage of fresh water resources is mainly caused by a population increase, by changing lifestyles, by a decreased availability of conventional water resources, by drought and by more stringent environmental legislation. It is even expected that the shortage of water will finally limit economical and social growth (Mujeirigo, 2000;

Ødegaard, 2000). The above-mentioned periods, in which the focus on wastewater treatment changed, are summarised in table 1.1.

Table 1.1 Overview of focus change in treatment of wastewater (Ødegaard, 2000)

Period	Focus	Name of period
1850 - 1950	Hygiene	Sanitary engineering
1950 - 2000	Environment	Environmental engineering
2000 - 2050	Reuse	Water Environment Management

Until the end of the 20th century the use of water has been supply driven. It has been an abundant commodity in most places and water has been supplied in large quantities at a very low price. Due to a lack of fresh water resources the situation will very soon change in many regions. Therefore, the policy of water management will change from supply driven to demand driven. In a demand driven situation, the price of water will increase and even the extensive treatment of wastewater may turn out to be cost effective in order to produce the necessary amounts of clean water (Ødegaard, 2000).

Compared to other water resources, treated wastewater has some benefits that require more attention. The following benefits of the reuse of wastewater are commonly recognised (Mujeirigo, 2000):

- An additional contribution to water resources; either as a new water resource or as an alternative water resource that can be used for applications that do not require drinking water quality, leaving water with a good quality available for the urban water supply;
- A reduction of costs of the treatment and the disposal of wastewater; reuse will offer an economical advantage when the quality requirements for the reclaimed water are lower than the water quality standards for discharging of the effluent into surface water;
- A reduction of the pollutant load to surface water, when reuse involves agricultural, landscape or forest irrigation; irrigation with reclaimed water provides an opportunity for organic substances to be degraded through

biochemical processes in the soil into its mineral components; this may eventually be assimilated by plants;
- A reduction, postponement, or cancellation of new drinking water treatment facilities, with the positive consequence on natural water courses and water costs;
- The beneficial use of nutrients (nitrogen and phosphorous) in reclaimed water, when it is used for agricultural and landscape irrigation (golf courses);
- A considerably higher reliability and uniformity of the available water flows; urban wastewater flows are usually much more reliable than most rivers and streams in semi-arid areas.

1.1.2 Constituents in wwtp-effluent and technologies for advanced treatment

Advanced treatment of wastewater is applied for the removal of constituents, which can be grouped into four categories: (1) the residual organic and inorganic colloidal and suspended solids, (2) dissolved organic constituents, (3) dissolved inorganic constituents and (4) biological constituents. The potential effects of the residual constituents in effluent of wastewater treatment plants (wwtp-effluent) may vary considerably. Some effects of these constituents in wwtp-effluent are listed in table 1.2.

For each group of residual constituents the available treatment techniques are summarised here (Metcalf & Eddy, 2003; Mujeirigo and Asano, 1999). In Appendix 1-A a complete summary of the various treatment techniques is presented.

- The residual organic and inorganic colloidal and suspended solids: various filtration techniques, like multi-media (depth) filtration, surface filtration and membrane filtration; if necessary in combination with coagulation and flocculation;
- The dissolved organic constituents: adsorption techniques like activated carbon and ion exchange; reverse osmosis and other membrane filtration techniques;
- The dissolved inorganic constituents: chemical precipitation, ion exchange, ultrafiltration;
- The biological constituents: chlorination, ozonisation, UV-disinfection and membrane filtration (Kirkpatrick and Asano, 1986).

Table 1.2 Typical residual constituents found in wwtp-effluent and their impacts (Metcalf & Eddy, 2003)

Residual constituent	Effect
Inorganic and organic colloidal and suspended solids	
Suspended solids	- may cause sludge deposits or interfere with receiving water clarity
	- can impact disinfection by shielding organisms
Colloidal solids	- may affect effluent turbidity
Organic matter (particulate)	- may shield bacteria during disinfection, may deplete oxygen resources
Dissolved organic matter	
Total organic carbon	- may deplete oxygen resources
Refractory organics	- toxic to humans; carcinogenic
Volatile organic compounds	- toxic to humans; carcinogenic; form photochemical oxidants
Pharmaceutical compounds	- impact aquatic species (e.g. endocrine disruption, sex reversal)
Surfactants	- cause foaming and may interfere with coagulation
Dissolved inorganic matter	
Ammonia	- increases chlorine demand for disinfection
	- can be converted to nitrates and can deplete oxygen resources
	- with phosphorous, may lead to undesirable aquatic weed growth
	- unionised form toxic to fish
Nitrate	- stimulates algal and aquatic growth
Phosphorus	- stimulates algal and aquatic growth
	- interferes with coagulation
	- interferes with lime-soda softening
Calcium and magnesium	- increase hardness and total dissolved solids
Total dissolved solids	- interfere with agricultural and industrial processes
Biological	
Bacteria	- may cause diseases
Protozoan cysts and oocysts	- may cause diseases
Viruses	- may cause diseases

1.1.3 Examples of full-scale plants for water reclamation

In some parts of the world is the advanced treatment of wastewater already implemented in large-scale facilities. In arid and semi-arid areas the effluent of wastewater treatment plants is an essential alternative for the conventional water resources like ground- and surface water (STOWA, 2001). One well-known example is Water Factory 21 in Orange County, California (USA), where since 1976 the secondary effluent is treated in a series of treatment steps: flocculation, multi-media filtration, activated carbon adsorption, reverse osmosis and chlorination. The reclaimed water is recharged into the groundwater in order to stop salt intrusion with a total flow of 60,000 m^3 per day (Wehner, 1992). Recently also micro- and ultrafiltration were evaluated (Arviv *et al.*, 2002). Another example in the USA is found in St. Petersburg (Florida), where since 1972 almost 25% of the total effluent flow, about 65,000 m^3 per day, is reclaimed and used for irrigation (STOWA, 2001).

In Windhoek (Namibia) the wwtp-effluent is used as a resource for the potable water supply. About 24,000 m^3 of water per day is reclaimed by a series of treatment steps such as ozonisation (for disinfection), activated carbon (for removal of micropollutants) and finally ultrafiltration as a second disinfection step. Chlorine is added to prevent the growth of bacteria in the water supply and distribution system (Haarhoff and van der Merwe, 1996).

The indirect potable use of wwtp-effluent is applied in Belgium (Van Houtte *et al.*, 1998; Van Houtte and Verbauwhede, 2003). About 7,500 m^3 of effluent per day is treated and recharged into the groundwater aquifers. After a residence time of one to two months the recharged groundwater is used for the production of drinking water.

WWTP-effluent is used at the Peterborough Power Station (United Kingdom) for the production of boiler feed water. The effluent is purified with microfiltration, ion exchange and reverse osmosis (Murrer and Latter, 2003).

More examples of the advanced treatment and reuse can be found all over the world (Lazarova *et al.*, 2000; Lazarova *et al.*, 2001): in Israel (Brenner *et al.*, 2000; Soffer *et al.*, 2000), in Japan (Asano *et al.*, 1996; Maeda *et al.*, 1996; Ogoshi *et al.*, 2001), in Australia (Gibson and Apostolidis, 2001; Patterson, 2001) as well as in Europe (Arviv *et al.*, 2002; Lazarova *et al.*, 2000; Lazarova *et al.*, 2001).

1.2 Possibilities for water reclamation in the Netherlands

1.2.1 Wastewater treatment in the Netherlands

General

In the Netherlands, biological treatment of wastewater was initiated around 1970, with the Act on Pollution of Surface Waters (*Wet Verontreiniging Oppervlaktewater, WVO*). Until that moment the discharge of untreated or mechanically treated wastewater into surface water led to serious problems in the receiving rivers and lakes. The organic load and nutrients in the wastewater led to oxygen deficit in the surface water, leaving fishes and plants to die (Dirkzwager, 1997). At first, biological treatment of wastewater focused mainly on the reduction of organic oxygen consuming substances (Biochemical Oxygen Demand, BOD). From 1978 also ammonia had to be removed and the discharge standards for Kjeldahl-nitrogen (ammonia plus organically bound nitrogen) were defined. From 1985 onwards the prevention of eutrophication became important and standards for nutrients (phosphorus and nitrogen) were added. Nowadays, 98% of all houses are connected to the sewer system, and about 96% of the wastewater is treated in a wastewater treatment plant (CBS, 2003). In the year 2000 the total volume of wastewater treated in wwtp's in the Netherlands was about 2,100 Mm^3.

The wastewater transported to and treated in a wwtp consists of municipal and industrial (pre-treated) wastewater, and storm water. Although the quality of the untreated wastewater (influent) is specific for each wwtp, an average influent quality for all wwtp-influents in the Netherlands is presented in table 1.3. Also the average effluent quality, the discharge standards, the (potential) future standards (based on Maximum Tolerable Risk, MTR) and, for comparison, the composition of surface water of the river Meuse and Rhine are presented for COD, BOD, suspended solids (SS) and nutrients.

Table 1.3 Average concentrations of contaminants in influent, wwtp-effluent, river Meuse and Rhine in the year 2000 in the Netherlands, as well as the standards for effluent discharge and future standards.

Parameter		Influent[a]	WWTP-effluent[a]	Discharge standards[b]	Future standards[c]	River Meuse[d]	River Rhine[d]
COD	(mg O_2/l)	470	45	125	< 40	10	10[e]
BOD	(mg O_2/l)	180	6	20	< 5	2.1	< 1[e]
Suspended solids	(mg ss/l)	586	33	30	< 5	18.3	24.5
total-P	(mg P/l)	7	2	1 – 2	0.05 – 0.15	0.30	0.18
total-N	(mg N/l)	44	11	10 – 15	1 – 2.2	4.2[f]	3.2[f]

[a] Data 2000 (CBS, 2003); [b] VROM (1996); [c] COD and BOD (van der Graaf, 2003); SS, P, N values for Maximum Tolerable Risk (MTR), in: MinVenW (1998); [d] Data 2000, river Meuse at Eijsden and river Rhine at Lobith (RIZA/RIKZ, 2003); [e] Data 1999, location Lobith (RIZA/RIKZ, 2003); [f] $N_{Kjeldahl} + NO_3^- + NO_2^-$

Pathogenic microorganisms

Raw, untreated wastewater contains by its nature high concentrations of pathogenic microorganisms. In table 1.4 the concentration ranges of pathogenic microorganisms found in untreated wastewater, wwtp-effluent, groundwater and in surface water are presented. The influent has the highest concentration of pathogenic microorganisms; about 2 log units are removed in a wastewater treatment plant (wwtp). The concentrations in surface water are different for each location but generally much lower than in wwtp-effluent. Groundwater is hygienically reliable and has a maximum pathogenic concentration lower than 1 per litre.

Table 1.4 Range of concentrations of pathogenic microorganisms in influent, wwtp-effluent, surface- and groundwater in the Netherlands

Pathogenic microorganism		Influent[a]	WWTP-effluent[b]	Surface water[c]	Ground-water[d]
Total coliform	(amount/l)	$10^7 – 10^9$			< 1
E.Coli	(amount/l)		~ 10^6	$10^0 – 10^4$	< 1
Thermo tolerant coliform	(amount/l)		$10^5 – 10^7$	$10^4 – 10^5$	< 1
Faecal coliform	(amount/l)	$10^6 – 10^8$	~ 10^7		< 1
Faecal streptococcus	(amount/l)	$10^4 – 10^7$	$10^4 – 10^6$	10^3	< 1
Enteroviruses	(amount/l)	$10^3 – 10^4$	$10 – 10^3$		< 1
Giardia lamblia cysts	(amount/l)	$10^3 – 10^4$	$10 – 10^3$		< 1
Cryptosporidum oocysts	(amount/l)	$10^2 – 10^3$	$10 – 10^3$		< 1

[a] Metcalf & Eddy (2003); [b] Data 1997/1998 at wwtp Etten, in: STOWA (2001) and van der Graaf (1995); [c] Data 2000 (RIZA/RIKZ, 2003); [d] Data van Dijk (2003)

Micropollutants

Some typical micropollutants that are found in wastewater are pesticides, endocrine disrupters, residues of medicines and heavy metals. In table 1.5 a summary of some selected micropollutants and their concentration in wwtp-effluent and surface water is presented. For these constituents a wide range of concentrations can be found. Usually, the concentrations in wwtp-effluent are higher than in surface water.

Table 1.5 Range of concentrations of some selected micropollutants in wwtp-effluent and surface water in the Netherlands

Group of micropollutants	Found in	Concentration[a]	
		WWTP-effluent (ng/l)	Surface water (ng/l)
Fibrates and ß-blockers	Human medicine	< 10 – 9,710	< 0.5 – 3,100
Anti-epileptica		580 – 6,300	< 10 – 2,100
Analgetica		< 10 – 95,620	< 1 – 1,200
Oncolytica		< 6 – 60	< 5 – 17
Antibiotics		< 10 – 6,000	< 10 – 1,700
Anti-depressives		< 30 – 1,000	~ 0.01 – 30
Natural hormones	Hormones	< 0.3 – 11	< 0.3 – 7.3
Synthetic hormones		< 0.3 – 2.6	< 0.3 – 0.4
Alkylfenolen	Surfactants	< 450 – 2,200	< 50 – 4,100
Ftalaten	Plastics	< 1 – 20,000	< 1.9 – 5,000
Bisfenol-A		< 43 – 4,090	< 8.8 – 1,000

[a] Data of human medicines based on RIWA (2001), other data on RIZA/RIKZ (2002)

Table 1.6 shows the same information for heavy metals, including the future standards for the short-term (MTR) and for the long-term. In order to reach the future standards, only the concentrations of copper and zinc in wwtp-effluent have to be decreased. Generally, in surface waters the concentrations of heavy metals are lower than in wwtp-effluent.

Table 1.6 Average concentrations of heavy metals in wwtp-effluent and surface water in the Netherlands, as well as the future standards for effluent discharge

Parameter		WWTP-effluent[a]	Future[b]	River Meuse[c]	River Rhine[c]
As	(µg/l)	1.3		1.1 (NA)	1.7 (NA)
Cd	(µg/l)	0.2	2.0 (0.4)	0.24 (0.46)	0.06 (0.11)
Cr	(µg/l)	2.6	8.4 (2.4)	1.4 (11.3)	1.3 (4.4)
Cu	(µg/l)	8.4	3.8 (1.1)	5.1 (7.9)	4.1 (5.3)
Hg	(µg/l)	0.1	1.2 (0.07)	0.02 (0.04)	0.02 (0.04)
Pb	(µg/l)	4.4	6.3 (4.1)	5.3 (6)	2.9 (3.6)
Ni	(µg/l)	6	220 (5.3)	3 (11.2)	2.3 (4.8)
Zn	(µg/l)	50.8	40 (12)	30 (58)	15 (22)

[a] Data 2000 (CBS, 2003); [b] High concentrations: short-term ambitions (MTR); low concentrations between brackets: long-term ambition (MinVenW, 1998); [c] Average values in 2000, river Meuse at Eijsden, river Rhine at Lobith (RIZA/RIKZ, 2003); behind brackets the 90-percentile concentrations are given (10% of all samples had a higher concentration) (CIW, 2002)

1.2.2 Reclaimed water in the Netherlands

Wastewater can be (re)used as a water resource for various purposes including a whole range of less advanced to more highly advanced purposes. In Stowa (2001) a summary of the various reuse options is presented in the 'Compendium for use of wwtp-effluent', focusing on the quality aspects, the production costs, the quantity and other important aspects. In this section the various options for reuse are presented, referring to water quantity figures of 1996/1997, which are at this moment the most recent overall figures. Therefore, in this section the total annual flow of wwtp-effluent of the year 1997 is used as a comparison, which was 1,700 Mm^3 (2,100 Mm^3 in the year 2000). The following reuse options for wwtp-effluent are considered for the Netherlands:

- Process water in the industry. Process water includes a whole range of applications from water for cleaning purposes to ultra pure water. In 1996, Dutch industries used a total volume of process water of about 2,500 Mm^3. In the production of electricity an additional volume of about 6,000 Mm^3 was used, mainly as cooling water (CBS, 2003);

- Household water. This is defined as water with a lower quality than drinking water for non-potable use in households. When using dual water distribution systems, household water may account for maximum 50% of the total water demand, including toilet flushing and washing of clothes. Large-scale application is not expected, but if implemented it may account for 275 Mm^3 a year. Household water must be hygienically safe, but the quality standards are not yet available. Almost all Dutch projects on the implementation of household water were cancelled in 2002 due to hygienic problems;
- Agricultural water. Water for this purpose might be used for irrigation on land or in greenhouses. In the Netherlands, farmers use about 400 Mm^3 for this purpose. Additionally, about 200 Mm^3 per year is used for watering cattle. In all cases disinfection of the treated wastewater is necessary;
- Replenishment of natural and recreational waters. Usage for landscaping and recreation ponds might be a good option. Also groundwater recharge is considered as a viable option. The total quantity of the water for natural use is maximised by the total available volume of effluent. About 1,700 Mm^3 is available as wwtp-effluent, but even more water could be used (for comparison: at least an additional 1,000 Mm^3);
- Drinking water, the ultimate challenge for technologists. In areas lacking clean water resources like groundwater and (clean) surface water, reclaimed water can be used as a source for drinking water. In the Netherlands drinking water accounted for 1,270 Mm^3 in 1996. In the coming years wwtp-effluent will not be used as a water resource for drinking water, because cleaner resources are widely available. But nearby, in Belgium (Van Houtte and Verbauwhede, 2003), wwtp-effluent is indirectly used for the production of drinking water.

For the Dutch situation a figure of the volume of available wwtp-effluent versus the maximum volume of used water for various applications is shown in figure 1.1, using data from 1996/1997. Figure 1.1 shows that household and agricultural water account for a relatively small amount of the total volume and both industrial and natural use account for large quantities of water.

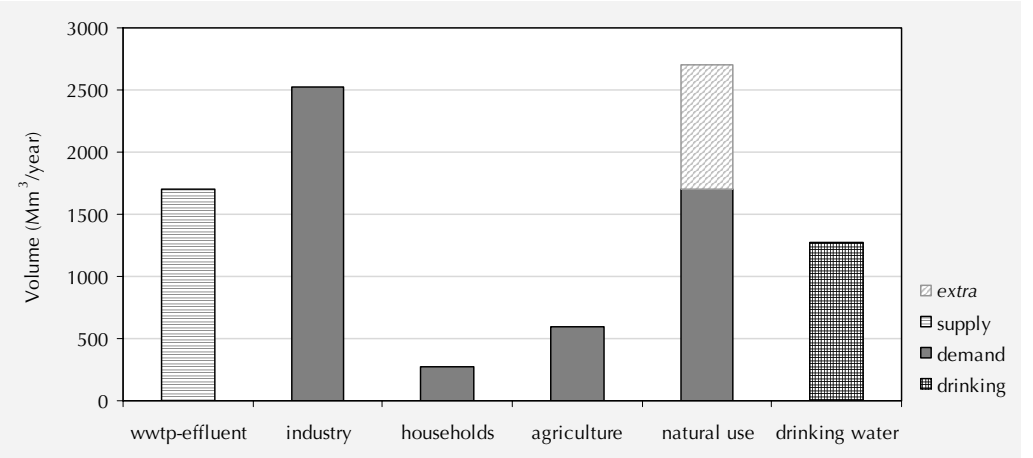

Figure 1.1 **Volume of wwtp-effluent in the Netherlands together with the volume of water for possible applications using data from 1996-1997; the amount of water used in industry is presented without cooling water for the production of electricity, which accounts for an additional 6,000 Mm^3 per year (CBS, 2003; STOWA, 2001)**

1.3 Dead-end ultrafiltration of wwtp-effluent

1.3.1 Ultrafiltration membranes

Classification of membrane filtration processes

Membrane filtration is a filtration technique in which a membrane acts as a selective barrier between two phases (Mulder, 1997). As a result of a driving force across the membrane, components are transported towards the membrane surface, where some components pass the membrane and others are retained at the membrane surface. Membrane processes are available for numerous applications, each with its own driving force and separation characteristics:

- Pressure driven processes, e.g. micro-, ultra- and nanofiltration, reverse osmosis;
- Concentration driven processes, e.g. gas separation, pervaporation, dialysis;
- Temperature driven processes, e.g. membrane distillation;
- Electrically driven processes, e.g. electrodialysis.

Pressure driven membrane processes are subdivided in microfiltration, ultrafiltration, nanofiltration and reverse osmosis. In table 1.7 a summary is presented of the main characteristics of these processes.

Table 1.7 Membrane filtration and application for water treatment (Mulder, 1997)

Membrane process	Pressure (bar)	Pores (nm)	Removable components
Microfiltration	0.1 – 2	100 – 1,000	Suspended solids, bacteria
Ultrafiltration	0.1 – 2	10 – 100	Macromolecules, viruses, proteins
Nanofiltration	4 – 20	1 – 10	Micropollutants, bivalent ions (Ca^{2+}, Mg^{2+}, SO_4^{2-}, CO_3^{2-})
Reverse Osmosis	10 – 30	0.1 – 1	Monovalent ions (Na^+, K^+, Cl^-, NO_3^-), hardness

Figure 1.2 gives a comparison of the size of the constituents found in wastewater and the operating size ranges for membrane processes, including conventional depth filtration (multimedia or deep-bed).

Ultrafiltration versus microfiltration membranes

Microfiltration and ultrafiltration membranes are operated under similar process conditions, but differ in pore size characteristics (Durham *et al.*, 2001; Kunikane *et al.*, 1995; Wakeman and Williams, 2002; Wiesner and Aptel, 1996). An ultrafiltration membrane is, due to its smaller membrane pores, better capable to remove small components than a microfiltration membrane. A complete rejection of viruses is found for ultrafiltration membranes, whereas microfiltration membranes do not completely remove viruses (Madaeni *et al.*, 1995; Madaeni, 1999). In practice, also other differences related to the pore diameter of the membranes are found, especially in the use of microfiltration or ultrafiltration prior to reverse osmosis. Pre-treatment of reverse osmosis feedwater with ultrafiltration shows lower operating pressures across reverse osmosis membranes and longer cleaning intervals than with microfiltration (Kim *et al.*, 2002; Tchobanoglous *et al.*, 1998).

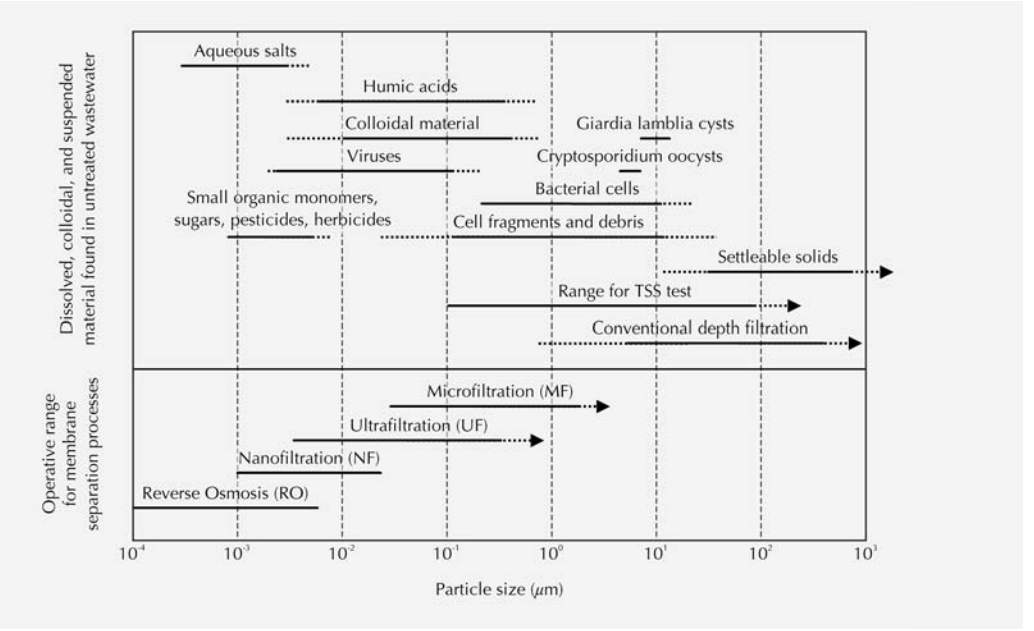

Figure 1.2 Comparison of the size of the constituents found in wastewater and the operating size ranges for membrane technologies; the operating size for conventional depth filtration is also shown (Metcalf & Eddy, 2003)

In the research described in this dissertation ultrafiltration membranes are used because of their good capability for the removal of effluent constituents and the complete removal of viruses compared to microfiltration membranes. Also the similarity in process conditions (and related costs per m^3 of treated feedwater) has been taken into account.

Ultrafiltration membranes for water treatment

The ultrafiltration membrane separates wwtp-effluent in a purified water flow called permeate and a concentrated flow called concentrate or retentate. Membranes typically consist of a porous support layer (100 μm) and a thinner toplayer of 0.1 to 1.0 μm (Mulder, 1997). Most membranes used in water treatment are organic membranes that are made of polypropylene, cellulose acetate, aromatic polyamides, or thin-film

composite (TFC). Inorganic membranes include additional layers of especially aluminiumoxide (Al_2O_3) and zirconiumoxide (ZrO_2).

The term membrane module is used to describe a complete unit comprised of membranes, pressure support structure, feed inlet, concentrate outlet and an overall support structure. The principal types of membrane modules used for wastewater treatment are (Aptel and Buckley, 1996; Mulder, 1997):

- Tubular membranes; having an internal diameter larger than 3 mm, which are bundled in a module;
- Hollow fiber or capillary membranes; having an internal diameter of less than 3 mm, which are bundled in a membrane module with hundreds to thousands of fibers;
- Spiral wound membranes are flat membranes wound around a spacer;
- Plate and frame membranes, comprised of a series of flat membrane sheets and support layers.

The flow direction in tubular and hollow fibre membranes can be inside-out or outside-in. In inside-out mode the feedwater flows from the inside of the membrane tube to the outside of the tube and the cleaned water (permeate) is collected. The outside-in configuration shows the opposite flow direction.

Definitions

The most common terms that are used in ultrafiltration processes are shortly described in this section. An important property of a membrane is its flux, which is defined as the permeate volume (or mass) through the membrane per unit of membrane area. The permeate flux or simply the flux J through the membrane is given by the general equation 1.1 (Mulder, 1997) in $m^3/m^2.s$. In practice the flux J is represented as litre filtered volume per m^2 membrane area per hour as $l/m^2.h$. The fluxes in dead-end ultrafiltration for the treatment of effluent are in the range of 50 to 200 $l/m^2.h$, depending on the Trans Membrane Pressure.

$$J = \frac{dV}{dt} \cdot \frac{1}{A_{memb}} \qquad \text{(eq. 1.1)}$$

where J = flux ($m^3/m^2.s$)
V = filtered volume (m^3)
t = time (s)
A_{memb} = membrane area (m^2)

The pressure difference over a membrane is called the Trans Membrane Pressure (TMP) and is the difference between the pressures at the feedwater side and the pressures at the permeate side. The relationship between flux J and TMP is defined by a modified form of Darcy's law (Wiesner and Aptel, 1996) and is introduced in equation 1.2.

$$J = \frac{\Delta P}{\eta_T \cdot R_{tot}} \qquad \text{(eq. 1.2)}$$

where ΔP = pressure difference, TMP (N/m^2, Pa),
η_T = dynamic viscosity ($N.s/m^2$, Pa.s)
R_{tot} = total resistance over membrane (1/m)

The dynamic viscosity η_T is related to the feedwater temperature T (°C). In this dissertation the following empirical relationship is used (Huisman, 1996):

$$\eta_T = \frac{497 \cdot 10^{-3}}{(T + 42.5)^{1.5}} \qquad \text{(eq. 1.3)}$$

Another characteristic of a membrane is its selectivity. Selectivity can be expressed as the retention R that is defined in equation 1.4. When solutes are completely retained by the membrane, the membrane has a retention of R = 1. The term retention is especially used in nanofiltration and reverse osmosis processes.

$$R = 1 - \frac{c_p}{c_f} \qquad \text{(eq. 1.4)}$$

where R = retention (-)
c_p = concentration in the permeate (kg/m^3)
c_f = concentration in the feedwater (kg/m^3)

1.3.2 Filtration and fouling mechanisms

Membrane fouling

During membrane filtration some constituents of the feedwater deposit on the membrane surface and/or in the membrane matrix. This retention process is often referred to as fouling of the membrane and causes a decrease of the flux. The common definition of membrane fouling is provided by the International Union for Pure and Applied Chemistry (IUPAC), which defined fouling as (Koros *et al.*, 1996): '*Fouling is the process resulting in loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surfaces, at its pore openings, or within its pores*'. Although other definitions exist[1], the definition of IUPAC is used in this dissertation.

The easily removable part of the retained material is called the reversible part of the fouling layer, the remaining part is called the irreversible fouling layer. The feedwater constituents that are retained on or in the membrane surface are called foulants. The retention of feedwater constituents causes an increase of the total resistance over the membrane, resulting at a constant TMP in a decreased flux. The decrease in flux that is found during membrane filtration is schematically drawn in figure 1.3.

[1] Mulder (1997) gave a second definition of fouling and relates it to the deposition of material: 'The (ir)reversible deposition of retained particles, colloids, emulsions, suspensions, macromolecules, salts etc. on or in the membrane'; Wiesner and Aptel (1996) gave a third definition and define an irreversible flux reduction as fouling: 'A reduction in permeate flux that cannot be reversed'

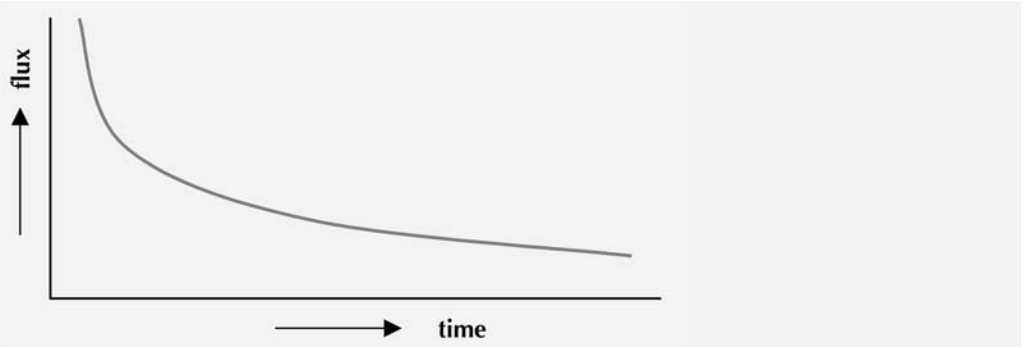

Figure 1.3 Flux development in time due to the retention of feedwater constituents during ultrafiltration at constant a Trans Membrane Pressure

Fouling mechanisms

Feedwater constituents that are retained can be found on several places near the membrane surface. Essentially, five so-called 'fouling mechanisms' can be distinguished, each mechanism contributes to the total resistance over the membrane. These fouling mechanisms are schematically drawn in figure 1.4 (van den Berg, 1988):

- Adsorption inside the membrane pores (R_a);
- Blocking of the membrane pores (R_p);
- High concentration of foulants near the membrane, concentration polarisation (R_{cp});
- Deposition on the membrane surface forming a cake layer (R_c);
- Compression of the cake layer (R_{cc}) (*not shown in figure 1.4*).

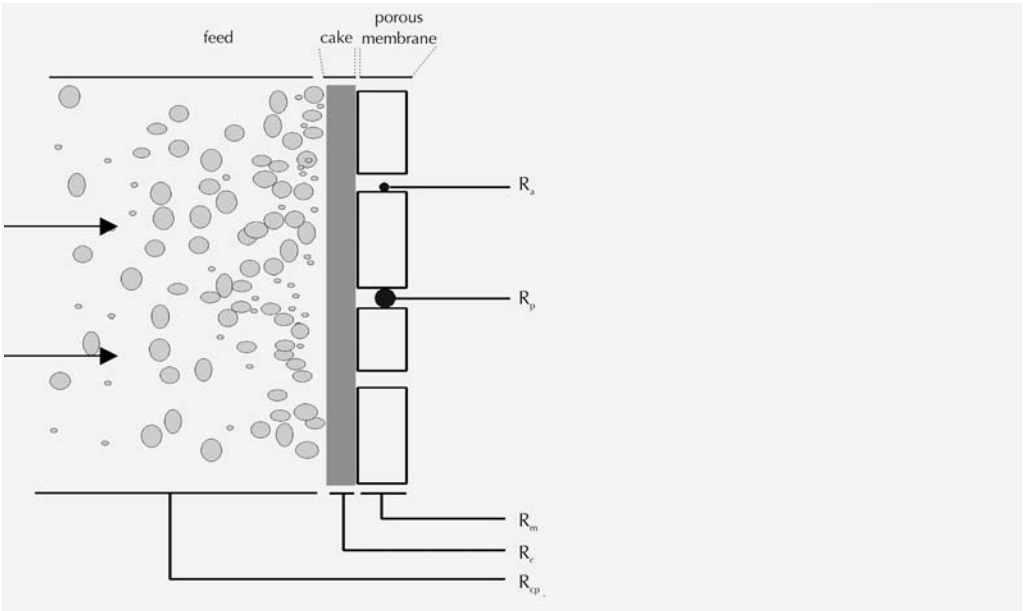

Figure 1.4 **The resistances of a fouled membrane by various fouling mechanisms, the driving force is from the left to the right: R_a = adsorption, R_p = pore blocking, R_m = initial membrane resistance, R_c = cake filtration, R_{cp} = concentration polarisation (van den Berg, 1988)**

During membrane filtration these mechanisms may occur simultaneously. The initial membrane resistance is mainly determined by the average pore diameter and porosity of the membrane. According to data obtained in the research described in this dissertation, the total resistance may exceed the membrane resistance up to three times under conditions in practice (Roorda and van der Graaf, 2000).

1.3.3 Configuration and process design

General information

In figure 1.5 a schematic drawing of the ultrafiltration process for the treatment of wwtp-effluent is given, showing the influence of four different aspects on the filtration characteristics. Firstly, the properties of the feedwater (wwtp-effluent) influence the membrane filtration process. The properties of the feedwater can be changed by pre-treatment either with physical processes or with chemical processes. Physical processes include pre-filtration (Botes *et al.*, 1998; Bourgeous *et al.*, 2001; Drage *et al.*,

2001; van der Graaf and van Nieuwenhuijzen, 1998), chemical processes include pH-adjustment, precipitation-coagulation-flocculation (Adin *et al.*, 1998; Al-Malack and Anderson, 1996; Doyen *et al.*, 2002; Minegishi *et al.*, 2001), adsorption on activated carbon (Park *et al.*, 1999; Seo *et al.*, 1996; Snoeyink *et al.*, 2000) and disinfection (Cornelissen, 1997; Milisic and Bersillon, 1986; Wakeman and Williams, 2002). By coagulation a pre-coat layer can be formed on the membrane surface. The pre-coat layer may act as a secondary filtration layer that may result in an increased performance of the membrane process (Galjaard *et al.*, 2001; Guigui *et al.*, 2002; Wiesner and Laîné, 1996). In the current research is pre-treatment examined for the reduction of membrane fouling.

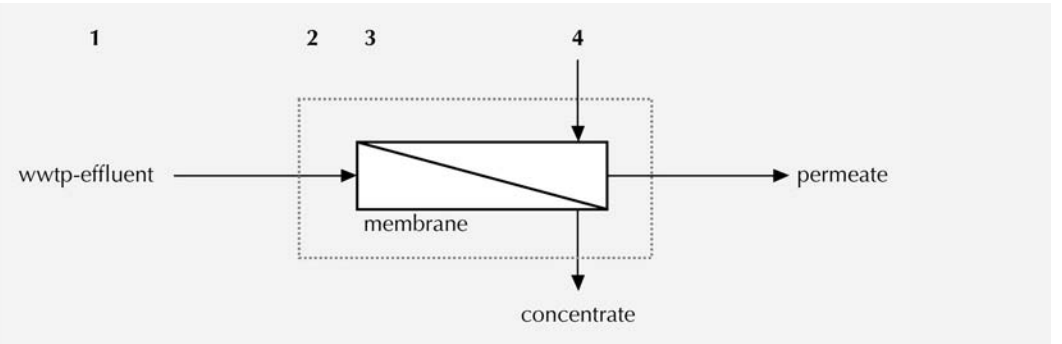

Figure 1.5 **Schematic drawing of a membrane process and factors that influence the filtration characteristics: 1 - Feedwater properties; 2 - Process configuration; 3 - Membrane characteristics; 4 - Cleaning methods**

Secondly, the process configuration influences the membrane filtration process. In cross-flow systems the concentrate is constantly transported with a recirculation loop, as is shown in figure 1.6 (a). In dead-end systems the total volume of the feedwater passes the membrane, leaving all components that are larger than the membrane pores in or on the membrane material (see figure 1.6 (b)). Cross-flow systems are widely used (Al-Malack and Anderson, 1996), but these systems use more energy than dead-end configured systems (Parameshwaran *et al.*, 2001). Therefore is a dead-end configuration used in the research presented here. Other parameters that influence the process configuration like the TMP, temperature, etc., may also be changed.

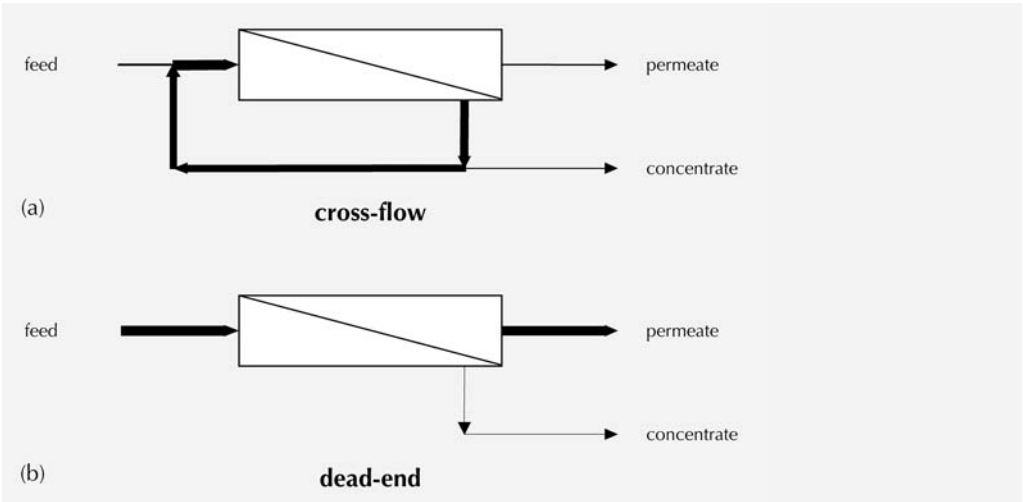

Figure 1.6 **Typical operational modes for ultrafiltration membrane processes: (a) cross-flow configuration, and (b) dead-end configuration**

Thirdly, the filtration behaviour is influenced by the membrane characteristics. Generally, minimal membrane fouling is found for membranes with a narrow pore size distribution, with a hydrophilic rather than a hydrophobic surface and with a negative surface charge (Cornelissen, 1997; Fane and Fell, 1987). In the current research the membrane is not modified, but commercially available membranes have been used in the filtration experiments.

The fourth aspect of membrane filtration is the cleaning of the membrane, which is described in the next section.

Methods for removal of retained material

One way to remove a layer of retained material is by cross-flushing of the membrane. If this is done regularly, it is called forward flushing. The effect of a forward flush may be improved by the addition of air bubbles, and is called AirFlush® (Verberk *et al.*, 2002).

Another commonly applied method for the removing of retained material is back flushing. In this case the flow is reversed and permeate is flushed through the membrane pores. As a result, the retained material in the membrane pores and on the

membrane is released, lifted up and is flushed out of the membrane module. If components are strongly adsorbed onto the membrane, back flushing is usually not very effective. Typical back flush periods of 30 to 60 seconds at every 10 to 30 minutes are mostly found to be effective during filtration (Mulder, 1997). By regular back flushing during ultrafiltration under constant TMP a typical curve is found, which is drawn in figure 1.7 (black curve). Initially the flux decreases, but after a back flush the flux is increased to its initial value.

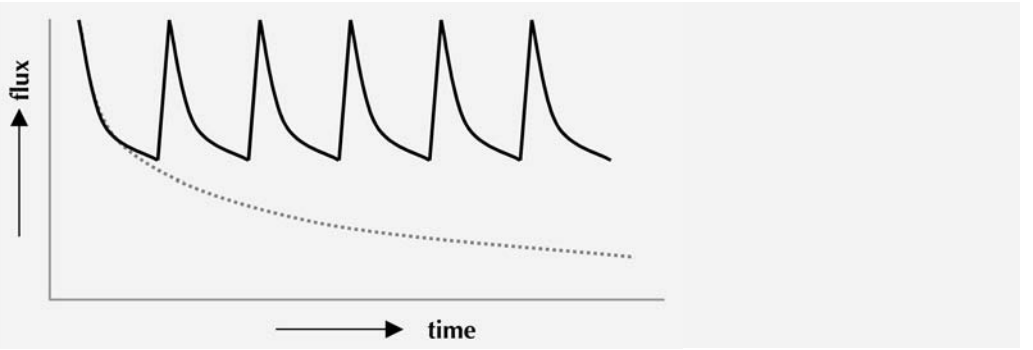

Figure 1.7 **Effect of a back flush on the flux development during dead-end ultrafiltration at a constant TMP: the dotted line shows a continuous flux decrease without back flushing of the membrane, the black line shows a decrease in flux followed by a flux increase due to a back flush; the average flux is higher in the latter**

Relatively new cleaning methods for ultrafiltration membranes are ultrasound-associated cleaning (at 45 kHz) (Chai *et al.*, 1999) or vibration (50-1000 Hz) of the module (Vigo *et al.*, 1993). However, these cleaning methods are not yet applied on full-scale.

Finally, chemicals might be used to displace the foulants, to dissolve the foulants or to chemically modify the foulants. The concentration of the chemicals and the cleaning time are important parameters for efficient use of a chemical cleaning procedure. In order to prevent membrane degradation during a chemical cleaning, the chemical properties of the membrane should be known. Effective cleaning must inhibit the redeposition of the foulants back on the membrane surface. The chemicals

that are used for cleaning can be classified in the following way (Zeman and Zydney, 1996):

- Acids, which are used to dissolve calcium salts and metal oxides;
- Alkalis, which are used to remove silica, inorganic colloids and many biological/organic foulants. The working mechanisms consist of neutralization of acidic material, saponification (hydrolysis) of fat and oil and dispersion/emulsification of colloidal material;
- Surfactants, which are used to displace foulants, to emulsify oils and to dissolve hydrophobic foulants. Surfactants can possess neutral (non-ionic) groups, negatively charged (anionic) groups or positively charged (cationic) groups. For effective cleaning a good balance between hydrophobic and hydrophilic characteristics is required;
- Oxidants, which are used for oxidation of organic material and bacteria (disinfection);
- Sequestrates (chelating agents), which are used for removal of metal cations from a solution;
- Enzymes, which are used to degrade foulants. Proteases are used to degrade proteins, amylases are used to degrade polysaccharides and lipases hydrolyse fatty acids.

Stable ultrafiltration performance

Long-term stable operation of an ultrafiltration process for the treatment of wwtp-effluent may only be met if both the filterability of the effluent and the reversibility of the fouling layer are taken into account. In figure 1.8 the development of the total resistance of the (fouled) membrane or the Trans Membrane Pressure against time is shown at constant flux for two situations. In (a) the resistance (or TMP) increases only slightly during filtration and the additional resistance caused by fouling of the membrane is completely removed during a cleaning procedure. In (b) the increase in resistance is much higher and the removal of the fouling layer during the applied cleaning procedure is insufficient. This is presented as the resistance at A_3' is higher than the initial resistance A_3. The repeating effect is shown in the remaining part of the curve, whereby A_3'' is higher than A_3'.

Stable ultrafiltration performance will only be found if the reversibility of the fouling layer is complete. This is shown in figure 1.8 (a) as the initial resistance A_1 is the same as the resistance after filtration and cleaning A_1'. This is continued for the following filtration steps (A_1'' is the same as A_1).

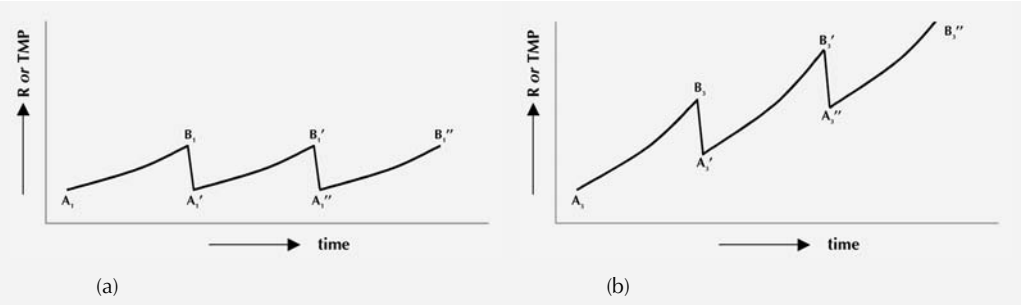

Figure 1.8 **The development of the resistance and/or TMP during ultrafiltration of wwtp-effluent; line A to B shows the increase in resistance during filtration and line B′ to A shows the decrease of resistance during cleaning of the membrane; in (a) the fouling layer is completely removed by cleaning; the following filtration step starts again at the initial resistance ($A_1'=A_1$); (b) shows a higher increase in resistance (*i.e.* lower filterability) with a subsequent cleaning procedure that is insufficient to remove the fouling layer completely**

1.3.4 Theoretical description of membrane filtration behaviour

The wwtp-effluent contains of a broad range of constituents, varying in both chemical and physical conditions. The composition of the effluent is even more complex when considering the continuous variation in the concentrations and the nature of the constituents. A theoretical description of filtration behaviour has to take these aspects into account. In the current research other criteria for a description of the filtration process are the mode of operation (dead-end filtration) and the applied membranes (ultrafiltration membranes).

In most references that review fouling phenomena (van den Berg and Smolders, 1990; Bowen and Jenner, 1995; Fane and Fell, 1987; Jönnson and Trägård, 1990; Matthiasson and Sivik, 1980; Mulder, 1995; Wessling, 2001) the focus is on cross-flow systems and/or reverse osmosis of single-solute feedwater solutions. Bowen and Jenner (1995) reviewed the theoretical filtration models for colloidal and fine particle dispersions, in both cross-flow and dead-end operational mode. In the gel

polarization model it is suggested that when the concentration at the membrane surface increases, the macro-solute reaches its solubility limit and precipitates on the membrane surface and forms solid gels. Solutes that do not precipitate are not taken into account. The osmotic pressure model describes flux behaviour when solutes do no not precipitate or gelate. However, in ultrafiltration processes the osmotic pressure is negligible. These two models can be modified in various ways (van den Berg and Smolders, 1990; Bowen and Jenner, 1995), but are still only applicable for well-defined solutions.

Particle interactions are taken into account by a different group of filtration models. Of special interest is the work by Wessling (2001) on stochastic modelling, in which fouling of microfiltration membranes was described as a function of nano-particles (like proteins). Although these particles are much smaller than the membrane pores, it was found that these particles could foul the membrane heavily. The stochastic modelling showed that interaction of the particles might lead to the formation of agglomerates of particles forming bridges over a pore. Verification of the theoretical results was found to be very difficult. Also Bowen and Jenner (1995) described various particle interaction models.

Ultrafiltration performance can also be interpreted by a resistance-in-series relationship (Bowen and Jenner, 1995). The resistance-in-series modelling is based on Darcy's law (see equation 1.2) and neglects the osmotic pressure term (as is the case in ultrafiltration). Each deposition layer (inside, outside the membrane pores, adsorption, etc.) relates to an additional resistance. The sum of all resistances refers to the total membrane resistance (as defined in Darcy's law).

In the current research the approach introduced by Hermia (1982) is used for the description of filtration phenomena in dead-end ultrafiltration. At constant pressure and assuming straight cylindrical pores and laminar flow through the pores, the filtration process is described for pore sealing (complete blocking, *i.e.* no flow at all through that pore), complete blocking with superimposition (intermediate blocking), deposition of particles inside the membrane pore (standard blocking) and finally cake filtration. Below, the formulae derived by Hermia (1982) are presented.

For complete blocking it is assumed that (a) each particle reaching the membrane participates in the blocking phenomenon by pore sealing, and (b) particles

are not superimposed one upon the other. Summarised, each particle blocks one pore and forms a single particle layer. These assumptions lead to the following equation (1.5) as the relation between the total permeate volume V and the total filtration time t:

$$K_b \cdot V = Q_o \cdot \left(1 - e^{-K_b \cdot t}\right) \qquad \text{(eq. 1.5)}$$

and

$$\frac{d^2t}{dV^2} = K_b\left(\frac{dt}{dV}\right)^2 \qquad \text{(eq. 1.6)}$$

where $K_b = u_o \cdot \sigma$

K_b = constant for complete blocking (s^{-1})

u_o = filtrate velocity (m/s)

σ = blocked area per unit filtrate volume (m^{-1})

Q_o = initial flow (m^3/h)

For intermediate blocking it is assumed that when a particle enters a pore it is completely blocked, but particles are able to superimpose one upon the other and a multi-layer of particles might occur. This leads to the following equation (1.7) as the relation between the total permeate volume V and the total filtration time t:

$$K_i \cdot V = \ln\left(1 + K_i \cdot Q_o \cdot t\right) \qquad \text{(eq. 1.7)}$$

and

$$\frac{d^2t}{dV^2} = K_i\left(\frac{dt}{dV}\right)^2 \qquad \text{(eq. 1.8)}$$

where $K_i = \dfrac{\sigma}{A_{memb}}$

K_i = constant for intermediate blocking (m^{-3})

For the standard blocking filtration law the equations 1.9 and 1.10 are derived, assuming that the pore volume decreases proportionally to filtrate volume by particle deposition on the pore walls. This implicates a reduction of the pore diameter, only inside the membrane pores. The relation between the total permeate volume V and the total filtration time t is shown in equation 1.9 and 1.10. As can be seen, the standard blocking filtration law results in a linear relationship between t and t/V.

$$\frac{K_s}{2} \cdot t = \frac{t}{V} - \frac{1}{Q_o} \qquad \text{(eq. 1.9)}$$

and $$\frac{d^2t}{dV^2} = K_s \left(\frac{dt}{dV} \right)^{\frac{3}{2}} \qquad \text{(eq. 1.10)}$$

where $K_s = \dfrac{2 \cdot C}{L \cdot A_o}$

K_s = constant for standard blocking (m^{-3})
C = volume of solid particles retained per unit filtrate volume (-)
L = membrane thickness (m)
A_o = initial active filter membrane surface (m^2)

Finally, Hermia (1982) derived for cake filtration the filtration laws, assuming resistance in series and a constant superimposition of particles. These assumptions lead to the following equation (1.11 and 1.12) as the relation between the total permeate volume V and the total filtration time t. As can be seen, the cake filtration law results in a linear relationship between V and t/V:

$$\frac{K_c}{2} \cdot V = \frac{t}{V} - \frac{1}{Q_o} \qquad \text{(eq. 1.11)}$$

and $$\frac{d^2t}{dV^2} = K_c \qquad \text{(eq. 1.12)}$$

where $K_c = \dfrac{\alpha \cdot \gamma \cdot s \cdot \eta}{A_{memb}{}^2 \cdot \Delta P \cdot (1 - m \cdot s)}$

K_c = cake filtration constant ($s.m^{-6}$)
α = specific cake resistance (m/kg)
γ = filtrate density (kg/m^3)
s = mass fraction of solids (-)
m = mass ratio of wet to dry cake (-)

Similar derivations haven been presented for constant flux filtration (Hlavacek and Bouchet, 1993), as well as for varying flux and TMP (Agustin Suarez and Veza, 2000). These are not used in the current research.

Madaeni *et al.* (1995) used the standard blocking law and the cake filtration law successfully to distinguish between particle deposition within the membrane and cake filtration on the membrane surface in unstirred and stirred batch microfiltration of virus suspensions (without *E.Coli*). Unstirred operation was related to dead-end filtration. The stirred cell operation was done at 400 rpm, resulting in similar results as a thin channel cross-flow cell at a Reynolds number of about 2200. The microfiltration membranes were hydrophobic Millipore (GVHP) membranes (0.22 μm). The ultrafiltration membranes were polysulfone Amicon (PM30) membranes (MWCO 30 kDa). The bacteria created a much higher fouling layer resistance, indicating blocking and pore obstruction. The viruses probably adsorbed on and inside the membrane.

Lojkine *et al.* (1992) reviewed cross-flow microfiltration of cell suspensions, focusing mainly on models for flux prediction, emphasising on particle size effects. The main focus was the application of the standard blocking filtration law and the cake filtration law. Various models based on the film theory were also reviewed, but these were mainly useful for cross-flow systems. Lojkine *et al.* (1992) stated that models have various drawbacks. They are often derived for dilute solutions of rigid, spherical and neutrally buoyant particles. The following effects are generally ignored:

- Particle interactions, especially in tubular pinch models (in cross flow systems);
- Particle-membrane interactions;
- Membrane fouling;
- Cake compression (particularly important for deformable particles);
- Feed properties, especially viscosity depending on concentration.

According to Lojkine *et al.* (1992), most reviewed research suggested that flux decreases with decreasing particle size (for polystyrene lattices, kaolin clay), although also the opposite has been found (colloidal silica, ~ *Aspergillus Niger* (reduction of particle size distribution)). Addition of anionic and cationic resins used as flocculants increased fluxes due to the reduction of a concentration polarisation layer and an

increased cake porosity. It might be possible that particle properties like shape, surface roughness and adhesion forces will have a more pronounced effect on filtration behaviour than particle size (Lojkine *et al.*, 1992).

1.4 Literature review on micro- and ultrafiltration of wwtp-effluent

Research on microfiltration and ultrafiltration for advanced treatment of wastewater focuses on performance, achievable permeate quality and costs but also on characterisation of foulants and filtration mechanisms. Advanced treatment of wastewater is performed as a polishing step before discharge, as well as a treatment technique before reuse of the water. Most experience has been obtained with microfiltration in a cross-flow configuration. In this section a review is presented of research on microfiltration and ultrafiltration of municipal effluent; advanced treatment of effluent from industrial wastewater treatment plants is not considered here.

1.4.1 Micro- and ultrafiltration as advanced treatment of municipal wastewater

Optimisation studies

About twenty-five years ago the first publications on ultrafiltration of effluent appeared in scientific journals. Inoué *et al.* (1982) presented the results of large scale research on ultrafiltration for advanced treatment of wastewater. One hollow fibre type UF module (polyvinyl alcohol; A_m = 70 m^2; pore diameter: 0.04 μm) and one tubular type UF module (polyacrylonitrile; A_m = 22 m^2; pore diameter: ~0.01 μm (MWCO 13 kDa)) were examined for the treatment of effluent (5.4 mg SS/l) for reuse as process water in industry. Cross-flow ultrafiltration performance was compared with the performance of (1) a micro strainer (21 μm) plus reverse osmosis and of (2) coagulation double-layer filtration plus reverse osmosis. Ultrafiltration produced clean water with a low turbidity that might be additionally treated with reverse osmosis. No comments were made on the filtration characteristics of the effluent.

The first experiments in the Netherlands on cross-flow microfiltration of wwtp-effluent were performed around 1990 (Oesterholt and Bult, 1993). On lab-scale the performance of a microfiltration unit (STORK Friesland; A_m = 0.1 m^2; pore diameter: 0.2 μm) was compared to a continuous sand filtration pilot plant (DynaSand; 0.7 m^2) for polishing of wwtp-effluent. Stable performance was found at a maximum permeate flux of 135 $l/m^2.h$ at a TMP of 1.0 bar. Permeate was free of bacteria and the total costs were estimated at €0.94 per m^3 for the cross-flow microfiltration unit and at €0.07 per m^3 for the continuous sand filtration plant.

These two early examples already show the capabilities of micro- and ultrafiltration as advanced treatment for **reuse** (Inoué *et al.*, 1982) and for **effluent polishing** (Oesterholt and Bult, 1993). Worldwide, the most common applications for reuse are found in agriculture for irrigation of crops (Messalem *et al.*, 2000; Vera *et al.*, 1998) and in industry for use as process water. Micro- and ultrafiltration are commonly used as a pre-treatment step that is followed by reverse osmosis[1] (Durham *et al.*, 2001; van Hoof *et al.*, 1998; Kim *et al.*, 2002, Naerssen *et al.*, 2002). Effluent polishing to improve the water quality (Duin *et al.*, 2000; van der Graaf *et al.*, 1999) focused on disinfection (Dorau, 1998; Gnirss, 2000; Jolis *et al.*, 1999; Langlais *et al.*, 1992 and 1993; Madaeni, 1998; Sadr Ghayeni *et al.*, 1998) and on P-removal before discharge of the polished effluent to the surface water (Dittrich *et al.*, 1996).

Most studies are optimisation studies in which the filtration properties of the effluent are improved by pre-treatment with coagulants (Decarolis *et al.*, 2001) or with multi-media filtration (Bourgeous *et al.*, 2001; Tchobanoglous *et al.*, 1998). The cleaning strategy is changed accordingly. Vial *et al.* (1992) suggested optimisation of the ultrafiltration process by applying an experimental matrix for maximum information with a minimum number of experiments and Agustin Suarez and Veza (2000) used the blocking filtration models to optimise the system.

Filtration characteristics

All studies on micro- and ultrafiltration of effluent showed declining membrane fluxes, but still many uncertainties regarding the fouling mechanisms exist (Vera *et al.*, 1998). Only a few studies showed some insight in occurring mechanisms. Decarolis *et*

[1] An example of direct reverse osmosis (without pre-treatment of the effluent with MF or UF) of effluent can be found in Abdel-Jawad *et al.* (2002)

al. (2001) studied dead-end ultrafiltration of tertiary effluent. The wastewater was treated biologically and polished with sand filtration. This sand-filtered effluent was used as the feedwater for ultrafiltration experiments (UF: capillary (0.8 mm); polyethersulfone; MWCO of 150 kDa). Also the impact of coagulation on ultrafiltration performance was investigated (0, 7 and 14 mg Fe^{3+}/l). Organic matter was measured as UV254 and Total Organic Carbon (TOC) and was only rejected for 4.1% (UV254) and 5.6% (TOC). It was concluded that, although organic matter passed the membrane for about 95%, organic substances seemed to play an important role in membrane fouling during ultrafiltration of wwtp-effluent. From other research (Bersillon, 1989; Wiesner and Aptel, 1996) it was concluded that organics like polyphenolic compounds, proteins, and polysaccharides bind together colloids that deposit on the membrane; this may cement the fouling layer to the membrane surface, which causes an increase in fouling layer resistance.

Next, in Decarolis *et al.* (2001), at increasing fluxes from 34 up to 102 l/m².h the membrane was increasingly fouled, leading to structural changes, probably compression, of the fouling layers. For cleaning a backflush was used, showing at a large backflush interval (> 30 minutes) a decrease in flux that was attributed to internal fouling and cake filtration. Finally, addition of ferric chloride improved the performance of the system, which might be attributed to the aggregation of colloidal particles (< 1 μm). The larger aggregates were supposed to result in a lower specific resistance of the cake layer. It was suggested that filtration characteristics are a function of both organics and colloids concentration.

Tchobanoglous *et al.* (1998) investigated dead-end ultrafiltration (polysulfone; 100 kDa) of secondary effluent (4.6 mg SS/l) and tertiary effluent (secondary effluent treated with continuous sand filtration, 1.9 mg SS/l)). The performance of an ultrafiltration unit treating effluent of two wwtp's was compared. The results showed that particles smaller than 1.0 μm had a greater effect on filtration characteristics than larger particles. These investigations were continued with the same effluents and extensively described by Bourgeous *et al.* (2001). It was shown that a thin cake layer developed for ultrafiltration of effluent after pre-filtration (tertiary effluent), while a thicker cake layer developed for secondary effluent. Fractionation experiments showed that pre-filtration removed only particles > 5 μm (more than 50% reduction), and particles > 20 μm for 100%. The thinner cake layer (pre-filtered effluent) showed a

better filterability and was completely removed by a regular back flush. The cake layer found for secondary effluent could not be removed completely with a backflush, because of clogging of the membrane fibers (with an internal diameter of 0.76 mm).

Bourgeous *et al.* (2001) found, by comparing results of experiments at three different wwtp's, that the filtration mechanisms were more related to the particle size distribution of the effluent than to the suspended solids load. Again small particles (< 5 μm) were found to play a more important role in the filtration characteristics than larger particles. It was suggested that this was caused by a larger amount of submicron particles for effluents with a relatively high fraction of particles smaller than 5 μm. Removing these small particles with a backflush is more difficult than removing larger particles. The remaining particles caused on the long run a decreased filterability, which is similar to an increased total membrane resistance. At an increased resistance the TMP was higher for the same flux, which caused the growth of a more embedded and less removable cake layer. The best performance was found for the ultrafiltration experiments with pre-filtered effluent (tertiary effluent).

Composition of foulants

Bourgeous *et al.* (2001) and Tchobanoglous *et al.* (1998) related the occurring fouling phenomena mainly to the particle size distribution. Decarolis *et al.* (2002) related it to organics interacting with colloids. Abdessemed *et al.* (2002) found that COD retention increased after coagulation with 20 mg Fe^{3+}/l, as well as the permeate flux. Ferric chloride stimulated flocculation and subsequently increased the cake porosity.

Jarusutthirak and Amy (2001) and Jarusutthirak *et al.* (2002; 2002a) described an extensive study on the characterization of effluent constituents that foul ultrafiltration membranes. Fouling characteristics were related to feedwater constituents as well as to membrane characteristics. The main foulants were detected as polysaccharides and/or amino sugars from colloids (Jarusutthirak *et al.*, 2002).

1.4.2 Membrane bioreactor

A relatively new treatment concept for wastewater treatment is the Membrane Bioreactor (MBR). In a MBR the biological process is combined with a physical process for solid liquid separation, combining a suspended growth bioreactor with a membrane filtration device. The membrane is used in a recirculation stream (cross-

flow configuration) or immersed in the bioreactor. The membrane acts as a barrier that retains colloidal and macromolecular materials, including bacteria. The bioreactors can then be operated with very high concentrations of biomass, typically up to 20 g/l. This process has been improved during the last 30 years and its effluent has a similar composition as wwtp-effluent after a polishing step with ultrafiltration (Ben Aïm and Semmens, 2002; Jefferson *et al.*, 2000). The MBR system has especially advantages for small-scale treatment of wastewater (Jefferson *et al.*, 2000), where compactness and effluent with a high quality are necessary (Rosenberger *et al.*, 2002). It is expected that its compactness and the high quality of its effluent will lead to an increase of MBR applications.

Similar as in effluent filtration, also in MBR-systems much is unclear about the filterability of the feedwater. A better understanding of the filterability will improve the MBR system and will make it economically more feasible (Rosenberger and Kraume, 2002). Filterability depends on the concentration of suspended extra cellular polymer substances (EPS) in the water phase. The impact of suspended solids concentration and sludge viscosity seems to be of minor importance. More research on this topic might lead to a better understanding of the factors influencing filterability (Evenblij and van der Graaf, 2003).

1.5 Research objectives

The scientific work described in this dissertation was carried out within the scope of the research project entitled 'Membrane filtration of effluent'[1]. The objective of the research project was to develop low-cost ultrafiltration systems for advanced treatment of municipal wastewater. This included pre-treatment of the effluent for optimal performance of the ultrafiltration plant, as well as post-treatment of the ultrafiltration

[1] The Delft University of Technology (TUDelft) initiated the research project 'Membrane filtration of effluent' in 1998 in cooperation with Rossmark Watertreatment (a Veolia Water company) and Witteveen+Bos consulting engineers; the Dutch Ministry of Economic Affairs supported the project financially (BTS 99112); from the early 1990s the research group of prof.ir. J.H.J.M. van der Graaf of the department of Sanitary Engineering (faculty Civil Engineering and Geosciences, TUDelft) focused on the advanced treatment of wastewater; various advanced treatment processes including multi-media filtration and membrane filtration have been subject of research (van der Graaf *et al.*, 1998; van der Graaf and van Nieuwenhuijzen, 1998)

permeate to meet reuse criteria. The performance of the ultrafiltration step was investigated in on-site pilot-scale studies and additional lab-scale research. The pilot-scale studies provided insight in the water quality of the produced water, the optimal process configuration and conditions and finally yielded insight in design characteristics for full-scale application of the ultrafiltration technology.

Within the scope of the research project, two main topics were determined for research on dead-end ultrafiltration of wwtp-effluent. The first topic dealt with the filterability of the wwtp-effluent and is the subject of this dissertation. The second topic dealt with the reversibility of the fouling phenomena during ultrafiltration, and is the subject of the ongoing research (see te Poele *et al.*, 2003).

The research described in this dissertation focused on the filterability of wwtp-effluent and the characteristics of dead-end ultrafiltration as a technique for advanced treatment of municipal wastewater. The objective of the current research is:

Determination of filtration characteristics of wastewater treatment plant effluent in dead-end ultrafiltration.

The following research topics are covered:

- Optimisation of performance of pilot-scale ultrafiltration plants, in terms of net fluxes and applied TMP's;
- Determination of filtration mechanisms;
- Determination of proper methods for the characterisation of effluent filterability;
- Development of a parameter for the prediction of filtration characteristics;
- Influence of pre-treatment on the filtration characteristics of wwtp-effluent;
- Translation of results from pilot-scale research into a fundamental understanding of filtration characteristics.

1.6 Structure of the dissertation

This thesis continues in chapter 2 with a description of the pilot-plant tests on dead-end ultrafiltration of wwtp-effluent that were performed at various wwtp's in the

Netherlands (Ede, Kaffeberg, Tilburg-Noord and Emmtec). The most important results are presented for the pilot-plant tests, focusing on the performance of the ultrafiltration pilot-plant installations. The influence of pre-treatment of the effluent is investigated, as well as the influence of various methods for membrane cleaning.

At some wwtp's more detailed experiments were performed, which are presented in chapter 3. These experiments were done with the pilot-plant for investigation of the filterability of the effluent and of the reversibility of the fouling layer. The effect of pre-treatment of the effluent on the filterability and on the reversibility was studied. Also the effect of various methods for membrane cleaning on the reversibility of the fouling layer was studied. With these experiments a rough estimate of the influence of pre-treatment on the filtration characteristics of wwtp-effluent could be made.

The filterability of the effluent was studied in detail with lab-scale ultrafiltration experiments. In these experiments a new parameter was developed for description of the filtration characteristics, the Specific Ultrafiltration Resistance (*SUR*). With the *SUR* it is possible to measure even small differences in filtration characteristics of the effluent.

The *SUR* was used for measurement of the filterability of size fractions of the wwtp-effluent. The effluent was fractionated by pre-filtration over filters with various pore diameters. The results are presented in chapter 5.

Many researchers applied theoretical models for the description of the ultrafiltration process and the fouling characteristics. In chapter 6 some existing cake filtration models based on the resistance-in-series concept are applied to the ultrafiltration data of (various size fractions of) wwtp-effluent.

Finally, the main findings of the current research are discussed and presented in chapter 7. The conclusions are drawn and some suggestions are made for improvement of future ultrafiltration installations that treat wwtp-effluent.

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2 Pilot-plant research at wastewater treatment plants in the Netherlands

2.1 Introduction

Pilot-plant tests were performed at three different wastewater treatment plants (wwtp's) that treat municipal wastewater (wwtp Ede, Kaffeberg, and Tilburg-Noord), and at one wwtp that treats wastewater of an industrial site (wwtp Emmtec). The pilot-plant tests were done for at least six months at each wwtp and aimed at determining the optimal configuration and process conditions of an ultrafiltration pilot-plant installation for the treatment of wwtp-effluent. The results of the tests were used for the design of full-scale installations for ultrafiltration of effluent at each wwtp.

Ultrafiltration has been applied for the treatment of raw water coming from many different sources. Anselme and Jacobs (1996) showed that each raw water source and each membrane type have specific and unique interactions. Therefore, pilot-plant tests are necessary to determine the optimal combination of process conditions and configuration of an ultrafiltration installation treating a (new) raw water source (Bersillon and Thompson, 1996; Metcalf and Eddy, 2003).

In this chapter the pilot-plant tests are presented. The experimental facilities that were used are presented in §2.2. The pilot-plant tests are described in the following sections per wwtp (Ede, Kaffeberg, Tilburg-Noord and Emmtec). The results presented in this chapter mainly focus on the performance of the ultrafiltration pilot-plant; some results that provide information for a better understanding of the filtration process are described in more detail. In the last sections the results are summarized (§2.7) and some general conclusions are drawn (§2.8).

2.2 Pilot-plant configuration

The pilot-plant installations consisted of an ultrafiltration pilot-plant and a multi-media filtration pilot-plant for pre-treatment of the wwtp-effluent. In-line coagulation was used in some cases as an additional pre-treatment technique. At each wwtp various combinations of pre-treatment techniques were investigated as to their effect on the filterability of the wwtp-effluent and on the reversibility of the fouling layer.

2.2.1 Ultrafiltration pilot-plant

The ultrafiltration pilot-plant is shown in figures 2.1 and 2.2. The membrane module is situated at the back of the pilot-plant, which could be placed both horizontally (2.2 (a)) and vertically (2.2 (b)). The total length of the horizontally placed module was 3 m with a diameter of 8 inch; the vertically placed modules were three modules of one meter each. The pilot-plant installation was automatically controlled with a PLC. A personal computer was combined with the PLC for data logging and direct data analysis (except at wwtp Ede). The pilot-plant installation was operated with the pc and the process configuration could be changed on-line. The installation could also be handled manually.

The ultrafiltration pilot plant was used in dead-end mode, although it was possible to use the cross-flow mode. The installation had a capacity of maximum 15 m^3/h, but mostly the flow did not exceed 5-10 m^3/h. Constant flow filtration as well as constant pressure filtration could be applied. Details about the various components can be found in the schematic drawing (Appendix 2-A). During the pilot-plant test the applied (constant) flux was changed between 40 to 150 $l/m^2.h$, with a subsequent TMP of 0.2 to maximum 1.5 bar. The membranes were back flushed for every 10 to 30 minutes with a flux of 225-300 $l/m^2.h$. A chemical cleaning was applied every two to six hours. Usually, a solution of sodium hypochlorite (100 ppm NaOCl) was used. Once a while, also other chemicals were used: HCl for dissolution of salts, and membrane cleaning chemicals like soaps and enzymatic cleaning agents for further membrane cleaning.

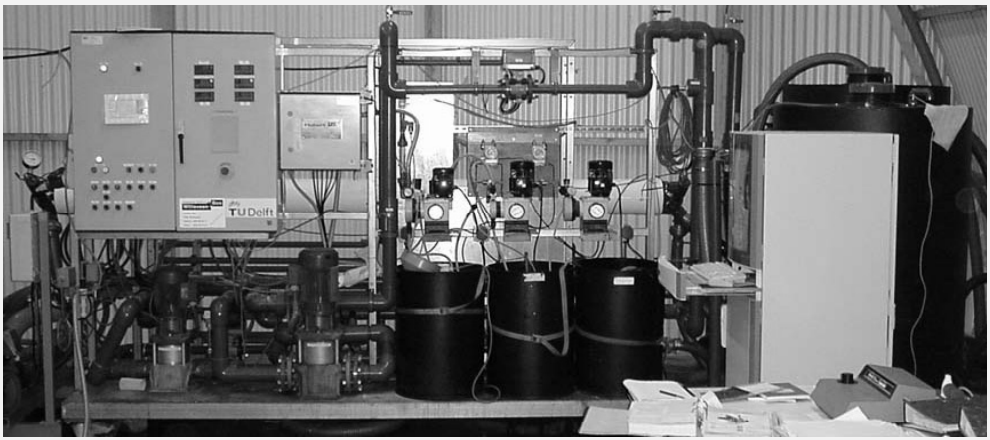

Figure 2.1 **Picture of the ultrafiltration pilot-plant that was used at wwtp Kaffeberg and Tilburg-Noord, a similar pilot-plant was used at wwtp Ede and Emmtec**

Filtration data were logged every 5 seconds, thus providing the basis for a detailed visualisation and analysis of the filtration data. The following parameters were logged: time, flow (feedwater and back flush), temperature, turbidity of the feedwater and pressure in the two permeate tubes and in the two feedwater tubes. The filtration data were recalculated to the permeability, *i.e.* the flux at $T = 15°C$ and TMP = 1 bar.

The feedwater and the permeate were regularly analysed for quality parameters like COD, N-total, P-total and bacteria. The analysing programmes were designed at each pilot-plant test in close co-operation with the principals of the tests.

The wwtp-effluent was supplied both with and without pre-treatment to the ultrafiltration plant. The effluent could be pre-treated with multi-media filtration (see following section) and with in-line coagulation. Other researchers have successfully used both pre-treatment techniques (Bourgeous *et al.*, 2001; Wiesner and Laîné, 1996; see also previous chapter). For coagulation mostly ferric chloride or aluminum chloride (as poly aluminum chloride, PACl) were used in low concentrations of 0.5 to 2.0 mg Fe^{3+} or Al^{3+}/l. In-line coagulation was done by addition of the coagulant to a tube flocculator with a static mixer, providing enough energy input for the development of small particles (G-value is about 800 s^{-1}).

(a) (b)

Figure 2.2 Picture of two ultrafiltration membrane modules: (a) horizontally placed module, (b) vertically placed modules in the front

2.2.2 Pilot-plant for multi-media filtration

Multi-media filtration is in this research mainly used as a treatment technique for the separation of particles from a feedwater. The filter bed comprises one or more granular media, which have each a specific density and particle size distribution. Pre-treatment of wwtp-effluent with multi-media filtration before ultrafiltration may improve the ultrafiltration performance (Botes *et al.*, 1998; Bourgeous *et al.*, 2001; Drage *et al.*, 2001; de Koning and van Nieuwenhuijzen, 1999). A filter bed with two or three different layers of filter media was applied in the current research.

The wwtp-effluent enters the filter bed on top of the filter and leaves the filter bed at the bottom. Particles are retained and, as particles accumulate in the filter bed, the head loss over the filter bed increases. The filter bed is cleaned either after a certain period (24 hours) or at a pre-determined head loss or at a decreased quality of

the filtrate. The latter can be evaluated by the measuring the turbidity of the filtrate. Under ideal conditions, the time required for the build up of the head loss to the set point corresponds to the time that the filtrate quality decreases to its set point. In practice one of the two will govern the cleaning frequency (Metcalf & Eddy, 2003). Cleaning of the filter bed is done by backwashing of the filter. The granular filter media expands, which causes scouring of the filter media and the removal of retained particles. Air may be added for improved scouring. The accumulated particles are then removed on top of the filter bed.

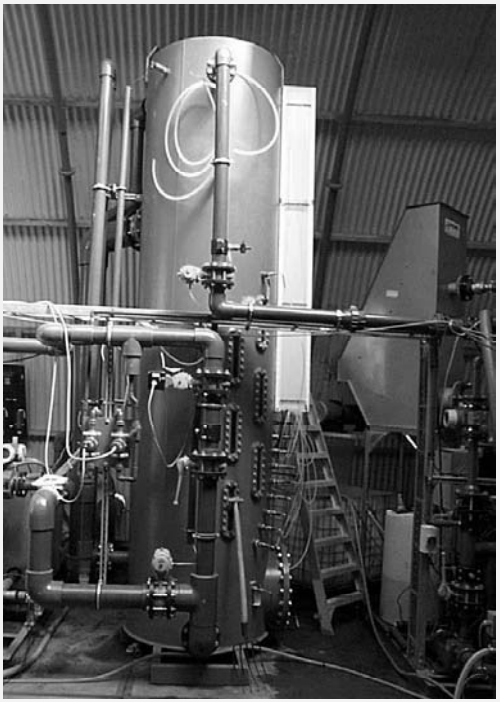

Figure 2.3 Picture of the multi-media filter used in pilot-plant tests

Coagulants may be dosed for improved removal of particles from the wwtp-effluent. Addition of metal salts (like ferric chloride, aluminium sulphate) for coagulation before multi-media filtration might improve the filtrate quality for suspended solids

(Ghosh *et al.*, 1994; van der Graaf *et al.*, 2001) as well as nutrients (Jonsson, 1999; van der Graaf and van Nieuwenhuijzen, 1998). The concentration of coagulants was in the pilot-plant tests in most cases 0.5 to 2 mg Fe^{3+} or Al^{3+}/l. The coagulants were dosed before a static mixer and the overflow on top of the filter bed provided enough energy input for coagulation (G-value was 500-1000 s^{-1})

Figure 2.3 presents the multi-media filter with a height of 4 meters and a diameter of 1 meter (a schematic drawing is presented in Appendix 2-A). The pilot-plant unit was supplied with feedwater from a continuously refreshed feedwater buffer tank and the filtrate was sent to the filtrate buffer tank. The filtrate was used for backwashing of the filter, with a flow up to 80 m^3/h. The pilot-plant installation was fed with a constant flow of 0-12 m^3/h, which related to a velocity of 10 to 30 m/h. The installation could be operated both manually and automatically and was provided with a data-logger for flow, feedwater temperature and turbidity (in and out). The head loss was measured manually by determining the water pressure at different filter bed heights. This was enabled by fifteen sampling points over the total height of the filter bed.

2.2.3 Additional lab-scale research

Additional research on lab-scale was performed with effluent from the wwtp's where pilot-plant tests were performed. Also effluent of other wwtp's (Berkel, Hoek van Holland, Vlaardingen and Zaandam-Oost) was used for lab-scale experiments. These lab-scale experiments were performed at the laboratory of Sanitary Engineering of the Delft University of Technology. A short description of these wwtp's together with some quality figures of their effluent is presented in Appendix 2-B.

2.3 Pilot-plant tests at wwtp Ede

2.3.1 Effluent quality

WWTP Ede treats the wastewater of 300,000 population equivalents, using the Bio-Denipho system for biological nitrate and phosphate removal. A short description of the wwtp, together with the plant layout is presented in Appendix 2-C. Organics, nutrients and suspended solids are almost completely removed in the wwtp. The

quality of the effluent during the first part of the pilot-plant tests (1/1998 – 10/1998) is presented in table 2.1.

Table 2.1 Quality of the effluent at wwtp Ede during pilot-plant tests[a] (STOWA, 1999)

Parameter		Average	Range	Household water[b]
Flow	10^3 m^3/day	35.9	17.3 – 114.4	
Temperature	°C	14	4 – 23	
pH	–	7.7	7.0 – 8.3	7.5 – 9.0
Colour	mg Pt/l	57	50 – 60	< 15
COD	mg O_2/l	40	21 – 59	–
BOD_5	mg O_2/l	2.9	0.5 – 7.6	
N-Kjeldahl	mg N/l	3.1	1.4 – 9.9	
NO_3^-	mg N/l	3.0	1.3 – 7.3	< 50
NO_2^-	mg N/l	0.1	0.0 – 0.8	< 0.1
NH_4^+	mg N/l	0.3	0.1 - 3.5	< 0.2
Total-P	mg P/l	0.6	0.1 – 3.9	< 2
Suspended solids	mg/l	4.0	0.5 – 8.6	< 1
Al	µg/l	200	50 – 400	200
As	µg/l	0.6	0.3 – 1.0	50
Cd	µg/l	0.5	0.01 – 1.00	5
Cr	µg/l	11.4	2.4 – 24.0	50
Cu	µg/l	12.3	2.0 – 35.6	100
Hg	µg/l	0.1	0.02 – 0.5	1
Ni	µg/l	6.6	– 16.0	50
Pb	µg/l	10.1	0.01 – 21.6	50
Zn	µg/l	113	73 – 205	100
Chloride	mg/l	98	33 – 137	< 150
Sulphate	mg/l	51	22 – 78	< 150
E.Coli	amount/100 ml	$> 10^5$	10^4 – 10^6	$< 10^4$

[a] Based on flow proportional 24-hrs samples (21 in total); [b] Indicative as no standards were defined at that time (STOWA, 1999)

2.3.2 Pilot-plant tests

Waterschap Vallei & Eem (waterboard)[1] commissioned Witteveen+Bos consulting engineers in 1997 to investigate the possibilities for the supply of household water produced from wwtp-effluent. The quality standards for household water are presented in table 2.1. Comparing the quality of the wwtp-effluent and the required quality standards for household water showed that the following parameters needed to be improved: suspended solids, colour, ammonium, heavy metals and *E.Coli* (bacteria). Two filtration techniques were chosen for improvement of these parameters: multi-media filtration (with post-treatment for *E.Coli* removal) and ultrafiltration.

Multi-media filtration

The research on multi-media filtration started in December 1997 with a double layer filter; a third layer was added in April 1998. The composition of the filter bed is presented in table 2.2. Research was carried out on optimisation of the removal of phosphorus and suspended solids by changing the coagulant dose. Also the relation between the filtrate quality and the filtration rate was investigated.

Table 2.2 Composition of filter bed during pilot-plant tests at wwtp Ede

Layer	Typical bed depth (mm)	Medium	Grain size (mm)	Density (kg/m^3)
Top layer	800	Anthracite	2.0 – 4.0	1,400
Second layer	400	Quartz sand	1.5 – 2.25	2,600
Third layer[a]	300	Garnet	0.5 – 0.8	3,500

[a] Third layer was added in the last test period

[1] Waterschap Vallei & Eem (waterboard) in cooperation with NV NUON Water (Drinking water supply company), RIZA (Institute of the Ministry of Transport, Public Works and Watermanagement) and STOWA commissioned Witteveen+Bos to investigate the production of household water using wwtp-effluent; part of the research was performed by the TUDelft (multi-media filtration, ultrafiltration) and by the RIVM (Institute of Public Hygiene and the Environment of the Ministry of Housing, Spatial Planning and the Environment; research on hygienic parameters like bacteria and viruses)

Dead-end ultrafiltration

Research on dead-end ultrafiltration started in April 1998, using the pilot-plant unit described before. Extensive tests were performed on various aspects of ultrafiltration. The research emphasized in the period on the criteria for the design of a full-scale ultrafiltration installation for treatment of wwtp-effluent. Below, a short description of the various periods is given:

Period 1

From April to August 1998 the pilot-plant tests focused mainly on the design criteria for a full-scale ultrafiltration installation for the production of household water from wwtp-effluent. The tested membrane was a horizontal, tubular PVDF membrane (STORK WF4285, tube diameter 5.2 mm; detailed information is presented in Appendix 2-D).

Period 2

From August to November 1998, a vertical, capillary PES/PVP membrane was used in the pilot-plant tests (STORK E015-010, fibre diameter 1.5 mm; see Appendix 2-D for details). The research focused in this period mainly on optimisation of the process configuration.

Period 3

From December 1998 – November 1999, a horizontal, capillary PES/PVP membrane was used (X-flow S-225-FSFC, fibre diameter 0.8 mm; see Appendix 2-D for details). The tests focused until April 1999 mainly on the optimisation of the process. Experiments to determine the filtration characteristics were performed from April to November 1999. A detailed description of these experiments is presented in Chapter 3 of this dissertation.

2.3.3 Results

Water quality

Multi-media filtration improved the quality of the effluent for suspended solids and turbidity, as well as for components related to suspended solids (like total P). The filtrate quality did not improve, with respect to the removal efficiency of particle

volume (as calculated form particle size distributions (PSD's)), by the addition of a low dosage of coagulant (1 to 2 mg Al^{3+} or Fe^{3+}/l) before the filter bed. It was shown that the addition of coagulants resulted in a decrease of the total particle volume. This indicates that the density of the particles increased, which was attributed to shrinkage of the particles. This finding is comparable with the findings in sludge dewatering. Coagulant dosage causes a decrease in water binding capacity of the flocs, which is due to neutralisation of the negatively charged surface layers (van der Graaf *et al.*, 2001). The hygienic quality improved in terms of *E.Coli* numbers with a log 1.5. The optimal process conditions were found for a dosage of 2 mg Al^{3+}/l as a coagulant at a filtration rate of 10 m/h. More details can be found in van der Graaf *et al.* (2001), Kramer *et al.*, (2000) and STOWA (1999).

For the ultrafiltration pilot-plant, the final permeate quality was independent of pre-treatment by in-line coagulation or by multi-media filtration. Suspended solids concentration decreased down to 0 mg/l and Ammonia was not removed; colour was removed for 30%, whereas bacteria were removed for 100%.

It was concluded that for the reuse of wwtp-effluent (Ede) as household water, an additional treatment step of the ultrafiltration permeate would still be necessary. Colour, as well as organic micropollutants, odour and biological stability need to be removed adequately; whereas an additional disinfection step is needed to ensure hygienic quality (STOWA, 1999).

Ultrafiltration performance

In figure 2.4 the results of the first period of the pilot-plant tests at wwtp Ede are presented. This figure shows the development of the TMP in time for ultrafiltration of effluent with and without pre-treatment and at different fluxes. It shows an uncontrollable TMP increase at fluxes higher than 40 l/m².h. Relatively stable ultrafiltration performance was found for a flux of 35-45 l/m².h at a TMP of 0.3-0.4 bar. The interval between two back flushes was 15 minutes and a chemical cleaning with 100 ppm NaOCl was performed every 90 minutes. Pre-treatment with coagulation or multi-media filtration did not improve the performance of the ultrafiltration pilot-plant.

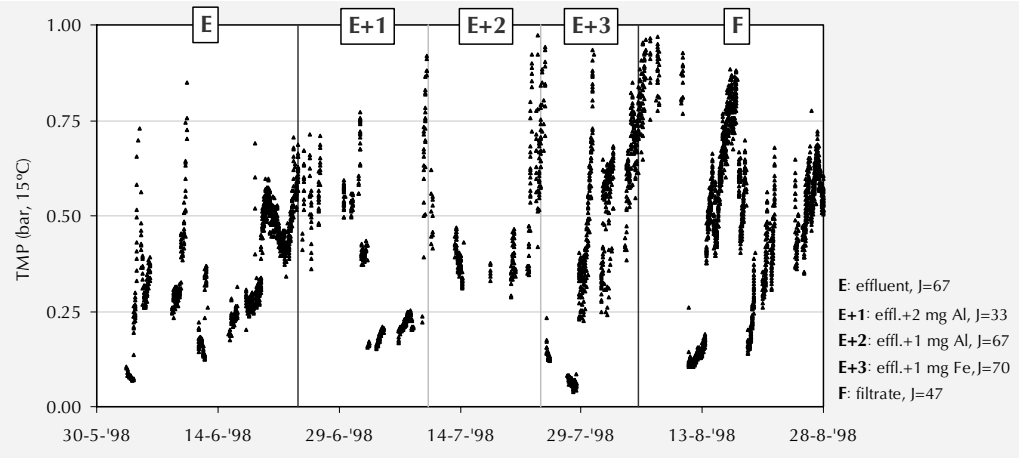

Figure 2.4 Results of pilot-plant tests at wwtp Ede on ultrafiltration of (pre-treated) effluent in the first period using a PVDF ultrafiltration membrane

In the second period of the pilot-plant test a stable performance was found at a flux of 60 l/m².h and a TMP of 0.3-0.6 bar. The cleaning intervals were similar to those in the first period. Again, pre-filtration did not result in a major improvement of the ultrafiltration performance.

The results of the pilot-plant tests that were obtained in the third period are presented in figure 2.5. No major differences in ultrafiltration performance were found for pre-filtered effluent compared to raw effluent. Relatively stable performance of the ultrafiltration pilot-plant was found at a flux of 60 l/m².h with a TMP of 0.3-0.4 bar. However, variations in the Trans Membrane Pressure were found continuously during the last three months of pilot-plant testing. This was probably related to changes in the composition of the wwtp-effluent.

The results presented here, show that the cleaning procedure is not strong enough to reduce the additional resistance, which develops when the TMP increases due to changes in feedwater composition and not in process conditions (*i.e.* the flux did not change). This induces for the following filtration steps an increased resistance. Once the TMP starts to increase (at a constant flux), the only way to return to the initial membrane resistance is the use of a chemical cleaning.

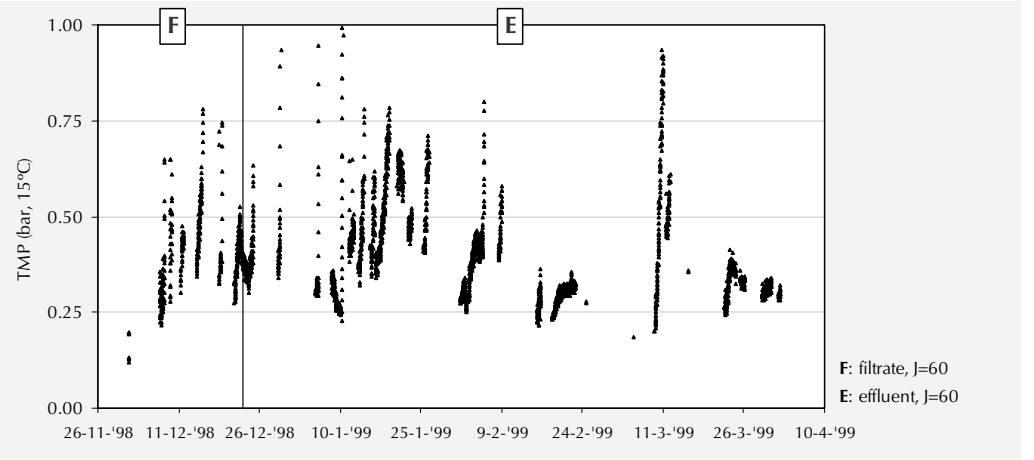

Figure 2.5 **Results of pilot-plant test at wwtp Ede on ultrafiltration of (pre-treated) effluent in third period using a PES/PVP ultrafiltration membrane**

An important aspect in this respect may be the compressibility of the fouling layer. At constant flux filtration a higher resistance will lead to an increased TMP. For compressible fouling layers an increase of the TMP will induce the compression of the fouling layer, which results in an even higher resistance of the fouling layer. If this is the case, it is unavoidable to reduce the flux or to apply chemical cleaning of the membrane.

2.4 Pilot-plant tests at wwtp Kaffeberg

2.4.1 Effluent quality

WWTP Kaffeberg treats since 1990 the wastewater of 150,000 population equivalents. The treatment plant consists of two oxidation ditches; the layout of the wwtp is presented in Appendix 2-B. The quality of the effluent during the pilot-plant tests is presented in table 2.3.

Table 2.3 Quality of the effluent at wwtp Kaffeberg during pilot-plant tests[a]

Parameter		Average	Range	e-water[c]
Flow[b]	10^3 m^3/day	13.7	8 – 42	
Temperature[b]	°C	14	4 – 23	< 25
pH[b]	-	7.5	6.7 – 8.0	
colour	mg Pt/l	60	28 – 92	< 20
COD	mg O_2/l	36	10 – 57	< 20
BOD_5[b]	mg O_2/l	3.6	0 – 31	
N-Kjeldahl	mg N/l	7.2	1.7 – 14	
NO_3^-	mg N/l	4.9	0 – 20	
NO_2^-	mg N/l	0.3	0 – 1.3	
NH_4^+	mg N/l	3.8	0 – 18	< 0.2
Total-P	mg P/l	2.8	0.6 – 8.3	< 2.0
Suspended solids	mg/l	3.4	< 2 – 6	< 1.0
Al	µg/l	75	48 – 120	
As[b]	µg/l		< 0.5	
Cd[b]	µg/l		< 5.4	
Cr[b]	µg/l	22.1	< 47	
Cu[b]	µg/l	9.8	1 – 61	
Hg[b]	µg/l		< 0.05	
Ni[b]	µg/l	31	< 120	
Pb[b]	µg/l	2.5	< 18	
Zn[b]	µg/l	73	< 220	
Chloride	mg/l	113	40 – 570	< 150
Sulphate	mg/l	105	54 – 160	< 250
E.Coli	amount/100 ml	NA		

[a] During research from October 1998 to April 1999; [b] Average of data from 1995 to 1998; [c] Basic quality for process water (defined by the e-Water group)

2.4.2 Pilot-plant tests

Pilot-plant tests were performed from October 1998 to April 1999 at wwtp Kaffeberg in Kerkrade, for research on the advanced treatment of wwtp-effluent. These tests were performed by order of a drinking water company (Waterleiding Maatschappij Limburg) and a waterboard (Zuiveringsschap Limburg). The objective of these tests was to define criteria for the design of an advanced treatment scheme for the production of process water. In these tests the process water was called 'e-water', in which the 'e' relates to ecological, economic, and efficient. E-water might be used as process water in various industries. Comparing the quality of the wwtp-effluent and the required quality standards (table 2.3) showed that the following parameters had to be reduced: suspended solids, COD and ammonium. Three different treatment techniques were investigated for this: multi-media filtration, microfiltration and ultrafiltration. Additionally, coagulation was investigated as a pre-treatment step prior to ultrafiltration.

Multi-media filtration

In October 1998 the pilot-plant tests on multi-media filtration started with a double layer filter; in February 1999 a third layer was added. The composition of the filter bed is presented in table 2.4. Research was carried out on the optimisation of the coagulant dosage for the removal of suspended solids. Also the relation between the filtrate quality and the filtration rate was investigated.

Table 2.4 Composition of filter bed during pilot-plant tests at wwtp Kaffeberg

Layer	Typical bed depth (mm)	Medium	Grain size (mm)	Density (kg/m^3)
Top layer	900	Anthracite	2.0 – 4.0	1,400
Second layer	300	Quartz sand	1.5 – 2.25	2,600
Third layer[a]	450	Garnet	0.5 – 0.8	3,500

[a] Third layer was added in February 1999

Memcor microfiltration

At the research site a Memcor microfiltration unit was tested. Two membrane modules with hydrophobic polypropylene membranes (pore diameter 0.2 μm) were tested as outside-in configuration in dead-end mode. Filtration data were automatically stored, together with the applied cleaning procedures. Air was introduced during membrane back flushing to flush retained material from the membrane pores out of the membranes.

Dead-end ultrafiltration

Research on dead-end ultrafiltration was performed with a focus on the various aspects of ultrafiltration. Research emphasized on permeate quality and design criteria for a full-scale installation. Raw effluent, as well as wwtp-effluent pre-treated with in-line coagulation and multi-media filtration was fed to the ultrafiltration unit. Three membrane modules (STORK E015-010, PES/PVP, 1 meter length; 1.5 mm capillary; pore size 10 nm) were placed vertically; next to other cleaning methods it was possible to forward flush with air (Airflush®, see Verberk *et al.*, 2002).

2.4.3 Results

Water quality

Multi-media filtration produced filtrate that did not reach the e-water quality; special attention was given to the ammonium and phosphorus concentrations. It was found that by injecting extra oxygen in the filter bed, the ammonium was nitrified completely. The microfiltration unit showed complete removal of suspended solids, but concentrations of ammonium, phosphorus, manganese, COD and colour were too high to reach the required quality. The same was found for ultrafiltration; pre-treatment of the effluent with poly aluminium chloride resulted in an extra 20% reduction of colour (Hoeijmakers, 1999).

Pilot-plant performance

The microfiltration plant showed high fluxes of 90 l/m^2.h for filtration of raw effluent at a temperature of 13°C, with the TMP ranging from 0.4 to 1.5 bar. The effect of pre-treatment of wwtp-effluent on the performance of the microfiltration pilot-plant was not investigated.

Ultrafiltration of raw wwtp-effluent resulted in a stable performance at fluxes of 40 l/m².h. Coagulation with 1 mg Al^{3+}/l (PACl) showed an improved and stable performance at fluxes up to 70 l/m².h. Also pre-filtration of the wwtp-effluent followed by coagulation of the filtrate resulted in relatively stable operation at a flux up to 80 l/m².h. The latter is illustrated in figure 2.6; although the TMP is rather high (between 0.5 and 0.7 bar), the increase in resistance was relatively low by applying regular chemical cleanings. Every 10 minutes an Airflush® of 40 seconds was applied, followed by a 60 seconds back flush; a chemical cleaning (NaOH; NaOCl) was applied every hour. These cleaning cycles were necessary to stabilise the resistance increase. Every week a more intensive chemical cleaning was applied in order to decrease the TMP to its initial value.

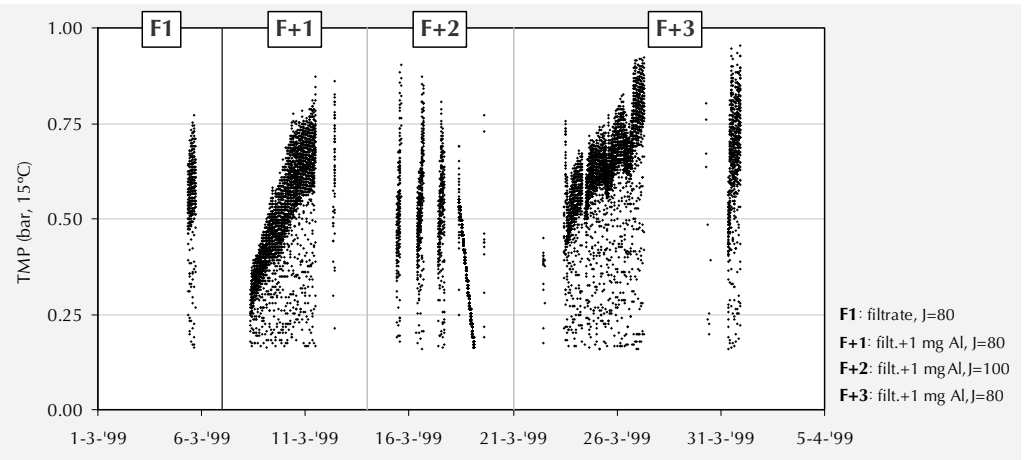

Figure 2.6 Results of pilot-plant tests at wwtp Kaffeberg on ultrafiltration of pre-filtered effluent; a PES/PVP ultrafiltration membrane was used

Some results of the pilot-plant tests are presented in figure 2.7, in which the resistance increase is shown for four types of feedwater: (1) raw effluent, (2) effluent + 1 mg Al^{3+}/l (PACl), (3) effluent after pre-filtration, and (4) pre-filtered effluent + 1 mg Al^{3+}/l (PACl). The actual resistance over the membrane was calculated by using Darcy's law and the development of resistance versus time is calculated for filtration intervals between cleaning.

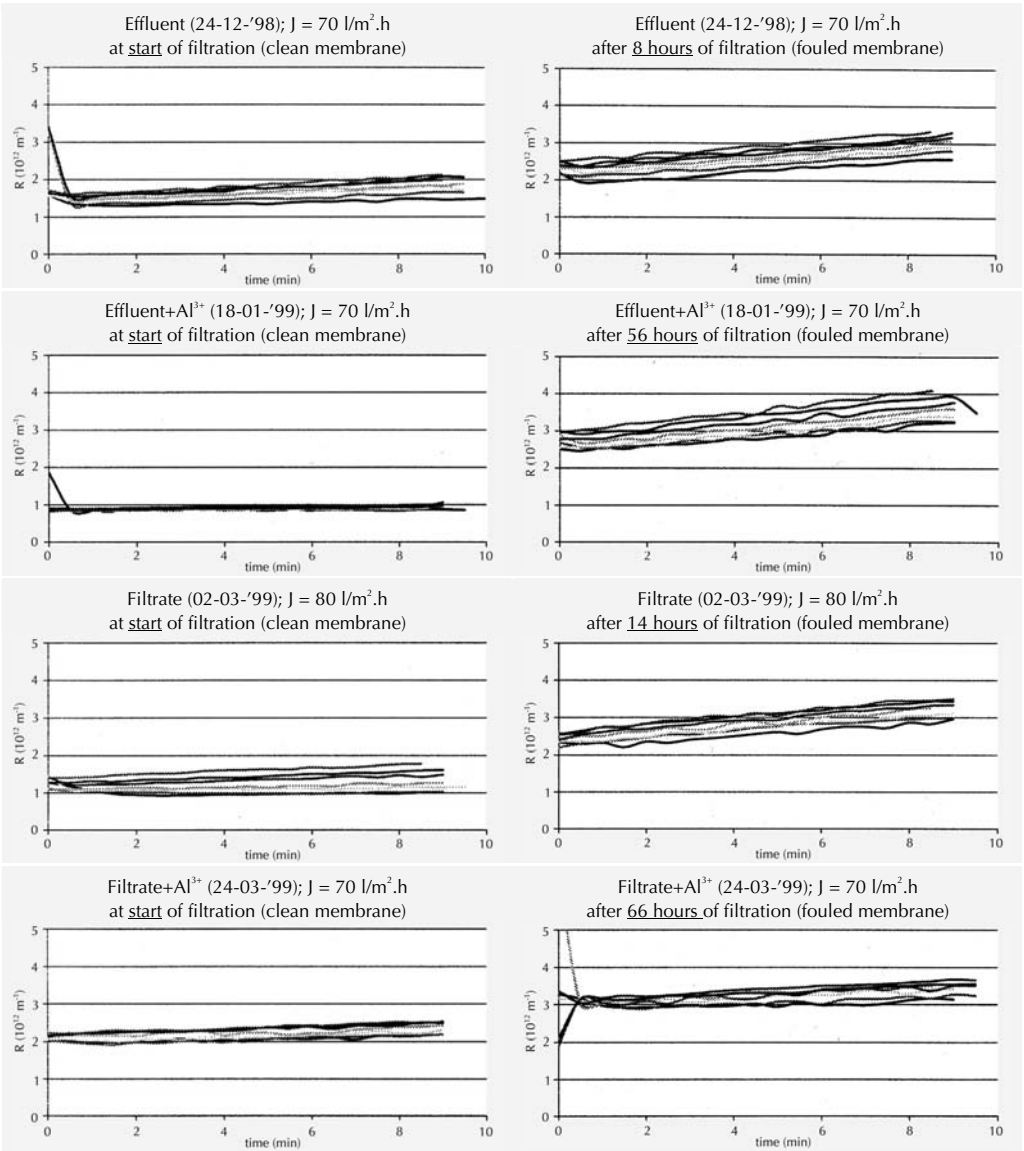

Figure 2.7 **Results of resistance increase during pilot-plant tests on dead-end ultrafiltration of (pre-treated) wwtp-effluent (Kaffeberg); on the left: the resistance increase of the first seven filtration intervals between hydraulic cleaning (FF+A40+15 BF60); on the right: the same for filtration intervals after hours of filtration**

The applied cleaning procedure was the same for all tests presented in figure 2.7: a forward flush of 40 seconds is applied every ten minutes with an additional Airflush during the last 15 seconds (FF+A40+15); after this a back flush was performed for 60 seconds (BF60). These tests were started after a thorough chemical cleaning, which provided relatively clean membranes at the start of the test. All graphs on the left are the filtration curves measured during the first seven subsequent filtration intervals. The graphs on the right show the seven subsequent filtration intervals after hours of filtration.

The results that were found with effluent and filtrate without additional coagulation showed that at the beginning of the test the increase in resistance was about $1.0 \cdot 10^{12}$ m^{-1} per filtration interval. The resistance increased for every subsequent filtration interval, indicating that the developing fouling layer was not completely reversible. After 8 hours and 14 hours of filtration of respectively raw effluent and pre-filtered effluent the resistance increased up to $3.5 \cdot 10^{12}$ m^{-1}. The tests that were done with coagulated effluent and coagulated filtrate, resulted in a much lower increase in resistance during the first seven filtration cycles, compared to the tests with raw effluent and raw filtrate. The resistance increase during one filtration cycle seems for coagulated feedwater to be better reversible than without coagulation. This resulted in a longer filtration period, respectively 56 and 66 hours of filtration. These results show that effluent of wwtp Kaffeberg needed additional coagulation for the reduction of its fouling properties.

2.5 Pilot-plant tests at wwtp Tilburg-Noord

2.5.1 Effluent quality

WWTP Tilburg-Noord is designed for the biological treatment of the wastewater of 445,000 population equivalents. In Appendix 2-C the layout of the treatment plant is presented. The effluent has a very high quality, which is presented in table 2.5.

Table 2.5 Quality of the effluent (right after the pond system), during pilot-plant tests at wwtp Tilburg-Noord[a]

Parameter		Average	Range	Classification[b]		
				Quality 1	Quality 2	Quality 3
flow	10^3 m^3/day	46				
Temperature	°C		8 – 25	< 20	< 20	< 20
pH	–	7.6				
Colour	mg Pt/l	56		20	20	10
COD	mg O_2/l	36	20 – 40	< 30	< 30	< 10
BOD_5	mg O_2/l	1.8	1.8 – 4.7			
N-Kjeldahl	mg N/l	3	2 – 5			
NO_3^-	mg N/l		3 – 5			
NO_2^-	mg N/l	< 0.1	< 0.1			
NH_4^+	mg N/l	< 0.5		< 0.5	< 0.5	< 0.1
Total-P	mg P/l	0.5	0.1 – 1.0			
Suspended solids	mg/l	< 1		< 0.5	< 0.1	<< 0.1
Al	µg/l					
As	µg/l	1.5				
Cd	µg/l	< 2				
Cr	µg/l					
Cu	µg/l	4	1 – 14			
Hg	µg/l	< 0.1				
Ni	µg/l	5.5	4 – 8			
Pb	µg/l	< 40				
Zn	µg/l	70	60 – 100			
Chloride	mg/l	120	50 – 200			
Sulphate	mg/l					
EGV	m^3/m				< 125	< 10
Hardness	mmol				< 2	< 1
E.Coli	amount/100 ml		10^4 – 10^5			

[a] Data of 1999; [b] Classification of water quality for various use: quality 1 – low-quality, cooling water, household water; quality 2 – groundwater infiltration, process water; quality 3 – best quality, resource for drinking water, boiler feed water

After biological nutrient removal and final sedimentation, the effluent is discharged in a pond system with a residence time of one day[1]. After this, the effluent is discharged into the surface water.

2.5.2 Pilot-plant tests

In the year 2000 pilot-plant tests were started at wwtp Tilburg-Noord for the production of 'other water', which was defined as water for non-potable use that is cheaper than drinking water. The water supply company (Tilburgsche Waterleiding Maatschappij) together with the waterboard (Waterschap de Dommel) and the local municipality (gemeente Tilburg) initiated these tests. Other water was defined for three different water classes, ranging from a relatively low quality (quality 1 in table 2.5) to a high water quality (quality 3). In the pilot-plant tests special attention was given to some critical micropollutants like diuron, atrazin and AMPA (Maas, 2003). The wwtp-effluent was investigated for treatment with multi-media filtration and ultrafiltration, as well as with capillary nanofiltration. With nanofiltration additionally small constituents like bivalent ions will be removed.

The water for the pilot-plant tests was withdrawn from the outlet of the pond system. To prevent various biological materials like small fishes, water plants, etc., to enter the pilot-plant installations a sieve was placed in front of the pilot-plant installations. The diameter of the sieve openings was 5 mm.

Multi-media filtration

The pilot-plant tests at wwtp Tilburg-Noord started in May 2000 for the multi-media filtration with two layers of granular media, the dimensions of which are shown in table 2.6. In some tests a coagulant (poly aluminium chloride, PACl) was dosed to the effluent before entering the filter bed. It was beforehand expected that the treatment of the wwtp-effluent with multi-media filtration would be appropriate to reach the required water quality for quality 1 (table 2.5).

[1] The average flow is 46,000 m^3 and the volume of the pond is 180,000 m^3, which results in a maximum residence time of three to four days; during the pilot-plant test a shortcut in the pond system resulted in a residence time of one day

Table 2.6 Composition of filter bed during pilot-plant tests at wwtp Tilburg-Noord

Layer	Typical bed depth (mm)	Medium	Grain size (mm)	Density (kg/m^3)
Top layer	700	Anthracite	2.0 – 4.0	1,400
Second layer	500 (+200)[a]	Quartz sand	1.5 – 2.25	2,600

[a] On the 11th of October 2000 extra quartz was added for improvement of the performance

Dead-end ultrafiltration

Pilot-plant tests on dead-end ultrafiltration were performed using two horizontal membrane modules of 1.5 m in series (X-flow; PES/PVP; pore diameter 20 nm; capillaries 1.5 mm). The wwtp-effluent was fed to the ultrafiltration pilot-plant both without pre-treatment and with pre-treatment by in-line coagulation (1 and 2 mg Al^{3+}/l PACl) and/or by multi-media filtration.

Capillary nanofiltration

At wwtp Tilburg-Noord also direct treatment of (pre-treated) effluent with capillary nanofiltration was investigated. The capillary nanofiltration membranes were from X-flow (NR015-500) and had a capillary diameter of 1.5 mm. It was expected that with nanofiltration the wwtp-effluent could be upgraded to the required water quality for quality 3 (table 2.5).

2.5.3 Results

Water quality

The tests showed that it was possible to produce water with quality 1 by applying the multi-media filter; still critical were temperature, colour and *E.Coli*. With dead-end ultrafiltration colour and ammonia were insufficiently removed to reach the water quality for quality 2, whereas also the temperature was too high. With nanofiltration a similar quality as for ultrafiltration was found with respect to the parameters mentioned in table 2.5. Micropollutants were not found in the wwtp-effluent, accordingly also no micropollutants were found in the filtrate and the permeate of the ultrafiltration and nanofiltration pilot-plants.

The results for ultrafiltration (including the performance of the pilot-plant, see the next paragraphs) made the water supply company decide to build a full-scale

treatment plant based on ultrafiltration, which will produce process water for the industries located nearby wwtp Tilburg-Noord, as well as for irrigation of a golf course located nearby the wwtp (Maas, 2003).

Ultrafiltration performance

Pre-treatment of the wwtp-effluent had a positive effect on the performance of the ultrafiltration pilot-plant. Without pre-treatment a stable ultrafiltration performance was found at a flux of 80 l/m².h with a TMP of 0.7 bar. After in-line coagulation with 2 mg Al^{3+}/l stable performance was found at a flux of 110 l/m².h and a TMP of 0.65 bar (back flush interval 15 minutes, every 2 hours chemical cleaning (two times HCl, third time NaOCl)). The recovery was about 90% and the net flux was 90 l/m².h. At a flux of 100 l/m².h the TMP stabilised at 0.55 bar for more than one month. These results are presented in figure 2.8, showing the development of the TMP during one month of filtration.

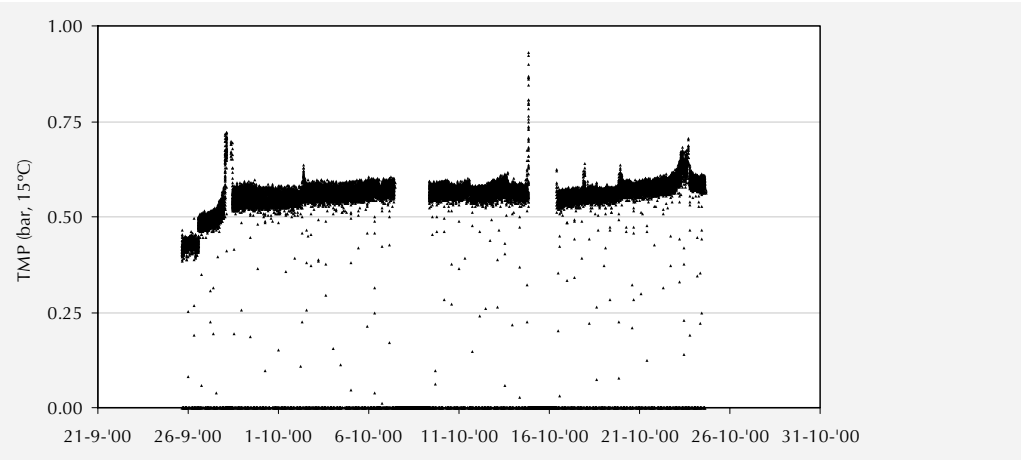

Figure 2.8 Results of pilot-plant experiments at wwtp Tilburg-Noord on ultrafiltration of effluent + 2 mg Al^{3+}/l at a flux of 100 l/m².h; a PES/PVP ultrafiltration membrane was used

Also pre-treatment with multi-media filtration (and pre-coagulation with 1-2 mg Al^{3+}/l) improved the ultrafiltration performance and resulted in a TMP of 0.5-0.6 bar at a constant flux of 100 l/m².h. This result seemed to be a function of the performance of

the multi-media filter; figure 2.9 shows the TMP and the turbidity of the filtrate during ultrafiltration of filtrate. The upper curve represents the TMP where the lower curve represents the turbidity of the filtrate.

After washing of the double layer filter the turbidity in the filtrate decreased to a low value (0.4 NTU). This indicated an improvement of the filter bed (ripening). This situation continued for approximately 10 hours. Then, the turbidity gradually increased, indicating a loss in performance of the filter bed. The highest value (0.8 NTU) was reached after 24 hours when the filter cycle was ended and the filter was washed again. In response to this cycle the ultrafiltration process showed a slight increase of the TMP at the start (from 0.5 to 0.6 bar), while the turbidity improved. The TMP gradually dropped down to 0.5 bar in response to an increasing turbidity. This phenomenon was reproduced a couple of times, also at lower fluxes of 80 l/m².h and at similar fluxes of 100 l/m².h.

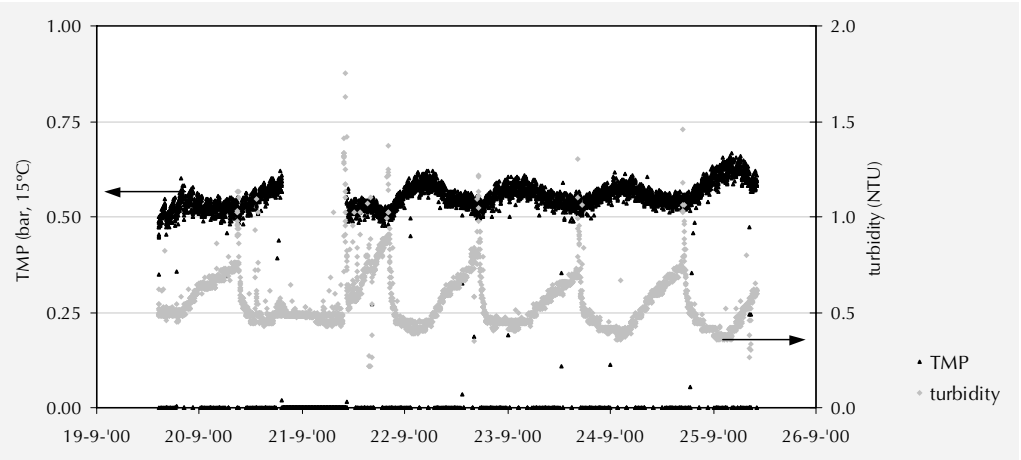

Figure 2.9 Results of ultrafiltration tests at wwtp Tilburg-Noord for effluent pre-treated with a double layer filter; flux is 100 l/m².h; the upper curve represents the TMP, the lower curve represents the turbidity measured in the filtrate (*i.e.* the feedwater of the ultrafiltration pilot-plant)

This phenomenon illustrated the sensitivity of the ultrafiltration process to changes in the composition of the feedwater. Sizes, concentrations, chemical and physical

properties of particles and their size distributions seem to play an important role in the final behaviour near the ultrafiltration membrane surface.

For an explanation of these occurring phenomenons (decreasing TMP at increasing filtrate turbidity) the following should be considered (Lawler, 1997): Understanding of the behaviour of particles in multi-media filters is incomplete due to the complexity of the process. Filters will never reach a steady-state situation because of ripening of the filter bed and breakthrough of particles; it is a dynamic process. As seen in figure 2.9 in the first period after cleaning of the filter bed, the turbidity decreased, which could be attributed to this ripening process. Until a certain concentration, more and smaller particles are caught by the decreasing pore sizes in the filter bed.

The effect of in-line coagulation before the filter bed was shown when, at one time, the coagulation before the filter bed was stopped. This caused an immediate increase in TMP over the ultrafiltration unit. This finding is presented in figure 2.10. From this result it was concluded that coagulation played a major role in the particle size distribution that was captured by the filter bed.

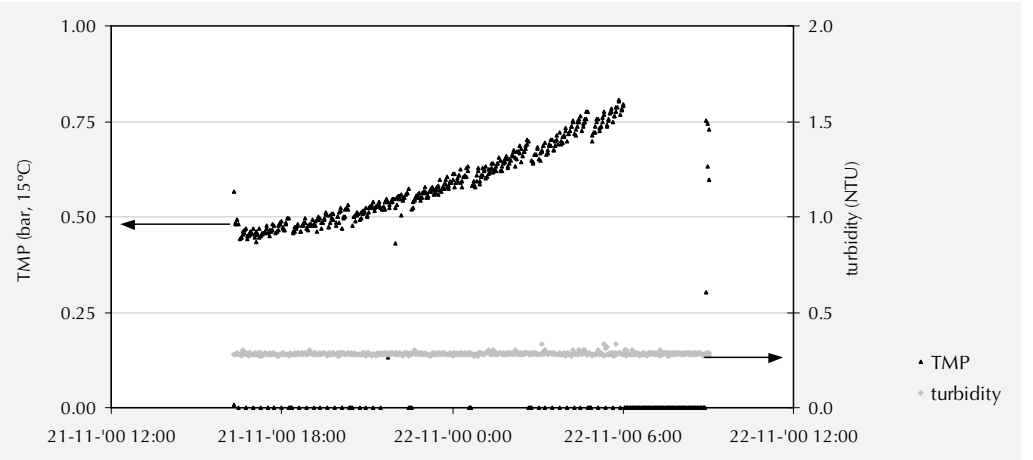

Figure 2.10 Results of ultrafiltration tests at wwtp Tilburg-Noord for effluent that is pre-treated with a double layer filter without pre-coagulation; the flux is 100 l/m^2.h; the upper curve shows the TMP, the lower curve represents the turbidity measured in the filtrate (*i.e.* the feedwater of the ultrafiltration plant)

2.6 Pilot-plant tests at the wwtp of Emmtec Services

In Emmen at the wwtp of Emmtec Services (industrial park) the advanced treatment of effluent was investigated. Effluent might be used as a source for the production of boiler feed water. The pilot-plant tests were performed in cooperation with Witteveen+Bos and NUON Water, by order of Emmtec Services. The wwtp treats industrial wastewater and is presented without details. A detailed description of the results can be found in Witteveen+Bos (2002).

Table 2.7 Quality of effluent Emmtec and treated effluent, as well as reuse criteria

Parameter		Effluent	Sandfiltrate with PACl	UF – permeate	RO – permeate	Criteria Boiler feed water
Temperature	°C	22 – 28	–	18 – 24	18 – 24	
pH	–	7.4	7.2 – 7.4	7.1 – 7.3	7 – 7.8	7 – 9
COD	mg O_2/l	3 – 23	4 – 10	< 1 – 15	< 1 – 4	
BOD_5	mg O_2/l	–	–	–	–	
N-Kjeldahl	mg N/l	0.2 – 7.7	0.6 – 7.5	0.1 – 6.9	< 0.01 – 0.3	
NH_4^+	mg N/l	0.03 – 5.4	0.01 – 5.4	< 0.01 – 5.3	< 0.01	
SS[a]	mg/l	4 – 10	1 – 2	0	0	
SiO_2 total	mg/l	14.1 – 18.9	14 – 16.5	13.7 – 18	0.02 – 0.13	< 0.02
EGV	μS/cm	–	–	980 – 1280	3 – 11	< 0.1

[a] SS = Suspended solids

The following treatment techniques were investigated in parallel as well as in series: a continuous sand filter (Dynasand) with and without pre-treatment of the effluent, a dead-end ultrafiltration unit and a reverse osmosis unit. Table 2.7 shows the effluent quality of the biologically treated industrial wastewater. Next to this, the quality of the water is presented for effluent treated with sandfiltration (with and without pre-coagulation), with ultrafiltration and with reverse osmosis. Table 2.7 is finalised with the quality criteria for boiler feedwater.

In the pilot-plant tests at the wwtp of Emmtec Services a stable ultrafiltration performance was found for effluent pre-treated by the continuous sand filter (pre-coagulated) with in-line coagulation of 2 mg Al^{3+}/l of the sandfiltrate. At a constant flux of 100 l/m².h the TMP was in the range of 0.3 to 0.45 bar. The back flush interval was 30 minutes with a chemical cleaning for every 6 hours (intermittent NaOCl and Divos2).

2.7 Discussion

The results of the pilot-plant tests on dead-end ultrafiltration of wwtp-effluent show great differences in the applied process conditions for long-term stable performance. Various pre-treatment techniques were tested and were combined with various cleaning strategies. In table 2.8 the results of the four pilot-plant tests are summarised. These results were within the same range as found for dead-end ultrafiltration of wwtp-effluent by others (Bourgeous *et al.*, 2001; Decarolis *et al.*, 2001; van der Graaf *et al.*, 1998; van Hoof *et al.*, 1998). Treatment of the effluent of wwtp Ede and Kaffeberg was only possible by applying an intense cleaning strategy.

Table 2.8 Summary of the results in four pilot-plant tests

WWTP	Effluent composition			Pre-treatment	Cleaning[a]	Flux	TMP_n	Permeability
	COD (mg/l)	SS (mg/l)	Colour (mg Pt/l)			(l/m².h)	(bar, 15°C)	(l/m².bar, 15°C)
Ede	21–59	0.5–6	50–60	No; Filtration; 1 mg Fe^{3+}/l	BF*15*; CF*90*	60	0.3–0.4	150–200
Kaffeberg	10–57	< 2–6	28–92	Filtration + 1 mg Al^{3+}/l	BF*10*; CF*70*	80	0.4–0.7	115–200
Tilburg-Noord	20–40	< 1	56	Filtration; 2 mg Al^{3+}/l	BF*15*; CF*120*	>100	0.55	< 180
Emmtec[b]	3–23	4–10		Filtration + 2 mg Al^{3+}/l	BF*30*; CF*360*	100	0.3–0.45	330–220

[a] BF10 = Back Flush every 10 minutes; CF90 = Chemical Flush every 90 minutes; [b] Industrial wwtp

The variations can be related both to the properties of the effluent (feedwater of the ultrafiltration pilot-plant) and the properties of the membrane. In the current research PES/PVP membranes were tested on the various wwtp's. The tests that were performed at wwtp Tilburg-Noord resulted in relatively high fluxes (> 100 l/m².h). The

effluent from wwtp Tilburg-Noord is treated in pond system with a retention time of one day before it is discharged to the surface water. It seems that the amount of foulants is reduced by the pond system. Kampf *et al.* (2003) found that in natural systems like constructed wetlands the organic content of effluent can be reduced, which might as well be the case in the pond system at wwtp Tilburg-Noord.

Furthermore, it was shown that the ultrafiltration installation is sensitive towards changes in composition of the effluent. This is also the case towards changes in composition of effluent that is pre-treated with in-line coagulation or multi-media filtration (for example figure 2.9). The size of effluent constituents, the concentrations, the chemical and physical properties and the size distribution play an important role. The differences that were found at the various wwtp's can not be related directly to the parameters that were measured at the wwtp's (see table 2.8). This shows the need for parameters that can be used for prediction of fouling properties of ultrafiltration membranes and for prediction of the performance of such systems.

In all pilot-plant tests the performance of the ultrafiltration pilot-plant was improved by pre-treatment. Bourgeous *et al.* (2001), Decarolis *et al.* (2001) and van der Graaf *et al.* (1998) found that both sand filtration and in-line coagulation improve the performance of the ultrafiltration installations. It seems that by sand filtration foulants are removed, whereas in-line coagulation seems to be effective by increasing the particle size of foulants (Decarolis *et al.*, 2001). The results that were found at wwtp Ede and Kaffeberg show that pre-treatment with in-line coagulation or multi-media filtration resulted in higher fluxes, but compared to the results of wwtp Tilburg-Noord and Emmtec the applied cleaning strategy was more intense. Also the performance of the ultrafiltration plant found at Tilburg-Noord and at Emmtec was more stable compared to wwtp Ede and Kaffeberg. The optimum combination of pre-treatment and cleaning strategy showed still a gradual increase in TMP (figure 2.4 for wwtp Ede and figure 2.6 for wwtp Kaffeberg). It seems that pre-treatment by in-line coagulation or multi-media filtration is not strong enough to reduce all fouling problems. Improved pre-treatment techniques and/or cleaning strategies are necessary to successfully treat wwtp-effluent that has an insufficient filterability, as was the case in Ede and Kaffeberg.

The current research showed that, in order to obtain an optimal performance of the ultrafiltration pilot-plants, at all sites it was necessary to perform pilot-plant tests for at least 6 months. This finding is similar to Bersillon and Thompson (1996), who insisted on doing long-term pilot-plant tests for the optimisation of the process conditions of ultrafiltration. At this moment, no parameter is available for a quick evaluation of filtration characteristics and for the prediction of long-term fouling tendencies.

2.8 Conclusions

The pilot-plant tests that were performed at four different sites on ultrafiltration of wwtp-effluent showed that great differences exist in the treatability of wwtp-effluent.

The performance of ultrafiltration pilot-plants shows a direct relation with the composition of the (pre-treated) wwtp-effluent, but at this moment no parameters exist that can be related directly to the performance of the ultrafiltration plants.

Pre-treatment of wwtp-effluent with in-line coagulation or multi-media filtration improves the performance of an ultrafiltration installation and is a pre-condition for stable performance of the ultrafiltration plant. However, raw wwtp-effluent that has a low initial filterability will show lower fluxes than effluent with a high filterability even when effluent is pre-treated and extensive cleaning procedures are applied.

Pilot-plant testing of dead-end ultrafiltration of wwtp-effluent is necessary in order to obtain the conditions for long-term stable ultrafiltration performance.

More insight in filtration characteristics might help to improve the performance of ultrafiltration systems.

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3 Filterability and Reversibility in pilot-plant experiments

3.1 Introduction

As shown in the previous chapters the filtration mechanism(s) responsible for flux decline in dead-end ultrafiltration of wwtp-effluent are still unclear. Pilot-plant experiments confirm that, due to a lack of knowledge of the occurring filtration and fouling mechanisms, the applied process conditions for long-term stable performance cannot be fully optimised (van der Graaf *et al.*, 1999; van Hoof *et al.*, 1998).

Stable process conditions may be realised if the characteristics of both the filterability of the feedwater (*i.e.* filtration characteristics during a certain time period) and the reversibility of the fouling layer (*i.e.* removal efficiency of the fouling layer by various cleaning techniques) are identified. The actual changes in the flux are mainly determined by the filterability, while both the filterability and the reversibility greatly affect the long-term net flux. Fouling mechanisms, like adsorption, pore blocking, particle deposition and concentration polarisation, may influence the filterability and the reversibility. The first two mechanisms can take place inside the membrane pores or the membrane structure. The latter two occur only on the top layer of the membrane. The filterability and the reversibility greatly depend on the quality of the feedwater and are influenced both by pre-treatment of the feedwater and by the characteristics of the membrane (Bourgeous *et al.*, 2001; Doyen *et al.*, 2002; van der Graaf *et al.*, 1999).

It is rather unusual to analyse the filtration mechanisms and the fouling phenomena in ultrafiltration pilot-plant experiments, because such experiments are difficult to perform due to the scale of research as well as the fluctuations of the wwtp-effluent composition. As shown in chapter 1 only few researchers presented research

results dealing with the relationship between occurring filtration phenomena and (theoretical) filtration mechanisms (Bourgeous *et al.*, 2001; Decarolis *et al.*, 2001).

In this chapter[1] a new method is presented for analysis of the filtration characteristics in pilot-plant experiments, in which the total membrane resistance is related to the volume of filtered feedwater and the process conditions. The influence of the applied flux and of pre-treatment of the wwtp-effluent, with in-line coagulation and multi-media filtration, is evaluated.

3.2 Definitions of Filterability (F) and Reversibility (R_x)

In order to provide a better understanding of the fouling phenomena that are found on ultrafiltration membranes in a pilot-plant, both the filterability of the effluent and the reversibility of the occurring fouling layer are defined. Filtration and cleaning experiments were performed with a pilot-plant for dead-end ultrafiltration. From the filtration curves and the changes in the total membrane resistance, both the Filterability F and the Reversibility R_x were calculated. The basis for all calculations is the modified form of Darcy's law, which shows the relationship between the permeate flux J, the Trans Membrane Pressure (TMP), the dynamic viscosity (temperature) and the total membrane resistance (eq. 3.1).

$$J = \frac{\Delta P}{\eta \cdot R_{tot}} \qquad \text{(eq. 3.1)}$$

where J = permeate flux ($m^3/m^2.s$)
ΔP = Trans Membrane Pressure, TMP (N/m^2)
η = dynamic viscosity ($N.s/m^2$)
R_{tot} = total resistance (m^{-1})

Filterability of wwtp-effluent

The Filterability of a feedwater is determined by the interaction between the feedwater and the membrane. If the Filterability is low, the membrane fouls rapidly. A high Filterability will be found if the membrane is slowly fouled. In this chapter the

[1] Part of this work has been presented in Roorda and van der Graaf (2000)

Filterability of wwtp-effluent is defined as the ratio between permeate volume per m² membrane area (V_{tot}/A_m) and the resistance of the fouling layer (ΔR_{fo}). The fouling layer is formed during dead-end ultrafiltration of wwtp-effluent without cleaning of the membrane for at least one hour of filtration. The equation for the Filterability F is shown in equation 3.2.

$$F = \frac{V_{tot}}{A_m \cdot \Delta R_{fo}} \qquad \text{(eq. 3.2)}$$

where F = Filterability of wwtp-effluent (m²)
V_{tot} = total permeate volume (m³)
A_m = membrane area (m²)
ΔR_{fo} = resistance of fouling layer after ultrafiltration of a feedwater, without cleaning; $\Delta R_{fo} = R_{tot,end} - R_{tot,start}$ (m^{-1})
$R_{tot,end}$ = total membrane resistance after filtration (m^{-1})
$R_{tot,start}$ = initial membrane resistance before experiment (m^{-1})

The total membrane resistance is measured by filtration of (clean) permeate over the ultrafiltration membrane and is the average value determined at five different fluxes (20, 40, 60, 80 and 100 l/m².h), using equation 3.1. During filtration of effluent the total membrane resistance will increase, which results in a lower permeability. The increase in total membrane resistance can be attributed to the formation of a fouling layer.

The Filterability F of wwtp-effluent was found as low as $1 \cdot 10^{-14}$ m², which indicated rapid membrane fouling. The highest value of the Filterability F for wwtp-effluent was about $15 \cdot 10^{-14}$ m², which indicated a small decline in flux and only little membrane fouling.

Reversibility of fouling layer

An indication of the removal characteristics of a fouling layer can be derived from permeability data that are measured with permeate at the end of the filtration period and right after cleaning of the membrane. The removal characteristics of the fouling layer and the effectiveness of the cleaning procedures relate to the Reversibility of the fouling layer. The Reversibility R_x of the fouling layer is defined as the ratio between

the change in resistance caused by cleaning of the membrane (ΔR_{fo} - $\Delta R_{fo,x}$) and the resistance of the fouling layer (ΔR_{fo}) as a function of various cleaning procedures ($_x$). The equation for the Reversibility is presented in equation 3.3.

$$R_x = \frac{\Delta R_{fo} - \Delta R_{fo,x}}{\Delta R_{fo}} \cdot 100\% \qquad \text{(eq. 3.3)}$$

where R_x = Reversibility of fouling layer as a function of cleaning procedure x (%)

$\Delta R_{fo,x}$ = Resistance of the fouling layer after cleaning with procedure x (m^{-1})

The reversibility of the fouling layer is determined by a combination of foulants (retained material at the membrane surface), by the membrane properties and by the applied cleaning procedures. The cleaning procedures vary in the cleaning frequency, the cleaning time, the physical cleaning methods (back flush, forward flush, AirFlush®) and the chemical cleaning methods. A cleaning method is sufficient when after filtration and the subsequent cleaning the total membrane resistance is at its initial value ($\Delta R_{fo,x}$ becomes zero). A sufficient cleaning method relates to a Reversibility of 100%.

3.3 Experimental set-up

The experiments were performed in a pilot-plant ultrafiltration installation with a flow of 5 to 10 m^3/h. A detailed description of the pilot-plant is presented in chapter 2 of this thesis (§2.2). The pilot-plant was equipped with an 8-inch module consisting of hollow fibre ultrafiltration membranes. The total membrane area was 30 to 70 m^2, depending on the fibre diameter. Filtration was carried out under constant flux in dead-end filtration mode. The main specifications of the ultrafiltration membranes that were used in these experiments are presented in table 3.1.

Table 3.1 Characteristics of the membranes used in the experiments.

Membrane type[a]	Arrangement	Average pore size[b] (nm)	Internal diameter (mm)	Membrane area (m^2)
PVDF	Horizontal	30	5.2	30
PES/PVP (1)	Vertical	20	1.5	45
PES/PVP (2)	Horizontal	10 – 30	0.8	70

[a] PVDF – hydrophilic poly vinyl idene fluoride; PES/PVP – hydrophilic poly ether sulfone/poly vinyl pyrrolidone; [b] As indicated by manufacturer

Effluent of wwtp Ede (experiment 1-1: May to June 1998; experiments 3-1 to 3-4: May to June 1999) and effluent of wwtp Kaffeberg (experiments 2-1 and 2-2: November 1998 to January 1999) were used as the feedwater for the experiments. The wwtp-effluent was fed to the pilot-plant with and without pre-treatment by in-line coagulation with poly aluminium chloride (PACl; 0.5 mg Al^{3+}/l) or multi-media filtration. More results on multi-media filtration of wwtp-effluent were presented in van der Graaf *et al.* (1999) and van der Graaf *et al.* (2001). The average quality of the feedwater during the experiments is summarised in table 3.2.

Table 3.2 Average quality of feedwater used in experiments

		wwtp Ede			wwtp Kaffeberg		
		Effluent	Effluent+[a]	Filtrate[b]	Effluent	Effluent+	Filtrate
COD	mg COD/l	22	36	16	36	*nd*[c]	32
Turbidity	FTE	1.8	2.5	0.5	4	*nd*	< 1
Suspended Solids	mg SS/l	5.3	3.0	1.3	> 4	*nd*	< 2
P_{tot}	mg P/l	0.15	0.15	0.15	2.8	*nd*	2.4

[a] Effluent + PACl (0.5 mg Al^{3+}); [b]Filtrate of multi-media filtration; [c] Not determined

Before the start of each experiment the membrane was soaked in a sodium hypochlorite solution (200-300 ppm) for one night and back-flushed with ultrafiltration permeate (200-300 l/m^2.h). Each experiment started with the determination of the permeability using permeate, after which filtration of the (pre-treated) effluent started. Filtration was stopped when the TMP reached a value of 0.8

bar, or in some experiments after the filtration of a specified feedwater volume (V_{tot} per membrane area (A_m) = 85 l/m²).

Most experiments were performed at three different fluxes, repeating some of the experiments up to four times on different days. In the first experiments (exp. 1 and 2) the intervals between two experiments are more than a week. In the last series of experiments (exp 3-1 to 3-4) one experiment is repeated with the same process conditions on the next day. By this the influence of fluctuations in effluent quality was minimised. An overview of the filtration experiments is shown in table 3.3.

Table 3.3 Overview of experiments in ultrafiltration of wwtp-effluent

Exp.	Date	Membrane	WWTP	Feed	End at fixed TMP or V_{tot}/A_m	Applied flux (l/m².h)
1-1	5/'98 to 6/'98	PVDF	Ede	Effluent	TMP = 0.8 bar	35; 70; 100
2-1	10/'98 to 1/'99	PES/PVP (1)	Kaffeberg	Effluent	TMP = 0.8 bar	80
2-2	2/'99	PES/PVP (1)	Kaffeberg	Effluent+	TMP = 0.8 bar	80
3-1	3/5/'99 to 11/5/'99	PES/PVP (2)	Ede	Effluent	TMP = 0.8 bar	40; 70; 100
3-2	10/6/'99 to 15/6/'99	PES/PVP (2)	Ede	Effluent	V_{tot}/A_m = 85 l/m²	40; 70; 100
3-3	1/6/'99 to 9/6/'99	PES/PVP (2)	Ede	Effluent+	V_{tot}/A_m = 85 l/m²	40; 60
3-4	20/4/'99 to 27/4/'99	PES/PVP (2)	Ede	Filtrate	TMP = 0.8 bar	40; 70; 100

At the end of the filtration period the permeability was measured using ultrafiltration permeate and consequently the resistance of the fouling layer was calculated. The relationship between the different cleaning procedures and the decrease in the total membrane resistance was studied by applying various cleaning procedures and by determining the permeability in between two cleaning procedures. The following cleaning procedures were applied subsequently: a *forward flush*, a forward flush with air (*Airflush*®; experiment 2-1 and 2-2) and a *back flush*. The next cleaning procedure was a *chemical flush*, in which the membrane was soaked in a NaOCl solution (200-300 ppm) during 15 minutes, which was followed by a back flush for removal of the chemicals. In experiments 3-1 to 3-4 an additional *thorough chemical cleaning* was applied, in which the membrane was soaked in a NaOCl solution (200-300 ppm) for

more than 16 hours, which was followed by a back flush. The effectiveness of the applied cleaning procedure for the removal of the fouling layer was evaluated afterwards by the Reversibility as a function of the applied cleaning procedure.

3.4 Results

3.4.1 General

The results obtained in the experiments are presented in two series: first the resistance of the fouling layer is shown in tables 3.4 to 3.6, together with some graphical presentation (figures 3.1 and 3.2); in Appendix 3-A all data of tables 3.4 to 3.6 are presented graphically. In the subsequent tables (tables 3.7 to 3.9) the feedwater Filterability and the fouling layer Reversibility are presented. In this section some comments are made on the first series of results, concerning the increase of the total membrane resistance. In the following sections the feedwater Filterability (§3.4.2) and the Reversibility of the fouling layer (§3.4.3) are commented.

Table 3.4 Resistance increase found during experiments on dead- end ultrafiltration of effluent (wwtp Ede; PVDF membrane) at 15°C[a]

Exp.	Feed	J	V_{tot}/A_m	R_{cl}	$\Delta R_{fo,fwf}$	$\Delta R_{fo,cwf}$	ΔR_{ff}	ΔR_{bf}	ΔR_{cf}
		(l/m^2.h)	(l/m^2)	(10^{12} m^{-1})	(10^{12} m^{-1})	(10^{12} m^{-1})	(10^{12} m^{-1})	(10^{12} m^{-1})	(10^{12} m^{-1})
1-1	Effluent[b]	35	440	0.9	8.1	5.9	5.9	3.4	0.8
	Effluent[b]	70	280	0.9	3.9	3.3	3.6	2.6	0.9
	Effluent	100	136	1.0	2.1	2.2	1.5	1.1	0.4

[a] Applied cleaning methods did ***not*** include *forward flush with air* and *thorough cleaning*; [b] Based on one experiment

Tables 3.4 to 3.6 show for each experiment the applied flux J, the filtered feedwater volumes relative to the membrane surface area (V_{tot}/A_m) at the end of the run and the initial resistance of the clean membrane (R_{cl}). In the following two columns the increase in resistance due to the filtration of (pre-treated) effluent is presented, which was measured with the feedwater ($\Delta R_{fo,fwf}$) and with permeate ($\Delta R_{fo,cwf}$). The last series of columns show the fouling layer resistance; each column represents the remaining

fouling layer resistance right after cleaning the membrane with a *forward flush* ($\Delta R_{fo,ff}$), a *forward flush with air* ($\Delta R_{fo,ff+a}$) and a back flush ($\Delta R_{fo,bf}$). Finally, the remaining resistance of the fouling layer after a *chemical flush* ($\Delta R_{fo,cf}$) and after a thorough chemical cleaning ($\Delta R_{fo,tc}$) are shown.

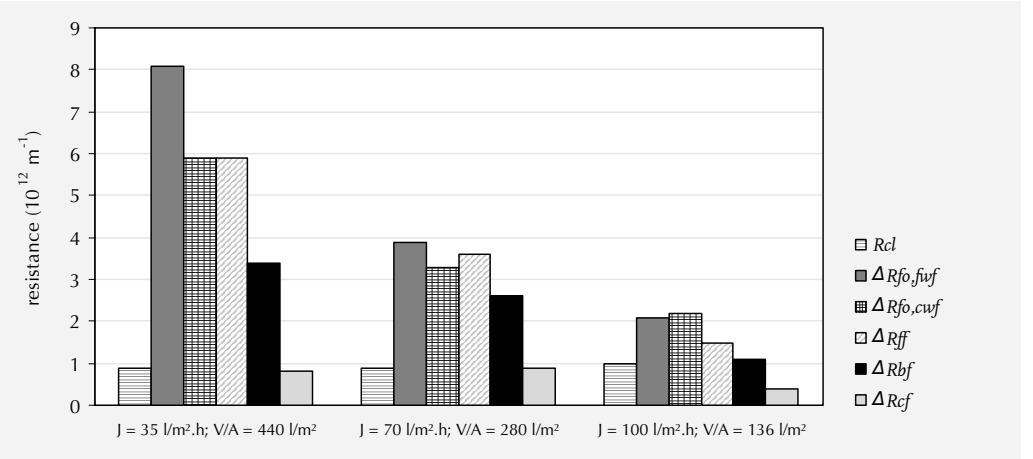

Figure 3.1 **Resistance build up in experiment 1-1; feedwater was effluent from wwtp Ede and membrane is PVDF 5.2 mm; measured at various fluxes**

The standard deviation of the measured resistances is in all experiments 20% or less. The data in table 3.4 to 3.6 show that the initial resistance of the ultrafiltration membranes in all experiments was about 0.9 to $1.2 \cdot 10^{12}$ m^{-1}, resulting in a membrane permeability of 350 to 265 l/m².h.bar at 15°C.

Table 3.5 **Resistance increase found during experiments on dead-end ultrafiltration of effluent (wwtp Kaffeberg; PES/PVP(1) membrane) at 15°C** [a]

Exp.	Feed	J (l/m².h)	V_{tot}/A_m (l/m²)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{ff+a} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})
2-1	Effluent	80	36	0.9	1.9	1.2	0.6	0.4	0.3	0.2
2-2	Effluent+ [b]	80	59	0.9	1.6	1.1	1.0	0.8	0.1	0.2

[a] Applied cleaning methods did ***not*** include *thorough cleaning*; [b] Based on one experiment

The resistance of the fouling layer was strongly influenced by the filtered volume of feedwater. This is illustrated in figure 3.1 showing experiment 1-1 in which the resistance was about 3 to 7 times higher than the initial membrane resistance and the filtered volume was reasonably large. In experiments 2-1 up to 3-4 the resistance of the fouling layer was 1 to 2 times higher than the initial membrane resistance. In figure 3.2 the results of experiment 3-2 are shown. This experiment was stopped at a fixed volume of 86 l/m², resulting in a similar fouling behaviour for all applied fluxes.

Table 3.6 Resistance increase found during experiments on dead-end ultrafiltration of effluent (wwtp Ede; PES/PVP(2) membrane) at 15°C [a]

Exp.	Feed	J (l/m².h)	V_{tot}/A_m (l/m²)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})	ΔR_{tc} (10^{12} m^{-1})
3-1	Effluent	40	160	1.2	3.1	2.5	2.1	0.3	0.2	0.0
	Effluent	70	70	1.0	2.7	2.0	1.8	0.8	0.5	0.1
	Effluent	100	96	0.9	1.7	1.0	0.8	0.1	0.1	0.0
3-2	Effluent	40	86	1.1	0.8	0.8	0.7	0.1	0.0	0.0
	Effluent	70	86	1.2	0.9	0.8	0.7	0.2	0.2	0.0
	Effluent	100	86	1.0	1.0	0.7	0.7	0.2	0.1	0.1
3-3	Effluent+	40	86	0.9	0.9	0.8	0.7	0.1	0.0	0.1
	Effluent+	60	86	0.9	1.0	0.9	0.8	0.0	0.0	-0.1
3-4	Filtrate	40	180	1.1	1.9	2.3	2.2	0.0	0.0	0.0
	Filtrate	70	210	1.0	2.3	2.1	2.1	0.1	0.1	0.0
	Filtrate	100	142	1.0	1.6	1.1	1.0	0.1	0.1	0.0

[a] Applied cleaning methods did ***not*** include *forward flush with air*

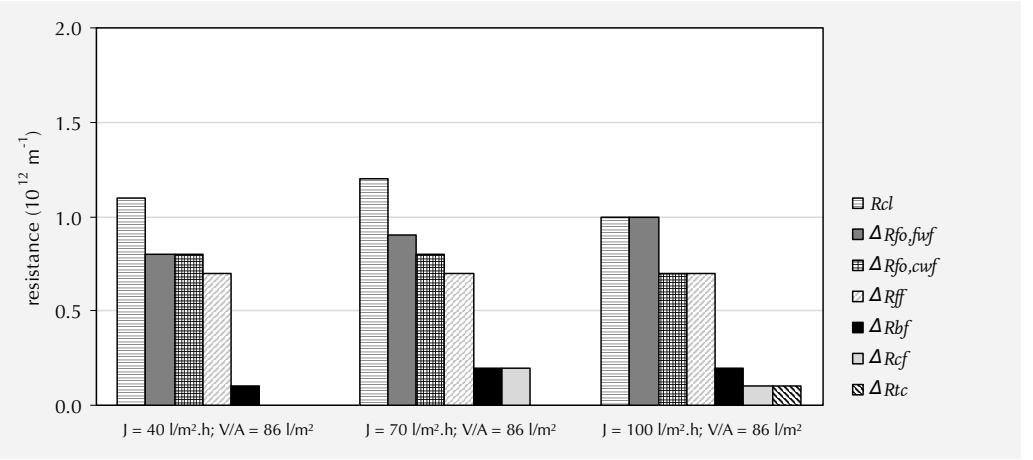

Figure 3.2 Resistance build up in experiment 3-2; feedwater was effluent from wwtp Ede and membrane is PES/PVP 0.8 mm; measured at various fluxes and ended at fixed volume (86 l/m²)

The results presented in table 3.4 to 3.6 showed that in all experiments the resistance of the fouling layer measured with the feedwater ($\Delta R_{fo,fwf}$) had a higher value than the resistance measured with permeate ($\Delta R_{fo,cwf}$), indicating that the resistance of the fouling layer just before the end of the filtration period was higher than the resistance measured with ultrafiltration permeate at the start of the cleaning experiments. This may be attributed to the higher applied TMP in the measurement of the $\Delta R_{fo,fwf}$, namely 0.8 bar in experiments 1, 2, 3-1 and 3-4. The $\Delta R_{fo,cwf}$ has been determined from the permeability (measured with permeate) that is measured at various TMP values, resulting in an average TMP that was lower than 0.8 bar.

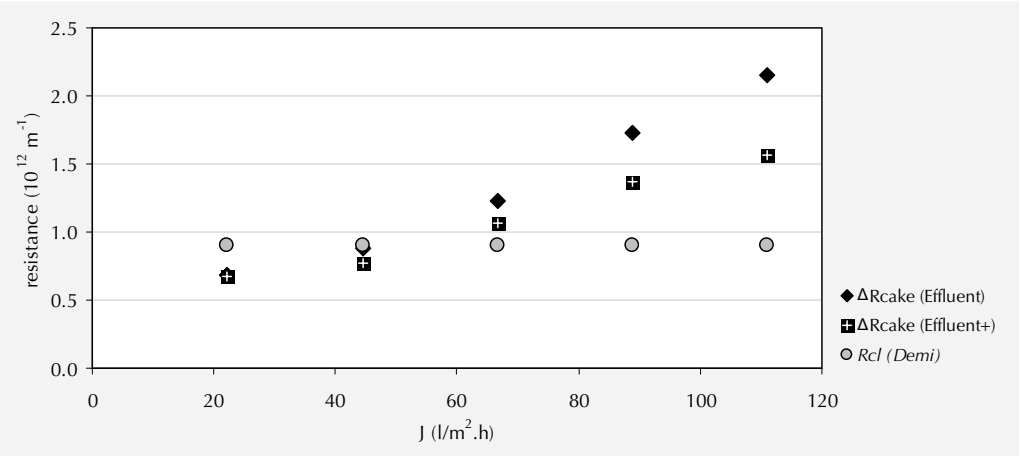

Figure 3.3 **Resistance of the fouling layer (ΔR_{cake}) as a function of the applied flux (or TMP) as found during the measurement of the permeability with ultrafiltration permeate; these data were measured at wwtp Kaffeberg after ultrafiltration of effluent and ultrafiltration of coagulated effluent (effluent+); similar results were found at wwtp Ede; for comparison the resistance of the clean membrane (R_{cl}) is presented as found during the permeability measurement**

Another finding from the measurement of the permeability is that an increase in total filtration resistance was found at increasing TMP (*i.e.* increasing flux). This is shown in figure 3.3, which presents the resistance of the fouling layer as a function of the applied flux (*i.e.* applied TMP). The resistance increased at an increased flux (or TMP). The increase is less for effluent that is pre-treated with a coagulant. The resistance of the clean membrane as a function of the applied flux or TMP is presented for comparison. These results are an indication for the compressibility of the fouling layer, which may also (partly) explain the higher value of $\Delta R_{fo,fwf}$.

3.4.2 Feedwater Filterability

Tables 3.7, 3.8 and 3.9 present the Filterability of the feedwater and the Reversibility of the fouling layer for each experiment. The Filterability (F) was calculated according to equation 3.2, using the data from table 3.4, 3.5 and 3.6. The Reversibility (R_x) was calculated for each of the applied cleaning methods, using also these data and equation 3.3. Firstly, flux and filtered feedwater volume are presented, followed by the Filterability. In the subsequent columns the fouling layer Reversibility is presented for

a *forward flush* (R_{ff}), a *forward flush with air* (R_{ff+a}) and a *back flush* (R_{bf}). Finally, the resistance of the fouling layer after a *chemical flush* (R_{cf}) and thorough chemical cleaning (R_{tc}) are shown.

Table 3.7 **Filterability of feedwater (*F*) and Reversibility (R_x) of the fouling layer in experiments on dead-end ultrafiltration of effluent using various cleaning methods (wwtp Ede; PVDF membrane)**[a]

Exp.	Feed	J (l/m^2.h)	V_{tot}/A_m (l/m^2)	F (10^{-14} m^2)	R_{ff} (%)	R_{bf} (%)	R_{cf} (%)
1-1	Effluent[b]	35	440	7.4	0	42	87
	Effluent[b]	70	280	8.5	-9	20	73
	Effluent	100	136	6.3	29	50	81

[a] Applied cleaning methods did ***not*** include *forward flush with air* and *thorough cleaning*; [b] Based on one experiment

The results found in experiment 1-1 (table 3.7) showed no clear relationship between flux and Filterability *F*, nor between filtered feedwater volume and Filterability *F*. The experiments at differend fluxes were performed with a time interval between two experiments of at least one week. As the composition of wwtp-effluent shows variations in time, the results might better be related to these variations. These findings indicate the difficulty to reproduce results in these pilot-scale experiments.

The results found in experiment 2 (table 3.8) show an increase in Filterability *F* from $3.0 \cdot 10^{-14}$ m^{-2} for effluent to $5.3 \cdot 10^{-14}$ m^{-2} for coagulated effluent (0.5 mg Al^{3+}/l). This can only be seen as an indication for improvement of the Filterability by coagulation of the effluent, as the Filterability is also influenced by the composition of the feedwater (experiment 1-1).

Table 3.8 **Filterability of feedwater (*F*) and Reversibility (R_{ff}) of the fouling layer in experiments on dead-end ultrafiltration of effluent using various cleaning methods (wwtp Kaffeberg; PES/PVP(1) membrane)**[a]

Exp.	Feed	J (l/m^2.h)	V_{tot}/A_m (l/m^2)	F (10^{-14} m^2)	R_{ff} (%)	R_{ff+a} (%)	R_{bf} (%)	R_{cf} (%)
2-1	Effluent	80	36	3.0	48	63	72	84
2-2	Effluent+[b]	80	59	5.3	9	23	93	86

[a] Applied cleaning methods did ***not*** include *thorough cleaning*; [b] Based on one experiment

Experiments 3-1 to 3-4 were performed within one week each, *i.e.* experiment 3-1 consists of sub-experiments (two to three for each flux) that were performed within five days in total. The same procedure is followed for experiment 3-2, 3-3 and 3-4. Between each experiment (3-1, 3-2, 3-2 and 3-4) the time interval amounted to more than one week. The time between two sub-experiments was decreased in order to minimize the change in composition of the effluent. However, the results from these experiments provided no clear results either.

Table 3.9 Filterability of feedwater (*F*) and cleaning dependent Reversibility (R_x) of the fouling layer in experiments on dead-end ultrafiltration of effluent (wwtp Ede; PES/PVP(2) membrane)[a]

Exp.	Feed	J (l/m^2.h)	V_{tot}/A_m (l/m^2)	F (10^{-14} m^2)	R_{ff} (%)	R_{bf} (%)	R_{cf} (%)	R_{tc} (%)
3-1	Effluent	40	160	6.4	16	88	94	98
	Effluent	70	70	3.6	8	58	74	95
	Effluent	100	96	9.5	19	86	94	101
3-2	Effluent	40	86	11.3	8	87	95	103
	Effluent	70	86	11.0	6	68	78	101
	Effluent	100	86	11.6	3	77	80	91
3-3	Effluent+	40	86	11.3	3	91	100	92
	Effluent+	60	86	9.8	9	95	99	108
3-4	Filtrate	40	180	7.9	1	98	99	102
	Filtrate	70	210	10.1	-1	96	97	101
	Filtrate	100	142	12.7	12	95	94	103

[a] Applied cleaning methods did ***not*** include *forward flush with air*

The results of experiments 3-1 and 3-2 (table 3.9) show a different profile, although the feedwater in both experiments r is aw effluent of wwtp Ede and the process conditions

are the same. In experiment 3-1 no relationship was found between flux and Filterability *F*, but the results seem to indicate a relationship between filtered feedwater volume (V_{tot}/A_m) and Filterability. Experiment 3-2 indicates that Filterability is independent of flux. Experiment 3-3 shows for coagulated effluent a slight decrease in *F* at an increasing flux. The results of pre-filtered effluent show an increase in *F* at increasing flux. In this experiment (3.4) again the filtered feedwater volume varied at varying fluxes.

The Filterability *F* in all experiments (1-1 to 3-4) might be a function of filtered volume, rather than flux or pre-treatment. In figure 3.4 the Filterability *F* is shown for all experiments as a function of the filtered feedwater volume (V_{tot}/A_m). The data in this figure show no clear relationship between filtered volume and *F*. This shows that parameters other than the filtered feedwater volume influence the Filterability, most probably by the feedwater composition.

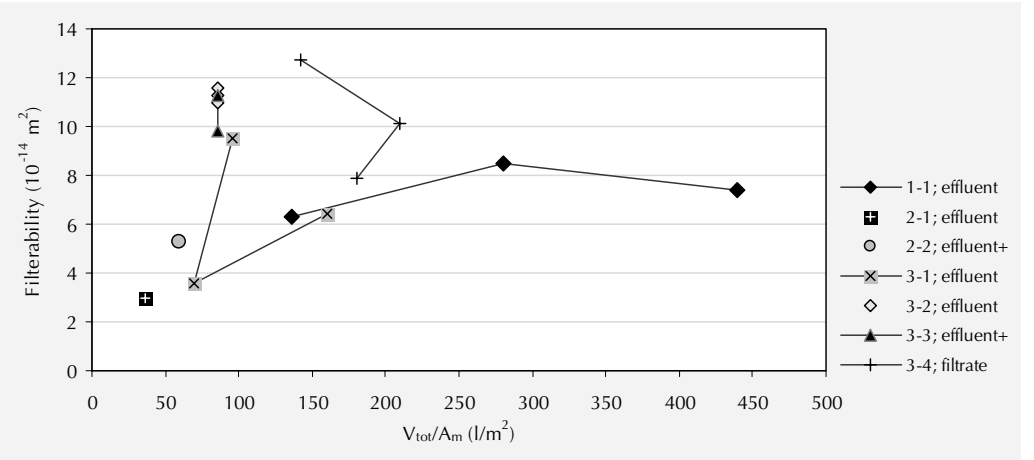

Figure 3.4 Filterability versus filtered feedwater volume per m^2 membrane area (V_{tot}/A_m)

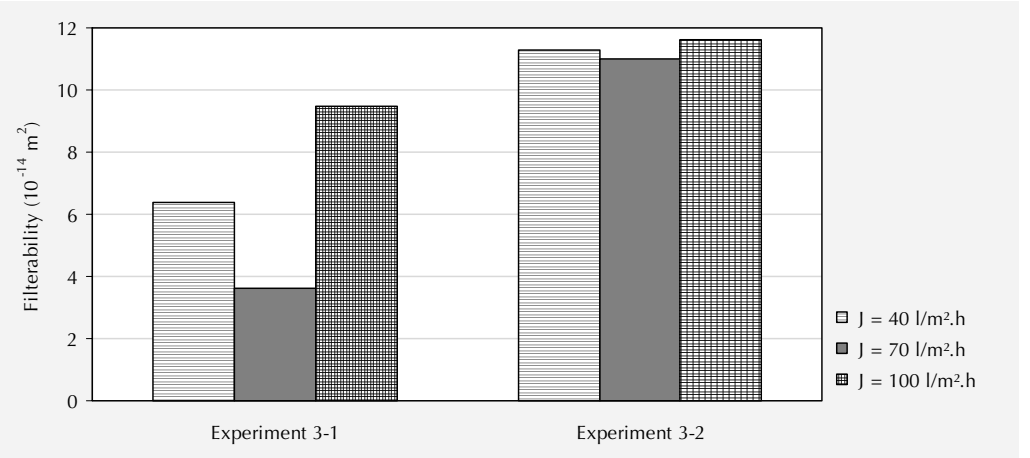

Figure 3.5 Filterability of effluent (wwtp Ede) as found in experiment 3-1 (filtration ends at fixed TMP) and 3-2 (filtration ends at fixed volume)

As shown in figure 3.5 for experiments 3-1 and 3-2 the Filterability showed large variations for the effluent of the same wwtp (Ede), ranging from $3.6 \cdot 10^{-14}$ to $11.6 \cdot 10^{-14}$ m^2. This might be due to variations in the water composition, which results in differences in fouling layer composition. In figure 3.6 the particle numbers that were measured in effluent at wwtp Ede are shown as an example of the effluent water composition. The particle numbers showed during these two months a standard deviation of more than 50%.

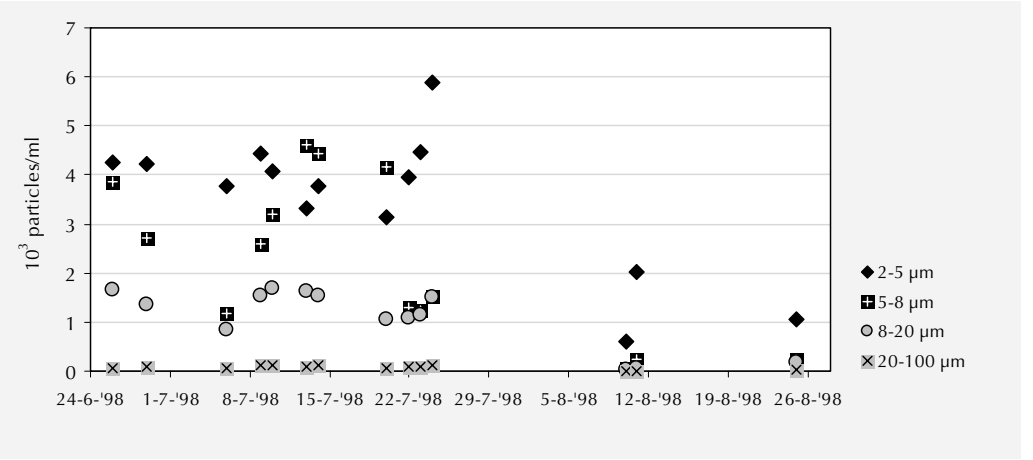

Figure 3.6 **Particle numbers measured in the effluent of wwtp Ede in the period 6/'98 to 8/'98, showing fluctuations in water quality during this period; similar fluctuations were found during other time periods**

Similar fluctuations were found for COD, suspended solids and other parameters. The effluent quality at wwtp Ede is shown as an example in table 3.10, again showing large variations of COD and suspended solids.

Table 3.10 **Water quality of the effluent at wwtp Ede (1/'98 to 10/'98)**

Parameter		Average	Minimum	Maximum
COD	mg COD/l	40	21	59
Suspended Solids	mg SS/l	4	0	9

No relationship was found between the applied fluxes and the Filterability, which indicates that the fouling layer composition was independent of the flux (*i.e.* the TMP). This was confirmed in experiment 3-2 in which the Filterability had the same value of $11.3 \cdot 10^{-14}$ m^2.for all applied fluxes

The Filterability of effluent from wwtp Kaffeberg (experiment 2-1) differed from the Filterability of effluent from wwtp Ede (experiments 3-1 and 3-2), although the membrane types were the same. This indicates that the Filterability was mainly

determined by the feedwater quality and not by the membrane material. However, when comparing the effluent quality at wwtp Kaffeberg and at wwtp Ede (as presented in table 3.2), the differences seem to be small. This indicates that parameters other than Filterability are necessary to explain the filtration and cleaning characteristics of wwtp-effluent (Roorda and van der Graaf, 2001).

The membrane material used in the experiments might partly explain the differences in Filterability found in experiment 1-1 and experiment 3-1 and 3-2, all determined on effluent of wwtp Ede. Doyen *et al.* (1998) showed that different membrane types might have implications on filtration and cleaning characteristics.

In experiments 2-1 and 2-2 the Filterability increased from $3.0 \cdot 10^{-14}$ m² to $5.3 \cdot 10^{-14}$ m², indicating that addition of PACl (0.5 mg Al^{3+}/l) improved the Filterability of the feedwater. However, the pilot-plant experiments at wwtp Ede (3-2 and 3-3) showed no significant improvement of the Filterability after in-line coagulation of PACl. Also the Filterability of effluent pre-treated with a multi-media filter was in the same order of magnitude. Both results indicate that the Filterability was not a direct function of the bigger particles (5-100 μm) that were removed by multi-media filtration or decreased in size by coagulant dosage (as shown by van der Graaf *et al.* (2001)). Filterability was probably more influenced by smaller particles (presumably < 10 μm).

3.4.3 Reversibility of fouling layer

In all experiments the Reversibility of the fouling layer after a forward flush (R_{ff}) was low (maximum was 20%); the forward flush resulted only in experiment 2-1 (table 3.8) in a 48% Reversibility. The membrane module in experiment 2-1 was arranged vertically and the total filtered volume was very low, which might both explain the high reversibility. The low Reversibility of the fouling layer by a forward flush that was found in experiments 3-1 to 3-4 (table 3.9) might be attributed to the configuration of the membrane module. According to the manufacturer, the module was developed for optimised back flushing without the use of a forward flush.

In all experiments the back flush showed a high removal efficiency of the fouling layer, sometimes resulting in a Reversibility of the fouling layer that is higher than 95% (experiment 3-4). However, back flushing in these experiments was not effective enough for complete removal of the fouling layer. The results of experiment 3-1 at a flux of 70 l/m².h, as well as of experiment 3-2 at a flux of 70 and 100 l/m².h,

showed a lower value for R_{bf} than at the other fluxes in these experiments. For these findings no reasonable explanation could be found. Back flushing had to be followed by a chemical cleaning; sometimes the membrane had to be soaked with chemicals for a very long time. In experiments 3-3 and 3-4 the back flush removed 91 to 98% of the fouling layer, which related to a remaining fouling layer with a resistance of only $0.0 \cdot 10^{12}$ to $0.1 \cdot 10^{12}$ m^{-1}. In such cases additional chemical cleaning is not necessary. Next to this, a fouling layer with a low resistance was more effectively removed by a back flush than a fouling layer with a high resistance (see table 3.9).

Experiment 1-1 showed a low fouling layer Reversibility that might be related to the large volume of the filtered feedwater, which caused a high fouling layer resistance of $5.9.10^{12}$ m^{-1}. This indicates that the Reversibility decreased with increasing filtration volume. The Reversibility was low for all cleaning methods that were applied when the filtered volume was large ($V_{tot}/A_m > 200$ l/m²).

In all experiments it was found that the fouling layer Reversibility did not depend on the applied flux.

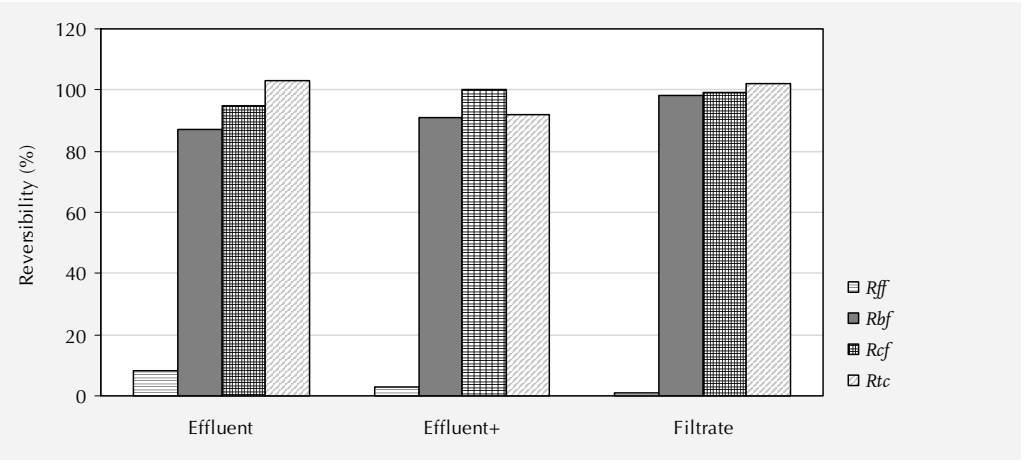

Figure 3.7 Reversibility of the fouling layer in ultrafiltration of wwtp-effluent (Ede), effluent with PACl (effluent+) and filtrate as found in experiment 3-2, 3-3 and 3-4 at a flux of 40 l/m².h

In figure 3.7 and 3.8 the Reversibility of the fouling layer is shown as a function of pre-treatment (coagulation and multi-media filtration). Experiments 2-1 and 2-2 and

experiments 3-2 up to 3-3 showed an improvement of the Reversibility of 5 to 20% after in-line coagulation PACl (0.5 mg Al^{3+}/l) of the wwtp-effluent. Also pre-treatment of the effluent with multi-media filtration improved the Reversibility of the fouling layer, which resulted in a Reversibility of more than 95%.

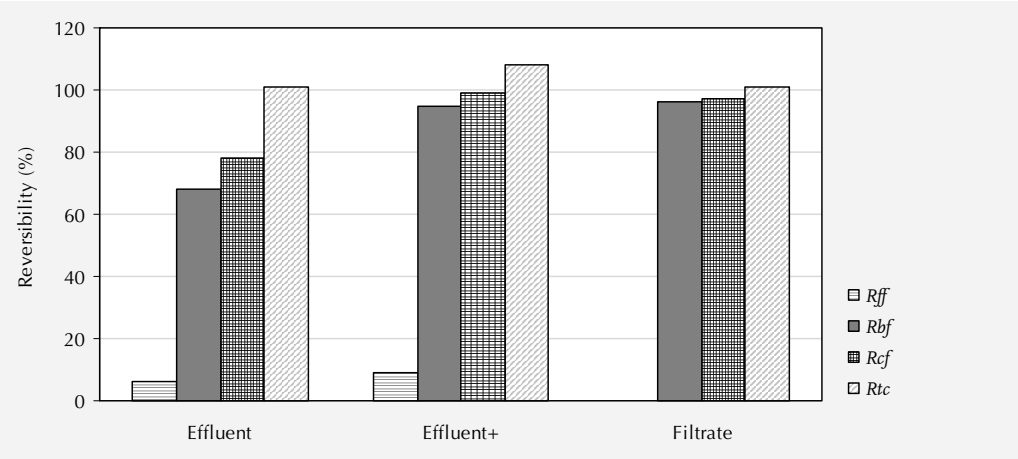

Figure 3.8 **Reversibility of the fouling layer in ultrafiltration for (pre-treated) effluent of wwtp Ede as found in experiment 3: effluent at a flux of 70 l/m².h (exp. 3-2), effluent+ (effluent pre-treated with PACl) at a flux of 60 l/m².h (exp. 3-3) and filtrate (effluent pre-treated with multi-media filtration) at a flux of 70 l/m².h (exp. 3-4)**

3.5 Discussion

The experiments that were done for analysis of the filtration characteristics provided information about the influence of the flux and of pre-treatment on the Filterability of the effluent and the Reversibility of the fouling layer. However, the use of the Filterability F and the Reversibility R_x for characterisation of the ultrafiltration process shows some disadvantages.

Firstly, the experiments are time consuming. One experiment lasts at least a few hours, in most cases leaving not enough time for a duplicate measurement on the same day with the same effluent and the same process conditions. The effect of

changes in process conditions on the filtration characteristics is therefore rather difficult to measure.

Next, the filtration time that is needed in these experiments is rather long compared to the filtration time in pilot-plant tests. The filtration time in pilot-plant tests amounts for 15 to 30 minutes, which relates to a filtered feedwater volume of about 15 to 30 l/m^2. Whereas the filtration time in the characterisation experiments is is mostly a few hours, which relates to 90 to 180 l/m^2. The fouling layer will have a different composition for a large volume of filtered feedwater, than for a smaller volume of filtered feedwater.

Also the composition of the wwtp-effluent shows great variations in time. These variations induce changes in filtration characteristics and therefore influence the reproducibility of the characterisation experiments.

These findings urge the need for other parameters that can be used for characterisation of these filtration phenomena. Measurement of this parameter should be less time consuming and should easily be duplicated. If so, on-line adaptation of the process conditions might lead to a more stable performance of ultrafiltration plants.

Although the information provided by the characterisation experiments is incomplete, some general remarks can be made on the filtration characteristics of wwtp-effluent. The minor effect of pre-treatment on the Filterability of effluent indicates that particles larger than 10 μm have a minor effect on the Filterability. The particles that predominantly influence the effluent Filterability are probably smaller than 10 μm.

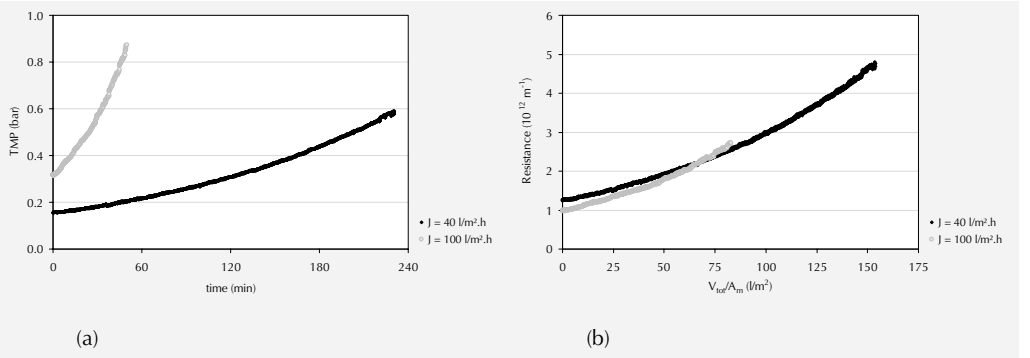

Figure 3.9 **Graph (a) shows the TMP (bar) versus time (min) as measured in experiment 3-1, during dead-end ultrafiltration of wwtp-effluent at constant flux of 40 and 100 l/m².h; graph (b) shows the actual resistance as a function of filtered feedwater volume relative to membrane area (V_{tot}/A_m), as derived from the measured relationship between TMP and *t***

Furthermore, the results provide an indication of compression of the fouling layer. Figure 3.9 shows the TMP-increase as a function of time for two fluxes in experiment 3-1 (figure 3.9 (a)). These data were recalculated to a relationship between 'actual' resistance[1] and the filtered volume, as presented in figure 3.9 (b). This graph shows that, although the relationship between resistance increase and volume seems similar for the first 90 l/m², the slope of the curve increases. Larger volumes will therefore result in a higher resistance and, consequently, a lower Filterability. This complicates the comparison of the results of Filterability experiments that were stopped at a fixed TMP, rather than a fixed volume. The slope of the grey line (figure 3.9 (b)) for a flux of 100 l/m².h is steeper than the slope of the black line that was found for a lower flux of 40 l/m².h. This indicates that the fouling layer compresses at an increasing flux.

The results of experiment 3-2, which was stopped at the same filtered volume, indicate the same. At a flux of 40 l/m².h the resistance of the fouling layer ($\Delta R_{fo,fwf}$) was $0.8 \cdot 10^{12}$ m^{-1}. At a flux of 70 l/m².h the $\Delta R_{fo,fwf}$ was $0.9 \cdot 10^{12}$ m^{-1} and at a flux of 100 l/m².h $\Delta R_{fo,fwf}$ was $1.0 \cdot 10^{12}$ m^{-1}. Since the filtered feedwater volume is the same and the

[1] The 'actual' resistance was calculated with Darcy's law (eq. 3.1) from the filtration data that were measured during filtration of (pre-treated) effluent; in this chapter, most resistances were calculated from permeability data that were measured during filtration of ultrafiltration permeate

TMP increases with increasing flux, the increase of the resistance of the fouling layer should be related to compression of the layer at an increased TMP.

3.6 Conclusions

The Filterability of wwtp-effluent (Ede) was independent of the applied fluxes. The actual quality of the effluent had a great influence on the Filterability. Pre-treatment of wwtp-effluent by in-line coagulation with aluminium (0.5 mg Al^{3+}/l) and by multi-media filtration induced only a small improvement of the Filterability. This finding indicates that filtration characteristics are predominantly determined by small particles (< 10 μm), rather than by larger particles (> 10 μm).

The resistance of the fouling layer increased as a function of the TMP, which relates to compression of the fouling layer. Compression of the fouling layer during ultrafiltration of wwtp-effluent was found both with and without pre-treatment.

The Reversibility of the fouling layer was independent of the applied fluxes in dead-end ultrafiltration of wwtp-effluent (Ede). The effect of pre-treatment was more distinct for the Reversibility than for the Filterability. Pre-treatment of the wwtp-effluent by in-line coagulation with aluminium (0.5 mg Al^{3+}/l) and by multi-media filtration improved the Reversibility of the fouling layer with at least 10% (for a back flush).

Back flushing of the membrane resulted in a high Reversibility of the fouling layer, especially when the resistance of the fouling layer was low. However, the fouling layer was not completely removed.

It is concluded from these experiments, that analysis of pilot-plant data of filtration and cleaning experiments by calculation of both the Filterability (F) and the Reversibility (R_x), provides some indicative information about the filtration characteristics of (pre-treated) wwtp-effluent during dead-end ultrafiltration. However, these parameters provide only limited information that can be used for the

improvement of the performance of ultrafiltration installations for the treatment of wwtp-effluent.

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4 Specific Ultrafiltration Resistance (*SUR*), parameter for evaluation of ultrafiltration characteristics

4.1 Introduction

As shown in Chapter 1 wwtp-effluent is a complex mixture with varying biological, chemical and physical properties. WWTP-effluent contains, as well as other raw water sources, dispersed particles, macromolecules, biological active substances and various ions (Brauns *et al.*, 2002a). Even a single group of substances may represent thousands of chemical species, like for instance the group of humic substances (Drewes and Croue, 2002). The variations of constituents in the effluent in both quantity and quality make it difficult to relate one or a few components to the occurring fouling phenomena.

Wright *et al.* (2001) suggested the use of Flow Field Flow Fractionation (FlFFF) as a method to fingerprint feedwater for membrane filtration. FIFFF is based on solute characteristics and membrane solute interactions. FlFFF is a method to determine particle and macromolecule characteristics, most notably size and diffusion coefficient, in natural and synthetic systems. Brauns *et al.* (2002; 2002a) proposed a multi-value approach of fouling characterization using filtration data (Volume and Permeability), presented in specific graphical formats and tables. That data-presentation provides additional information to other fouling parameters like the Silt Density Index (SDI) and the Modified Fouling Index (MFI), both used for fouling prediction in reverse osmosis systems. The SDI and MFI are measured by filtering a feedwater sample over a flat sheet membrane with a pore size of 0.45 μm at a Trans Membrane Pressure (TMP) of 2.0 bar. The SDI is calculated from the time required to

filter a fixed volume over the 0.45-μm membrane (Wiesner and Aptel, 1996). The MFI is calculated using the same filtration data as the SDI, but interpreting these with the cake filtration theory (Schippers and Verdouw, 1980). To incorporate colloidal particles into the MFI measurement Boerlage *et al.* (2002; 2003) proposed the application of ultrafiltration (UF) membranes and named this parameter the MFI-UF.

Jarusutthirak *et al.* (2002) analysed Effluent Organic Matter (EfOM) in wwtp–effluent and distinguished different fractions of EfOM (colloids and hydrophobic and transphillic fractions) by different techniques. Jarusutthirak *et al.* concluded that the colloidal fraction of wwtp-effluent plays an important role in fouling of the membranes.

In all of these examples a mix of (filtration) properties of the feedwater was related to the occurring fouling phenomena in membrane filtration. This approach may be useful for the prediction of the fouling properties of a single feedwater.

The ultrafiltration process could be optimised when it would be possible to measure the filtration and fouling properties of the feedwater, *i.e.* wwtp-effluent in this research. Until now, there has not been any viable parameter for the determination of filtration characteristics in dead-end ultrafiltration of wwtp-effluent. The results presented in the previous chapter show that it is not easy to measure differences in filterability by performing pilot-plant experiments. The results underline the need for a viable parameter that can be related to the occurring fouling phenomena.

Next to the complex composition of the effluent, also (dis)continuous changes in composition have a distinct effect on ultrafiltration. The composition of the wwtp-effluent is subject to regular changes due to weather influences, discontinuous discharges in the raw wastewater and other aspects (Roorda and van der Graaf, 2001).

Other important aspects in determination of the fouling properties of wwtp-effluent are membrane-solute interactions, which are specific for each combination of membrane type and wwtp-effluent. Fouling parameters like the MFI-UF have to be measured with a specific membrane (Boerlage *et al.*, 2002) and therefore do not take into account the membrane-solute interactions.

Finally, in dead-end ultrafiltration the filtration time is 15 to 30 minutes, after which the membrane is backflushed. The filtration characteristics of the effluent have to be optimised for this time interval, as a high increase of resistance during this time

interval frequently refers to increased fouling of the membrane on the long run (Doyen *et al.*, 1998; Galjaard *et al.*, 1998).

The objective of the research described in this chapter[1] is to develop a parameter for evaluation of filtration characteristics during dead-end ultrafiltration of wwtp-effluent. The Specific Ultrafiltration Resistance (*SUR*) is proposed as a parameter. This parameter provides useful information about the filterability of the wwtp-effluent and can be measured within a short time (30 minutes). This enables on-line evaluation of the filtration properties and adjustment of the feedwater, which may therefore result in a more stable ultrafiltration process.

4.2 Theoretical basis of the Specific Ultrafiltration Resistance (*SUR*)

Darcy's law describes the relationship between flux, TMP, viscosity of the fluid and the total membrane resistance (eq. 1.2 in §1.3.1). In dead-end ultrafiltration of wwtp-effluent cake filtration is assumed to be the predominant filtration mechanism for the increase in resistance; therefore the total filtration resistance (R_{tot}) is the sum of membrane resistance (R_m) and cake resistance (R_c). Combining Darcy's law with the definition of flux (eq. 3.1 in the previous chapter), the following relationship for the total filtration resistance can be derived[2] (eq. 4.1).

$$R_{tot} = R_m + R_c = \frac{\Delta P}{\eta_T} \cdot A_m \cdot \frac{dt}{dV} \qquad \text{(eq. 4.1)}$$

where R_{tot} = total filtration resistance (m^{-1})
R_m = membrane resistance (m^{-1})
R_c = cake resistance (m^{-1})
ΔP = Trans Membrane Pressure, TMP (N/m^2 or Pa)
η_T = dynamic viscosity ($N.s/m^2$ or Pa.s)
A_m = membrane area (m^2)

[1] Part of this work has been presented in Roorda and van der Graaf (2001)

[2] The complete derivation is presented in Appendix 4-A`

t = time (s)
V = filtered volume (m^3)

The membrane resistance R_m is assumed to be constant. The cake resistance is not constant but increases with increasing cake layer thickness. The following equation shows the cake resistance as a function of the average specific cake resistance, the solids concentration and the filtered feedwater volume per m^2 of membrane area (Mulder, 1997). The resistance of the cake layer is the sum of the resistance caused by all particles retained within the cake layer.

$$R_c = \alpha_{av} \cdot c_v \cdot \frac{V}{A_m} \qquad \text{(eq. 4.2)}$$

where α_{av} = average specific cake resistance (m/kg)
c_v = solids concentration in feedwater (kg/m^3)

When equation 4.1 and 4.2 are combined and integrated, assuming constant TMP and temperature ($t_o = 0$; $t_t = t$; $V_o = 0$; $V_t = V$), the relation for t/V and V can be derived (see Appendix 4-A for the complete derivation). Equation 4.3 presents the linear relationship between t/V and V, depending on TMP and viscosity (temperature).

$$\frac{t}{V} = \frac{\eta_T \cdot R_m}{\Delta P \cdot A_m} + \frac{\eta_T \cdot \alpha_{av} \cdot c_v}{2 \cdot \Delta P \cdot A_m^{\,2}} \cdot V \qquad \text{(eq. 4.3)}$$

Except for the average specific cake resistance (α_{av}) and the solids concentration in the feedwater (c_v), all parameters in equation 4.3 are known or can be measured in experiments on dead-end ultrafiltration of wwtp-effluent. The slope of the curve (t/V versus V) may therefore be used to calculate the product of the average specific cake resistance and the solids concentration ($\alpha_{av} \cdot c_v$). In figure 4.1 the theoretically characteristic filtration curve is presented (according to Schippers and Verdouw, 1980). Three filtration mechanisms can be distinguished from this graph: initially pore blocking occurs, which is followed by cake filtration and finally cake filtration with compression of the cake layer.

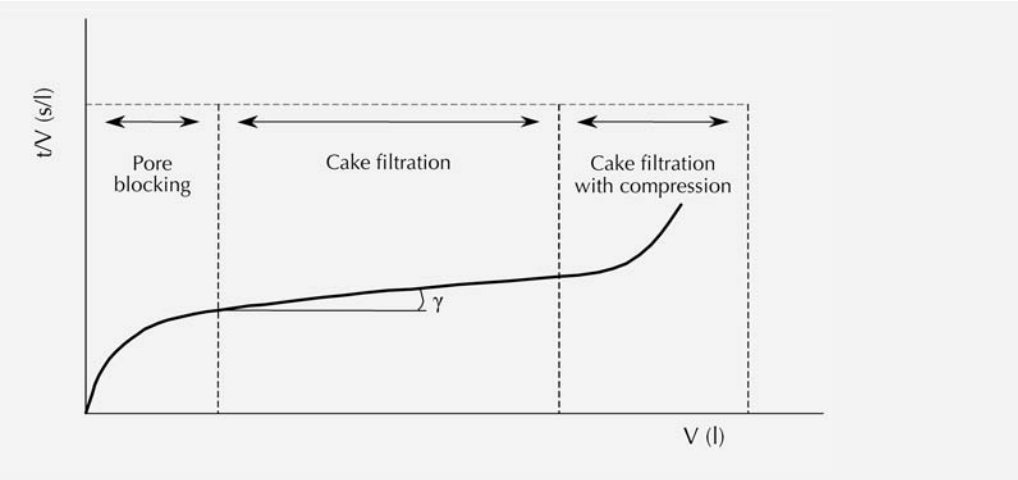

Figure 4.1 **Ratio of filtration time and filtered volume (*t*/*V*) as a function of the total volume of filtered feed water (*V*) at constant TMP; this graph indicates three filtration mechanisms that occur theoretically: pore blocking, cake filtration and cake filtration with compression; tan γ is used to calculate the Specific Ultrafiltration Resistance (*SUR*) (adapted from Schippers and Verdouw, 1980).**

In this dissertation the product of the average specific cake resistance α_{av} and the solids concentration of the feedwater c_v is defined as the Specific Ultrafiltration Resistance (SUR) in m^{-2}. The SUR is used for characterisation of filtration behaviour.

$$SUR = \alpha_{av} \cdot c_v = \frac{d(\frac{t}{V})}{d(V)} \cdot \frac{2 \cdot \Delta P \cdot A_m{}^2}{\eta_T} \qquad \text{(eq. 4.4)}$$

where SUR = Specific Ultrafiltration Resistance, the cake layer resistance per unit of filtered feedwater per m^2 membrane area (m^{-2})

Schippers and Verdouw (1980) used the relation shown in equation 4.3 for the Modified Fouling Index (MFI) that was applied as a parameter for prediction of fouling in RO-systems. The MFI has been normalised to a TMP of 2.0 bar and a membrane area of a standard 0.45 μm microfilter ($13.8 \cdot 10^{-4}$ m^2). Boerlage *et al.* (2002) used the same equation to calculate the MFI-UF using ultrafiltration membranes by

using the same conditions as Schippers and Verdouw. Roorda and van der Graaf (2001) presented the normalised MFI-UF (MFI-UF_n) as a standardised measurement for the prediction of filtration behaviour (more details can be found in Appendix 4-A).

4.3 Experimental set-up and configuration

Experiments were performed to investigate the optimal procedure for measurement of the filtration curve and calculation of the *SUR*. In the experiments described in this section the lab-scale set-up and the process configuration were optimised, focusing on the following aspects: membrane module, constant pressure device, and total filtration time. In section 4.4 the influence of the process conditions (TMP and temperature) on the *SUR* is presented and the description of a procedure for measuring the *SUR* is suggested.

4.3.1 Membrane module for *SUR* measurement

Filtration and fouling phenomena in ultrafiltration of wwtp-effluent depend both on membrane and feedwater characteristics (Galjaard *et al.*, 1998). The *SUR* will be especially useful when this interaction is taken into account. Many membrane types varying in physical and chemical properties are commercially available for full-scale application of ultrafiltration. For measurement of the *SUR* a membrane module was made in which the same membrane type was used as in the pilot- or full-scale plant to which it refers. Two different membrane modules were tested. The first module is shown in figure 4.2 in which the membrane is not supported. In this module the membrane was easily damaged during measurement of the filtration curve. Therefore a second module type has been tested, as shown in figure 4.3.

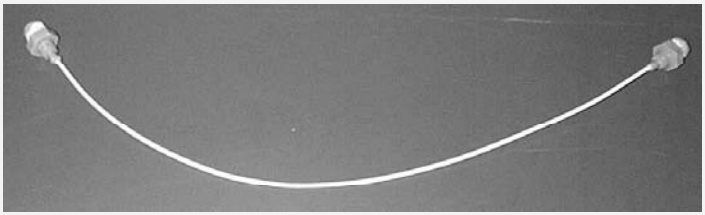

Figure 4.2 **First membrane module with a membrane without module support for measurement of the filtration curve during dead-end ultrafiltration; the membrane was easily damaged during an experiment**

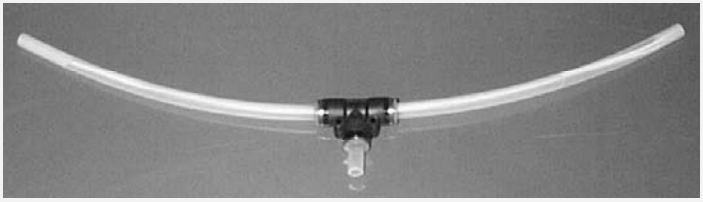

Figure 4.3 **Second membrane module with PVC-tube supported membrane for measurement of the filtration curve during dead-end ultrafiltration; this module showed good results and is proposed as the standard module for SUR measurement**

The membrane in the second module (figure 4.3) was glued in a PVC tube (6 mm inner diameter), in which permeate is collected and continuously discharged through the outlet in the middle of the module. This module with the supported membrane showed good results and is proposed as the standard module for SUR measurements.

4.3.2 Constant pressure difference device

The relationship between t/V and V, used for calculation of the SUR was derived by using the cake filtration theory at a constant TMP. In this research three different devices for a constant TMP were tested. The first device was the lab-scale unit shown in figure 4.4 in which constant pressure is provided by a pump that recirculates feedwater with a sub flow over the membrane. The feedwater temperature was measured with sensor T before and after an experiment, TMP was manually kept at a constant value and was analogically measured. Filtration data were sent to a personal

computer (PC) that calculated the curve for t/V - V relationship. Experiments using this unit showed an increase of feedwater temperature with several degrees Celsius within 10-15 minutes. Next to this, the pump caused turbulence resulting in changes of the feedwater composition. Therefore, this lab-scale unit was transformed into the second device shown in figure 4.5.

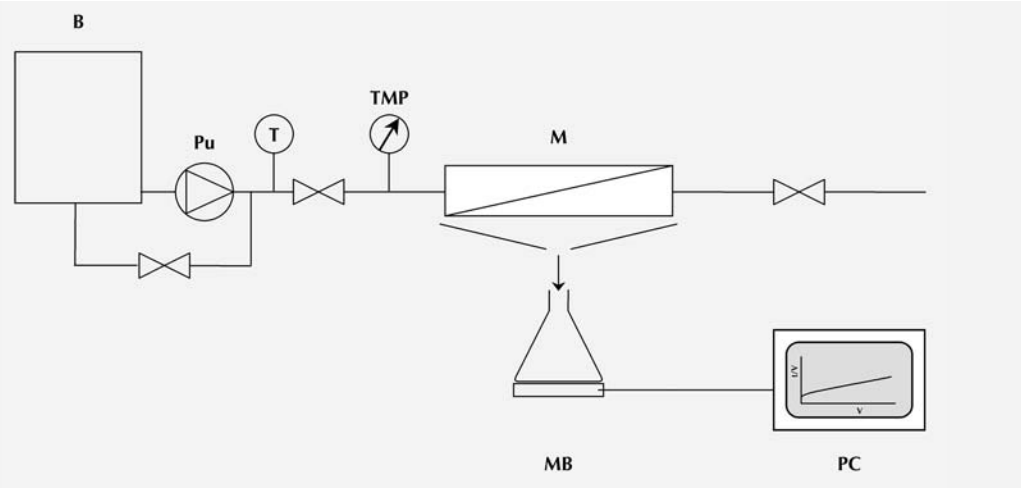

Figure 4.4 **First lab-scale unit with a feedwater pump and recirculation of the feedwater through the pump; this unit is used for measuring filtration curves and calculating *SUR*; in which B: buffer tank, T: temperature sensor, Pu: pump, TMP: pressure sensor, M: membrane module, MB: analytical balance, PC: personal computer**

The air-pressured buffer tank (volume is 10 dm^3) provided a constant pressure difference over the membrane. Initially the TMP was measured analogically, and manually kept constant at 0.5 bar. Permeate was collected in a beaker on an analytical balance (Mettler Toledo, 0-2000 g) that was connected to a computer with data-analyses software. Some experiments were performed with a digital TMP measurement device, the data of which were sent to the PC together with the filtration data. The buffer tank was filled to a maximum of 50% with feedwater, the minimum sample volume was 1.5 dm^3. The feedwater is continuously stirred in the buffer tank.

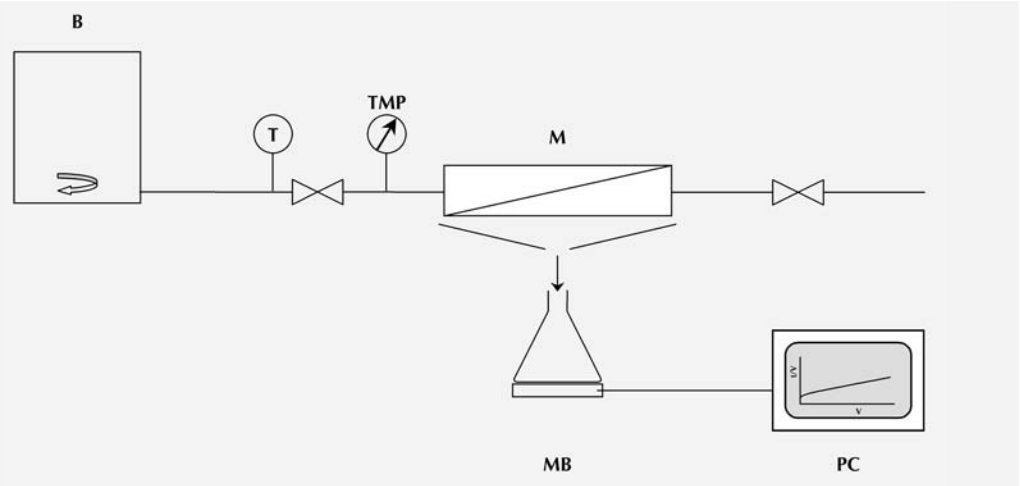

Figure 4.5 **Second lab-scale unit with a pressurised buffer tank (B) collecting permeate in a separate permeate collector used for measuring filtration curves and calculating SUR; B: continuously stirred pressurised buffer tank, T: temperature sensor, TMP: pressure sensor, M: membrane, MB: mass balance and PC: personal computer**

Finally, the lab-scale unit was modified by putting two pressurised buffer tanks in parallel. As shown in figure 4.6, buffer tank B1 was filled with demineralised water and the buffer tank B2 with feedwater. The use of two pressurised buffer tanks enabled initial filtration with demineralised water until TMP was constant. When TMP was constant valve 1 (V1) was changed from buffertank 1 to buffertank 2 providing constant TMP at the start of the feedwater filtration experiment. This led to more accurate filtration data. TMP was measured digitally and feedwater temperature was measured before and after an experiment. The device shown in figure 4.5 (one pressurised buffer tank) has been used in most experiments presented in the subsequent part of this chapter. The device with two pressurised buffer tanks in parallel was used in the experiments described in chapters 5 and 6 of this dissertation.

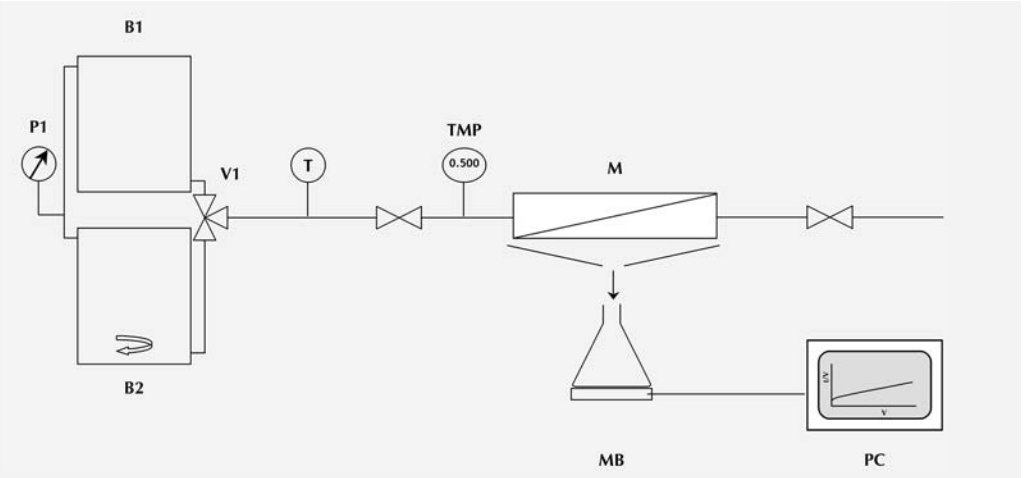

Figure 4.6 **Lab-scale unit with two pressurised buffer tanks and used for measuring filtration curves and calculation of the *SUR*; B1: pressurised buffer tank 1 filled for 50% with demineralised water, B2: continuously stirred pressurised buffer tank 2 filled with feedwater, V1: valve choosing between Buffertank 1 or 2, T: temperature sensor, P1: constant pressure device before buffer tank (~0.5 bar), TMP: digital pressure sensor, M: membrane, MB: mass balance and PC: personal computer**

4.3.3 Total filtration time

In dead-end ultrafiltration of a feedwater the most frequently used cleaning method is the back flush (see chapter 1). The time interval between two back flushes is normally 15 up to 30 minutes of filtration. It has been suggested that the filtration and fouling phenomena that occur during this short time interval influence the long-term fouling (Doyen *et al.*, 1998; Galjaard *et al.*, 1998). To characterise the short-term effect of interaction between feedwater and membrane with the *SUR*, the calculated value has to be stable within a time interval of 15 up to 30 minutes. For example, in figure 4.7 a graphical presentation is shown of the filtration curve and related *SUR* curve, which was found in a lab-scale experiment using effluent from wwtp Berkel (sample date: 14-5-'02). In the first minutes of filtration the *SUR* increased due to the development of a cake layer. After ten minutes of filtration the *SUR* was more stable and showed a value of $(15.8 \pm 0.3)\cdot10^{12}$ m^{-2} (a deviation of 1.8%) in the following twenty minutes of filtration, from 10 to 30 minutes of filtration.

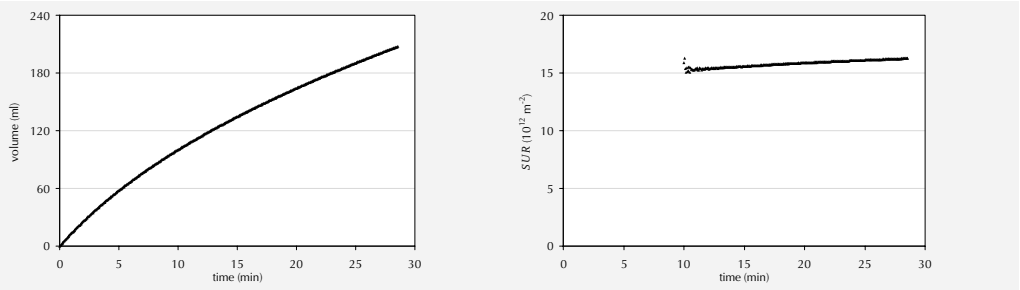

Figure 4.7 **Result of lab-scale experiment on filtration of effluent from wwtp Berkel (14-5-'02) using the device with two buffer tanks in parallel (shown in figure 4.6); the filtration curve is shown in the graph at the left side, the calculated *SUR* curve is shown in the graph at the right side; between *t* = 10 and *t* = 30 minutes the *SUR* is determined (15.8 ± 0.3)·10^{12} m^{-2} (standard deviation of 1.8%)**

Similar filtration experiments were performed on effluent from wwtp Berkel for a longer period, *i.e.* more than ten hours (figure 4.8). These long-term experiments showed a continuously increasing *SUR*, with a large increase within the first two hours of filtration. During the last ten hours of filtration the *SUR* still increases, but at a lower rate. In figure 4.8 the results are presented for the first thirty minutes of filtration (graph left) showing an average *SUR* of (8.7 ± 0.3) ·10^{12} m^{-2}. After two hours of filtration the *SUR* increased to an average of (10.4 ± 0.1) ·10^{12} m^{-2}, as shown in the graph on the right in figure 4.8.

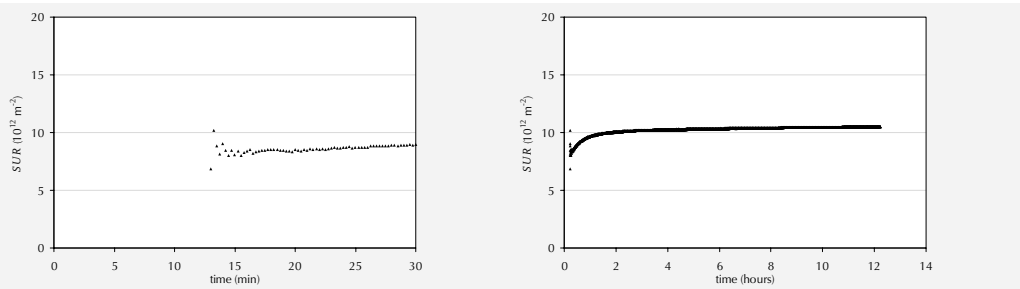

Figure 4.8 **Results of long-term lab-scale experiment on filtration of effluent from wwtp Berkel (26-10-'00) using the device with one buffer tank (shown in figure 4.5); the development of the *SUR* is presented for the first 30 minutes of filtration graph in the graph on the left side, with *SUR* is (8.7 ± 0.3) ·10^{12} m^{-2}; the *SUR* is presented for twelve hours of filtration in the graph on the right side, after two hours the *SUR* increased to an average of (10.4 ± 0.1) ·10^{12} m^{-2}**

In the research presented in this dissertation the *SUR* was calculated from data of a lab-scale experiment with dead-end ultrafiltration performed for about 30 minutes. The standard deviation of the value of the *SUR* was similar in all experiments. The *SUR* had a standard deviation to a maximum of 3% within the first 30 minutes of filtration (for more details on the calculation of the *SUR* in practice see the description of the experimental procedure in the following section (§4.4.1)).

4.4 Influence of process parameters on the *SUR*

In this section the general experimental procedure that was followed is described. Next, the influence of the following process parameters on the *SUR* is investigated: Trans Membrane Pressure (TMP), feedwater temperature, and membrane type. All experiments described in §4.4 have been performed using the device with one pressurised buffer tank (figure 4.5). In cases where the device with two buffertanks (shown in figure 4.6) was used, this is mentioned explicitly.

4.4.1 Experimental procedure for measuring *SUR*

The applied experimental procedure for measuring the *SUR* was as follows. By gluing a commercially available capillary membrane (0.8 or 1.5 mm inner diameter) in a PVC tube (6 mm inner diameter) the membrane module was made. In order to remove conditioning liquid and other impurities, the membrane module was then soaked for 30 minutes in a NaOCl solution (200-400 ppm).

After soaking in a NaOCl solution the membrane module was placed in the lab-scale filtration unit (figure 4.5) and the buffer tank was filled with demineralised water (5 dm^3, 50% of the volume of the buffer tank). Next, the membrane module was cross-flushed with demineralised water (for about 2 minutes). Back flushing of the membrane was not possible with the lab-scale unit. Finally, the permeability (in $l/m^2.h.bar$ at 15 °C) was determined by filtering demineralised water over the membrane at a TMP of 0.5 bar. Before and after filtration the temperature of the demi-water was measured.

The demineralised water was removed from the buffer tank after determination of the permeability. The feedwater for measurement was added to the

buffer tank. The whole system was flushed with feedwater (for 2 minutes) and by closing the valve right after the module (see figure 4.5) the filtration mode could be transformed from cross-flow to dead-end filtration. The TMP was manually set to 0.5 bar and the feedwater was filtered over the membrane. After 30 minutes of filtration the system was cross-flushed with feedwater and the membrane module was removed from the lab-scale unit and soaked in a sodium hypochlorite solution. After soaking for 30-60 minutes the permeability for demi-water was measured. If the change in permeability for demi-water was less than 10% as compared to the initial value, the module was (re)used in the next experiment.

The collected data were stored in a computer and were analysed in a spreadsheet model for calculation of the *SUR*. An example of this spreadsheet is presented in Appendix 4-B. The slope of the $t/V - V$ curve was evaluated and the interval for which the slope was constant was noted. Next, the *SUR* was calculated from the difference between the highest and the lowest value in this interval for which the slope was constant. This simplification of the calculation of the *SUR* led to an overestimation of the *SUR* of less than 3%[1]. The overestimation was demonstrated by calculating the *SUR* for each point of the $t/V - V$ curve (< 500 points)[2]. The average value of the *SUR* calculated accordingly showed this minor overestimation (whereas the standard deviation was less than 3%). The measurement of the *SUR* was done in duplicate for each sample.

4.4.2 Trans Membrane Pressure (TMP)

Material and methods

The TMP is an important process parameter in membrane filtration processes. Increasing the TMP will increase the flow through the membrane and at the same time increase the deposition of particles. The fouling layer may be compressed at

[1] This minor overestimation (1-3%) was not taken into account in the results presented in this thesis

[2] The *SUR* was calculated as follows, which resulted in an overestimation compared to the average *SUR*:

$$SUR = \frac{(t_2/V_2) - (t_1/V_1)}{V_2 - V_1} = \frac{\Delta(t/V)}{\Delta V}$$

the average *SUR* measured for *n* points of the $t/V - V$ curve, could be calculated as follows:

$$SUR_{average} = \frac{\sum_{n=0}^{n} \left(\frac{\Delta(t/V)}{\Delta V} \right)_n}{n}$$

increasing TMP. Boerlage *et al.* (2003) found a non-linear relationship between TMP and MFI-UF, which was attributed to cake layer compression. Three series of experiments have been performed to investigate the influence of the TMP on the *SUR* during dead-end ultrafiltration of wwtp-effluent. In all experiments a 1.5 mm capillary X-flow membrane (PES/PVP) was used. In table 4.1 an overview of these experiments is shown.

The first series of experiments (TMP-1) were performed with effluent from wwtp Berkel at a TMP of 0.5, 1.0 and 2.0 bar. The experiment started by measuring the *SUR* at a TMP of 0.5 bar. After 30 minutes of filtration, the membrane was cross-flushed (without soaking in chemicals) and the TMP was set to 1.0 bar. The same procedure was applied for measurement of the *SUR* at a TMP of 2.0 bar. Finally, at the end of the experiment, the membrane was soaked for 5-20 hours (during the night) with NaOCl solution (200 ppm). This experiment was repeated on the following three days, using the same effluent sample that was stored in a refrigerator (at 4°C).

Table 4.1 Three series of experiments for the study of the influence of TMP on *SUR* using effluent from wwtp Berkel

Experiment	TMP (bar)	Increasing/decreasing TMP	NaOCl cleaning	No. of experiments
TMP-1	0.5, 1.0, 2.0	Increasing (0.5 -> 2.0)	No	4
TMP-2	0.25, 0.5, 1.0, 1.5, 2.0	Increasing (0.25 -> 2.0)	Yes	2
TMP-3	0.25, 0.5, 1.0, 1.5, 2.0	Decreasing (2.0 -> 0.25)	Yes	2

The second series of experiments (TMP-2) was also carried out with effluent from wwtp Berkel for five different TMP's (0.25, 0.5, 1.0, 1.5, and 2.0 bar) and started by measuring the *SUR* at the lowest TMP (0.25 bar). The experiment was finished by determination of the *SUR* at the highest TMP (2.0 bar). The membrane was soaked for 15 minutes with 200-400 ppm NaOCl and cross-flushed before increasing the TMP for the following experiment. This experiment was repeated on the following day, using the same effluent sample.

The last series of experiments (TMP-3) was performed under similar conditions as TMP-2, but now with decreasing TMP. The first *SUR* determination was

done at a TMP of 2.0 bar and the last measurement of the *SUR* on a day was done at a TMP of 0.25 bar. This experiment was also repeated on the following day.

Finally, the membrane permeability for demineralised water was determined at 0.5, 1.0 and 2.0 bar. The permeability of the membrane was used for calculation of the membrane resistance (equation 1.2).

Results

The results of the experiments are summarised in table 4.2, which shows the average *SUR* measured at the various applied TMP's. A graphical presentation is given in figure 4.9, showing on the X-axis the applied TMP and on the Y-axis the average *SUR* found in the experiments. Before starting the experiment, the permeability of the membrane was measured by filtration of permeate. Contrary to Persson *et al.* (1995), Huisman *et al.* (1997) and Boerlage *et al.* (2003) no change in membrane resistance was found at increasing TMP.

Table 4.2 Results of three series of experiments for the study of the influence of TMP on *SUR* using effluent from wwtp Berkel

Experiment	TMP-1[a]	TMP-2[b]	TMP-3[c]
Applied TMP (bar)	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})
0.25	*nd*[d]	6.2 ± 2.0	4.6 ± 0.0
0.5	7.7 ± 1.0	8.4 ± 0.3	6.4 ± 0.0
1.0	12.3 ± 1.7	12.9 ± 0.3	11.4 ± 0.7
1.5	*nd*	18.3 ± 1.7	13.7 ± 1.1
2.0	21.7 ± 1.9	21.3 ± 0.8	17.8 ± 1.0

[a] Measured at increasing TMP; measured four times; [b] Measured at increasing TMP; measured twice; [c] Measured at decreasing TMP; measured twice; [d] Not determined

Increasing the TMP with 100% from 0.5 to 1.0 bar, showed a 55% to 80% increase of the *SUR*. Similar results were found for a TMP increase from 1.0 to 2.0, which resulted in an additional *SUR* increase of 56% to 75%. The results of experiment TMP-1 and TMP-2 were the same, the results of experiment TMP-3 showed some difference

for TMP of 1.5 and 2.0 bar. This might be due to the differences in the experimental procedure. Experiment TMP-3 started *SUR* measurement at the highest TMP, whereas TMP-1 and TMP-2 started at the lowest TMP. It is possible that the cross-flushing (TMP-1) and chemical cleaning (TMP-2) were not effective enough to remove all retained material within and on the membrane structure. The remaining material on the membrane could have promoted the development of a cake layer and thereby the increase of the *SUR*.

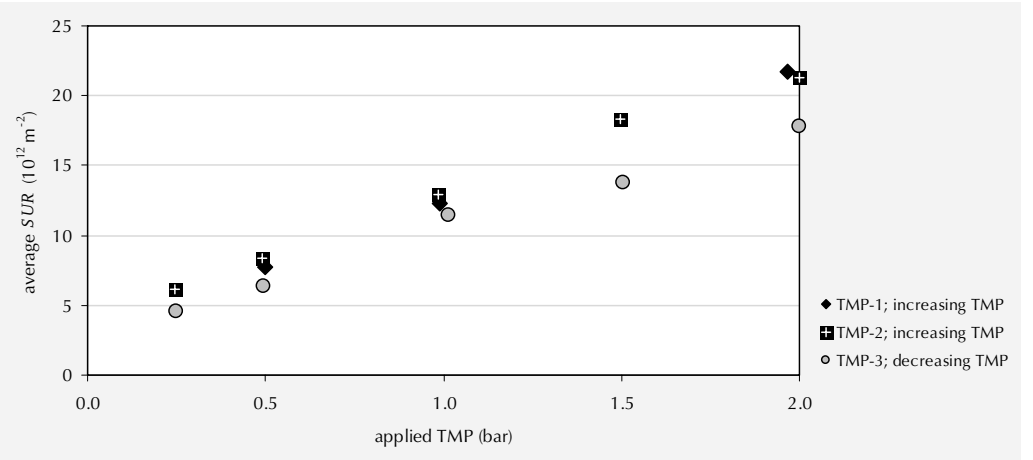

Figure 4.9 ***SUR*** **of wwtp-effluent (Berkel) measured at variable TMP**

The *SUR* was defined as the product of the average specific cake resistance (α_{av}) and the solids concentration in the feedwater (c_v). As the latter (the composition) was kept constant, the increase in *SUR* at increasing TMP should have been caused by an increase of the average specific cake resistance (α_{av}). This might be due to cake layer compression (Hermia, 1982; Mulder, 1997; Boerlage *et al.*, 2003).

Compressibility of the cake layer can be expressed as a variation in the specific cake resistance as a function of the pressure (Boerlage *et al.*, 2003; Lee *et al.*, 2003; Matsumoto *et al.*, 1999; Tiller and Yeh, 1987):

$$\alpha = \alpha_{\circ} \cdot \Delta P^{s} \qquad \text{(eq. 4.5)}$$

where α = specific cake resistance (m/kg)
$\alpha_{\circ}$ = specific cake resistance at reference pressure, *i.e.* 0.5 bar (m/kg)
s = compressibility coefficient, expressing degree of compressibility
(when s = 0, no compression occurs; when s = 1, compression is complete)

This relationship is used for calculation of the compressibility of the cake layer in these experiments, assuming that the *SUR* (specific cake resistance * solids concentration) shows an identical relationship. The log-log curve of the *SUR* versus the TMP is presented in figure 4.10. The compressibility coefficient is the slope of each curve. For the three series of experiments the compressibility coefficient for TMP-1, TMP-2 and TMP-3 are 0.75, 0.61 and 0.66 respectively, indicating a highly compressible cake layer.

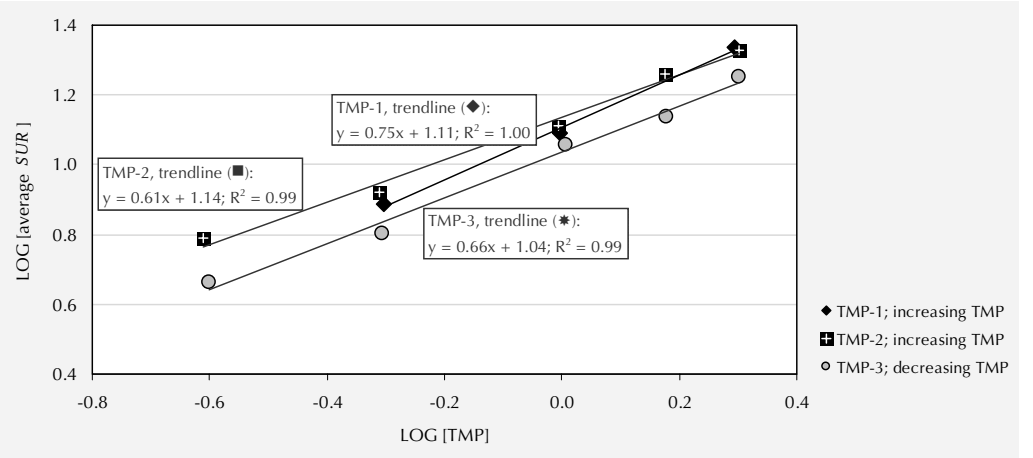

Figure 4.10 LOG-LOG curve of *SUR* of wwtp-effluent (Berkel) against measured TMP

As shown in these results the *SUR* strongly depends on the applied TMP. To use the *SUR* as a parameter for filtration characteristics of effluent, the TMP for *SUR* measurement should be defined as a constant value. In dead-end ultrafiltration of wwtp-effluent in practice, the TMP is generally in the range of 0.2 to 0.8 bar with an average of 0.5 bar. Therefore in this dissertation a TMP of 0.5 bar has been used for the determination of the *SUR* value of (pre-treated) wwtp-effluent.

4.4.3 Temperature of the feedwater

Material and methods

Since the (dynamic) viscosity of water is related to temperature (see equation 1.3), the feedwater temperature is an important factor in the calculation of the *SUR*. In the following experiments the relation between temperature and *SUR* has been investigated for temperatures of 10°C (exp. T-1), 20°C (exp. T-2) and 30°C (exp. T-3). Experiments at a feedwater temperature of 10°C were done with effluent stored in a refrigerator. At 20°C the effluent was first adapted to the ambient temperature. Finally, for experiments with a feedwater temperature of 30°C the effluent was heated with a thermo regulator.

Table 4.3 Three experiments to study the influence of feedwater temperature on *SUR* using effluent from wwtp Berkel

Experiment	Feedwater Temperature (°C)	Effluent age[a] (days)
T-1	10	1[b] and 2[c]
T-2	20	0[c] and 1[b]
T-3	30	2[d] and 3[e]

[a] The effluent had been stored in a refrigerator (at 4-6°C); [b] Measured twice; [c] Measured four times; [d] Measured once; [e] Measured five times

On the first day effluent is taken from wwtp Berkel, after which the first experiment was performed at 20°C. The next day two experiments were performed at 10 and 20°C. The following day the temperature was set to 10 and 30°C and on the final day to 30°C. In table 4.3 a summary of the experiments is presented.

The membranes used in these experiments were 0.8 mm X-flow membranes (PES/PVP). Before the start of each experiment the membrane was soaked in a 50-100 ppm NaOCl solution.

Results

The results of experiments T-1, T-2 and T-3 are presented in table 4.4 and figure 4.11 and show that the *SUR* increased with increasing effluent temperature. As found in these experiments, the *SUR* increased about $9 \cdot 10^{12}$ m^{-2} for every 10°C. Accordingly, for every degree Celsius the *SUR* of effluent from wwtp Berkel changes about $1 \cdot 10^{12}$ m^{-2}. Te Poele and van der Graaf (2002) found similar results for increasing the temperature of wwtp-effluent (Berkel) from 13 to 30°C (*SUR* changed from $18.3 \cdot 10^{12}$ to $30.4 \cdot 10^{12}$ m^{-2}. At even higher temperatures (60 and 90°C) similar results were found.

Table 4.4 Results of experiments on the influence of feedwater temperature in the *SUR* for effluent from wwtp Berkel, together with results of te Poele and vd Graaf (2002) and Meezen (2002)

Experiment	Sampling date	Average temperature feedwater (°C)	*SUR* (10^{12} m^{-2})
T-1	9-2000	9.6 ± 1.3	15.4 ± 0.7
T-2		20.4 ± 0.4	24.4 ± 5.3
T-3		31.2 ± 1.6	33.3 ± 4.4
te Poele and vd Graaf (2002)	4-2001	13.0	18.3
		30.0	30.4
Meezen -1 (2002)	4-2002	13.0	12.4
		14.0	12.7
		15.0	13.1
Meezen -2 (2002)	4-2002	8.0	11.6
		11.0	13.1
		15.5	14.0

These results are in contradiction to the findings for the permeability of Chiemchaisri and Yamamoto (1994) for membrane bioreactor sludge, where at a constant flux an increase of the feedwater temperature resulted in a decrease of the TMP. Also the results of Boerlage *et al.* (2003) on the permeability of tap water showed the opposite, *i.e.* increasing temperature increased the permeability.

The effect of temperature on the *SUR* was only determined for effluent of wwtp Berkel. Also te Poele and van der Graaf (2002) used Berkel effluent, the results of which are added to figure 4.11. The effect on demineralised water is not determined, but no increase of *SUR* is expected at increasing temperature. Te Poele and van der Graaf (2002) found no change in filtration characteristics of demineralised water after increasing the temperature from 30 to 60°C and even from 60 to 90°C. Also no change in membrane resistance was found, an observation that corresponds to the findings of Huisman *et al.* (1997).

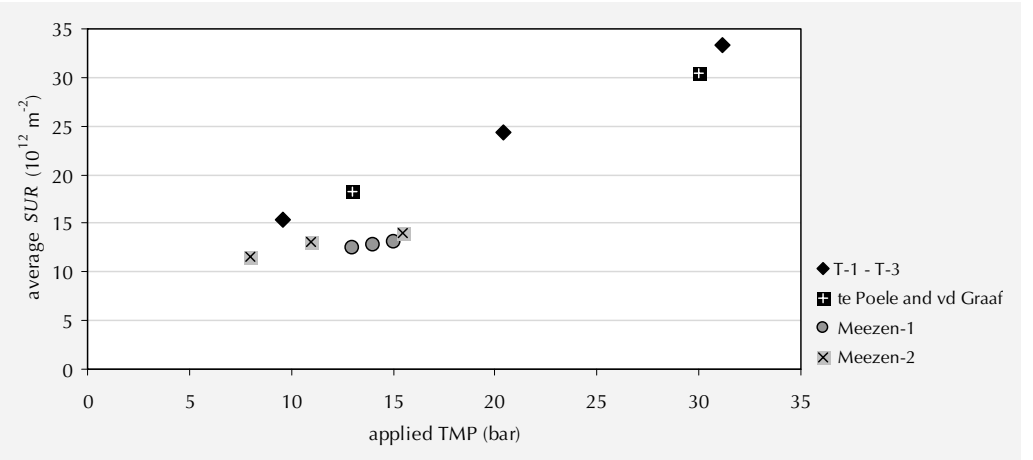

Figure 4.11 Graphical presentation of results of experiments studying the influence of feedwater temperature on *SUR* for effluent of wwtp Berkel

These experiments indicate that the temperature of the wwtp-effluent influences the filtration characteristics, which must be due to changes in the specific cake resistance. As the *SUR* values for experiments T-1 to T-3 are relatively high, it is expected (see data of Meezen (2002)) that effluent with a lower *SUR* will show a smaller change for a

temperature increase. The results of the experiments on wwtp-effluent (Berkel) showed that a pre-defined feedwater temperature is important for using the *SUR* as a parameter for the prediction of filtration behaviour.

4.4.4 Membrane type

Material and methods

As found by various researchers filtration characteristics are specific for different membrane types (Doyen *et al.*, 1998; Galjaard *et al.*, 1998). The influence of the membrane type on the *SUR* was investigated for four types of capillary PES-PVP membranes (X-flow). These membranes differ in internal diameter and Molecular Weight Cut-off MWCO. In table 4.5 the data of the four membranes are presented.

Table 4.5 Specifications of PES/PVP membranes used in experiments determining the relation between membrane type and *SUR*

Name	Internal Diameter[a] (mm)	MWCO[a] (kDalton)	Permeability[a] (l/m².h.bar)
Membrane A	1.5	150	1150
Membrane B	1.5	70	850
Membrane C	0.8	150	1130
Membrane D	0.8	70	800

[a] Data from manufacturer

Experiments were performed to investigate the influence of the internal fiber diameter (0.8 and 1.5 mm) and the MWCO (70 and 150 kDa). Two similar experiments were done, using wwtp-effluent (Berkel) as feedwater. In the first experiment (M-1) the *SUR* has been measured once, using all four membranes and repeating this on the following three days with the same effluent sample (stored in a refrigerator). In the second experiment (M-2) the *SUR* was measured once every two days. The membranes were cleaned before and after the experiment with a NaOCl solution (50-100 ppm).

Results

In table 4.6 the *SUR* is shown as measured in the two experiments for the four different membranes. Membranes A and B showed similar *SUR* values with the same internal diameter but different MWCO. This was also the case for membranes C and D. The membrane with the smallest internal diameter (0.8 mm) resulted in a 15-30% higher value than the membrane with the larger internal diameter (1.5 mm), irrespectively of the MWCO. Membranes A and C have a similar chemical composition and MWCO, the only difference is the internal diameter. The same counts for membrane B and D.

Table 4.6 ***SUR*** **measured for four different capillary membranes with wwtp-effluent (Berkel) as feedwater**

			Experiment M-1[a]		Experiment M-2[b]	
Membrane	ID[c] (mm)	MWCO (kDa)	*SUR* (10^{12} m^{-2})	Corrected (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	Corrected (10^{12} m^{-2})
Membrane A	1.5	150	10.4 ± 0.9	10.4 ± 0.9	7.0 ± 0.0	7.0 ± 0.0
Membrane B	1.5	70	10.1 ± 1.3	10.1 ± 1.3	6.9 ± 0.3	6.9 ± 0.3
Membrane C	0.8	150	12.1 ± 0.7	11.2 ± 0.7	8.2 ± 0.8	7.6 ± 0.8
Membrane D	0.8	70	12.7 ± 1.0	11.8 ± 1.0	8.8 ± 0.4	8.2 ± 0.4

[a] Measured on four consecutive days using the same sample effluent in the whole experiment (M-1); [b] Measured on two consecutive days using the same sample effluent in the whole experiment (M-2); [c] ID = internal diameter

The difference in *SUR* might be explained partly by the accuracy of the internal diameter. According to the membrane manufacturer (Molemaker, 2003) the 0.8 mm membrane is in reality 0.77 ± 0.01 mm, and the 1.5 mm membrane is 1.5 ± 0.05 mm. In table 4.6 the corrected values are presented next to the measured *SUR* values. With the corrected values the results of the membranes with the largest Internal Diameter (ID) are approximately 10% lower than the membrane with the smallest ID. It might be possible that the differences could be related to the differences in hydraulic conditions, but no clear explanation was found.

4.5 *SUR* for evaluation of filtration characteristics

In this section the feasibility of the *SUR* as a parameter to evaluate filtration characteristics and predict the flux decline in dead-end ultrafiltration systems is investigated. To determine the relation between the foulants/particle concentration and *SUR* in wwtp-effluent, the *SUR* was measured for diluted samples of wwtp-effluent. Next, the possibility to measure differences in filterability of wwtp-effluent due to pre-treatment of the effluent by in-line coagulation and multi-media filtration was examined. Finally, this section summarises *SUR* values found for effluent from eight wwtp's in the Netherlands.

4.5.1 Foulants concentration

Material and methods

The relationship between foulants concentration and the *SUR* value was investigated in five experiments. The *SUR* was measured using the lab-scale device with one pressure vessel (figure 4.5) at a TMP of 0.5 bar. The *SUR* of wwtp-effluent (Berkel, T = 20°C) was measured, as well as the *SUR* after diluting the wwtp-effluent (ratio wwtp-effluent : dilution water is 100:0, 50:50, and 25:75).

Table 4.7 Experiments to determine the effect of effluent dilution with demineralised water on the *SUR*

Experiment	Order[a]	No. of experiments
D-1	Demi, 100%, 50%, 25%	Two (over two days)
D-2	Demi, 100%, 50%, 25%	One
D-3	Demi, 100%, 50%, 25%	Two (over two days)
D-4	Demi, 100%, 50%, 25%	Two (over two days)
D-5	Demi, 25%, 50%, 100%	Four (over two days)

[a] Each experiment started with measurement of the *SUR* of demineralised water, followed by the (diluted) effluent samples; *100%* indicates undiluted effluent, *50%* indicates 50% effluent and 50% demi water, *25%* indicates 25% effluent and 75% demi water

In all experiments demineralised water was used for dilution. Experiments D-1 to D-3 were started by measurement of the *SUR* of demineralised water. Next, the undiluted effluent was measured, followed by 50%-effluent and 25%-effluent. In experiments D-4 and D-5 first the *SUR* of demineralised water was measured, followed by 25%-

effluent, 50%-effluent and finally undiluted effluent. The membrane was soaked in NaOCl (200 ppm) in between the *SUR* measurements. In table 4.7 an overview of the five experiments is presented.

Results

In table 4.8 the measured *SUR* values found in the dilution experiments are presented together with the relative value of the *SUR* compared to the *SUR* value for undiluted effluent (100% effluent). In figure 4.12 the relative values of the *SUR* are graphically presented as a function of the percentage of effluent for all dilution experiments. The *SUR* of demineralised water was between 0 and $0.4 \cdot 10^{12}$ m^{-2}. In figure 4.11 the *SUR* of a 0%-effluent (100%-demi) is defined as 0% of the *SUR* of a 100%-effluent sample.

Table 4.8 ***SUR*** **measured as function of foulants concentration (effluent diluted with tap water (D-1, D-2) and demineralised water (D-3, D-4, D-5)**

	Experiment D-1		Experiment D-2		Experiment D-3		Experiment D-4		Experiment D-5	
Foulants (%)	*SUR* (10^{12} m^{-2})	(%)	*SUR* (10^{12} m^{-2})	(%)	*SUR* (10^{12} m^{-2})	(%)	*SUR* (10^{12} m^{-2})	(%)	*SUR* (10^{12} m^{-2})	(%)
100	10.1 ± 0.9	100	8.3	100	9.4 ± 0.1	100	15.3 ± 0.7	100	13.9 ± 0.4	100
50	4.5 ± 0.2	45	3.7	45	3.8 ± 0.3	40	6.2 ± 0.2	41	5.4 ± 0.7	39
25	1.5 ± 0.1	14	1.2	15	1.6 ± 0.1	17	2.5 ± 0.2	16	2.3 ± 0.1	17

The *SUR* is defined as the product of the average specific cake resistance and solids concentration in the feedwater. A decrease in solids concentration should therefore result in a proportional decrease of the *SUR*. The results presented in table 4.8 and figure 4.12 showed a non-linear decrease in *SUR*. In all experiments a 50% dilution showed a more than 50% decrease of the *SUR* (55-61% decrease). Another 50% effluent concentration decrease showed again a decrease of more than 50% (percentage not presented in table 4.8; decrease of 57-68%). The *SUR* value of 100%-effluent was different in all experiments and reflects the variations in quality of wwtp-effluent (Berkel) in a period of a month.

An almost linear relation between *SUR* decrease and effluent dilution has been found in all experiments. In the dilution experiments the flux of 25%-diluted

effluent was 0-20% higher than for the undiluted effluent. This might have caused a change in average specific cake resistance, which could explain the small deviation of the measured data compared to a linear relationship. Another explanation for this might be that the dilution caused a different cake layer build-up.

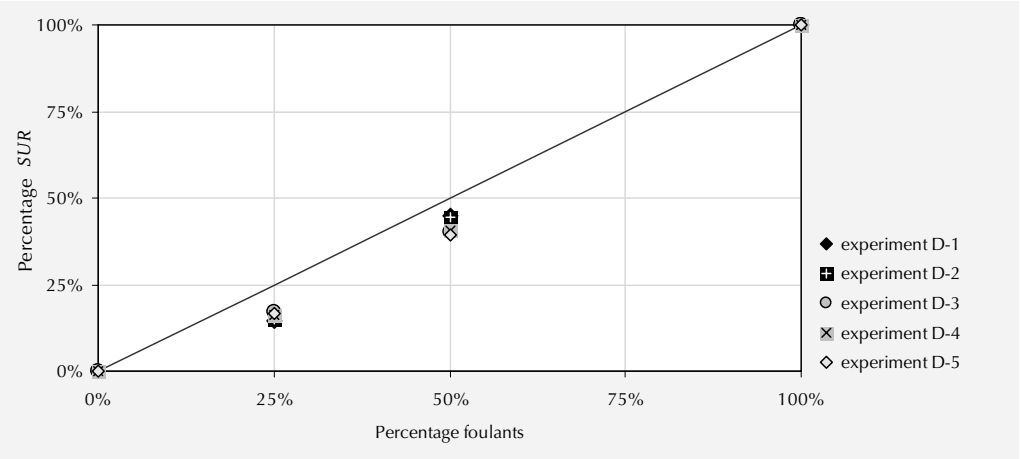

Figure 4.12 Percentage of *SUR* as a function of dilution for five experiments using wwtp-effluent (Berkel) as feedwater

4.5.2 Evaluation of feedwater pre-treatment

Material and methods

The possibilities for application of the *SUR* in evaluation of the effect of wwtp-effluent pre-treatment on the filtration characteristics are described in this section. The effect of pre-treatment by dual-media filtration and coagulation with ferric chloride on the value of the *SUR* was investigated in the following experiment. Firstly, the *SUR* of raw wwtp-effluent (Berkel, T = 20°C) was measured. Next, ferric chloride was added and the *SUR* was measured. The last measurement was performed using effluent that was pre-treated with dual-media filtration.

The first experiment (Pre-1) was performed for four consecutive days; all measurements were done in duplicate. The second experiment (Pre-2) was performed on two consecutive days. The (pre-treated) effluent was analysed for turbidity (Hach, model 2100N). The *SUR* was measured of:

1. Untreated effluent;
2. Effluent pre-treated with 1.0 mg Fe^{3+}/l. Ferric chloride (1.0 mg Fe^{3+}/l) was added to effluent (V = 4 dm^3) and stirred for 10 minutes. The *SUR* was determined after stirring the sample;
3. Filtrate of dual-media filtration of wwtp-effluent. The dual-media filter was a double media layer (V = 13 dm^3; A = 0.64 dm^2 and H = 20 dm) made of 0.5 m sand (d = 1.0 – 1.6 mm) and 1.5 m anthracite (d = 1.4 – 2.5 mm). The filtration rate was controlled with a valve in the filtrate line on the bottom of the filter bed and set at 5 m/h (32 l/h). After filtering 1.5 times the volume of the filter bed (~ 20 dm^3 of effluent) filtrate was sampled for the *SUR* measurement.

Results

In table 4.9 and figure 4.13 the results of the experiments for evaluation of pre-treatment on the *SUR* are presented, which show a decrease of the *SUR* for both dual-media filtration and coagulation with 1 mg/l of ferric chloride. This showed that differences in filterability could be measured by the *SUR*.

Table 4.9 ***SUR*** **measured of raw and pre-treated wwtp-effluent (Berkel, 20°C)**

Experiment	Pre-1				Pre-2			
Feedwater	*SUR*	%*SUR*[a]	Turbidity	%Turbidity	*SUR*	%*SUR*[a]	Turbidity	%Turbidity
	(10^{12} m^{-2})		(NTU)		(10^{12} m^{-2})		(NTU)	
Effluent	12.4 ± 1.1	100%	3.4 ± 0.6	100%	17.0 ± 1.4	100%	3.4 ± 0.7	100%
Effluent + 1 mg Fe^{3+}/l	9.1 ± 0.5	73%	3.3	97%	12.9 ± 1.4	76%	2.9 ± 0.6	85%
Filtrate	10.6 ± 0.6	85%	1.1 ± 0.2	32%	13.9 ± 0.6	82%	1.4 ± 0.2	41%

[a] Relative to the value of effluent

Coagulation resulted in a turbidity decrease of 5% to 15%, whereas the dual-media filter removed 60% to 70% of the turbidity. But the largest decrease in the *SUR* was found for coagulation, about 25%, where the dual-media filter resulted in a *SUR* decrease of 14% to 18%. This showed that a decrease in turbidity of the effluent did not relate to a similar increase in filterability of the effluent (*i.e.* decrease of *SUR*). Coagulation of the effluent compared to dual-media filtration proved to be a better

technique for reduction of components that determine the filterability of the wwtp-effluent.

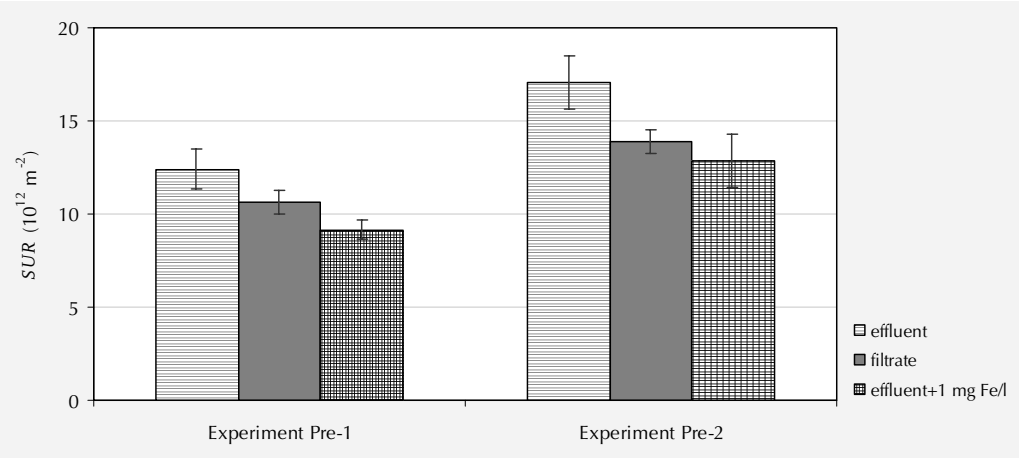

Figure 4.13 Diagram of *SUR* measured for raw and pre-treated wwtp-effluent (Berkel, 20°C)

4.5.3 *SUR* determination at various wwtp's in the Netherlands

Material and methods

During the research period described in this dissertation (1998-2003) the *SUR* has been measured at eight different wwtp's in the Netherlands. In 1999 the *SUR* was measured using the lab-scale unit equiped with a pump (figure 4.4) for wwtp Ede and Kaffeberg. In 2000 the second lab-scale unit, with one pressurised buffer tank (figure 4.5), was used and *SUR* was measured at wwtp Tilburg-Noord and Vlaardingen. All measurements in 2002 were performed with the lab-scale unit equipped with two pressurised vessels (figure 4.6) for the remaining wwtp's of Berkel, Emmtec, Hoek van Holland and Zaandam.

The *SUR* has to be measured at a feedwater temperature of 20°C. At some wwtp's the effluent or filtrate was coagulated (1.0 mg/l ferric chloride or aluminium chloride (poly aluminium chloride, PACl)). In some cases, the *SUR* was also measured for wwtp-effluent pre-treated with dual-media filtration.

Results

In table 4.10 the results of the *SUR* measurements are given. These results show large variations in *SUR* for effluent at the different wwtp's. The filterability of the effluent was specific for each wwtp. This shows the possible advantages of the use of *SUR* for actual investigation of the filtration characteristics in a pilot-plant or full-scale installation.

Table 4.10 ***SUR*** **values found for (pre-treated) effluent from eight different wwtp's in the Netherlands; for effluent the measured range is shown between brackets**

WWTP	Berkel[c]	Ede[a]	Emmtec[c]	HvHolland[c]	Kaffeberg[a]	Tilburg-N[b]	Vlaardingen[b]	Zaandam[c]
Year	2002	1999	2002	2003	1999	2000	2000	2000
Feedwater	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})	*SUR* (10^{12} m^{-2})
Effluent	11	18	9	21	29	5	15	20
(range)	(6-40)	(11-23)	(4-13)	(11-44)	(24-34)	(3-6)	(14-16)	(18-22)
Effluent + coag.	9	10	3			5		
Filtrate	10	4	7	16	28	4		
Filtrate + coag.			5	11				

[a] Measured using device with recirculation pump (figure 4.4); [b] Measured using device with one pressurised buffer tank (figure 4.5);
[c] Measured using device with two parallel pressurised buffer tanks (figure 4.6)

4.6 Discussion

4.6.1 Parameter for dead-end ultrafiltration

The *SUR* is the product of the specific cake resistance (α_{av} in m/kg) and the solids concentration (c_v in kg/m³). WWTP-effluent is a complex mixture of constituents that may vary in both physical and chemical properties. It is therefore very difficult to find the proper values for solids concentration and specific cake resistance. But the product of specific cake resistance and solids concentration can be found by applying the cake

filtration theory on filtration data that are measured in constant TMP experiments (Hermia, 1982). The *SUR* is then calculated from the slope of the t/V versus V curve[1].

To obtain reliable and reproducible data, a filtration device for constant pressure filtration was introduced. Firstly, a device with a pump that recirculates the effluent was tested, but changes in effluent composition made it necessary to transform this device into a lab-scale unit with a pressurised buffer tank. The turbulence in the recirculation pump changed the properties of the feedwater with rising temperatures (5°C for every 30 minutes of filtration). With the pressurised buffer tank (V = 10 dm^3) the composition of the feedwater sample could be kept constant throughout the experiment without increasing the temperature. At the start of the filtration cycle the TMP of the lab-scale unit with a single vessel was manually set to a constant TMP of 0.5 bar. This procedure took about 1 to 2 minutes and caused disturbances in the filtration data. By adding another buffer tank in parallel (filled with demineralised water), the TMP could be adjusted to a constant value within seconds. In this case no disturbance of the filtration data was found. The lab-scale unit with two buffer tanks in parallel worked properly and provided reliable filtration data for measurement of the *SUR*.

4.6.2 Process conditions

Total filtration time

The experiments that were performed for determination of the time needed for measurement of a reliable and reproducible *SUR* (§4.3.3) resulted in a total measurement time of 30 minutes. The *SUR* was calculated from the data that were found between 10 and 30 minutes of filtration. It was shown that the *SUR* increased slightly in time, about 1-3% within 30 minutes of filtration. This finding might be related to small particles that are retained by the fouling layer and penetrate the fouling layer. This will lead to a decrease in porosity of the fouling layer, which might increase the resistance of the fouling layer (*i.e.* an increase in *SUR*).

[1] The *SUR* is calculated without taking into account the membrane resistance; this does not influence the findings, which is shown in Appendix 4-C

Trans Membrane Pressure

The *SUR* was measured as a function of the Trans Membrane Pressure (TMP). An increase in the TMP resulted in an increased *SUR*, due to compression of the fouling layer. The compression coefficient was in the range of 0.61 to 0.75. The values for the compression coefficient reflect the compression of a whole range of constituents that are found in wwtp-effluent. The specific cake resistance and its compressibility depend both on the character of the foulants. This is shown in for instance Nakanishi *et al.* (1985) where it is concluded that the specific cake resistance depends on the species of microorganisms, their shape and size (Matsumoto *et al.*, 1999). Foulants in wwtp-effluent are expected to have a biological background and behave similar to, or are similar to (fractions of) microorganims.

Compression of the fouling layer should be taken into account in the design of the process conditions of full-scale ultrafiltration plants. The compressibility of a fouling layer determines the limits of the ultrafiltration process. The impact of compression is explained in the following figures. In figure 4.14 (a) the filtration cruve is drawn for a flux of 50 and 100 l/m².h. The resistance increase in this graph is a function of the amount of filtered feedwater (*i.e.* amount of foulants). The resistance over the membrane is doubled after 15 minutes of filtration at a flux of 50 l/m².h without compression of the fouling layer. At a flux of 100 l/m².h, the similar amount of foulants is filtered over the membrane within 7.5 minutes, resulting in the same resistance of the fouling layer. But a similar resistance over the membrane relates at a double flux to doubling of the TMP, from 0.2 bar for 50 l/m².h and 0.4 bar for 100 l/m².h (without compression).

In figure 4.14 (b) the effect of compression on the TMP is drawn. Compression is a function of the applied TMP and increases at increasing TMP. The subsequent increase in resistance over the membrane and fouling layer amplifies the TMP increase. Compression of the fouling layer is especially something to deal with at a higher TMP than at a low TMP. From these findings it is concluded that fouling layer compression is a very important aspect in determination of the process conditions of dead-end ultrafiltration systems.

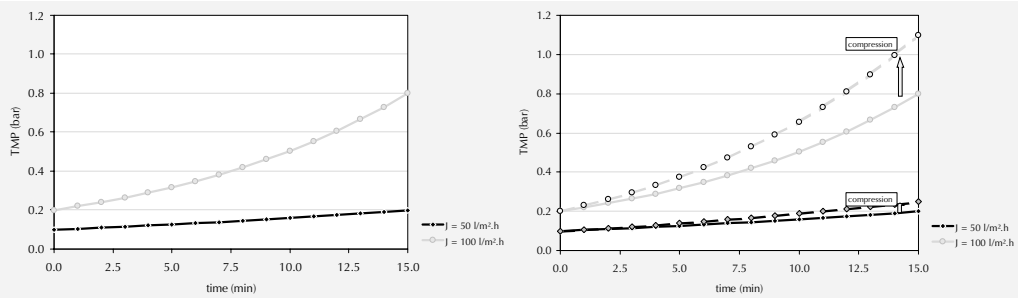

Figure 4.14 TMP as a function of the flux and its relationship with compression of the fouling layer

Temperature

The temperature of the (pre-treated) effluent influenced the *SUR*. Unexpectedly, no major flux increase was found at increasing temperature in the experiments described in §4.4.3. This effect of the temperature has also been found in other effluent ultrafiltration experiments in our laboratory (te Poele and van der Graaf, 2002; Meezen, 2002). In literature no similar results were found. The average specific cake resistance is the overall cake resistance of all foulants in the effluent. It is therefore expected that the temperature effect can be related to changes in the characteristics of most foulants. Maybe foulants stretch due to temperature increases (Campbell *et al.*, 1993).

The *SUR* values that were found in experiments T-1 to T-3 were rather high for effluent of wwtp Berkel ($24.4 \cdot 10^{12} \pm 5.3 \cdot 10^{12}$ m^{-2}, measured at $T = 20$°C), compared to the average *SUR* of Berkel effluent ($11.1 \cdot 10^{12}$ m^{-2}). This might also affect the filtration characteristics found in experiments T-1 to T-3. This is indicated by the research of Meezen (2002), in which a smaller *SUR* values were measured. This resulted in a lower increase of the *SUR* as a function of temperature.

Measurement of the *SUR*

From the TMP as well as the temperature experiments it became clear that the interpretation of *SUR* data is only possible if TMP and temperature are well defined. The *SUR* is introduced as a parameter for evaluation of filtration characteristics in ultrafiltration of effluent. In pilot-scale research (Chapter 2) as well as in full-scale

applications the TMP is found within the range of 0.2 to 0.8 bar. Measurement of the *SUR* is therefore proposed at a TMP of 0.5 bar. The temperature of the sample feedwater is measured at ambient temperature of 20°C which is proposed as the reference temperature.

Membrane type

Results of experiments M-1 to M-4 using four different membranes with the same chemical composition but a different pore size and diameter of the fibres, showed that the pore size of the membranes (Molecular Weight Cut-Off, MWCO) did not influence the *SUR*. Components with dimensions within the MWCO-range of 80-150 kDa have only minor influence on the filtration characteristics. These results indicate that the components retained on the membrane surface predominantly determine the filtration characteristics. It has been shown in §4.4.4 that differences found in the *SUR* for 0.8 and 1.5 mm capillaries were mainly related to inaccurate data of the internal diameter of the capillaries. Still the corrected values of the membranes with a larger inner diameter (1.5 mm) showed a 12% lower *SUR* than those with the smaller inner diameter.

In general, differences in membrane composition are related to differences in filtration characteristics (Boerlage *et al.*, 2003; Doyen *et al.*, 1998; Galjaard *et al.*, 1998). The *SUR* provides information of the interaction between the applied membrane and the feedwater. The information on filtration characteristics measured with the *SUR* might be related to experiments on a larger scale, using the same membrane type and wwtp-effluent. In that case also the measuring conditions have to be similar to the conditions in the pilot-plant or full-scale installation. The interaction between membrane and feedwater determines the filtration and fouling characteristics. Therefore the *SUR* is defined and measured at a TMP of 0.5 bar at a feedwater temperature of 20°C. The temperature of wwtp-effluent showed variations between 6-25°C, but evaluation of the influence of for instance pre-treatment should be performed under specified conditions. For on-line measurement of *SUR* it might be possible to incorporate the actual temperature of the feedwater.

SUR for characterisation of filtration

To investigate the relationship between the foulants concentration and the *SUR*, experiments were performed on the relation between *SUR* values and dilution of effluent samples (wwtp Berkel). The results showed an almost linear relationship between foulants concentration and *SUR*. By dilution of the effluent, only the solids concentration changes, which was found in these experiments. Boerlage *et al.* (2003) found similar results for the relationship between concentration and MFI-UF for dilution experiments on tap water and canal water.

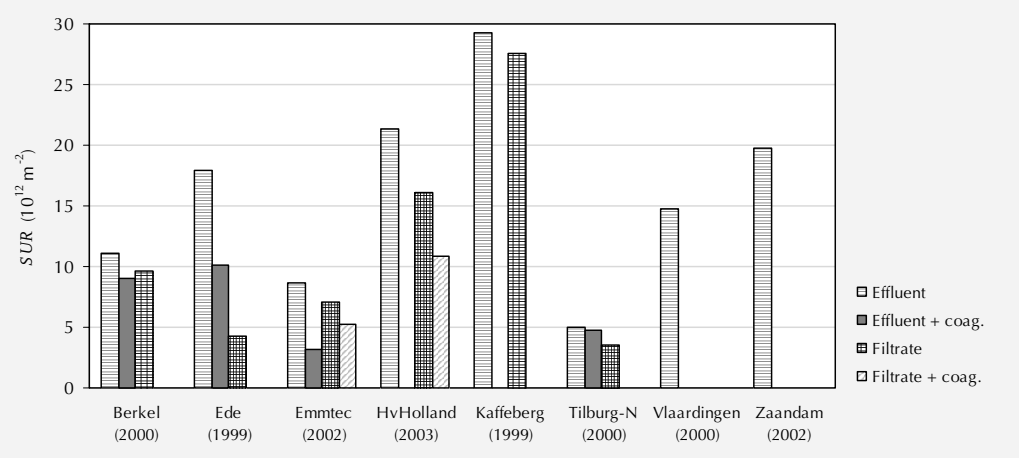

Figure 4.15 Average *SUR* values found for raw and pre-treated effluent from eight different wwtp's in the Netherlands

Pre-treatment of wwtp-effluent by multi-media filtration and in-line coagulation is sometimes found to increase the filterability of the effluent (Abdessemed *et al.*, 2000; Bourgeous *et al.*, 2001; Ghosh *et al.*, 1994; van der Graaf *et al.*, 1999). Experiments Pre-1 and Pre-2, as well as the results of experiments found at various wwtp's (data from table 4.10), resulted in changes in the *SUR* after pre-treatment of the effluent. In figure 4.15 the data of table 4.10 are graphically presented, showing a decrease of the *SUR* after in-line coagulation of 4% (wwtp Tilburg-Noord) to 63% (wwtp Emmtec). The decrease after multi-media filtration was for some effluents less than after in-line coagulation (wwtp's Berkel, Emmtec) and for some more than after coagulation (wwtp's Ede, Hoek van Holland, Tilburg). These results show that the choice of a

proper pre-treatment technique for improved filterability can be evaluated with the *SUR*.

Finally, it has to be emphasized that the performance of ultrafiltration in pilot- or full-scale installations depends only partly on the filterability. Although a high initial filterability (low *SUR*) indicates higher fluxes, also the removal efficiency of the retained material at the membrane surface is important. When the retained material is only partly removed during a cleaning interval, the overall flux will decrease. However, comparing the *SUR* of effluent with the results from the pilot-scale tests (Chapter 2), it is found that a low *SUR* (high filterability) is a requirement for good ultrafiltration performance.

4.7 Conclusion

The Specific Ultrafiltration Resistance (*SUR*) is proposed as a new parameter for measuring the filtration characteristics of effluent in dead-end ultrafiltration. The *SUR* is calculated from the filtration data measured with a lab-scale device. The *SUR* is calculated from the ratio of filtration time and filtrate volume (t/V) as a function of the total filtrate volume and is the product of the specific cake resistance and the solids concentration. The process conditions during measurement affect the *SUR* of the effluent, therefore the *SUR* is defined at a constant TMP of 0.5 bar and an effluent temperature of 20°C. The *SUR* has an accuracy of more than 95% and is measured within 30 minutes of filtration.

The experiments on the *SUR* revealed also additional information about the filtration characteristics of wwtp-effluent. The MWCO of a PES/PVP membrane did not influence the *SUR*. This indicates that effluent constituents larger than the pore sizes determine filtration characteristics. Experiments with varying Trans Membrane Pressure showed that the occurring fouling layer is highly compressible (s = 0.6-0.75). This implies that the fluxes should not increase too much, as the accompanying TMP increases more than linear. Pre-treatment induced an increase in filterability of 20% to 30%. Both pre-filtration and coagulation influenced mainly particles larger that 5-10 μm. This relatively small increase in filterability by pre-treatment indicates therefore that the filterability is only partly determined by particles larger than 5-10 μm.

The *SUR* was measured for effluent of various wwtp's in the Netherlands and showed great variations for the different wwtp's. The *SUR* was found to range from $5 \cdot 10^{12}$ to $30 \cdot 10^{12}$ m^{-2}. Although the effect of pre-treatment on the filterability (measured as *SUR*) was relatively small (see also Chapter 3), the change in filterability can be measured accurately with the *SUR*. In these tests coagulation as well as multi-media filtration showed a decrease of the *SUR* (of approximately 20% to 30%), greatly depending on the local conditions.

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5 Fractionation of wwtp-effluent

5.1 Introduction

As shown in Chapter 1 essentially five fouling mechanisms can be identified, which are summarised in two phenomena, *i.e.* as deposition inside the membrane pores and/or deposition on the membrane surface. These fouling mechanisms are influenced by the process conditions, as well as by the properties of the feedwater constituents. Ultrafiltration membranes reject material largely by mechanical sieving (Wiesner and Buckley, 1996; Starov *et al.*, 2002). However, electrostatic interactions, dispersion forces, and hydrophobic bonding may significantly affect the rejection of materials with dimension(s) similar to the size of the pores in UF membranes (Wiesner and Buckley, 1996; Fan *et al.*, 2001).

The process of mechanical sieving is influenced by the particle diameter and the diameter of the membrane pores (Meireles *et al.*, 1991). In this chapter the influence of particle fractions on filtration characteristics in relation to the pore diameter is investigated for dead-end ultrafiltration of wwtp-effluent.

The characteristics of filtration during dead-end ultrafiltration are influenced by the distribution of particle sizes of the feedwater. Bourgeous *et al.* (2001) performed research on dead-end ultrafiltration of primary and secondary effluent and hypothesised that particles in the same order of magnitude as the membrane pores are responsible for the membrane fouling problems. Tchobanoglous *et al.* (1998) concluded from research on dead-end ultrafiltration of secondary effluent that large particles (> 20 μm) did not significantly impact the performance of the ultrafiltration membrane system. But smaller particles (< 1 μm) seemed to have a considerable impact on membrane performance.

Vial *et al.* (1992) suggested in research on cross-flow ultrafiltration (plate and frame module) of tertiary effluent that filtration curves could be understood by an initial flow reduction by formation of a fouling layer and a subsequent slower flow reduction by cake modifications. Foulants are supposed to have a diameter larger than the membrane pores. Other researchers suggest the opposite: components in the feedwater, especially organic matter with a molecular weight smaller than the membrane pore size, are found to account for a significant irreversible membrane fouling (Crozes *et al.*, 1993). Jarusutthirak *et al.* (2002) defined for effluent the organic matter fraction Effluent Organic Matter (EfOM) and found especially that the colloidal fraction with large molecular weight compounds with a hydrophilic character foul the ultrafiltration membrane.

Doyen *et al.* (1998) found in comparing different ultrafiltration membranes that surface pores led to extensive membrane fouling and that slit shaped pores had a large 'Dirt Holding Capacity' (DHC). A large DHC induces that the membranes act as depth filters and not as surface filters.

In Chapter 3 (Filterability and Reversibility) of this dissertation it was indirectly concluded from pilot-plant experiments that particles with a small diameter (< 5 μm) largely influence filtration characteristics. From experiments using different MWCO membranes (experiments M-1 and M-2 described in Chapter 4 of this dissertation) it has been hypothesised that the similarity in filtration characteristics for membranes with different MWCO's was due to foulants larger than the MWCO's. These effluent constituents should have a diameter larger than the pore size and these constituents especially determine the occurring filtration phenomena.

The objective of the research described in this chapter[1] is to identify the relationship between the particle size of the effluent constituents and their filterability in dead-end ultrafiltration of wwtp-effluent.

Generally, information about particle size ranges has been used for the improvement of process steps in wastewater treatment (Levine *et al.*, 1985; Levine *et al.*, 1991; van Nieuwenhuijzen, 2002). Particle size distributions in wastewater also provide information about wwtp-effluent (and wwtp-influent) characteristics and its treatment

[1] Part of this work has been presented in Roorda *et al.* (2003)

possibilities. The effectiveness of treatment processes is strongly influenced by the size range distributions of constituents of wastewater. Levine *et al.* (1985) reviewed various treatment processes for their effectiveness in the removal of particles. Sedimentation is effective on material larger than 50 μm. Multi-media filtration is capable of removing particles larger than 1-5 μm. Coagulation can be used to aggregate wastewater constituents in the size range from less than 0.1 μm to about 10 μm. Biological treatment is capable to effectively treat wastewater constituents in all size ranges; larger molecules can be hydrolysed into smaller molecules that can be transported across the cell membrane and metabolised (< 0.1 – 1.0 nm).

Particles remaining in the wastewater after aerobic biological treatment are in the macro-colloidal range of 1-10 μm (Levine *et al.*, 1991). Abdessemed *et al.* (2002) showed that in effluent 58% of the organic load (COD) was found in a range larger than 0.1 μm, 13% of the COD was found within a range between 10 kDa (~ 5 nm[1]) and 0.1 μm and 29% was found in a fraction smaller than 10 kDa. Coagulation with 20 mg Fe^{3+}/l changed the composition to 71% of the COD in the largest fraction (> 0.1 μm), 5% in the smaller fraction (10 kDa – 0.1 μm) and only 24% in the smallest fraction.

In the research performed at wwtp Ede (see Chapter 2) the particle size distributions of effluent were measured within the range of 2-200 μm using a Met One PCX particle counter, based on laser light blocking. About 4% of the total volume of the particles (2-200 μm) was in the size range of 2-10 μm. Very low concentrations of coagulant (1 mg/l of Fe^{3+} and Al^{3+}) induced a shift towards smaller ranges, by shrinkage of the particles towards a decreased average diameter (van der Graaf *et al.*, 2001).

Particles in wastewater effluents are mostly colloidal in nature and negatively charged, thus repelling each other (Adin, 1999). In figure 5.1 some typical organic constituents for ranges of particles are presented for settled municipal wastewater. After biological treatment and settling, the remaining effluent constituents are probably related to (fragments of) activated sludge.

Van Nieuwenhuijzen (2002) defined the various fractions in a slightly different from Levine *et al.* (1985) in figure 5.1. In the current research the same fractions as used by van Nieuwenhuijzen were considered for lab-scale experiments on

[1] According to Granath and Kwist (1967), cited in Doyen (1997): $A = 0.33 \cdot MWCO^{0.46}$ in which A is the radius (in Å = 10^{-12} m) of Dextran molecules in a water solution related to the molecular weight (MWCO in Da)

filterability of fractions of (pre-treated) wwtp-effluent. The following fractions were considered: settleable suspended (> 63 μm), suspended (5 - 63 μm), supra colloidal (1.2 - 5 μm), colloidal (0.45 - 1.2 μm), semi-dissolved (< 0.45 μm), and dissolved (< 0.1 μm).

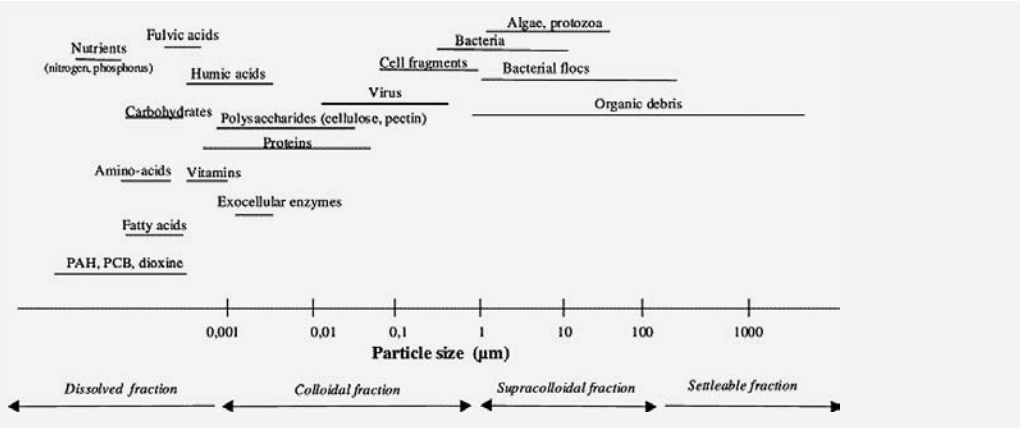

Figure 5.1 Typical organic constituents in settled municipal wastewater (Levine *et al.*, 1985)

5.2 Particle size distribution in wwtp-effluent

5.2.1 Material and methods

The particle size distributions (PSD's) of samples of effluent from wwtp Berkel were determined using a Met One PCX particle counter in the 2-100 μm range, based on laser light blocking. The data were produced in a particle number frequency function with intervals of 0.5 μm. Smaller particles were measured in a second particle counter, the HIAC Microcount 05. The HIAC measuring device was in the range of 0.5-2 μm based on laser light scattering with intervals of 0.1-0.2 μm and in the range of 2-90 μm based on laser light blocking.

Measurements with the Met One were also done on (pre-treated) effluent of wwtp Ede (ranges: 2-100 μm, with 0.5 μm intervals) and Kaffeberg (ranges: 1-3 μm; 3-5 μm; 5-10 μm; 10-15 μm; 15-30 μm; 30-75 μm). The HIAC Microcount was used for samples of effluent from wwtp Berkel, Hoek van Holland and Vlaardingen.

Measurement of particle size distributions with a laserlight blocking device underestimates the particle size of microorganisms for up to 50% (O'Shaugnessy *et al.*, 1997). Therefore, in the experiments described in this section the measured particle size was increased with 20% for further calculations. Furthermore, the particles were assumed to be spherical for calculations (O'Shaugnessy *et al.*, 1997). In order to simplify the calculations, the particles were assumed to be homogeneous in distribution. Besides, substances measured as particles can also change in shape and size by changing flow streams or changing pressures (O'Shaugnessy *et al.*, 1997).

Particle size measurements provided a histogram for numbers of particles versus particle size. The distribution function was transformed into a particle volume distribution function, assuming for the calculations that the particles had an ideal spherical shape. This function covered the range from 2-100 μm for effluent of wwtp Ede (the range 0.5-10 μm for wwtp Berkel, Hoek van Holland and Vlaardingen are not presented). With the particle volume distribution function several other calculations were performed (van der Graaf *et al.*, 2001):

- Cumulative particle volume distribution (in m^3/m^3) versus particle size;
- Relative cumulative particle volume distribution (in %) versus particle size.

By assuming an equal distribution of particles over the membrane surface, the maximum theoretical cake layer thickness (in μm) can be calculated for the PSD of a sample, as the total particle volume divided by the membrane surface area. The total particle volume was used to calculate the maximum theoretical cake layer thickness on the membrane surface, assuming a filtration period of 30 minutes at a flux of 100 $l/m^2.h$

At a similar flux and filtration period the maximum percentage of the surface area filled with particles was calculated assuming that no superimposition of particles occurs and one particle deposited next to another particle. The cross-section of a spherical particle was calculated and the sum of all cross-sections was divided by the total membrane surface area, resulting in the maximum percentage of the surface area that is filled with particles.

5.2.2 Results

In figure 5.2 the particle distribution curves found for (pre-treated) effluent from wwtp Ede are presented. Figure 5.2 presents the results for coagulation with ferric chloride, the results for coagulation with Alum chloride can be found in Appendix 5-A. These curves show that the total number of particles decreased by pre-treatment of the wwtp-effluent. The largest decrease was found for filtration of (not pre-treated) effluent. In the experiments using effluent from wwtp Ede, no differences were found between coagulation with ferric chloride (1 mg/l) and aluminium chloride (1 mg/l).

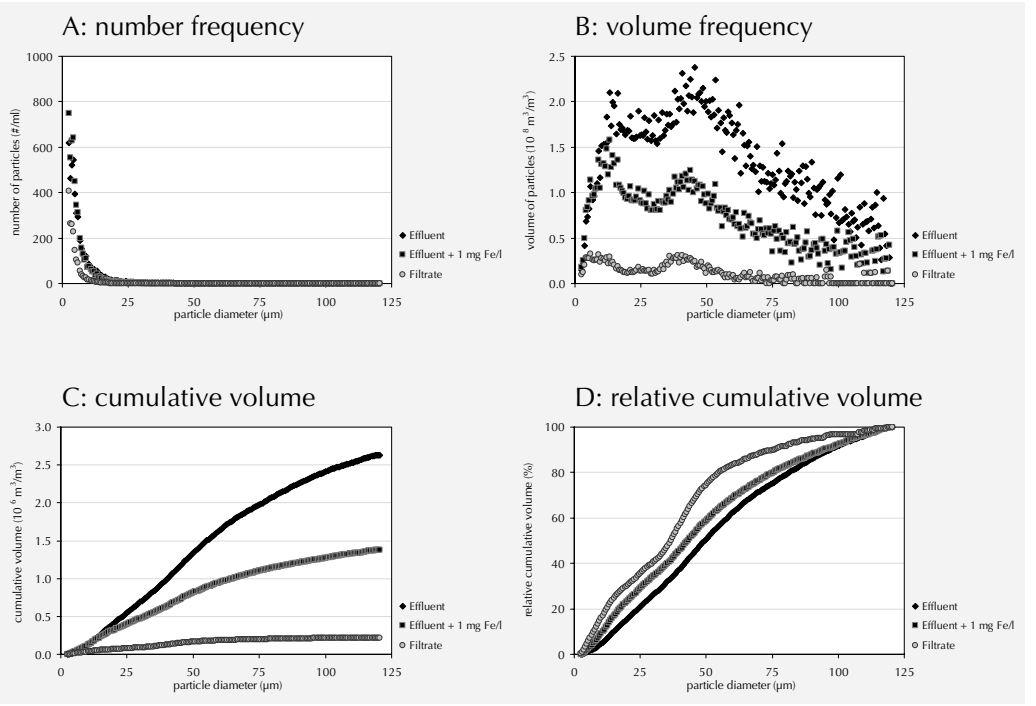

Figure 5.2 Particle distribution curves for effluent, effluent with 1 mg Fe^{3+}/l and filtrate from wwtp Ede; A: number frequency, B: volume frequency, C: cumulative volume, D: relative cumulative volume

In table 5.1 the maximum theoretical cake layer thickness and the percentage of fouled membrane area within 30 minutes of filtration at a flux of 100 l/m².h are presented. The thickness of the cake layer was 0.4 μm or less, which is very thin compared to the

counted particle sizes that were larger than 2 μm. This suggests that particles with a size larger than 2 μm can hardly affect the filtration characteristics. If a (realistic) membrane porosity of 10% is assumed and all particles are centred near the pores, the total membrane area that might be filled with particles would be about 20% of the total area. If so, the values in table 5.1 will increase with a factor 5, still showing a maximum cake layer thickness of 2 μm near the pores. If this is the case, particles with a diameter larger than 2 μm might only form a monolayer near the membrane pores.

Table 5.1 Maximum cake layer thickness and percentage of fouled membrane surface, as calculated from particle size distributions (2-100 μm) for (pre-treated) effluent of wwtp Ede

Coagulant	**1 mg Al^{3+}/l**		**1 mg Fe^{3+}/l**	
Feedwater	Cake layer thickness (μm)	% fouled membrane surface (%)	Cake layer thickness (μm)	% fouled membrane surface (%)
Effluent	0.4	1.7	0.4	1.9
Effluent + coag.	0.2	1.1	0.2	1.3
Filtrate	0.04	0.2	0.03	0.3

This was underlined by the calculations on the percentage of fouled membrane, showing a maximum 2 % of fouled membrane area, which in the case described above must be found in the 20% of the area near the pores. If so, this 20% area was only filled for 10% with particles (maximum). This showed that a monolayer could only be found in 10% of the area near the membrane.

But during dead-end ultrafiltration an increase in resistance was found that could hardly be related to 10% fouled area near the membranes (90% of the area around the pores is not fouled). This indicated a minor influence of particles with a size range >2 μm on the filtration characteristics in dead-end ultrafiltration of wwtp-effluent. For pre-treated wwtp-effluent the calculations showed even much lower cake layer thickness and fouled membrane area, which supported this indication.

In table 5.2 the results are shown for effluent of Berkel, Hoek van Holland and Vlaardingen as calculated from particle size distributions in the range of 0.5-10 μm.

The values for maximum cake layer thickness and percentage of fouled membrane area showed similar results as found for the effluent of wwtp Ede (2-100 μm). Also these results indicate a minor impact of particles within the size range of 0.5-10 μm on filtration characteristics.

Table 5.2 Maximum cake layer thickness and percentage of membrane surface fouled, as calculated from particle size distributions (0.5-10 μm) for effluent of various wwtp's

Feedwater	Cake layer thickness (μm)	% fouled membrane surface (%)
wwtp-effluent Berkel	0.13	1.9
wwtp-effluent HoekvHolland	0.03	0.6
wwtp-effluent Vlaardingen	0.03	0.6

5.3 Fractionation of wwtp-effluent

5.3.1 Material and methods

Fractionation procedure

Fractionation of wwtp-effluent was performed according to the procedure described by van Nieuwenhuijzen (2002). Feedwater samples are fractionated over clean, pre-flushed sieves (stainless steel) and filters with five different pore sizes. The feedwater was divided in the size fractions: < 200 μm, < 5.0 μm, < 1.2 μm, < 0.45 μm, < 0.2 μm and < 0.1 μm. The properties of filters used for fractionation are summarised in table 5.3.

Firstly, the pre-sieved effluent (200 μm) was filtered under a vacuum over 5.0 μm filter. After this, its 5.0 μm filtrate was filtered over the 1.2 μm filter, followed by the 0.45 and 0.2 μm. The last separation step using a 0.1 μm filter was filtered under constant pressure of 2.0 bar.

Table 5.3 Applied Sartorius filters used in fractionation experiments

Nominal pore diameter	Type	Type number
5.0 μm	Cellulose Nitrate	0200 11342 9905733
1.2 μm	Cellulose Nitrate	0888 11303 0913/7
0.45 μm	Cellulose Acetate[a]	0692 11106 9200263
0.2 μm	Cellulose Nitrate	0400 11090 9256223
0.1 μm	Cellulose Nitrate	0400 11358 9903623

[a] Cellulose Nitrate was not available

SUR measurement of fractions

For each fraction the filtration characteristics were evaluated by measuring the Specific Ultrafiltration Resistance (*SUR*). A minimum feedwater volume of 1.0 dm^3 was needed for one measurement of the *SUR* with the procedure described in Chapter 4. The lab-scale unit with two pressurised buffer tanks in parallel (figure 4.6 in section 4.3.2 of this dissertation) was used in the fractionation experiments.

The ultrafiltration membranes used for the *SUR* measurement in these experiments were capillary membranes with an internal diameter of 0.8 mm. The membranes were made of PES/PVP (type: UFC M5 ID 0.8 mm, X-flow) with a MWCO estimated at 50-80 kDa. This accounted for pore diameters of about 10-15 nm (Granath and Kwist, 1967). According to the membrane manufacturer the pore diameter was about 5-30 nm.

Before starting the *SUR* measurement, the ultrafiltration membrane was soaked for 30 minutes in NaOCl (200 ppm) and rinsed with demineralised water. First, the *SUR* of the smallest fraction was measured, followed by a forward flush and the filtration of demineralised water. This was done in order to check the membrane permeability. Subsequently, the following larger fraction was filtered and the *SUR* was determined. In figure 5.3 the procedure is shortly summarised:

Soak the membrane for 30 minutes with NaOCl-solution (200 ppm)

Forward flush with demineralised water (demi)

Measure the *SUR* of demi within 10 minutes

Measure the *SUR* of dissolved fraction (< 0.1 μm) within 20 minutes

Forward flush, Measure the *SUR* of demi within 10 minutes

Measure the *SUR* of fraction < 0.2 μm

Forward flush, Measure the *SUR* of demi within 10 minutes

Measure the *SUR* of fraction < 0.45 μm

Forward flush, Measure the *SUR* of demi within 10 minutes

Measure the *SUR* of fraction < 1.2 μm

Forward flush, Measure the *SUR* of demi within 10 minutes

Measure the *SUR* of fraction < 5.0 μm

Forward flush, Measure the *SUR* of demi within 10 minutes

Measure the *SUR* of fraction < 200 μm

Forward flush

Soak membrane in NaOCl solution (200 ppm)

Figure 5.3 Procedure for determination of the *SUR* for fractions of (pre-treated) wwtp-effluent

Feedwater for fractionation experiments

All experiments on fractionation were performed at the laboratory of the department of Sanitary Engineering (faculty Civil Engineering and Geosciences of the Delft University of Technology). Fractionation experiments were performed using effluent from wwtp Berkel, Hoek van Holland and Zaandam. The effluent was fractionated and measured on the sampling day, or stored in a refrigerator for a maximum of two days (T = 5°C). Effluent of wwtp Berkel was fractionated with and without pre-treatment. The pre-treatment procedures for Berkel effluent were:

1. Coagulation of effluent with 1.0 mg Al^{3+}/l. Poly aluminium chloride (PACl, 51 g Al^{3+}/l) was added to the effluent (V = 4 dm^3) and stirred for 10 minutes. After stirring and without settling the sample was fractionated and the *SUR* was determined;

2. Filtration of effluent over a one-layer sandfilter. The sandfilter (V = 13 dm³; A = 0.64 dm² and H = 10 dm) consisted of one layer of sand (d = 1.0 – 1.6 mm). The filtration rate was regulatedby a valve in the filtrate line on the bottom of the filter bed and was set at 10 m/h (64 l/h). Filtrate was sampled after 1.5 times the volume of the filter bed had passed the filter bed.

5.3.2 Results

In table 5.4 the results of the fractionation experiments are presented, which shows the absolute *SUR* values for the various fractions of (pre-treated) wwtp-effluent. In Appendix 5-B the relative *SUR* values are presented, based on the *SUR* of the unfractionated effluent (fraction < 200 μm). *SUR* values of demineralised water in all experiments were lower than $0.5 \cdot 10^{12}$ m^{-2}.

Table 5.4 ***SUR*** **measured for fractions of raw and pre-treated wwtp-effluent**

WWTP	**Berkel**			**HvHolland**	**Zaandam**
Feedwater fraction	Effluent *SUR* (10^{12} m^{-2})	Effluent+[a] *SUR* (10^{12} m^{-2})	Filtrate[b] *SUR* (10^{12} m^{-2})	Effluent *SUR* (10^{12} m^{-2})	Effluent *SUR* (10^{12} m^{-2})
< 200 μm	11.1 ± 2.4	9.0 ± 1.7	9.6 ± 2.3	28.8 ± 2.4	19.8 ± 2.3
< 5.0 μm	11.4 ± 1.2	7.4	9.9 ± 1.1	*nd*[c]	19.1 ± 1.7
< 1.2 μm	8.9 ± 1.0	7.4 ± 1.3	8.6 ± 0.8	*nd*	18.1 ± 1.7
< 0.45 μm	8.4 ± 0.8	5.8 ± 0.8	7.8 ± 1.0	*nd*	16.0 ± 1.4
< 0.2 μm	7.2 ± 1.2	5.8 ± 0.8	6.7 ± 1.0	19.9 ± 1.7	15.1
< 0.1 μm	1.9 ± 0.7	2.2 ± 0.3	2.8 ± 1.1	3.4 ± 0.9	*nd*

[a] Effluent + 1 mg Al^{3+}/l (PACl); [b] One layer sand filtration (10 m/h) without coagulation; [c] Not determined

In the fractionation experiments the highest *SUR* for unfractionated effluent was $28.8 \cdot 10^{12} \pm 2.4 \cdot 10^{12}$ m^{-2}, which was found at wwtp Hoek van Holland. Next highest was $19.8 \cdot 10^{12} \pm 2.3 \cdot 10^{12}$ m^{-2}, which was found at wwtp Zaandam, and the lowest values were found for effluent of wwtp Berkel $11.1 \cdot 10^{12} \pm 2.4 \cdot 10^{12}$ m^{-2}. Pre-treatment of Berkel

wwtp-effluent lowered the *SUR* (*i.e.* increased filterability) with a maximum of 19%, down to $9.0 \cdot 10^{12}$ m^{-2} for coagulated effluent.

In figure 5.4 and 5.5 a graphical presentation of the results is shown for effluents of respectively Zaandam and Hoek van Holland. Zaandam effluent was fractionated for fractions down to 0.2 μm; the 0.1 μm fraction was not determined. The results for fractionation experiments of Zaandam wwtp-effluent (figure 5.4) showed a slight decrease in *SUR* of 3-9% per fraction. The smallest fraction, 0.2 μm, of wwtp-effluent (Zaandam) showed a 23% lower *SUR* than the *SUR* of the largest fraction (< 200 μm), but the smallest fraction (< 0.2 μm) still showed a relatively high *SUR* of $15.1 \cdot 10^{12}$ m^{-2} (see for comparison figure 4.15 in Chapter 4).

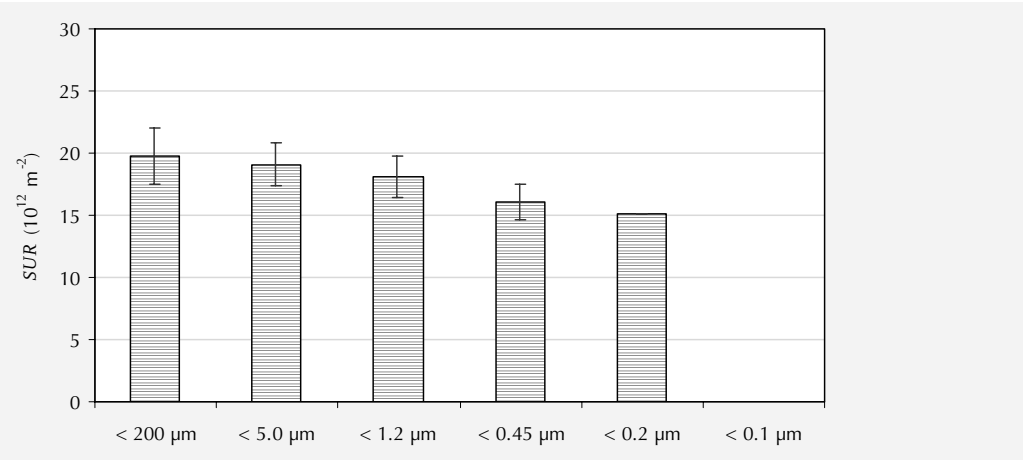

Figure 5.4 ***SUR*** **of various effluent fractions at wwtp Zaandam**

In fractionation experiments executed on effluent of wwtp Hoek van Holland (figure 5.5) the *SUR* decreased 31% relative to the unfractionated effluent (< 200 μm) for the fraction smaller than 0.2 μm, from $28.8 \cdot 10^{12}$ m^{-2} to $19.9 \cdot 10^{12}$ m^{-2}. An additional 57% decrease down to $3.4 \cdot 10^{12}$ m^{-2} was found for the smallest fraction (< 0.1 μm). These results indicate that the fraction retained by the 0.1 μm membrane, but permeated through the 0.2 μm membrane, determined to a large extent the filterability of the feedwater.

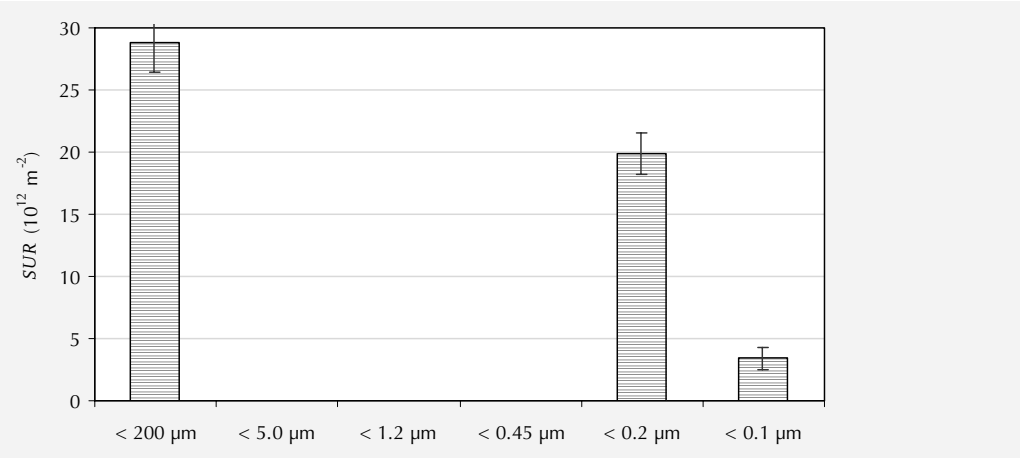

Figure 5.5 ***SUR*** **of effluent fractions at wwtp Hoek van Holland**

The results of effluent and pre-treated effluent from wwtp Berkel are graphically presented in figure 5.6. Contrary to figures 5.4 and 5.5 the Y-axis shows the *SUR* for a smaller range of 0 to $14 \cdot 10^{12}$ m^{-2}. As shown previously in Chapter 4, the *SUR* could be reduced by pre-treatment; in the fractionation experiments (figure 5.6) coagulation (1 mg Al^{3+}/l) reduced the *SUR* from $11.1 \cdot 10^{12}$ m^{-2} to $9.0 \cdot 10^{12}$ m^{-2}. The *SUR* of the subsequent fractions of coagulated effluent decreased with the lowest value found for the smallest fraction (< 0.1 μm).

Pre-treatment with sand filtration reduced the *SUR* from $11.1 \cdot 10^{12}$ m^{-2} down to $9.6 \cdot 10^{12}$ m^{-2}. For the subsequent fractions a decrease of -4% to 12% was found. Compared to the < 200 μm-fraction the decrease for the smallest fraction was 71%. Again the fraction retained by the 0.1 μm membrane and passed through the 0.2 μm membrane, seemed to predominantly determine the filterability of the feedwater.

A lower *SUR* was found for the smallest fraction of effluent compared to the smallest fraction of pre-treated effluent. This indicates that pre-treatment with coagulation (1 mg Al^{3+}/l) or sand filtration (one layer; 10 m/h; no coagulation) hardly influenced the smallest fraction (< 0.1 μm).

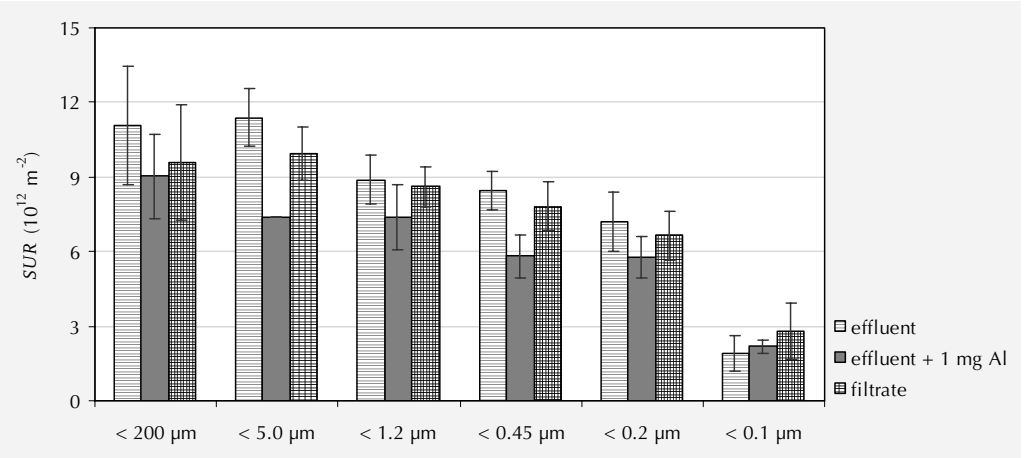

Figure 5.6 ***SUR*** **found for fractions of raw Berkel wwtp-effluent and effluent pre-treated with PACl (1 mg Al^{3+}/l) and sand filtration (one layer; 10 m/h; no coagulation)**

5.4 Discussion

Particle size distributions (PSD) of particles in the effluent can be used to calculate the theoretical cake layer thickness. In the experiments described in this chapter the theoretical cake layer thickness was calculated using PSD from 2-100 μm and from 0.5-10 μm. In both cases for a flux of 100 l/m².h and a filtration time of 30 minutes, the thickness was less than the diameter of the smallest particle. This suggested that particles larger than 0.5-2 μm do not have a great impact on the filterability of the wwtp-effluent. These particles are at least 20-50 times larger than the pore size of the ultrafiltration membrane, which is about 5-20 nm. The shape of particles in wwtp-effluent should not be considered as typically spherical and homogeneous. Furthermore, substances measured as particles might also change in shape and size by changing flow streams or changing pressures (O'Shaugnessy *et al.*, 1997). Nevertheless these simplifications, the conclusions still seem to be valid.

Filtration experiments of fractionated wwtp-effluent showed great differences in filterability of the fractionated particle ranges. The filterability was found to increase at decreasing particle range. Also in these experiments the term particle diameter had no

physical meaning with respect to the particles collected by filtration steps (Levine *et al.*, 1991). Particle multi-layers on top of the membrane surface might act as secondary filters that retain smaller material than expected from the pore diameter properties (Al-Malack and Anderson, 1997).

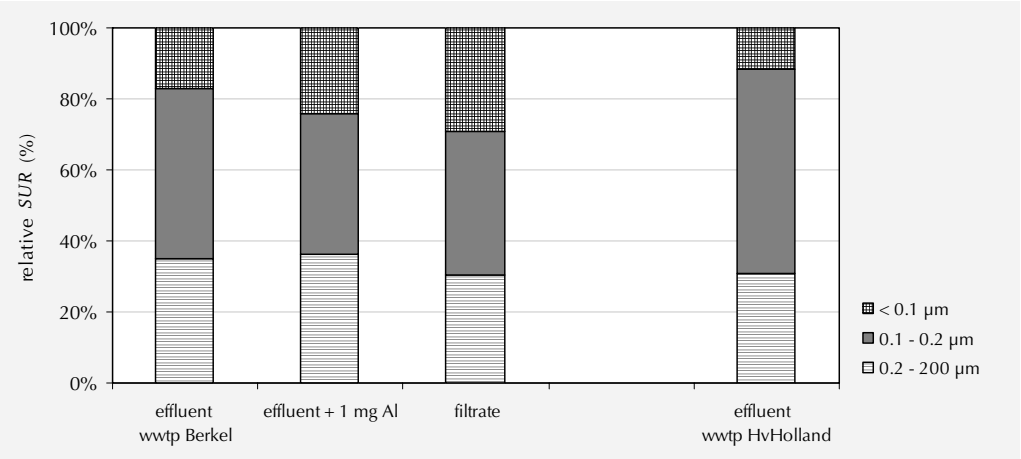

Figure 5.7 **Additional *SUR* relative to *SUR* value of < 200 µm fraction for three fraction, *i.e.* 0.2-200 µm, 0.1-0.2 µm and < 0.1 µm; calculated for fractions of effluent and effluent pre-treated with PACl (1 mg Al^{3+}/l) and sand filtration (one layer; 10 m/h; no coagulation); effluent samples were taken from wwtp Berkel (left) and wwtp Hoek van Holland (right)**

In figure 5.7 fractionation results are presented relative to the *SUR* of the unfractionated effluent, which is considered as the sum of the *SUR* for all fractions. These results showed that for effluent of wwtp Berkel the fraction smaller than 0.1 µm accounted for 12% to 29% of the *SUR* value for unfractionated effluent. The 0.1 to 0.2 µm fraction accounted for 41% to 57% and the fraction between 0.2 and 200 µm accounted for 30% to 36% of the *SUR* measured in unfractionated effluent. This showed that the 0.1 to 0.2 µm fraction accounted for the major part of the filterability and dominates filtration characteristics. This result was found for all experiments, for effluent of wwtp Hoek van Holland, for effluent of wwtp Berkel as well as for coagulated effluent (with 1 mg Al^{3+}/l) and pre-filtered effluent (with sandfilter without coagulation).

As the 0.1 μm to 0.2 μm fraction is five to twenty times larger than the membrane pore diameters, this fraction can hardly enter the membrane pores. These results therefore indicate that the filtration mechanism is sieving with the formation of a cake layer on top of the membrane.

In cross-flow systems treating surface waters, similar size-fractions were related to significant fouling problems. Thorsen (1999) found that particles within the range of 0.1-2 μm create problems with nanofiltration. Wiesner and Chellam (1992) assume that particles larger than 0.2 μm are back-transported from the membrane by the cross-flow stream. Wisniewski and Grasmick (1998) found for cross-flow microfiltration of activated sludge in a membrane bioreactor that the filtration characteristics were mainly determined by particles smaller than 0.05 μm. But the experiments described in this chapter were performed in dead-end configuration. The fraction between 0.1 and 0.2 μm is part of the semi-dissolved fraction defined in this chapter. Components found in this fraction might be, according to Levine *et al.* (1985), cell fragments, polysaccharides, proteins, and viruses, as well as humic acids, and colloidal material (Metcalf&Eddy, 2003). Research on the chemical properties of this fraction will provide information for a better understanding of the fouling phenomena (te Poele *et al.*, 2003).

The *SUR* of the effluent fraction < 0.1 μm was only slightly influenced by pre-treatment with coagulation or dual-media filtration. For the larger fractions (> 0.1 μm) pre-treatment had a positive effect on the filterability by reducing the *SUR*.

Pre-treatment of the effluent with coagulation or sand filtration showed only minor influence of the *SUR* of the fraction < 0.1 μm. The *SUR* values presented in figure 5.6 show that pre-treatment was able to increase the filterability for the fraction > 0.2 μm (*i.e.* lowering the *SUR*). The fraction between 0.1 and 0.2 μm might be reduced with other pre-treatment techniques, like for instance oxidation techniques (UV, ozonation, oxidising chemicals like NaOCl).

5.5 Conclusions

Calculations on the theoretical cake layer thickness and the fouled membrane area using particle counts of effluent within the range of 0.5 to 100 μm indicated that particles larger than 0.5-2 μm may have only little influence on the filtration characteristics. Particles smaller than 0.5-2 μm will therefore have a predominant influence on the filtration characteristics.

Fractionation experiments showed that particles within the range larger than 0.2 μm attribute to the filterability for only 30 to 36%. Particles within the 0.1 to 0.2 μm range showed a 40% to 57% contribution to the filterability. Particles within the latter range are about five to twenty times larger than the pore diameter (5-30 nm) of the ultrafiltration membrane. The smallest particles in the effluent passing a 0.1 μm membrane accounted for a 12% to 29 % of the filterability.

The results of the previous chapter (Chapter 4) indicated that a high filterability, measured as a *SUR* substantially lower than $10 \cdot 10^{12}$ m^{-2}, was needed for stable ultrafiltration performance with high fluxes. The results of the fractionation experiments indicated that these low *SUR* values will only be reached if the constituents in the effluent that are larger than 0.1 μm are considerably reduced, in this respect special attention should be given to the fraction of 0.1 to 0.2 μm size.

As the fraction of wwtp-effluent larger than 0.1 μm was responsible for 71% to 88% of the *SUR*, accumulation of effluent constituents on top of the membrane surface (*i.e.* sieving followed by cake layer filtration) is supposed to be the predominant filtration mechanism in dead-end ultrafiltration of wwtp-effluent.

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6 Theoretical analysis of filtration curves

6.1 Introduction

The findings from the experiments described in the previous chapters indicate that cake filtration is the predominant fouling mechanism in dead-end ultrafiltration of wwtp-effluent. Experiments on pilot-scale showed only a minor effect of pre-treatment on effluent filterability (Chapter 3). The applied pre-treatment techniques, coagulation (0.5 mg Al^{3+}/l) and multi-media filtration with pre-coagulation, generally should have a distinct effect on particles larger than 5 to 10 μm. This indicates that the particles smaller than 5 to 10 μm have a predominant effect on the filterability of the effluent. Assuming that the filtration process can be described by the cake filtration theory, experimental work was done on the development of the Specific Ultrafiltration Resistance (*SUR*) (Chapter 4). Filtration curves were measured in dead-end mode at constant TMP and for calculation of the *SUR* the filtration curves were transformed into t/V - V curves. These curves show a linear relationship between t/V and V, which indicates cake filtration to be the major fouling mechanism (Boerlage *et al.*, 2002; Schippers and Verdouw, 1980). The fractionation experiments described in Chapter 5 showed that a fraction of effluent constituents (sizing 0.1 to 0.2 μm), which are five to twenty times larger than the pore diameter of the ultrafiltration membranes, was predominantly responsible for the filtration characteristics.

Theoretical information about the fouling phenomena that occur in ultrafiltration systems was presented in the previous chapters (especially Chapter 1), but only limited knowledge is available about the fouling mechanisms occurring during dead-end ultrafiltration of wwtp-effluent (Bourgeous *et al.*, 2001; Decarolis *et al.*, 2001; Jarusutthirak *et al.*, 2002). In this chapter some filtration data are analysed

using the filtration laws that were theoretically derived by Hermia (1982). The objective of this chapter[1] is to verify the findings from the previous chapters, which assume the predominance of cake layer formation during dead-end ultrafiltration of wwtp-effluent. The usefulness of the filtration laws (Hermia, 1982) for identification of the predominant filtration mechanism is investigated.

6.2 General equation for filtration laws

The theoretical background of the filtration laws was presented in §1.3.4. As shown by Hermia (1982), the filtration laws can be rewritten into one characteristic form that is presented in equation 6.1. This equation represents[2] the resistance (inverse flux, dt/dV) related to the change in resistance (d^2t/dV^2).

$$\frac{d^2t}{dV^2} = k \cdot \left(\frac{dt}{dV} \right)^{\beta} \qquad \text{(eq. 6.1)}$$

where t = operation time (s)
V = cumulative permeate volume (m^3)
k = fluid characterisation constant ($s^{1-\beta} \cdot m^{3 \cdot (\beta-2)}$)
β = constant that indicates fouling mechanism (-)

When equation 6.1 is drawn in a log-scale graph, the slope of the curve is equal to the β-value (see equation 6.2). In table 6.1 the theoretically derived β-values are presented for the four filtration laws (Hermia, 1982).

$$\log\left(\frac{d^2t}{dV^2} \right) = \log(k) + \beta \cdot \log\left(\frac{dt}{dV} \right) \qquad \text{(eq. 6.2)}$$

[1] Part of this work has been presented in Roorda and van der Graaf (2002)

[2] The definition of flux is J = TMP / ($\eta_T \cdot R_{tot}$) (see eq. 1.2), for filtration at constant TMP and constant feedwater temperature (i.e. constant viscosity η_T) the resistance R_{tot} equals a constant value divided by the flux J (R_{tot} = constant/J); the flux is defined (eq. 1.1) as the difference in volume divided by the time difference ($J = dV/dt \cdot 1/A_m$), which shows that the resistance is a constant multiplied by dt/dV; the change in resistance is therefore the second derivate of the resistance (d^2t / dV^2)

Table 6.1 β values for various filtration mechanisms (Hermia, 1982)

Blocking filtration law	β
Complete blocking	2
Standard blocking	1.5
Intermediate blocking	1
Cake filtration	0

For identification of the occurring filtration mechanisms during micro- or ultrafiltration, various researchers analysed filtration data using equation 6.2. In the following sections a survey is presented of some of the results that have been found previously.

Protein fouling of microfiltration membranes can be simulated by microfiltration of solutions of bovine serum albumin, BSA[1]. The filtration laws have been applied on numerous BSA filtration data. Bowen *et al.* (1995) described flux decline at constant TMP for dead-end microfiltration of a solution with BSA (0.1 and 1.0 g/l). Four polycarbonate membranes with different pore diameters (0.1; 0.2; 0.4; 1.0 μm) were used in the experiments. Bowen *et al.* concluded that it was very difficult to distinguish specific filtration mechanisms, as these mechanisms occurred simultaneously. During the experiments the β-values decreased from 2 to 0, which might be explained as successive filtration steps or simultaneous presence of the different filtration mechanisms. In some experiments also negative β values were found, which Bowen *et al.* attributed to a limited cake layer resistance after which other (unspecified) mechanisms might occur.

Iritani *et al.* (1995) described similar experiments with BSA solutions (1.0; 2.0; 4.0 g/l) for four membranes with different pore sizes (polysulfone[2] – 13 nm; nitrocellulose – 25 nm; nitrocellulose – 50 nm; nitrocellulose – 0.1 μm). To explain all stages of filtration, none of the four different filtration laws could be applied. Although the β values were in the same range as in table 1, the values were different and also

[1] The shape of a BSA molecule is ellipsoid with the following dimensions: 140.9 x 41.6 x 41.6 Ångstrom (1 Ångstrom = 10^{-10} m²); its MWCO is approximately 67 kDa

[2] Chemical composition of the membrane and its pore diameter

negative β values were found (after about 2 hours of filtration). According to the authors these negative values might be explained by a combination of adsorption and detachment of the foulants. In a previous study Iritani *et al.* (1993) found that a compressible cake resistance model could accurately described filtration behaviour of BSA solutions.

Ho and Zydney (2000, 2001, 2002) performed similar experiments on microfiltration of BSA solutions. Ho and Zydney stated that none of the existing filtration models provided a full explanation for the complex range of fouling phenomena. Therefore, a new model that combines pore blockage and cake filtration was developed (Ho and Zydney, 2000). This model could accurately describe fouling behaviour in microfiltration of BSA solutions, even if negative β-values were measured. The model described the transition from pore blockage to cake filtration that arose because of a large reduction in the flux decline rate (dJ/dt).

Various authors used the filtration laws in a different way. For instance Jacob *et al.* (1998) performed research on both positively and negatively charged microfiltration membranes for filtration of BSA solutions (1 g/l). The filtration laws were rewritten to time dependent flux equations. It was concluded from the experiments that two or more filtration mechanisms coexisted simultaneously. Hlavacek and Bouchet (1993) derived the four filtration laws for constant flux experiments on dead-end microfiltration of BSA solutions. It was concluded that a good fit of experimental data by one of the filtration laws does not mean that the exact physical fouling mechanism is identified; but one of the filtration laws might reasonably explain the results and might therefore be used for quantification of membrane fouling.

In a study on fouling behaviour of silver dispersions, Kim *et al.* (1993) analysed filtration behaviour using the filtration laws. It was concluded from these analyses that initially pore blocking occurred, which was followed by cake filtration. No negative β-values were reported.

Wang and Lee (1999) reported filtration analysis on microfiltration of a polymer solution (PTMEG) containing solids. More than ten filtration equations with different β-values were derived. At the start of a filtration run low β-values were found

(cake filtration). This changed during filtration into higher β-values, which indicated a change from cake filtration to complete blocking at an increase in pressure drop.

Yuan *et al.* (2002) described constant pressure dead-end microfiltration (polycarbonate membranes; 0.2 μm) of humic acids (0.25; 1.0; 2.0; 4.0 mg/l) in solution. The results showed that β values decreased from 2 to 0, with finally even β values lower than 0. Apart from the classical models (Hermia, 1982) a combined pore blockage and cake filtration model was used to describe fouling behaviour (Yuan *et al.*, 2002). It was concluded that none of the classical fouling models were able to explain the negative values, but the model of Ho and Zydney (2000) combining pore blockage and cake filtration described filtration behaviour accurately.

Kennedy *et al.* (2002) calculated β values in dead-end ultrafiltration (MWCO = 150-200 kD) of canal water. The canal water had a turbidity of 16-23 NTU and a TSS of 19-21 mg/l. Experiments were performed for 100 minutes under constant pressure of 0.5 or 1.5 bar. The β values decreased rapidly from > 2 to around 0 in the first 20 minutes of filtration. During the last 80 minutes the β decreased further to 0 (0.02). Filtration of coagulated canal water showed similar results with a more rapid decrease from high values to 0. No specific conclusions were drawn on these β values.

Results of β-value-experiments on micro- and ultrafiltration of wwtp-effluent have been reported by Madaeni *et al.* (1995). Analysis of the filtration curves using the four filtration laws indicated that particle deposition within the membrane occurred, rather than the formation of a cake layer.

Fratila-Apachitei *et al.* (2001) investigated ultrafiltration of secondary effluent from refinery and petrochemical wastewater treatment plants with an average turbidity of 177 NTU. During constant pressure ultrafiltration with two different PES/PVP membranes (150 kDa ~ 6.6 nm; 50 kDa ~ 6.1 nm)[1] the β values were at the beginning of the filtration period as high as 10 and reached after some time values down to 0.2. According to the authors, this indicated a change in blocking mechanism from complete blocking towards intermediate and cake filtration. It was found that

[1] According to FESEM pictures showing the membrane surface (Fratila-Apachitei *et al.*, 2001)

membrane surface morphology had much more impact on the fouling rate than the pore diameter. Negative β values were not reported in this research.

6.3 Experimental set-up

Results of filtration experiments described in Chapter 4 (*SUR*) and Chapter 5 (Fractionation) were further analysed by calculation of the β-values. The filtration curve (filtered volume (V) versus filtration time (t)) was drawn using a spreadsheet program *Microsoft® Excel 2000*. The trending of the curve was fitted with a second order polynomial (t as a function of V), with calculation of the correlation coefficient R^2. The first derivative of this polynomial was used as dt/dV, and checked with the dt/dV of the raw data. The second derivative was d^2t/dV^2 and cannot be checked with the raw data, because of variations in interval. Furthermore, the relation between dt/dV and d^2t/dV^2 was drawn on log-scale axis. The slope of the curve on the log-scale axis was the β value. A more detailed description of this procedure is provided in Appendix 6-A.

Equation 6.1 is the general expression of all filtration laws for cake filtration and standard blocking. A linear relationship of t/V versus V and t/V versus t should theoretically be found respectively for cake filtration and standard blocking. All filtration data were also fitted to these two expressions (Chapter 1, eq. 1.11 and 1.9) with calculation of the correlation coefficient R^2. For each experiment the R^2 of the cake filtration curve and of the standard blocking curve were compared.

Filtration curves were measured with the lab-scale unit provided with one pressure vessel and the unit provided with two pressure vessels (see Chapter 4, §4.3.2). The unit with one vessel was pressurised from 0.0 bar to 0.5 bar within 2-3 minutes, which influenced the course of the filtration curve. In the analysis of the filtration mechanism these first minutes of filtration are neglected. In this chapter the analysis of the filtration curves measured with effluent from wwtp Tilburg-Noord and wwtp Berkel is presented. Filtration of effluent started from the beginning at a TMP of 0.5 bar for experiments using the lab-scale unit with two parallel pressure vessels. Filtration data of experiments on effluent of both wwtp Berkel and wwtp Emmtec were analysed.

6.4 Results

6.4.1 Lab-scale unit with one pressure vessel

All results are presented in detail in Appendix 6-B. In this section the results are summarised in two tables. In table 6.2 and 6-B.1 (Appendix 6-B) the results of the analysis on effluent of wwtp Tilburg-Noord are given. The *SUR* was relatively low for effluent and pre-treated effluent. The average β was found to be within the same order of magnitude as the theoretical values. In most experiments the β-values were not constant, but decreased during the 15 minutes of filtration. In some experiments the β was found above 2 or below 0 which values are theoretically impossible (Hermia, 1982). The effluent and the filtrate showed a β-value of about 1.5, which should indicate intermediate blocking (*i.e.* fouling on membrane surface without superimposition of particles). Although pre-treatment by multi-media filtration did not change the β-value, the *SUR* increased, showing its positive effect on the filterability of the effluent. Addition of a coagulant resulted in an average β-value of about 0.5, in some experiments even negative values were found. No improvement of the filterability was found, but a β-value of 0.5 and its linearity both indicate that cake filtration was the predominant filtration process for the coagulated effluent.

Table 6.2 Analysis of filtration curve of wwtp effluent (Tilburg-Noord)

feedwater	*SUR* (10^{12} m^{-2})	β-value (average)	range	Linearity of *t/V-t* or *t/V-V*[a]
Effluent	5.5 ± 0.9	1.4	0.7 – 5.2	Standard
Effluent + 1 mg Al^{3+}/l	5.6 ± 0.8	0.5	-1.3 – 5.2	Cake
Filtrate	3.7 ± 0.4	1.6	0.7 – 4.5	Standard

[a] Linearity of *t/V* vs. *t* indicates standard blocking, for *t/V* vs. *V* indicates cake filtration; the curve with the highest correlation coefficient (R^2) was chosen

In table 6.3 (and table 6.B.2 in Appendix 6-B) the results of experiments using effluent of wwtp Berkel are presented. The *SUR* was higher for effluent of wwtp Berkel than for effluent of wwtp Tilburg-Noord, which indicated a lower filterability for Berkel wwtp-effluent. The *SUR* was lowered by pre-coagulation for about 50%,

indicating an increase in filterability by pre-coagulation. The β-value changed too, for effluent from 0.5 to 0.2. This indicated that the filtration mechanisms changed into cake filtration during filtration. For pre-treated effluent negative β-values were found, after 4 hours of filtration the β-value was 0. Again this indicated that cake filtration was the predominant filtration mechanism.

In most experiments the β-value decreased slightly during 30 minutes of filtration. The highest correlation coefficients R^2 were found for the t/V vs. V curve, but the differences were mostly less than 1%. This showed that analysis of the linearity of the filtration data could hardly discriminate between the two filtration mechanisms.

Table 6.3 Analysis of filtration curves of wwtp-effluent (Berkel)

feedwater	time	*SUR* (10^{12} m^{-2})	β-value (average)	range	Linearity of *t/V-t* or *t/V-V*
Effluent	30 min	11.7 ± 2.4	0.5	0.1 – 0.4	Cake/ Standard
	4 hours		0.2	0.1 – 0.4	Cake
Effluent + 1 mg Al^{3+}/l	30 min	6.2 ± 0.5	-0.6	-1.0 – -0.3	Cake
	4 hours		0.0	-0.1 – 0.1	Cake

6.4.2 Lab-scale unit with two pressure vessels

Filtration curves were also measured by applying two pressure vessels in parallel. One was filled with demineralised water and one with the feedwater. This configuration showed a constant TMP from the start of a filtration until finishing the experiment. This should provide filtration curves without the influence of initial disturbances in TMP. Furthermore, fractionation of the (pre-treated) effluent was performed and for the various fractions the β-values were determined.

In table 6.4 a summary of the results is presented for (pre-treated) effluent of wwtp Berkel with a relatively high *SUR*. In table 6.5 the results are shown for Berkel effluent with a lower *SUR*. In these experiments also filtration curves of fractions down to 0.1 μm of the effluent were analysed. An overview of all results is presented in table 6-B.3 and 6-B.4 (Appendix 6-B).

Table 6.4 Analysis of filtration curves of (pre-coagulated) wwtp effluent (Berkel) with high *SUR* and for various fractions

Feedwater	Pre-filtration	*SUR* (10^{12} m^{-2})	β-value (average)	range	Linearity of *t/V-t* or *t/V-V*
Effluent	200 μm	12.2 ± 1.1	0.1	-2.0 – 0.6	Cake
	0.2 μm	9.1 ± 1.8	0.2	-0.7 – 0.6	Cake
Effluent + 1 mg Al^{3+}/l	200 μm	9.7 ± 2.3	-0.5	-0.9 – -0.2	Cake
	0.45 μm	6.2 ± 0.8	-0.4	-0.8 – -0.1	Cake

Table 6.4 shows that the *SUR* decreased by fractionating. These results indicated that still 65% to 75% of the filtration characteristics were determined by the fraction smaller than 0.2/0.45 μm (Chapter 5). No major change in β-values was found for the various fractions. The β-values were about 0 for effluent and for pre-coagulated effluent about -0.5. Also other researchers have reported negative β-values. Proposed explanations for the unexpected negative values were the occurrence of combined adsorption and detachment of foulants (Iritani *et al.*, 1995); the combination of pore blockage and cake filtration (Ho and Zydney, 2000; Yuan *et al.*, 2002); or even without a clear explanation, but relating the negative values to changes in the fouling layer characteristics (Bowen *et al.*, 1995). The complex mixture of constituents within the effluent is probably one of the main factors that determine the negative β-values. A possible explanation might be that smaller particles were constantly retained within the cake structure during the development of a cake layer, which is a filtration mechanism that is not covered by the filtration laws of Hermia (1982).

Table 6.5 Analysis of filtration curves of wwtp effluent (Berkel) with low *SUR* and for various fractions

Feedwater	Pre-filtration	*SUR* (10^{12} m^{-2})	β-value (average)	range	Linearity of *t/V-t* or *t/V-V*
Effluent	200 μm	7.2 ± 0.3	0.8	-0.4 – 4.0	Cake
	0.2 μm	5.6 ± 0.3	0.5	-1.1 – 3.9	Cake
	0.1 μm	1.9 ± 0.2	-0.6	-4.0 – 4.4	Standard

The results of experiments on effluent with a significant lower *SUR* (table 6.5) showed that the β-values decreased from 0.5 down to -0.6 for the respective fraction of 0.2 μm and 0.1 μm. This major decrease indicates that the filtration mechanism was mainly influenced by the 0.1 to 0.2 μm fraction. This might be due to a change from cake filtration (β = 0.5) to a different filtration process (β = -0.6).

Table 6.6 Analysis of filtration curves of wwtp effluent (Emmtec)

Feedwater	*SUR* (10^{12} m^{-2})	β-value (average)	range	Linearity of *t/V-t* or *t/V-V*
Effluent	10.3 ± 3.7	0.3	0.1 – 0.5	Cake
Filtrate	8.8 ± 2.5	0.3	-0.2 – 0.7	Cake
Filtrate + 1 mg Al^{3+}/l	6.1 ± 1.0	-0.1	-1.2 – 0.6	Cake

In table 6.6 the results are presented for analysis of filtration curves measured with (pre-treated) effluent of wwtp Emmtec. These results showed a decrease in β-value after pre-treatment. For pre-coagulated effluent the β-values even became negative.

6.5 Discussion

The models that were defined by Hermia (1982) for the description of various filtration laws were applied to filtration data that were found in the current research. The results of this approach showed that the β-values were in most experiments between 0 and 1. This seems to indicate that the filtration data can be described by the cake filtration, although no typical cake filtration (β-value is 0) was found. By plotting the data in a curve for t/V versus t ('standard blocking') and for t/V versus V ('cake filtration') the correlation coefficient of the linear relationship was almost the same. The deviation was in most experiments less than 1%, like in the example curve presented in Appendix 6-A. These results showed that different filtration laws could be used for description of the filtration data, which were found during dead-end ultrafiltration of wwtp-effluent.

In most experiments the β-values decreased from 2 to 0, and finally became negative. Other researchers also presented negative β-values (see §6.2), but the various

explanations could not be applied to the results found in this research. Here, it is hypothesised that the retention of small particles within the fouling layer results in gradual changes of the resistance of the fouling layer. In the first minutes of filtration a fouling layer is formed on top of the membrane surface. In the following minutes new particles can enter the fouling layer, or deposit on top of the fouling layer. For wwtp-effluent this is very well possible, as it comprises numerous different constituents varying in shape and size.

6.6 Conclusions

Analysis of the filtration mechanisms using the general expression for the filtration laws was done for dead-end ultrafiltration of wwtp effluent with and without pre-treatment. It showed that β-values are in the same order of magnitude as the theoretical values. The values did not clearly indicate one filtration mechanism, but β-values were mostly found between 0.0 and 1.0. This indicates that in most cases the cake filtration law could be used for description of the filtration mechanism.

Analysing filtration data by using the filtration laws is not strong enough to determine the filtration mechanisms that occur during dead-end ultrafiltration of wwtp-effluent.

The β values decreased slightly for fractions of smaller particles in the effluent. After pre-treatment with 1 mg Al^{3+}/l the β values even showed negative values. These values might be attributed to a fouling layer with the retention of small particles within the fouling layer.

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7 General discussion

7.1 Introduction

It has been shown from operational and economical point of view that ultrafiltration has great possibilities for the advanced treatment of wastewater. But, as described in Chapter 1, the filtration characteristics of wwtp-effluent are not well defined and the reason for decreasing membrane fluxes is hardly understood. The objective of the research presented in this dissertation was to determine the filtration characteristics of wwtp-effluent in dead-end ultrafiltration[1].

In this chapter the main findings from this research are presented (§7.2). Experiments were performed both on pilot-plant scale and on lab-scale. Combining the two made it possible to relate phenomena that occurred in pilot-plant scale to experimental results from the lab-scale research. The findings from the various researches are presented separately. In §7.3 the results are combined and some recommendations for improved ultrafiltration performance are done. In the last section the general conclusions are drawn.

7.2 Main findings

7.2.1 Filtration characteristics in theory

Performance of the ultrafiltration process is determined by both the filterability of the feedwater and by the reversibility of the fouling layer. Material retained during filtration can be found both inside the membrane structure, where it (partly) blocks

[1] Part of this work has been presented in Roorda and van der Graaf (2003)

the membrane pores, as well as on top of the membrane surface, where it also influences the characteristics of filtration. Long-term stable ultrafiltration performance will be found when the reversibility of the fouling layer is high. The filterability of the feedwater is important for the height of the applied flux. This is illustrated in figure 7.1, where line A to B shows the increase in the total membrane resistance during filtration, and B to A' show the decrease in resistance by cleaning of the membrane. This profile is similar if Trans Membrane Pressure (TMP) is considered. Figure 7.1 (a) shows the resistance or TMP profile that is found during stable operation; the increase in resistance during filtration is completely removed in the cleaning phase. Figures 7.1 (b) and (c) show the profile for unstable operation; the additional resistance is not completely removed in the cleaning phase, which results in a higher resistance at the start of the subsequent filtration phase.

Filterability and reversibility might be influenced by pre-treatment of the feedwater. In ultrafiltration of wwtp-effluent two commonly applied pre-treatment techniques are multi-media filtration and in-line coagulation. In several cases the filterability of the effluent and the reversibility of the fouling layer increased due to feedwater pre-treatment. Reversibility of the fouling layer is a function of the applied cleaning technique(s). Usually, tangential flow (cross-flow) and reversed flow (back flush) are used for the removal of retained material in or on the membrane matrix. Additional membrane cleaning is commonly applied with chemicals for oxidation (NaOCl) and/or dissolution of material (by changing the pH).

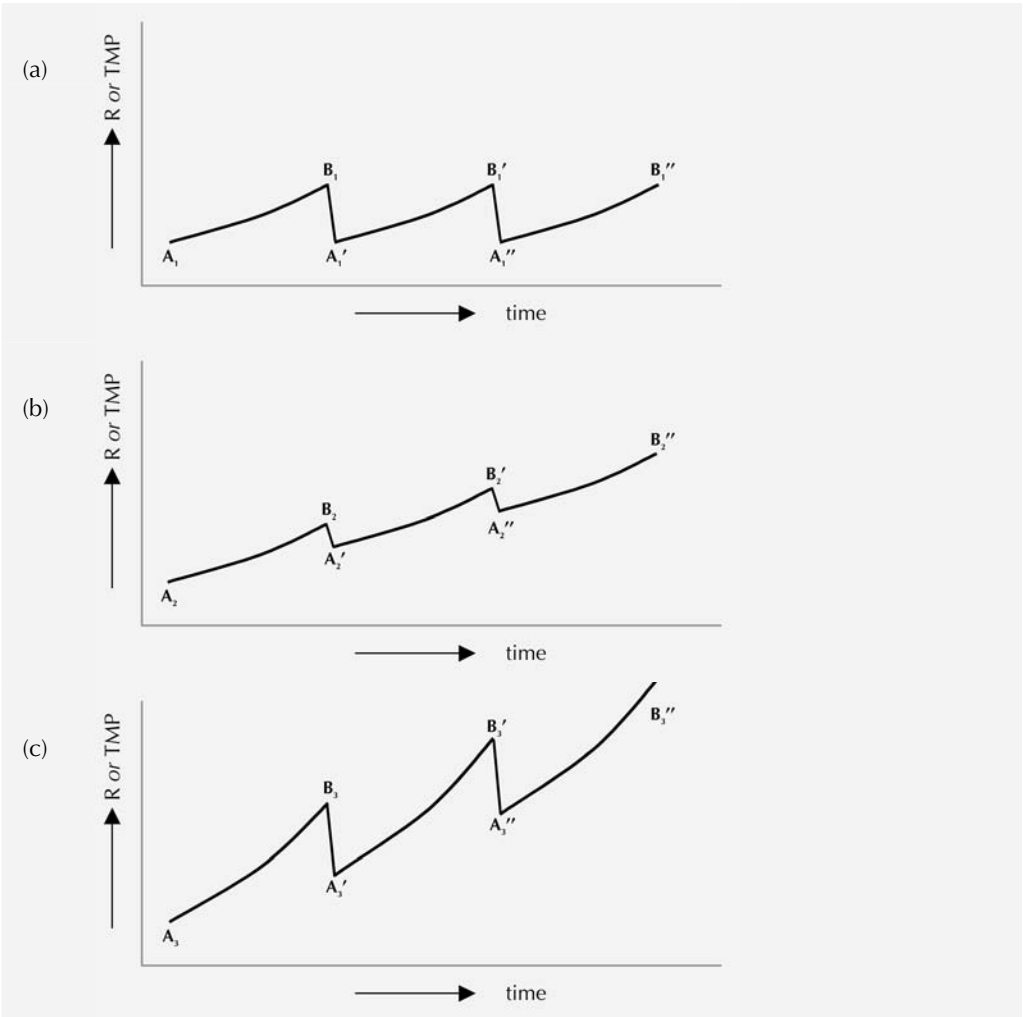

Figure 7.1 **Theoretical development of the resistance and/or TMP during constant flux ultrafiltration; line A to B shows the increase in resistance (ΔR) during filtration and line B to A′ shows the resistance decrease during cleaning of the membrane; in (a) the fouling layer is completely removed by cleaning, the following filtration step starts again at the initial resistance ($A_1' = A_1$); (b) shows a similar filterability as (a), but a lower reversibility, which results in an increase of resistance and/or TMP; (c) shows a larger resistance increase (*i.e.* lower filterability) with a subsequent cleaning step that is insufficient to remove the fouling layer completely; although the resistance decrease is higher than the resistance increase in (a), *i.e.* $\Delta R_3 > \Delta R_1$; a higher resistance and/or TMP is shown at the start of the subsequent filtration cycle ($A_3' > A_3$)**

7.2.2 Tests and experiments on pilot-scale

For more than six months each, pilot-plant tests were performed on dead-end ultrafiltration of effluent at different wwtp's in the Netherlands (Ede, Kaffeberg, Tilburg-Noord and Emmtec). In Chapter 2 an overview of these tests was presented. At wwtp Ede the use of effluent in the production of household water was investigated. At wwtp Kaffeberg, wwtp Tilburg-Noord and wwtp Emmtec the production of process water was studied. The research at wwtp Tilburg-Noord included also the production of water for irrigation.

Two different pre-treatment techniques were investigated, *i.e.* multi-media filtration and in-line coagulation ($\leq$ 2 mg Al^{3+}/Fe^{3+} per liter). In table 7.1 the process conditions are summarised for which stable performance of the ultrafiltration pilot-plant was found. Fluxes higher than those mentioned in table 7.1 led to serious fouling problems with increased membrane resistances. Permeate of the ultrafiltration plant was in all pilot-plant tests free of suspended solids (SS), particles and bacteria.

Table 7.1 Summary of process conditions for optimal performance at four pilot-plant tests on dead-end ultrafiltration of wwtp-effluent (at 15°C)

WWTP	Effluent composition			Pre-treatment	Cleaning[a]	Flux	TMP_n	Permeability
	COD (mg/l)	SS (mg/l)	Colour (mg Pt/l)			($l/m^2.h$)	(bar, 15°C)	($l/m^2.bar$, 15°C)
Ede	21–59	0.5–6	50–60	No; Filtration; 1 mg Fe^{3+}/l	BF*15*; CF*90*	60	0.3–0.4	150–200
Kaffeberg	10–57	< 2–6	28–92	Filtration + 1 mg Al^{3+}/l	BF*10*; CF*70*	80	0.4–0.7	115–200
Tilburg-Noord	20–40	< 1	56	Filtration; 2 mg Al^{3+}/l	BF*15*; CF*120*	>100	0.55	< 180
Emmtec[b]	3–23	4–10		Filtration + 2 mg Al^{3+}/l	BF*30*; CF*360*	100	0.3–0.45	330–220

[a] BF10 = Back Flush every 10 minutes; CF90 = Chemical Flush every 90 minutes; [b] Industrial wwtp

Although at all wwtp's the permeability with (pre-treated) effluent was in the same range (170-250 $l/m^2.h.bar$), the maximum flux for stable performance showed large differences. As the applied membrane systems were comparable, the major determining factor for the membrane fouling problems should be related to the feedwater composition. From the results of water quality analyses no direct relationship was found between feedwater composition and membrane performance.

For instance, the effluent at wwtp Kaffeberg had, compared to wwtp Ede, similar concentrations of SS, COD and colour, but the performance of the ultrafiltration plant was better than at wwtp Ede. These findings show that other parameters are necessary to relate feedwater composition to ultrafiltration performance. In the effluent of wwtp Tilburg-Noord low and constant concentrations of particles and SS were found, which was probably the result of the polishing step at this wwtp; the feedwater was secondary effluent polished in a pond system. At wwtp Tilburg-Noord the performance of the ultrafiltration pilot-plant was stable for both pre-filtered and pre-coagulated effluent at fluxes higher than 100 l/m².h. Without pre-treatment the performance could only be stabilised at a lower flux and higher TMP (J = 80 l/m².h; TMP = 0.7 bar). Maybe the high fluxes can be attributed to the additional polishing step (pond system) of wwtp Tilburg-Noord. As wwtp Emmtec treats industrial and not municipal wastewater, it is difficult to relate its results to the results found at the other wwtp's. The fluxes were kept stable at 100 l/m².h after pre-filtration combined with pre-coagulation. Generally, in all pilot-plant tests pre-treatment was necessary for stable performance of the ultrafiltration at higher fluxes compared to fluxes found for raw (not pre-treated) wwtp-effluent.

As described in Chapter 3, well defined experiments have been performed for characterisation of the feedwater Filterability F and the Reversibility of the fouling layer R_x. Filterability F was defined as the ratio between filtered volume (V/A_m) and fouling layer resistance. Reversibility R_x was defined as the relative change in resistance after a cleaning procedure ***x***. The R_x was calculated for the change in resistance after applying various cleaning methods (forward and back flush, chemical cleaning).

At wwtp Ede the Filterability of effluent was determined at a flux of 40, 70 and 100 l/m².h. As expected, the TMP increased after an increase of the flux. But the Filterability of effluent was independent of the applied flux.

The composition of the wwtp-effluent might be changed by pre-treatment with multi-media filtration or in-line coagulation. Multi-media filtration removes mainly particles larger than 10 μm. The majority of smaller particles will pass the filter bed and will be found in the filtrate. Pre-coagulation with a metal salt mainly influences particles larger than 5 μm. Pre-treatment of wwtp-effluent improved the Filterability ***F***

only slightly. These results indicated that particles smaller than 5-10 μm predominantly influence the filtration properties of the wwtp-effluent. At the same time the Reversibility (R_x) of fouling layer was increased by pre-treatment.

The pilot-plant research at wwtp Kaffeberg showed a low Filterability of the effluent, which was slightly improved by coagulation of the effluent. Reversibility of the fouling layer was also increased by coagulation. Comparison of the filtration curves of effluent and pre-treated effluent showed that without coagulation (and a backflush every 10 minutes) the resistance of the fouling layer was within 8 to 14 hours of filtration three times higher than the initial membrane resistance. Coagulation of the effluent as well as pre-treatment by multi-media filtration showed a similar increase in resistance after a much longer period of 55 to 66 hours. Although effluent of wwtp Kaffeberg had a relatively low Filterability, it was found that stable performance of the ultrafiltration plant with a flux of 80 l/m^2.h was only possible for pre-treated effluent combined with an intensive cleaning strategy.

At wwtp Tilburg-Noord an interesting relationship was found between the TMP profile and the turbidity of the multi-media filtrate, which was fed to the ultrafiltration plant. After cleaning of the multi-media filter bed the turbidity of the filtrate decreased for 10 hours, which indicated an improvement of the filter (ripening). After this, the turbidity increased again, indicating a loss of performance. The TMP over the ultrafiltration membrane increased at decreasing turbidity, whereas at increasing turbidity the TMP decreased. This illustrates the sensitivity of the ultrafiltration process to changes in chemical and physical properties of the feedwater.

The pilot-plant tests showed that the filtration properties of the effluent from different wwtp's were characteristic for each wwtp-effluent. The parameters commonly used to describe contaminants in wastewater like COD, turbidity, SS, nutrients, cannot be used for prediction of ultrafiltration performance. The pilot-plant tests on pre-treatment indicated that especially effluent constituents smaller than 5-10 μm determine filtration characteristics.

Furthermore, the pilot-plant experiments showed that extensive pilot-plant research is still necessary to find the optimal process configuration and pre-treatment steps. Pilot-plants might be optimised more easily if more information would be available about the Filterability wwtp-effluent and Reversibility of the fouling layer.

7.2.3 Parameter for evaluation of ultrafiltration characteristics: *SUR*

Until now, there is no parameter available that is able to relate feedwater properties to the filtration characteristics in dead-end ultrafiltration. In Chapter 4 the Specific Ultrafiltration Resistance, *SUR*, was introduced as a parameter for the evaluation of filtration characteristics. The *SUR* was calculated from filtration curves measured within 30 minutes with a lab-scale ultrafiltration unit equipped with one or two membrane fibres. The *SUR* was calculated from the filtration curve by applying the cake filtration theory (Hermia, 1982) and is defined as the product of the average specific cake resistance of the retained solids and the solids concentration. The *SUR* is expressed in m^{-2} and low *SUR* values indicate a relatively high feedwater filterability, whereas high *SUR* values indicate the opposite.

Various process conditions for optimised *SUR* measurement were investigated. The TMP influenced the filtration properties and was finally set to 0.5 bar, which is similar to the average TMP applied in full-scale plants for dead-end ultrafiltration of wwtp-effluent. An increased resistance of the fouling layer was found at increasing TMP. This increase was attributed to compression of the fouling layer. The temperature of the effluent greatly influenced the *SUR* measurement, which indicates that the temperature should be well defined. The temperature of the effluent for *SUR* measurement was proposed at ambient temperature of 20°C.

Each combination of feedwater and membrane showed specific filtration properties. To measure and to evaluate the filtration properties of a combination of feedwater and membrane, the same combination should be used for measurement of the *SUR*. In one experiment the *SUR* of wwtp-effluent was measured with two membranes, with the only difference being the MWCO (70 and 150 kDa). These two membranes showed a similar *SUR*. This indicates that effluent constituents with a size in the range of the pore diameter have a minor influence on filtration characteristics.

Dilution experiments of wwtp-effluent resulted in an almost linear relationship between the foulants concentration and the *SUR*, proving that the *SUR* actually measures filtration properties of wwtp-effluent. Furthermore, experiments on *SUR* measurement of pre-treated effluent clearly indicated that the *SUR* is a useful parameter for (on-line) evaluation of the effect of pre-treatment on filtration characteristics.

Finally, the *SUR* was measured for effluent of eight different wwtp's in the Netherlands. The *SUR* showed large variations for the different wwtp's. The range of the *SUR* was for these effluents between $5 \cdot 10^{12}$ to $30 \cdot 10^{12}$ m^{-2}.

7.2.4 Fractionation of wwtp-effluent

Results of pilot-plant tests (Chapter 3) indicated that effluent constituents smaller than 5-10 μm in diameter mainly determined filtration characteristics. Some results presented in Chapter 4 indicated that effluent constituents larger than the pore diameter had a large impact on filtration characteristics.

In Chapter 5 the theoretical cake layer thickness was calculated from particle size distributions (within the range of 2-100 μm and of 0.5-10 μm) in effluent and pre-treated effluent. These calculations showed that the concentration of particles larger than 2 μm (as well as 0.5 μm) is too low to form a cake layer on top of the membrane surface. Consequently, no significant increase in resistance can be expected from these particles.

Furthermore, wwtp-effluent was fractionated based on the size of its constituents. The *SUR* was measured for the resulting six fractions (< 200 μm, < 5 μm, < 1.2 μm, < 0.5 μm, < 0.2 μm, and < 0.1 μm). The *SUR* was measured using a membrane with a pore diameter of 5-20 nm. It was found that fractions of effluent passing the 0.2 μm filter were responsible for 64% to 77% of the *SUR*. On the other hand fractions of effluent passing the 0.1 μm filter are responsible for only 12% to 29% of the *SUR*. This showed that the fraction of effluent that passed a 0.2 μm filter and did not pass a 0.1 μm membrane predominantly determined the filtration characteristics.

Similar results were found for pre-treated effluent, which indicated that the 0.1-0.2 μm fraction was hardly influenced by coagulation (with 1 mg Al^{3+}/l) nor by sand filtration.

7.2.5 Filtration mechanism

The results of the fractionation experiments revealed that the feedwater constituents, which are larger than the pore diameter, have a major influence on filtration characteristics. These findings indicate that cake filtration should be the predominant filtration mechanism in dead-end ultrafiltration of wwtp-effluent. In Chapter 6 the

filtration mechanism was studied by analysis of filtration data. The theoretical filtration laws for complete blocking, standard blocking, intermediate blocking and cake filtration were used. Hermia (1982) derived a general equation for these filtration laws, which relates the fouling layer resistance to the change in resistance. The exponent of the formula is parameter β, which has a theoretical value for each filtration mechanism. As shown by others analysis of filtration data using this theoretical formula may provide insight in the occurring filtration mechanism.

In Chapter 6 filtration data that were measured in lab-scale experiments were analysed and the β-value was calculated. The β-values were found in the same order of magnitude as the theoretical values and resulted in a value close to zero, which is the theoretical value for cake filtration. For fractions of effluent with decreasing diameter the β-values decreased down to negative values. The results indicated that for effluent even for fractions down to 0.1 μm, the filtration data could be described with the cake filtration theory.

7.3 Discussion

7.3.1 The role of particle size

To investigate the possibilities of ultrafiltration for polishing of wwtp-effluent, it is necessary to study the filtration behaviour and pilot-plant performance in long-term tests (at least 4-6 months). This is due to uncertainties of the filtration characteristics of wwpt-effluent, and it is even amplified by variations in the composition of the effluent of one wwtp as well as of different wwtp's. More knowledge about the filtration characteristics and the influence of pre-treatment on filtration behaviour will enable more efficient pilot-plant tests.

Well-planned pilot-plant research provided useful information about the filtration characteristics of the wwtp-effluent. The pilot-plant experiments described in this dissertation indicated that filtration behaviour was predominantly influenced by effluent constituents with a particle diameter smaller than 5-10 μm. Subsequently, analyses of particle size distributions in the effluent (2-100 μm) showed that constituents larger than 2 (and 0.5) μm hardly influenced filtration characteristics. Finally, from lab-scale experiments on the filtration behaviour of size fractions of

effluent, it was found that the constituents that permeated through a 0.2 μm filter but that were retained by a 0.1 μm filter, predominantly (for more than 50%) determined filtration characteristics.

The pore diameter of the applied ultrafiltration membrane was still five to twenty times smaller than the 0.1-0.2 μm fraction. It must therefore be concluded that sieving, followed by cake filtration, determined the actual filtration behaviour in dead-end ultrafiltration of wwtp-effluent. No major influence was found of adsorption or any other mechanism than cake filtration. These findings were confirmed by results of lab-scale adsorption experiments that resulted in a constant Clean Water Flux for ultrafiltration membranes brought for more than twelve hours in contact with wwtp-effluent (Meezen, 2002). Also analysis of filtration data using theoretically derived formulas indicated that the filtration mechanism could be described by the cake filtration theory.

Similar observations were found for wwtp-effluent that was pre-treated with in-line coagulation (0.5 to 2 mg Al^{3+}/Fe^{3+} per liter) and multi-media filtration (with coagulation inside the filterbed). Contrary to the general opinion (Decarolis *et al.*, 2001; Wiesner and Laîné, 1996), coagulation did not result in the formation of larger particle aggregates. Although coagulation resulted in an increased suspended solids concentration, it was found that the particles which are larger than 2.0 μm showed a decreasing diameter. This was also found by van der Graaf *et al.* (2001) in experiments on multi-media filtration. The fractionation experiments (Chapter 5) showed a decrease of the *SUR* by pre-treatment of the effluent, but the *SUR* of the 0.1-0.2 μm fraction was hardly influenced by pre-treatment.

7.3.2 Surface structure

The surface structure of the membranes may play an important role in the occurring filtration mechanisms. The research of Doyen (1997) provides information for understanding of the filtration phenomena. If the membrane surface is flat with only pore openings of 5-20 nm in diameter, effluent constituents of five to twenty times larger than the pore diameter will form a cake on the membrane surface itself.
Figure 7.2 shows a membrane surface structure similar to the structure of the ultrafiltration membranes used in the experiments presented in this dissertation.
These ultrafiltration membranes have at the feedwater side a fibrous membrane

surface structure. The fibrous structure of the membrane surface may influence filtration characteristics (Doyen, 1997). The smallest pore openings are not found on top of the membrane, but can be found within the fibrous structure. Constituents with a diameter larger than the membrane pores will be retained within the fibrous structure, which will lead to blocking of the membrane openings.

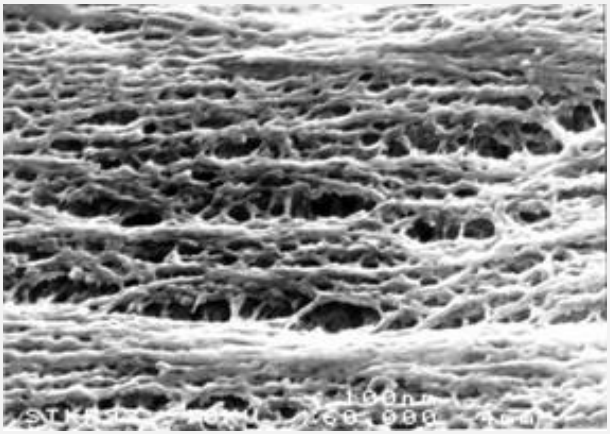

Figure 7.2 FE-SEM picture of skin layer of X-flow UFC membrane (PES/PVP) (adapted from Doyen (1997))

Next to this, the structure of the retained effluent constituents are probably very flexible and gel-like. The shape of the effluent constituents can therefore be influenced by the applied pressure gradient and might be adapted to the membrane structure (O'Shaugnessy *et al.*, 1997). This can even cause particles to get stuck inside the membrane matrix and these are hardly removed by cross-flushing or backflushing the membrane. According to Doyen *et al.* (1998), this may result in serious fouling problems because the Dirt Holding Capacity (DHC) of these membranes is relatively high.

Nevertheless, in the current research the size fraction that was retained by the 0.1 μm filter but passed the 0.2 μm filter predominantly determined filtration characteristics. The particles within this size range can be found between 50 to 200 nm in diameter. Even for the X-flow membrane the particles within this size range will be mostly be retained on top of the membrane surface, and not within the

membrane structure. So, the Dirt Holding Capacity of the membranes may not be relevant in the currrent research on wwtp-effluent.

7.3.3 Filtration cycle

It has commonly been stated that filtration always starts with pore blocking (Schippers and Verdouw, 1980). But from the current findings it was concluded that cake filtration occurred from the start of the filtration cycle. Although particles may enter the membrane pores, the filtration curve for dead-end ultrafiltration of wwtp-effluent was mainly influenced by the growing layer on top of the membane surface. The theoretical relationship presented in Chapter 4 (figure 4.1, adapted from Schippers and Verdouw, 1980) should therefore be redrawn.

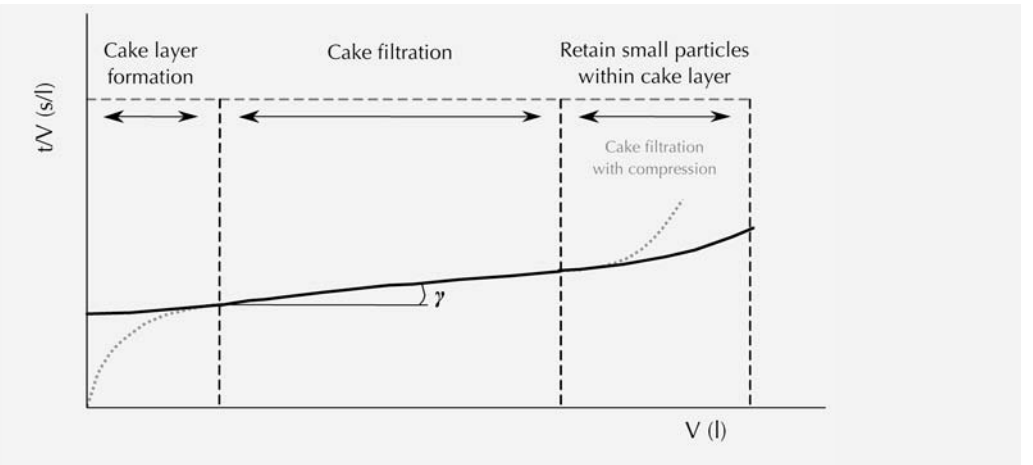

Figure 7.3 **Ratio of filtration time and filtered volume (t/V) as a function of the total volume of filtered effluent (V) at constant TMP; the black line indicates the filtration mechanism occurring during dead-end ultrafiltration of (pre-treated) wwtp-effluent: cake filtration with compression; compression is relatively small and can only be found if filtration is extended for a long period (more than hours); the dotted grey line indicates the theoretical relationship presented by Schippers and Verdouw (1980) for micro- and ultrafiltration of water samples; tan γ is used to calculate the Specific Ultrafiltration Resistance (SUR)**

In figure 7.3 the filtration curve is presented as found in this research on dead-end ultrafiltration of wwtp-effluent, showing initial cake layer formation gradually

changing to cake filtration with slowly emerging retention of smaller particles and maybe some cake layer compression. Compression was in the current research found at increasing TMP (Chapter 4), but in lab-scale experiments the TMP was constant and at increasing thickness of the fouling layer (after several hours of filtration) compression was not demonstrated.

7.3.4 Influence of pre-treatment

The research described in Chapter 4 showed that changes in the composition of the effluent could be measured as changes in the effluent filterability using the new parameter *SUR* (Specific Ultrafiltration Resistance). In most experiments the *SUR* decreased after pre-treatment of wwtp-effluent with coagulation or multi-media filtration. Lowering the *SUR* indicates that the filterability of the effluent increases. For most wwtp-effluents the decrease in *SUR* (*i.e.* an increase in filterability) is less than 25% for pre-treatment. However, pilot-plant tests showed that effluent pre-treatment led to a more stable ultrafiltration performance at higher fluxes, than was the case without pre-treatment. This underlines that in addition to the filterability of the effluent, the reversibility of the fouling layer should be taken into account.

The results of pilot-plant tests presented in table 7.1 showed that fluxes of 100 l/m².h and higher were found for pre-filtered and/or pre-coagulated effluent of wwtp Tilburg-Noord and wwtp Emmtec (with respectively a TMP of 0.55 and 0.3-0.45 bar). The *SUR* of untreated effluent from these wwtp's is $5 \cdot 10^{12}$ m^{-2} and $9 \cdot 10^{12}$ m^{-2} respectively. Only a small decrease of the *SUR* was found for these effluents after pre-treatment.

Much higher *SUR* values were measured for effluent of wwtp Ede and Kaffeberg, $18 \cdot 10^{12}$ m^{-2} and $29 \cdot 10^{12}$ m^{-2} respectively. Again, pre-treatment of the effluent resulted in the highest fluxes of 60 l/m².h (0.3-0.4 bar) for wwtp Ede and 80 l/m².h (0.4-0.5 bar) for wwtp Kaffeberg. Stable ultrafiltration performance for effluent of wwtp Kaffeberg was only found after extensive pre-treatment (multi-media filtration combined with coagulation of the filtrate) and intensive cleaning (backflush every 10 minutes; chemical cleaning every 70 minutes). Also at wwtp Ede the cleaning procedures were intensive (backflush every 15 minutes; chemical cleaning every 90 minutes) compared to those found at wwtp Tilburg-Noord and Emmtec.

From these results it was concluded that pre-treatment of wwtp-effluent with multi-media filtration or in-line coagulation is not always strong enough to substantially improve the filterability of wwtp-effluent. But additionally, pre-treatment influenced the material retained on the membrane, which led to an improved reversibility of the fouling layer.

7.3.5 Filtration characteristics in dead-end ultrafiltration of wwtp-effluent

These findings provided additional information to the findings of Bourgeous *et al.* (2001) on ultrafiltration of wwtp-effluent. Bourgeous *et al.* (2001) proposed a conceptual model on the impact of insufficient backwashing and operational mode on maintainable flux. In figure 7.4 their conceptuel model is presented, showing in figure 7.4 (a) the development of the flux in time versus TMP. Figure 7.4 (b) represents the flux development as a function of chemical cleaning interval. Point 1a to 1b in figure 7.4 (a) represents the flux development of a clean membrane during filtration. At the maximum allowable TMP the membrane was insufficiently backflushed. This will result in starting point 2a after which filtration will start at a higher TMP. The total membrane permeability will continuously decrease for the subsequent filtration intervals.

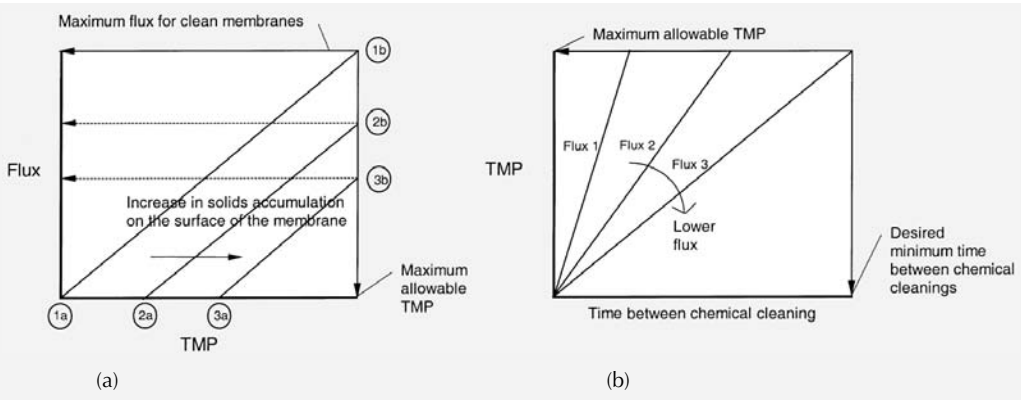

Figure 7.4 Conceptual model of impact of the insufficient backwash period and constant flux mode on maintainable flux (as proposed by Bourgeous *et al.* (2001))

In figure 7.4 (b) the occurring phenomena in pilot plant operation at constant flux mode is shown. If the chemical cleaning time is fixed in advance, low fluxes should be maintained. If a high flux (flux 1) is applied the maximum TMP will be exceeded before the minimum time between chemical cleanings. Because of this, only a low flux can be applied at constant flux. Therefore, Bourgeous *et al.* (2001) proposed operation of ulrafiltration plants at a variable flux and TMP.

In the research described in this dissertation a different pattern was found. From the results presented in the previous paragraphs the following hypothesis for dead-end ultrafiltration of wwtp-effluent is proposed: wwtp-effluent with a low initial filterability (the *SUR* exceeds $10 \cdot 10^{12}$ m^{-2}) will result in a slightly reversible fouling layer, even after pre-treatment of the effluent. This can be illustrated in figure 7.5 (which is similar to figure 7.1). Low initial filterability of wwtp-effluent (line A to B) will cause severe fouling problems, even if the effluent is pre-treated with in-line coagulation or multi-media filtration. The retained material can hardly be removed, which will result in a higher membrane resistance or TMP (A'). In the subsequent filtration cycle the fouling layer resistance is added to the higher membrane resistance A', which will result in a even higher total resistance or TMP (B'). The consequent cleaning procedure is again not sufficient enough and an even higher resistance will be found at the start of the following filtration cycle (A''). Because the fouling layer is highly compressible (Chapter 4) a higher TMP induces an even higher resistance of the fouling layer.

Bourgeous *et al.* (2001) described only insufficient cleaning intervals. The results in this dissertation showed additionally that long-term stable performance with fluxes as high as 100 $l/m^2.h$ will only be found for effluent with a high filterability, which is meausured as a low *SUR* ($< 10 \cdot 10^{12}$ m^{-2}). In that case, an increased reversibility of the fouling layer formation will be found for pre-treated effluent.

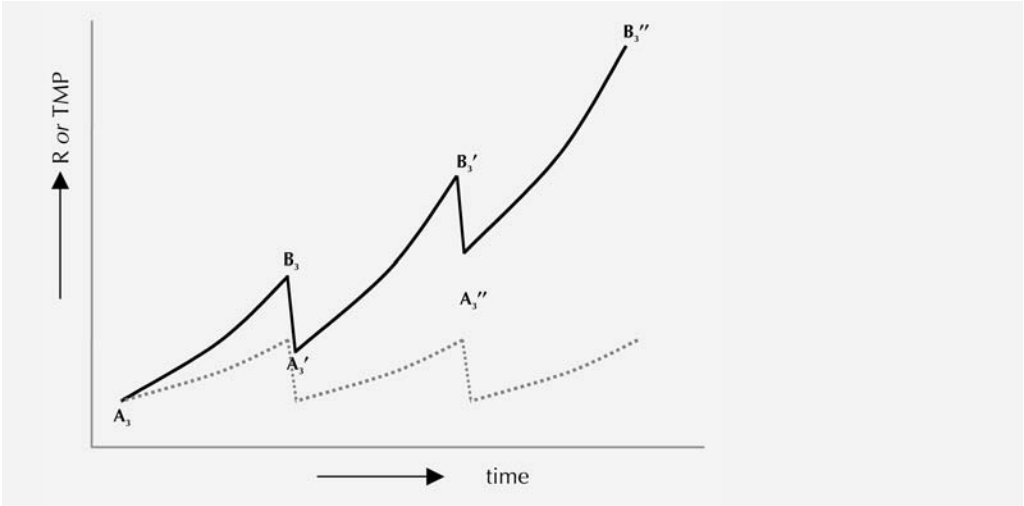

Figure 7.5 **Development of the resistance (R) and/or TMP during constant flux ultrafiltration of wwtp-effluent; line A to B shows the resistance increase during filtration and line B to A′ shows the resistance decrease by cleaning of the membrane; shows a larger resistance increase with a subsequent cleaning fase whicht is unable to remove the fouling layer completely, showing a higher resistance at the start of the subsequent filtration cycle (A′ > A), the increased TMP induces because of compression an even higher resistance; the dotted grey line represents the ideal situation in which a constant resistance or TMP can be obtained**

7.3.6 Recommendations for improved ultrafiltration performance

Pre-treatment with multi-media filtration or in-line coagulation did improve the filterability of the effluent with a maximum of 25%. But pre-treatment did not efficiently decrease (the effect of) the 0.1-0.2 μm fraction, which predominantly determines filtration characteristics. This fraction has a size 5-20 times larger than the pore diameter. Ultrafiltration performance will be improved if this fraction is removed or transformed. It is expected from the findings presented in this dissertation that only in that case effluent with a low filterability (i.e. *SUR* exceeds $10 \cdot 10^{12}$ m^{-2}) can form highly reversible fouling layers. Only than the performance of the ultrafiltration plant can be maximised up to fluxes of more than 100 $l/m^2.h$.

Removal of the 0.1-0.2 μm fraction might be done by the formation of a pre-coat on the membrane surface that acts as a secondary membrane (Galjaard *et al.*,

2001). Such a pre-coat should be chosen to preferentially remove effluent constituents within the size range of 0.05-0.4 μm.

Another possible removal concept could be biologically based. Kampf (2003) showed that water fleas (*Daphnia*) in a constructed wetland decreased the remaining organic content of the effluent. This might as well be the explanation for the good results found at wwtp Tilburg-Noord, where the secondary effluent is polished in a pond system. This polished effluent had a low *SUR* and stable performance with high fluxes was found in pilot-plant experiments.

Pre-treatment of effluent with activated carbon columns or the addition of powdered activated carbon (Clark *et al.*, 1996) might adsorb the 0.1-0.2 μm fraction preferentially.

Transformation of the 0.1-0.2 μm fraction might be achieved by addition of organic flocculants or coagulants that bind together effluent constituents (in the 0.1-0.2 μm range) and form larger and better filterable aggregates. Also oxidation of the 0.1-0.2 μm fraction might be possible by applying ozonation or UV-radiation.

If the cleaning procedure is optimised for the removal of the 0.1-0.2 μm fraction, an improved fouling layer reversibility may be found. It might in that case be possible to maintain high fluxes even for effluent with a low filterability. Until now, the application of such cleaning procedures is unknown.

Finally, analysis of the filtration characteristics of the different incoming flows in a wwtp, could make it possible to separate flows with a low filterability from the main influent flow. Specific foulants might be related to particular influent flows. When a particular flow with large concentrations of foulants is treated separately, this might improve the performance of the ultrafiltration plant.

7.4 Final conclusions

The objective of the research presented in this dissertation is 'the determination of filtration characteristics of wwtp-effluent in dead-end ultrafiltration'. It is shown that filtration characteristics are mainly determined by the fraction of wwtp-effluent with a size range (0.1-0.2 μm fraction) five to twenty times the pore diameter of the applied ultrafiltration membrane This fraction played a predominant in the formation of a cake layer on the membrane. This cake layer is highly compressible.

Pre-treatment of wwtp-effluent with in-line coagulation or multi-media filtration resulted in a small increase in filterability of the effluent. In pilot-plant tests a more distinct effect of pre-treatment on the reversibility of the fouling layer has been found. The reversibility of the fouling layer was improved by pre-treatment of the effluent.

A new parameter for evaluation of ultrafiltration characteristics of wwtp-effluent is called the Specific Ultrafiltration Resistance, *SUR*, and is measured using a lab-scale device within 30 minutes of filtration. The *SUR* is developed for evaluation of the filterability of wwtp-effluent and for evaluation of the impact of pre-treatment on the initial filterability. A low *SUR* indicates a high filterability. It was found that effluent with a low *SUR* resulted in stable ultrafiltration performance with fluxes higher than 100 l/m².h. On-line measurement of the *SUR* might be used for online adaptation of the process conditions of the ultrafiltration plant, which may improve ultrafiltration performance.

Pilot-plant tests at different wwtp's in the Netherlands showed that *SUR* values below $10 \cdot 10^{12}$ m^{-2}, *i.e.* with a high initial filterability, seem to be a precondition for stable ultrafiltration performance with high fluxes ($\geq$100 l/m².h). Effluent with high *SUR* values caused very extensive cleaning procedures in order to maintain high fluxes.

The research presented in this dissertation showed that polishing of wwtp-effluent with dead-end ultrafiltration is a viable option for further reuse of the effluent. The produced water is disinfected and free of particles and has a sufficient quality for direct use, or for use after a subsequent treatment step like desalination. Continued pilot-plant research will be necessary for analysis of the performance and for optimisation of the process configuration. Before starting pilot-plant tests a thorough analysis should be made of the filtration characteristics of the (pre-treated) effluent. This research showed that the *SUR* is a useful parameter for that purpose. If the *SUR* of effluent exceeds, even after pre-treatement, a value of $10 \cdot 10^{12}$ m^{-2} the pilot plant location should be reconsidered.

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Summary

FILTRATION CHARACTERISTICS IN DEAD-END ULTRAFILTRATION OF WWTP-EFFLUENT

Jelle Henderikus Roorda

With a growing need for water for many purposes on one side and an increasing degree of wastewater treatment producing water with an improved quality on the other side, the necessity of water reuse will grow, especially in highly industrialised countries with restricted natural water sources. Improvement of advanced treatment schemes for effluent will promote water reclamation and conservation in the near future. Membrane filtration will play an important role in such treatment schemes. The research presented in this dissertation focused on the application of ultrafiltration membranes for advanced treatment of wastewater treatment plant (wwtp) effluent. The filtration characteristics of wwtp-effluent are unknown and the mechanisms behind the decreasing fluxes are hardly understood. The objective of research was to determine the filtration characteristics of wwtp-effluent in dead-end ultrafiltration.

The research was performed both on pilot-scale and lab-scale. Pilot-scale tests were done at four different wwtp's for at least 6 months of research each. Two different techniques for pre-treatment of the wwtp-effluent were investigated, *i.e.* multi-media filtration and in-line coagulation ($\leq$ 2 mg Al^{3+}/Fe^{3+} per liter). Although the optimal process conditions were different at the various wwtp's, the permeate was in all tests free of particles and bacteria. The fluxes were found between 60 and > 100 $l/m^2.h$ at a TMP of 0.3-0.6 bar. The variations in effluent composition at different wwtp's urge the need for extensive pilot-plant studies to find the optimal process configuration and pre-treatment technique. These tests would be less time-consuming when proper parameters exist, which can describe the filterability of the wwtp-effluent.

Such experiments were done on pilot-scale, in which well-defined experiments were performed for the determination of the filterability of the wwtp-effluent and on the reversibility of the fouling layer. These experiments showed that pre-treatment of wwtp-effluent with multi-media filtration or coagulation had a minor influence on the filterability, indicating that filtration characteristics are determined by particles smaller than 5-10 μm. The reversibility of the fouling layer

was improved by pre-treatment. Nevertheless, these experiments were time consuming and underlined the need for quick tests for the determination of the filtration characteristics.

The Specific Ultrafiltration Resistance (*SUR*) is proposed as such a parameter. The *SUR* was developed in the current research and is determined in a lab-scale ultrafiltration set-up. Within 30 minutes a filtration curves is measured at a TMP of 0.5 bar, using a membrane module with a capillary membrane. The filterability of effluent was determined and the range for the *SUR* was for wwtp-effluent $5 \cdot 10^{12}$ to $30 \cdot 10^{12}$ m^{-2}. Low *SUR* values relate to high filterable water, whereas high *SUR* values relate to less filterable water. The TMP affected the *SUR*, which was caused by compression of the fouling layer (compression coefficient is 0.61-0.75).

The influence of the particle diameter from effluent constituents on the filterability was determined by fractionation of the effluent. Effluent constituents that passed a 0.2 μm filter, but were retained by a 0.1 μm filter determined the filterability for 39% to 57%. Pre-treatment of the effluent had hardly any influence on this fraction, which is five to twenty times larger than the pore diameter of the applied membrane. This shows that the formation of a fouling layer on top of the membrane is the main filtration mechanism. This was confirmed by analysis of the filtration data using the filtration laws. Because the filtration laws could not completely cover the filtration data, it is hypothesised that the occurring filtration mechanism is the formation of a fouling layer and the retention of smaller particles within this layer, thus responsible for an increased resistance of the fouling layer.

The results of pilot-plant tests were combined with the results of *SUR* measurements. Pre-treatment and membrane cleaning of effluent with a low filterability showed only partly improvement of ultrafiltration performance. Effluent with a high filterability (i.e. low *SUR*) is a requirement for highly reversible fouling layers that will result in stable ultrafiltration performance with fluxes as high as 100 $l/m^2.h$. The optimisation of the ultrafiltration process for treatment of wwtp-effluent should therefore include the influencing of the properties or removal of the 0.1-0.2 μm fraction.

Samenvatting

FILTRATIE EIGENSCHAPPEN VAN RWZI-EFFLUENT TIJDENS DEAD-END ULTRAFILTRATIE

Jelle Henderikus Roorda

De vraag naar water neemt nog steeds toe, maar het afvalwater wordt steeds beter gezuiverd. Afvalwater wordt daardoor steeds vaker beschouwd als een bron voor hergebruik, die kan voorzien in de toenemende vraag. Dit is vooral het geval in geïndustrialiseerde landen met een beperkte hoeveelheid natuurlijke waterbronnen. Vergaande behandeling van afvalwater heeft een positief effect op de terugwinning van water uit afvalwater en op de bescherming van de natuur. Membraanfiltratie speelt hierin een belangrijke rol. Het onderzoek dat in dit proefschrift is gepresenteerd gaat in op de toepassing van ultrafiltratie voor vergaande behandeling van afvalwater afkomstig van communale rioolwaterzuiveringsintallaties (rwzi's). De filtratie eigenschappen van rwzi-effluent zijn nog onbekend en de mechanismen die een rol spelen in het vervuilen van de membranen (en daarmee het afnemen van de prestaties) zijn onduidelijk. Het doel van het hier gepresenteerde onderzoek was om de filtratie eigenschappen te bepalen van rwzi-effluent tijdens dead-end ultrafiltratie.

Het onderzoek is uitgevoerd op pilot- en lab-schaal. Het pilot-schaal onderzoek vond plaats op vier verschillende rwzi's, per rwzi voor minimaal zes maanden. Twee verschillende technieken voor voorbehandeling van het rwzi-effluent zijn onderzocht: multi-media filtratie en in-line coagulatie ($\leq$ 2 mg Al^{3+}/Fe^{3+} per liter). De condities waaronder de beste prestaties gevonden werden verschilden per locatie, maar in alle gevallen was het gezuiverde effluent (het permeaat) vrij van bacteriën en deeltjes. De fluxen waarbij een stabiele bedrijfsvoering gevonden werd, varieerden tussen de 60 en de 100 $l/m^2.h$ bij en Trans Membraan Druk (TMD) van 0,3 tot 0,6 bar. De variaties in de samenstelling van het effluent van de verschillende rwzi's maakten het noodzakelijk dat het onderzoek op pilot-schaal werd uitgevoerd; zonder dit onderzoek op pilot-schaal is het welhaast onmogelijk om de optimale procesconfiguratie en voorbehandelingsstap te bepalen. Het onderzoek zou minder tijdrovend kunnen worden indien er goede parameters beschikbaar zouden zijn, die de filtreerbaarheid van het effluent goed kunnen voorspellen.

Op pilot-schaal is onderzoek uitgevoerd om in een korte, goed gedefinieerde test de filtreerbaarheid van het rwzi-effluent en de verwijderbaarheid van de vervuiling op het membraan te bepalen. Deze tests wezen uit dat voorbehandeling van het effluent met zowel multi-media filtratie als in-line coagulatie slechts een beperkte invloed heeft op de filtreerbaarheid. Daaruit is geconcludeerd dat de filtratie eigenschappen vooral bepaald worden door deeltjes die niet of nauwelijks door deze technieken beïnvloed worden, deze deeltjes zijn kleiner dan 5-10 μm. De verwijderbaarheid van de vervuiling werd wel positief beïnvloed door voorbehandeling van het rwzi-effluent. Een nadeel van deze gedefinieerde tests was de tijdrovendheid, waarmee nogmaals de noodzaak voor snelle tests, waarmee een goede parameter gemeten kan worden, duidelijk werd.
Een dergelijke snel te meten parameter is ontwikkeld tijdens dit onderzoek: de Specifieke Ultrafiltratie Weerstand (*SUR*). De *SUR* wordt bepaald in een lab-schaal opstelling. Daarbij wordt met een kleine membraanmodule met daarin één of enkele membraanrietjes gedurende 30 minuten de filtratiecurve gemeten bij een TMD van 0,5 bar. De filtreerbaarheid van het rwzi-effluent is bepaald als *SUR* in de range van $5 \cdot 10^{12}$ tot $30 \cdot 10^{12}$ m^{-2}. Een lage waarde wordt gevonden voor goed filtreerbaar water, bij een hogere waarde neemt de filtreerbaarheid af. De TMD beïnvloedde de *SUR*, wat veroorzaakt wordt door samendrukking van de vuillaag (een compressie coëfficiënt van 0,61 tot 0,75).

De invloed van de deeltjesgrootte van bestanddelen in het effluent op de filtreerbaarheid is bepaald met behulp van fractionerings experimenten. Hieruit bleek dat stoffen die worden tegengehouden door een 0,1 μm filter, maar die door een 0, 2 μm filter worden doorgelaten voor 39 tot 57% de filtreerbaarheid bepalen. Voorbehandeling had nauwelijks invloed op deze fractie die in ordegrootte vijf tot twintig maal groter is dan de diameter van de membraanporiën. De ordegrootte van deze stoffen tonen aan deze het belangrijkste filtratie mechanisme koekfiltratie was. Dit werd onderstreept door een analyse van de filtratie data met behulp van de filtratiewetten, zoals beschreven door Hermia (1982). Desondanks bleek dat deze filtratiewetten niet krachtig genoeg zijn om de gevonden resultaten volledig te verklaren. In dit proefschrift is daarom een nieuw filtratie mechanisme geïntroduceerd: het filtratiemechanisme is de vorming van een vuillaag op het membraan, waarin continue kleinere deeltjes in worden ingevangen, wat leidt tot een continue toename van de weerstand over het membraan.

Tenslotte zijn de resultaten van de pilot-schaal experimenten gecombineerd met de resultaten van de *SUR* metingen. RWZI-effluent met een slechte filtreerbaarheid (hoge *SUR*) kan slechts voor een deel met behulp van voorbehandeling en membraanreiniging leiden tot een verbetering van de ultrafiltratie installatie. Een hoge initiële filtreerbaarheid van het rwzi-effluent blijkt een vereiste te zijn voor een stabiele bedrijfsvoering met fluxen hoger dan 100 l/m².h. Dit kan ook gaan gelden voor slecht filtreerbaar effluent indien de eigenschappen van de 0,1-0,2 μm fractie worden veranderd of deze deeltjes worden verwijderd.

Terminology and abbreviations

BOD = biochemical oxygen demand (mg O_2/l)

the oxygen equivalent of the organic matter that can be oxidised biochemically by micro-organisms; mostly determined as BOD_5^{20} (oxygen demand over 5 days at 20°C)

COD = chemical oxygen demand (mg O_2/l)

the oxygen equivalent of the organic matter that can be oxidised by using a strong chemical oxidant in an acidic medium (usually potassium dichromate)

effluent

outgoing flow of treated wastewater

filtrate

outgoing flow from multi-media filter

influent

incoming flow of water or wastewater

MFI-UF

Modified Fouling Index using Ultrafiltration membranes; parameter for prediction of fouling properties of RO-feedwater, defined by Boerlage *et al.* (2002) in addition to the Modified Fouling Index (MFI) defined by Schippers and Verdouw (1980)

$MFI\text{-}UF_n$

The normalised MFI-UF, defined by Roorda and van der Graaf (2001) as a parameter for prediction of fouling properties in MF and UF membranes; in opposite to the MFI-UF is it measured within 30 minutes of filtration; for this dissertation the $MFI\text{-}UF_n$ is substituted by the *SUR* (see also Appendix 4-A)

p.e. = population equivalent

the oxygen equivalent to the amount of oxygen demand related to one inhabitant; for publicly owned wwtp calculated as 54 g BOD_5^{20} per day

permeate
: clean water coming out of a membrane

SS = suspended solids

SUR = Specific Ultrafiltration Resistance
: a parameter defined in the current thesis for evaluation of the filterability of the effluent; the *SUR* can be calculated from filtration curves found in dead-end, constant TMP ultrafiltration and is the product of the average specific cake resistance and the concentration of solids in the effluent

TMP = Trans Membrane Pressure
: pressure difference between permeate and feedwater in a membrane filtration installation

turbidity
: a measurement of light-transmitting properties of water indicating the quality of water with respect to residual suspended matter

wwtp = wastewater treatment plant
: generally a publicly owned municipal wastewater facility

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Curriculum Vitae

Jelle Henderikus Roorda was born on the 23rd of July 1972 in Den Helder, the Netherlands. After finishing secondary school (VWO: Middelburg) in 1990, he started his MSc.-study Environmental Hygiene at the Agricultural University Wageningen (nowadays Wageningen University). He finished his studies at the Sub-department of Environmental Technology with two subjects, 'Flotation in a zig-zag column' and 'Lignin removal in anaerobic-aerobic treatment of pulp wastewater'.

During a training period at the University of Amman (Jordan) he worked on the implementation of a two-stage UASB for the treatment of municipal wastewater at wwtp Khirbet As-Samra. In 1997 he finished his studies in Wageningen.

In 1998 he started as a researcher at the Delft University of Technology, Faculty of Civil Engineering and Geosciences. He worked at the research-group of prof.ir. J.H.J.M. van der Graaf on the department of Sanitary Engineering, within the project 'Membrane filtration of effluent', which was funded by Rossmark Watertreatment (a Veolia Water company) and Witteveen+Bos consulting engineers with support of the Dutch Ministry of Economic Affairs. The work carried out in this project formed the basis of this dissertation.

Since May 2003, he is employed at the engineering and consultancy firm Grontmij Water & Waste Management in de Bilt (the Netherlands). He is nowadays involved in various projects dealing with treatment and reuse of water and waste.

Appendices

Appendix 1 - Introduction

1-A Technologies used for advanced treatment

The most relevant types of unit operations for the removal of various components are presented in the following table. This table is adapted from Metcalf & Eddy (2003), table 11.2, p. 1040-1041.

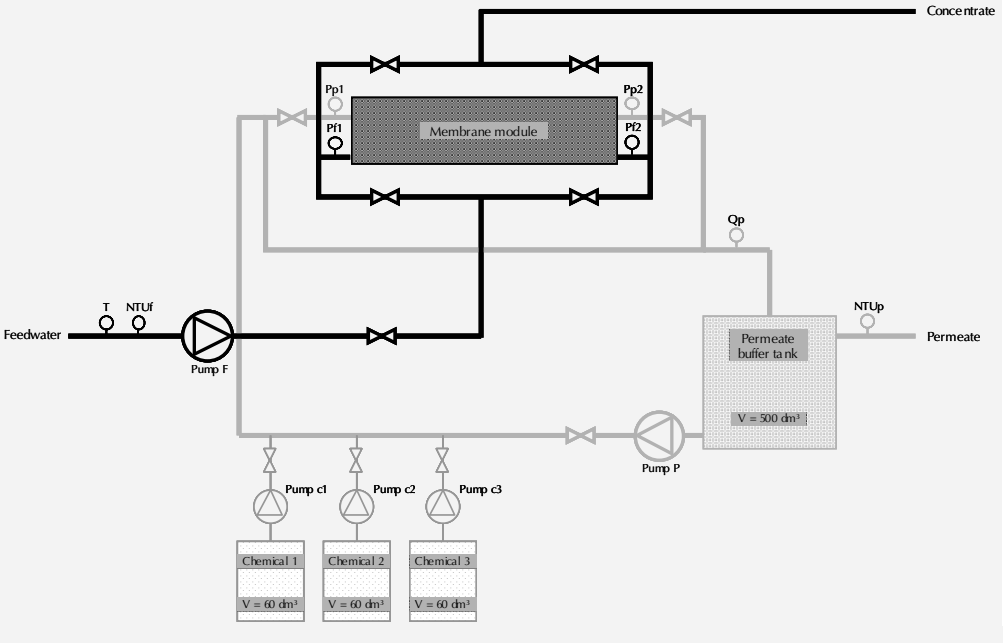
	Unit operation or process (1)					
Residual constituent	Multi-media filtration	Surface filtration	Micro- and ultrafiltration	Reverse osmosis	Electro-dialysis	Adsorption
Inorganic and organic colloidal and suspended solids						
Suspended solids	X	X	X	X	X	X
Colloidal solids	X	X	X	X	X	X
Organic matter (particulate)				X	X	
Dissolved organic matter						
Total organic carbon				X	X	X
Refractory organics				X	X	X
Volatile organic compounds				X	X	X
Dissolved inorganic matter						
Ammonia				X	X	
Nitrate				X	X	
Phosphorus	X			X	X	
Total dissolved solids				X	X	
Biological						
Bacteria			X	X	X	
Protozoan cysts and oocysts	X		X	X	X	X
Viruses				X	X	

	Unit operation or process (2)					
Residual constituent	Air stripping	Ion exchange	Advanced oxidation processes	Distillation	Chemical precipitation	Chemical oxidation
Inorganic and organic colloidal and suspended solids						
Suspended solids		X		X	X	
Colloidal solids		X		X	X	
Organic matter (particulate)				X		X
Dissolved organic matter						
Total organic carbon		X	X	X	X	X
Refractory organics			X	X		
Volatile organic compounds	X		X	X		
Dissolved inorganic matter						
Ammonia	X	X		X		
Nitrate		X		X		
Phosphorus				X	X	
Total dissolved solids		X		X		
Biological						
Bacteria				X		
Protozoan cysts and oocysts		X		X	X	
Viruses				X		

Appendix 2 – Pilot-plant research

2-A Schematic drawing of pilot plant installations

In the figure below the ultrafiltration pilot plant is schematically drawn. The membrane module can be fed from both ends and can also be backflushed from both sides. Chemical cleaning of the membrane module is possible and as well as forward flushing of the membrane module. Temperature, turbidity (in and out), flow, pressure and time intervals are logged for further data analysis.



Online measurement of:

T = Temperature feedwater

NTUf = Turbidity feedwater (0-15 NTU)

Pf1, Pf2 = manometer f1, f2 for pressure at feedwater side (0-2 bar)

Qp = permeate flow (0-15 m^3/h)

NTUp = Turbidity permeate (0-2 NTU)

Pp1, Pp2 = manometer p1, p2 for pressure at permeate side (0-2 bar)

Pumps UF pilot-plant:

Pump F = feedwater pump (0-15 m^3/h)

Pump P = backflush pump (0-25 m^3/h)

Pump c1, c2, c3 = pump cleaning chemicals (0-25 dm^3/h)

Figure 2-A.1 Schematic drawing of the ultrafiltration pilot plant

In figure 2-A.2 the multi-media pilot plant is schematically drawn. The filterbed consisted of one, two or three layers of filter media. Coagulants were dosed for flocculation inside the filterbed. The filterbed can be cleaned based on time interval or pressure increase (turbidity increase of the filtrate). The filter was equipped with sampling probes along the height of the filter bed (every 100 mm), with which samples could be taken for measurement of turbidity and for head loss measurements.

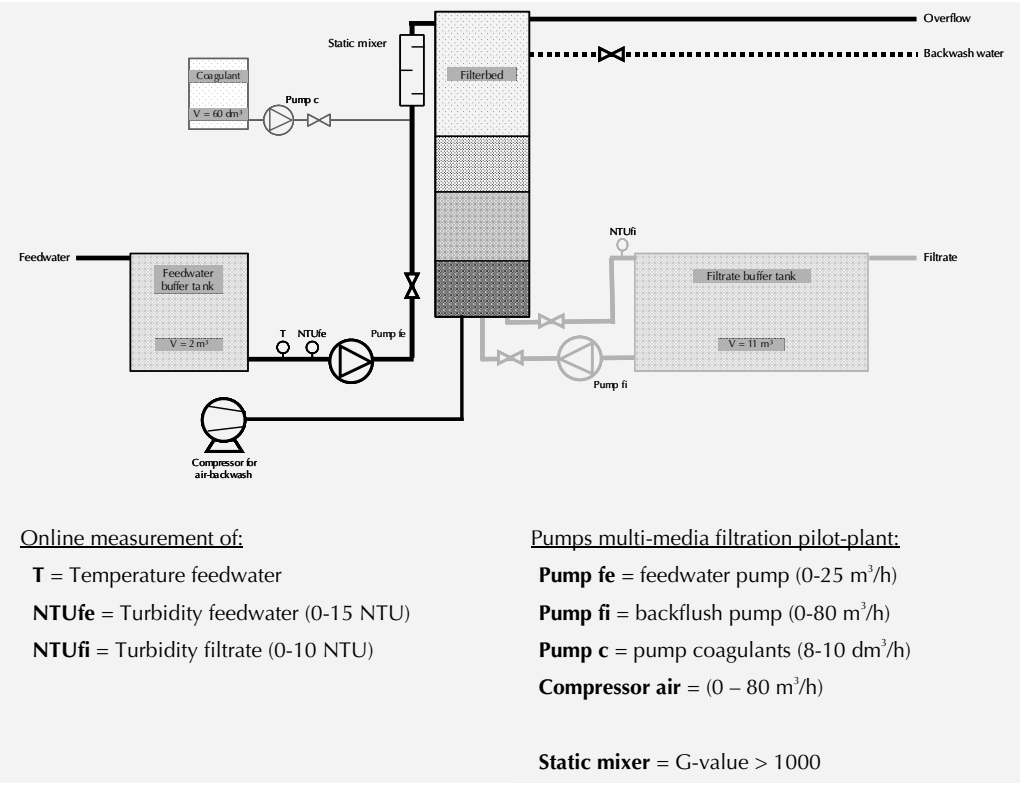


Figure 2-A.2 Schematic drawing of multi-media filter used in pilot-scale studies

2-B WWTP Berkel, Hoek van Holland, Vlaardingen and Zaandam-Oost

2-B.1 Wastewater treatment plant Berkel

The wastewater treatment plant in Berkel is designed for 27,000 population equivalents. The wastewater is treated in an oxidation ditch (see figure 2-B.1) and after final sedimentation the effluent is discharged into the surface water. In table 2-B.1 some quality parameters are presented.

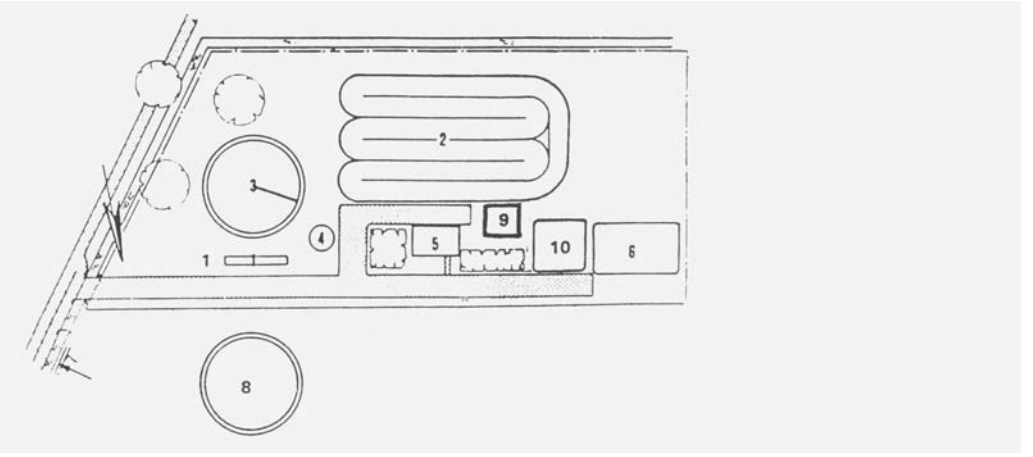


Figure 2-B.1 Schematic drawing of wwtp Berkel with the following treatment steps: 1 – grid, 2 – oxidation ditch, 3 – clarification tank 1, 4 – sludge thickening, 8 – clarification tank 2, other numbers correspond to non water treatment steps

Table 2-B.1 Effluent water quality at wwtp Berkel in 2002

parameter		Average concentration
COD	mg/l	38.8
BOD	mg/l	2.9
Suspended solids	mg SS/l	5.5
P-total	mg P/l	1.6
N-total	mg N/l	7.3

2-B.2 Wastewater treatment plant Hoek van Holland

The wastewater treatment plant in Hoek van Holland is called 'De Nieuwe Waterweg' and is designed for 110,000 population equivalents. The wastewater is treated in an oxidative-biological treatment facility (see figure 2-B.2). The wastewater comes from households, DSM-resins and a company treating garden waste (Stiching Verwerking Tuinafval Westland). In table 2-B.2 some quality parameters are presented.

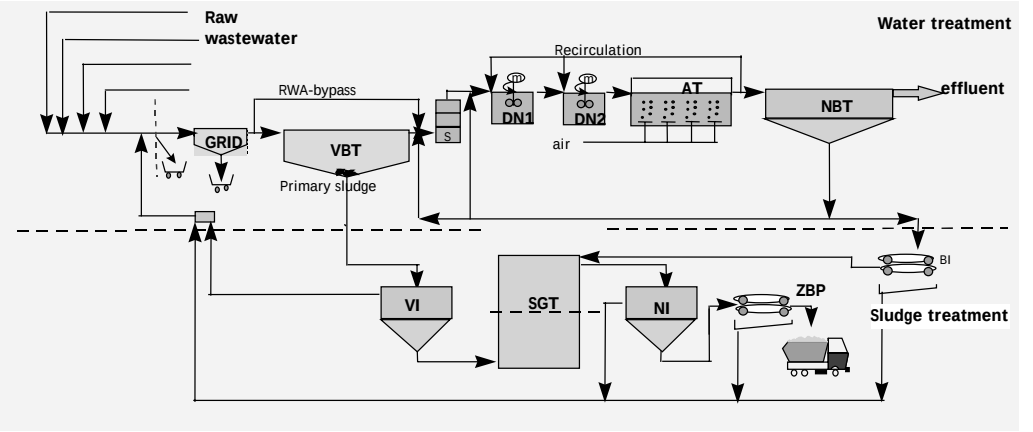


Figure 2-B.2 Schematic drawing of wwtp Hoek van Holland: VBT – pre-sedimentation, DN1,2 – denitrification tank, AT – activated sludge tank, NBT – clarification, all other abbreviations correspond to the sludge treatment.

Table 2-B.2 Effluent water quality at wwtp Hoek van Holland

parameter		Average concentration
COD	mg/l	63.6
BOD	mg/l	4.9
Suspended solids	mg SS/l	16.2
P-total	mg P/l	2.9
N-total	mg N/l	11.9

2-B.3 Wastewater treatment plant Vlaardingen

The wastewater treatment plant in Hoek van Holland is called 'De Groote Lucht' and is designed for 285,000 population equivalents. The wastewater is treated in a three step biological treatment facility with post-filtration for denitrification (see figure 2-B.3). This continuous filter is rather big and unique in the Netherlands. In table 2-B.3 some quality parameters are presented.

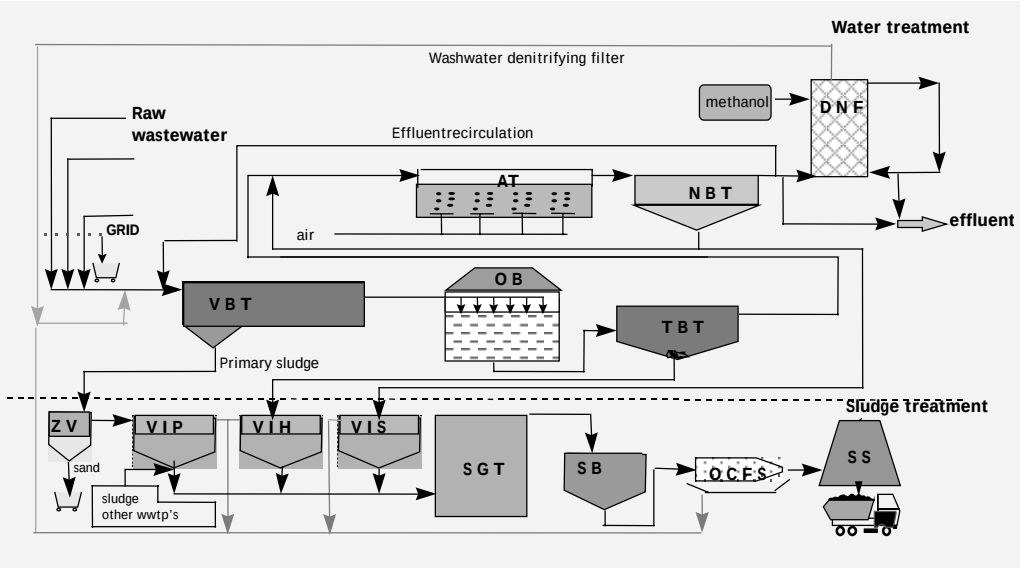


Figure 2-B.3 Schematic drawing of wwtp Vlaardingen: VBT – pre-sedimentation, OB – oxidation ditch, TBT – sedimentation, AT – activated sludge tank, NBT – clarification, DNF – denitrifying filter, all other abbreviations correspond to the sludge treatment.

Table 2-B.3 Effluent water quality at wwtp Vlaardingen

parameter		Average concentration
COD	mg/l	39.7
BOD	mg/l	3.6
Suspended solids	mg SS/l	7.7
P-total	mg P/l	1.4
N-total	mg N/l	7.1

2-B.4 Wastewater treatment plant Zaandam-Oost

The wastewater treatment plant in Zaandam-Oost is designed for 150,000 population equivalents. The wastewater is treated in two oxidation ditches (see figure 2-B.4). In table 2-B.4 some quality parameters are presented.

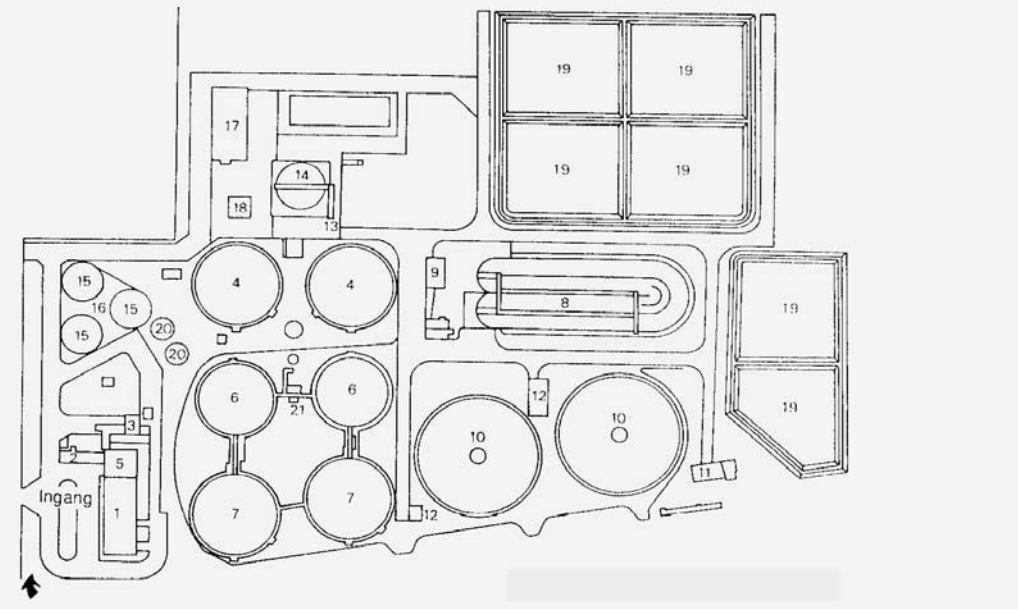


Figure 2-B.4 Schematic drawing of wwtp Zaandam-Oost: 3 – grid, 4 – pre-sedimentation, 6 – oxidation beds, 7 – sedimentation, 8 – aeration tanks, 10 – clarification, other numbers correspond sludge treatment or buildings.

Table 2-B.4 Effluent water quality at wwtp Zaandam-Oost

parameter		Average concentration
COD	mg/l	52.4
BOD	mg/l	6.4
Suspended solids	mg SS/l	11.4
P-total	mg P/l	3.7
N-total	mg N/l	23.9

2-C WWTP Ede, WWTP Kaffeberg, WWTP Tilburg-Noord

2-C.1 WWTP Ede

The capacity of wwtp Ede is 300.000 population equivalents. The wwtp is designed according to the Biodehipho system that combines biological nitrogen and phosphorus removal, by leaving the first compartment (selector) anaerobic for development of phosphate-bacteria. The following compartments are alternately aerated (aerobic) an not aerated (anoxic). By this are organic compounds degraded and is nitrogen removed and phosphorus bound to the activated sludge. The last compartment is aerated to ensure a good phosphorus binding. A schematic drawing of the wwtp is shown in figure 2-C.1.The numbers correspond to unit processes in the wwtp. Relating to the water stream: 1 shows the inlet of the water coming from the municipalities. 2. storage basin; 3. screen; 4. composting filters; 5. primary settling; 6. pump; 7. selector; 8. anaerobic tanks; 9. aeration tanks; 10. post-aeration; 11. post settling; 12. sludge recovery pumps; 13. discharge to surface water. Relating to the sludge stream: 14. grid removel; 15. primary thickening; 16. digestion; 17. heating; 18. buffers; 19. centrifuges; 20. storage; 21. gas holder; 22. energy building; 23 – 26 other buildings.

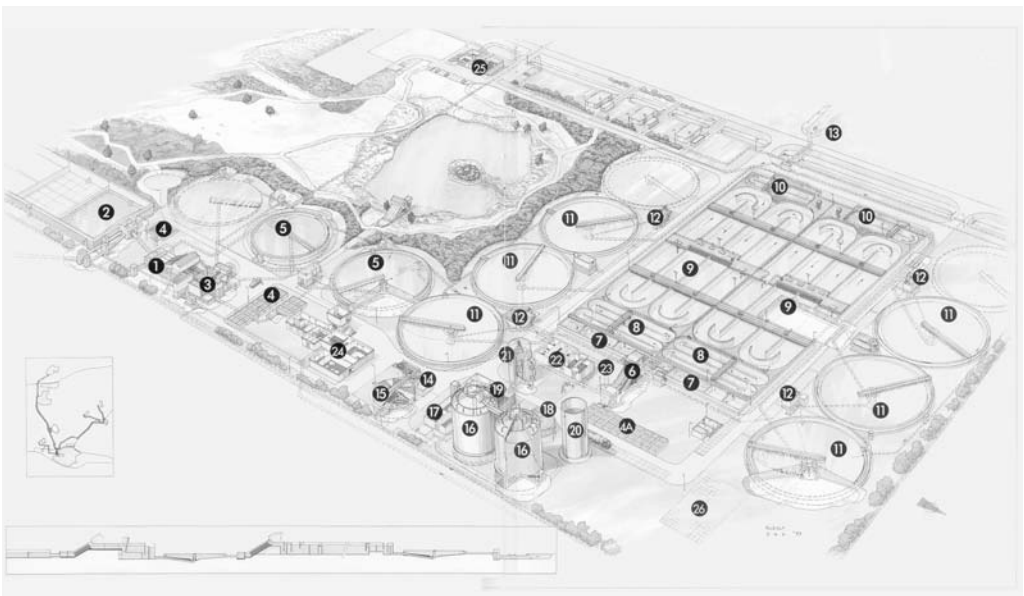


Figure 2-C.1 Treatment scheme of wwtp Ede, see text for the explanation of the numbers

2-C.2 WWTP Kaffeberg

The capacity of wwtp Kaffeberg is 150,000 population equivalents. The wastewater is treated biologically. A schematic drawing of the wwtp is shown in figure 2-C.2.The numbers correspond to unit processes in the wwtp. 1. inlet; 2. screen; 3. grid removal; 4. oxidation ditches; 5. aeration tanks; 6. clarification; 7. sludge recovery; 8. discharge of effluent to surface water; 9. operation building.

Figure 2-C.2 Treatment scheme of wwtp Kaffeberg; see text for the explanation of the numbers

2-C.3 WWTP Tilburg-Noord

The capacity of wwtp Tilburg-Noord is 445,000 population equivalents. The wastewater is treated biologically. A schematic drawing of the wwtp is shown in figure 2-C.3. The abbreviations correspond to: DET – denitrication tank, DNT – facultative tank, NBT – clarification tank, NIT – nitrification tank, SEL – selector, VBT – pre-sedimentation tank, ZV – grid, other abbreviations correspond sludge treatment or buildings

An important aspect of this wwtp is the polishing step of the effluent in a pond system. Effluent is discharged in a pond system with a residence time of one day (maximum 3 to 4 days). The pond system is not drawn on the figure below, but on the right bottom it can be found.

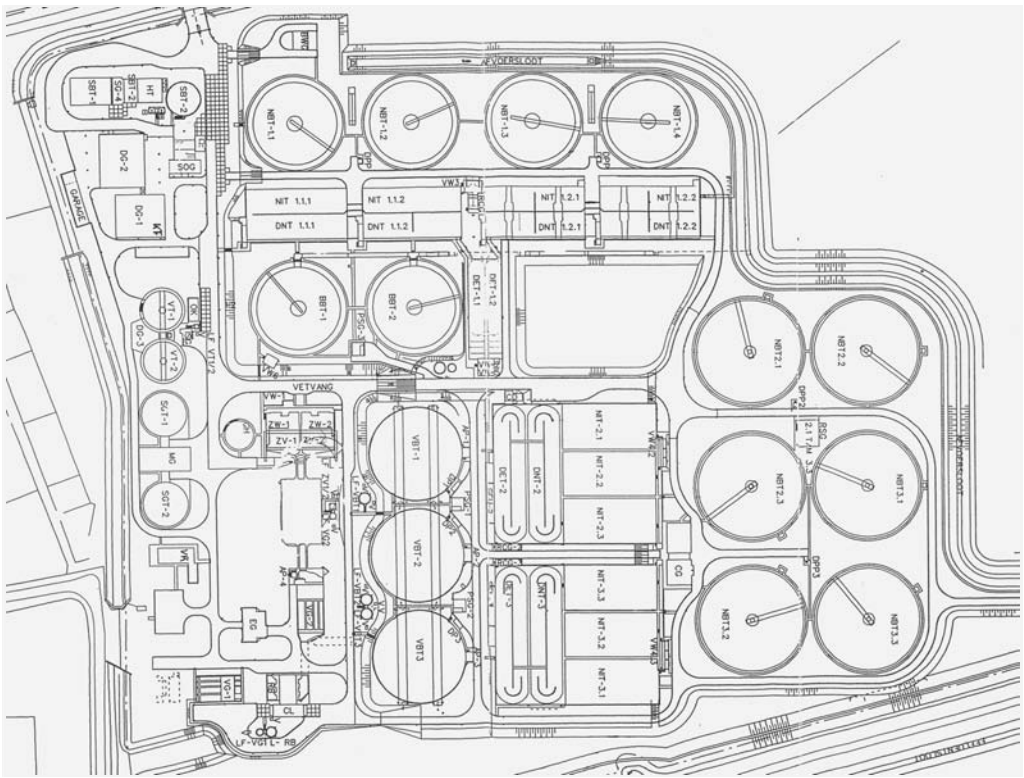


Figure 2-C.3 Treatment scheme of wwtp Tilburg-Noord; see text for the explanation of the numbers

2-D Membrane specifications

2-D-1 STORK WF 4285

STORK

Stork Friesland B.V.

PROVISIONAL

OPEN ULTRAFILTRATION MEMBRANE

Technical data

FX 4285, 4385, 4485

General description
These membranes are highly efficient hydrophilic tubular polyvinylidene fluoride membranes, cast on a polyester carrier, for use in a variety of filtration processes in food and non-food applications.
The hydrophilic properties of these membranes ensure a high performance and a good antifouling behaviour.
The FX-series compact membranes are only available as membrane filtration elements in a wide range of membrane areas.
The elements can be backflushed for efficient membrane cleaning.

Applications
Some of the major applications are:

Water treatment	- pretreatment of process and drinking water - recovery of back wash water
Biotechnology	- enzymes concentration/purification - concentration of yeast and bacteria slurries
Chemistry	- concentration of metal hydroxides and polymer suspensions - treatment of waste water
Oil emulsion	- concentration of oil-in-water emulsions

Physical and chemical properties

Membrane material	polyvinylidene fluoride
Structure	asymmetric/microporous
Membrane carrier	composite polyester fabric
Geometry	tubular

Performance data

parameter	unit	FX 4285	FX 4385	FX 4485	remarks
Initial flux	l/m².h	> 700	> 1000	> 1000	distilled water at 25°C and 100 kPa
Transmembrane pressure	kPa	-100..+300	-100..+300	-100..+300	
Mean pore size	nm	20	30	40	
pH		2 - 10	2 - 10	2 - 10	at 25°C
Chlorine exposure	ppm.h	250000	250000	250000	at 25°C
Temperature	°C	1 - 70	1 - 70	1 - 70	pH 7 and 100 kPa
Hydraulic diameter	mm	5.2	5.2	5.2	

Operation of membranes at any combination of maximum limits of pH, concentration, pressure or temperature, during cleaning or production, will severely influence the membrane life.

9750-FX

Solvent resistance
Since the resistance of the membrane to solvents strongly depends on the actual process conditions, the indications given below should only be considered as guidelines.

Acids	****
Bases	***
Organic esters, ketones, ethers	*
Aliphatic alcohols	****
Aliphatic hydrocarbons	****
Aromatic hydrocarbons	****
Halogenated hydrocarbons	****
Polar organic solvents	*
Oils	****
Formaldehyde	****

* = not resistant **** = 100% resistant

Cleaning
Depending on the nature of the feed solution the following cleaning agents can be chosen:

Chemical		
	: Chlorine	(500 ppm max.)
	: Hydrogen peroxide	(1000 ppm max.)
	: Sodium hydroxide	(pH 10.5 max.)
	: Nitric acid	(pH 1 min.)
	: Phosphoric acid	(pH 1 min.)
	: EDTA	(pH 10.5 max.)
	: Sodium tri-phosphate	
	: Citric acid	
	: Enzymatic compounds	

It is recommended to keep the pH between 1 and 10 and not to exceed a temperature of 35°C during cleaning and/or disinfection.
If those standard cleaning techniques fail to remove the foulants, more concentrated cleaning solutions can be tried. Please contact Stork Friesland for recommendations. It has to be stressed, however, that no warranty can be given on the efficiency of any cleaning nor on the membrane performance after such cleaning attempts.
Before using new membranes and after long term storage, we advise to do a cleaning and disinfection run in order to remove residues of conservation agents.
After cleaning we advise to discharge at least 25 liter of permeate per m² of membrane area.

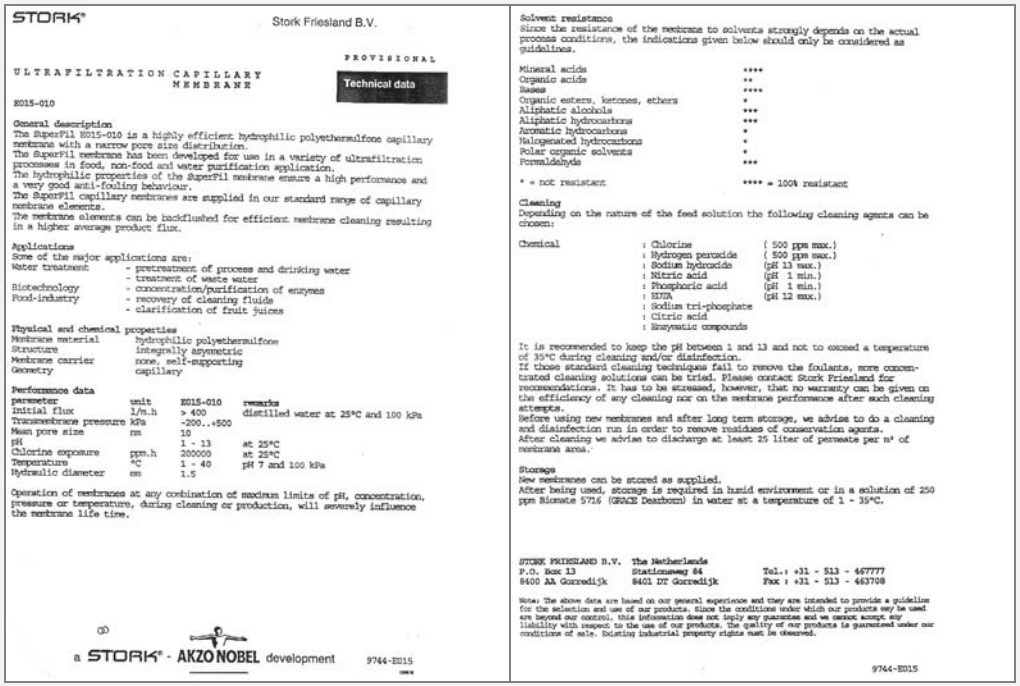
Storage
New membranes can be stored as supplied.
After being used, storage is required in humid environment or in a solution of 250 ppm Biocate 5716 (GRACE Dearborn) in water at a temperature of 1 - 35°C.

General
Non-standard dimensions (diameters, lengths) are available upon request.

STORK FRIESLAND B.V. **The Netherlands**
P.O. Box 13 **Stationsweg 84** **Tel.: +31 - 513 - 467777**
8400 AA Gorredijk **8401 DT Gorredijk** **Fax : +31 - 513 - 463708**

9750-FX

2-D-2 STORK E015-010



STORK®

Stork Friesland B.V.

PROVISIONAL

ULTRAFILTRATION CAPILLARY MEMBRANE

Technical data

E015-010

General description
The SuperPil E015-010 is a highly efficient hydrophilic polyethersulfone capillary membrane with a narrow pore size distribution.
The SuperPil membrane has been developed for use in a variety of ultrafiltration processes in food, non-food and water purification application.
The hydrophilic properties of the SuperPil membrane ensure a high performance and a very good anti-fouling behaviour.
The SuperPil capillary membranes are supplied in our standard range of capillary membrane elements.
The membrane elements can be backflushed for efficient membrane cleaning resulting in a higher average product flux.

Applications
Some of the major applications are:

Water treatment	- pretreatment of process and drinking water
	- treatment of waste water
Biotechnology	- concentration/purification of enzymes
Food-industry	- recovery of cleaning fluids
	- clarification of fruit juices

Physical and chemical properties

Membrane material	hydrophilic polyethersulfone
Structure	integrally asymmetric
Membrane carrier	none, self-supporting
Geometry	capillary

Performance data

parameter	unit	E015-010	remarks
Initial flux	l/m.h	> 400	distilled water at 25°C and 100 kPa
Transmembrane pressure	kPa	-200..+500	
Mean pore size	nm	10	
pH		1 - 13	at 25°C
Chlorine exposure	ppm.h	200000	at 25°C
Temperature	°C	1 - 40	pH 7 and 100 kPa
Hydraulic diameter	mm	1.5	

Operation of membranes at any combination of maximum limits of pH, concentration, pressure or temperature, during cleaning or production, will severely influence the membrane life time.

a STORK® - AKZO NOBEL development

9744-E015

Solvent resistance
Since the resistance of the membrane to solvents strongly depends on the actual process conditions, the indications given below should only be considered as guidelines.

Mineral acids	****
Organic acids	**
Bases	****
Organic esters, ketones, ethers	*
Aliphatic alcohols	***
Aliphatic hydrocarbons	***
Aromatic hydrocarbons	*
Halogenated hydrocarbons	*
Polar organic solvents	*
Formaldehyde	***

* = not resistant **** = 100% resistant

Cleaning
Depending on the nature of the feed solution the following cleaning agents can be chosen:

Chemical		
	: Chlorine	(500 ppm max.)
	: Hydrogen peroxide	(500 ppm max.)
	: Sodium hydroxide	(pH 13 max.)
	: Nitric acid	(pH 1 min.)
	: Phosphoric acid	(pH 1 min.)
	: EDTA	(pH 12 max.)
	: Sodium tri-phosphate	
	: Citric acid	
	: Enzymatic compounds	

It is recommended to keep the pH between 1 and 13 and not to exceed a temperature of 35°C during cleaning and/or disinfection.
If these standard cleaning techniques fail to remove the foulants, more concentrated cleaning solutions can be tried. Please contact Stork Friesland for recommendations. It has to be stressed, however, that no warranty can be given on the efficiency of any cleaning nor on the membrane performance after such cleaning attempts.
Before using new membranes and after long term storage, we advise to do a cleaning and disinfection run in order to remove residues of conservation agents.
After cleaning we advise to discharge at least 25 liter of permeate per m² of membrane area.

Storage
New membranes can be stored as supplied.
After being used, storage is required in humid environment or in a solution of 250 ppm Bicreate 5716 (GRACE Dearborn) in water at a temperature of 1 - 35°C.

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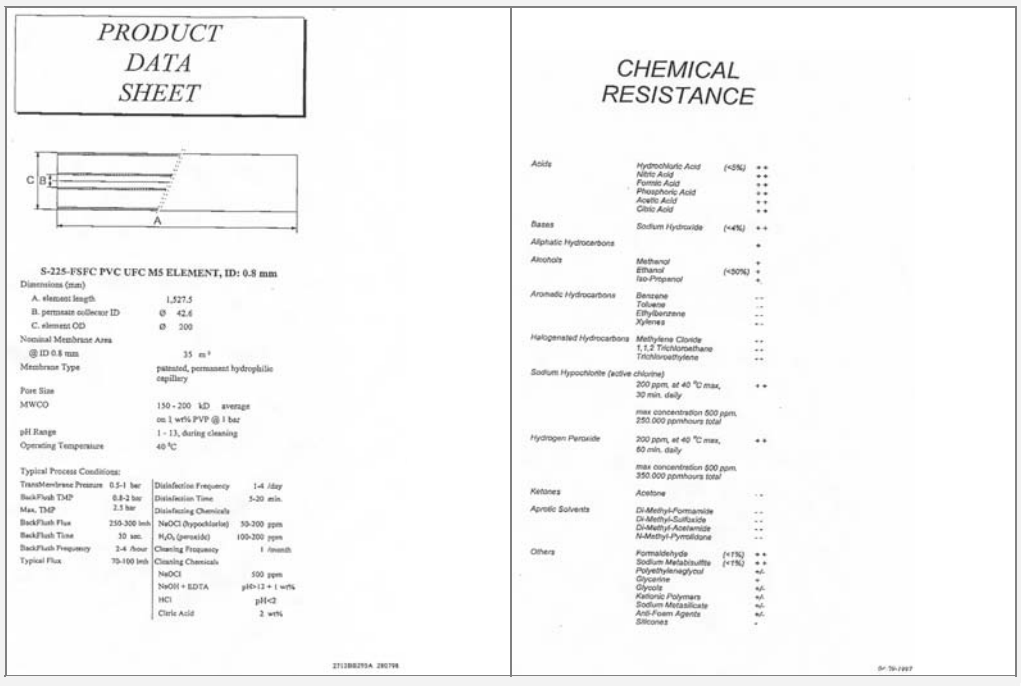
9744-E015

2-D-3 X-flow S-225-FSFC PVC UFM M5



PRODUCT DATA SHEET

S-225-FSFC PVC UFC M5 ELEMENT, ID: 0.8 mm

Dimensions (mm)

A. element length	1,527.5
B. permeate collector ID	Ø 42.6
C. element OD	Ø 200
Nominal Membrane Area @ ID 0.8 mm	35 m²
Membrane Type	patented, permanent hydrophilic capillary
Pore Size	
MWCO	150 - 200 kD average on 1 wt% PVP @ 1 bar
pH Range	1 - 13, during cleaning
Operating Temperature	40 °C

Typical Process Conditions:

TransMembrane Pressure	0.5-1 bar	Disinfection Frequency	1-4 /day
BackFlush TMP	0.8-2 bar	Disinfection Time	5-20 min.
Max. TMP	2.5 bar	Disinfecting Chemicals	
BackFlush Flux	250-300 lmh	NaOCl (hypochlorite)	50-200 ppm
BackFlush Time	30 sec.	H_2O_2 (peroxide)	100-200 ppm
BackFlush Frequency	2-4 /hour	Cleaning Frequency	1 /month
Typical Flux	70-100 lmh	Cleaning Chemicals	
		NaOCl	500 ppm
		NaOH + EDTA	pH>12 + 1 wt%
		HCl	pH<2
		Citric Acid	2 wt%

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CHEMICAL RESISTANCE

Acids	Hydrochloric Acid	(<5%)	+ +
	Nitric Acid		+ +
	Formic Acid		+ +
	Phosphoric Acid		+ +
	Acetic Acid		+ +
	Citric Acid		+ +
Bases	Sodium Hydroxide	(<4%)	+ +
Aliphatic Hydrocarbons			+
Alcohols	Methanol		+
	Ethanol	(<50%)	+
	Iso-Propanol		+.
Aromatic Hydrocarbons	Benzene		- -
	Toluene		- -
	Ethylbenzene		- -
	Xylenes		- -
Halogenated Hydrocarbons	Methylene Cloride		- -
	1,1,2 Trichloroethane		- -
	Trichloroethylene		- -
Sodium Hypochlorite (active chlorine)	200 ppm, at 40 °C max, 30 min. daily		+ +
	max concentration 500 ppm, 250.000 ppmhours total		
Hydrogen Peroxide	200 ppm, at 40 °C max, 60 min. daily		+ +
	max concentration 500 ppm, 350.000 ppmhours total		
Ketones	Acetone		- -
Aprotic Solvents	Di-Methyl-Formamide		- -
	Di-Methyl-Sulfoxide		- -
	Di-Methyl-Acetamide		- -
	N-Methyl-Pyrrolidone		- -
Others	Formaldehyde	(<1%)	+ +
	Sodium Metabisulfite	(<1%)	+ +
	Polyethyleneglycol		+/-
	Glycerine		+
	Glycols		+/-
	Kationic Polymers		+/-
	Sodium Metasilicate		+/-
	Anti-Foam Agents		+/-
	Silicones		-

Appendix 3 – Filterability and Reversibility

3-A Resistance increase in experiments on Filterability and Reversibility

Below the results of all experiments described in Chapter 3 are presented in tables and figures, showing the resistance of the clean membrane, the resistance increase due to filtration of feedwater, (a) measured with the feedwater ($\Delta R_{fo,fwf}$) and (b) with permeate ($\Delta R_{fo,cwf}$). The last series show the fouling layer resistance right after cleaning of the membrane with a *forward flush* ($\Delta R_{fo,ff}$), a *forward flush with air – AirFlush®* ($\Delta R_{fo,ff+a}$) and a *back flush* ($\Delta R_{fo,bf}$). Finally the resistance of the fouling layer after a *chemical flush* ($\Delta R_{fo,cf}$) and a *thorough chemical cleaning* ($\Delta R_{fo,tc}$) are shown. The fouling layer resistance was calculated as the total membrane resistance minus the initial membrane resistance of the clean membrane.

3-A-1 Experiment 1-1: effluent of wwtp Ede with PVDF (5.2 mm) membranes

Table 3-A.1 Resistance increase found during experiments on dead-end ultrafiltration of effluent (wwtp Ede; PVDF (5.2 mm) membranes), date 5/'98 to 6/'98

Exp.	Feed	J (l/m².h)	V_{tot}/A_m (l/m²)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})
1-1	Effluent[a]	35	440	0.9	8.1	5.9	5.9	3.4	0.8
	Effluent[a]	70	280	0.9	3.9	3.3	3.6	2.6	0.9
	Effluent	100	136	1.0	2.1	2.2	1.5	1.1	0.4

[a] Based on one experiment

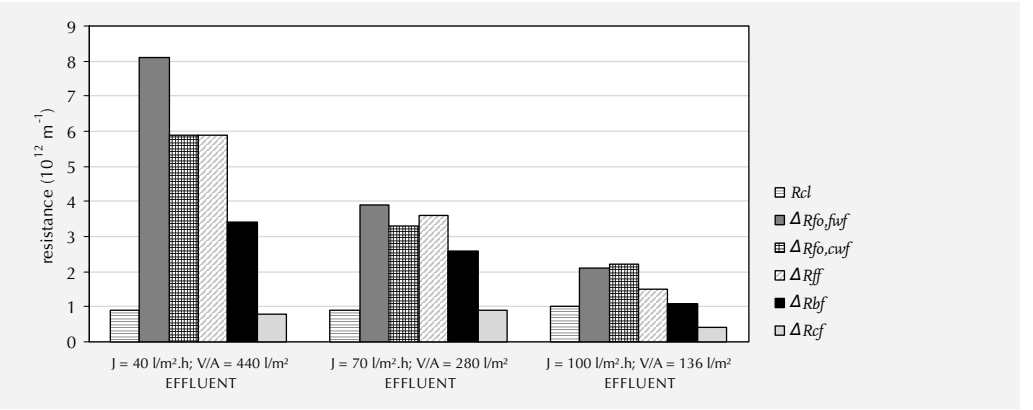


Figure 3-A.1 Graphical presentation of table 3-A.1; feedwater was effluent of wwtp Ede and membrane was PVDF 5.2 mm; measured for various fluxes

3-A-2 Experiment 2-1 and 2-2: effluent and coagulated effluent of wwtp Kaffeberg with PES/PVP (1.5 mm) membranes

Table 3-A.2 Resistance increase found during experiments on dead-end ultrafiltration of effluent and coagulated effluent (wwtp Kaffeberg; PES/PVP (1.5 mm) membranes), date 10/'98 to 1/'99 (effluent) and 2/'99 (coagulated effluent)

Exp.	Feed	J ($l/m^2.h$)	V_{tot}/A_m (l/m^2)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{ff+a} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})
2-1	Effluent	80	36	0.9	1.9	1.2	0.6	0.4	0.3	0.2
2-2	Effluent+[a]	80	59	0.9	1.6	1.1	1.0	0.8	0.1	0.2

[a] Based on one experiment

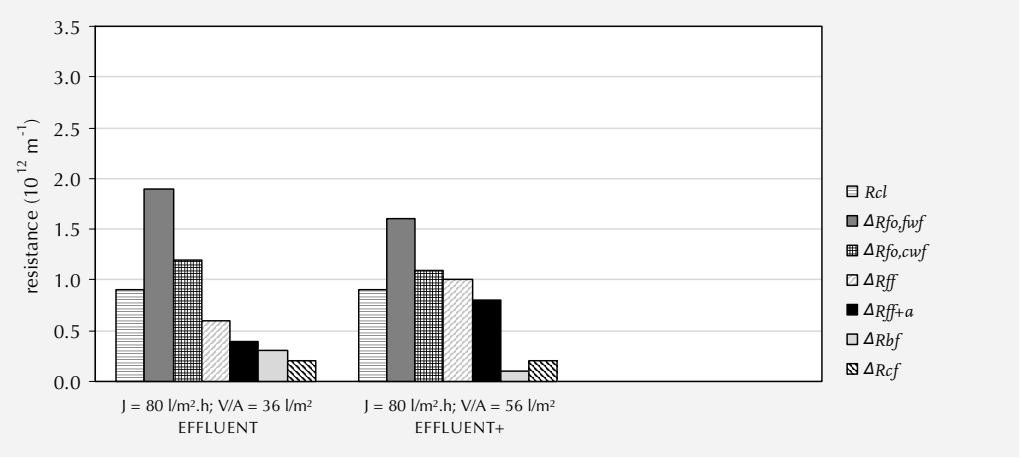


Figure 3-A.2 Graphical presentation of table 3-A.2; feedwater was effluent and coagulated effluent of wwtp Kaffeberg and membrane was PES/PVP 1.5 mm; measured for one flux

3-A-3 Experiment 3-1: effluent of wwtp Ede with PES/PVP (0.8 mm) membranes; filtration ends at standard TMP

Table 3-A.3 Resistance increase found during experiments on dead-end ultrafiltration of effluent (wwtp Ede; PES/PVP (0.8 mm) membranes), date 3/5/'99 to 11/5/'99

Exp.	Feed	J (l/m^2.h)	V_{tot}/A_m (l/m^2)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})	ΔR_{tc} (10^{12} m^{-1})
3-1	Effluent	40	160	1.2	3.1	2.5	2.1	0.3	0.2	0.0
	Effluent	70	70	1.0	2.7	2.0	1.8	0.8	0.5	0.1
	Effluent	100	96	0.9	1.7	1.0	0.8	0.1	0.1	0.0

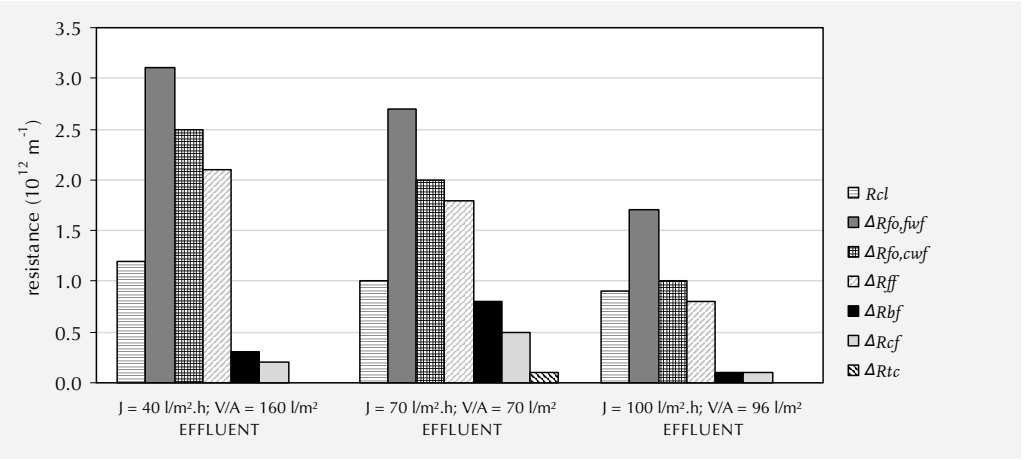


Figure 3-A.3 Graphical presentation of table 3-A.3; feedwater was effluent of wwtp Ede and membrane was PES/PVP 0.8 mm; measured at various fluxes, filtration was stopped at the same TMP

3-A-4 Experiment 3-2: effluent of wwtp Ede with PES/PVP (0.8 mm) membranes; filtration ends at standard volume

Table 3-A.4 Resistance increase found during experiments on dead-end ultrafiltration of effluent (wwtp Ede; PES/PVP (0.8 mm) membranes), date 10/6/'99 to 15/6/'99

Exp.	Feed	J (l/m².h)	V_{tot}/A_m (l/m²)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})	ΔR_{tc} (10^{12} m^{-1})
3-2	Effluent	40	86	1.1	0.8	0.8	0.7	0.1	0.0	0.0
	Effluent	70	86	1.2	0.9	0.8	0.7	0.2	0.2	0.0
	Effluent	100	86	1.0	1.0	0.7	0.7	0.2	0.1	0.1

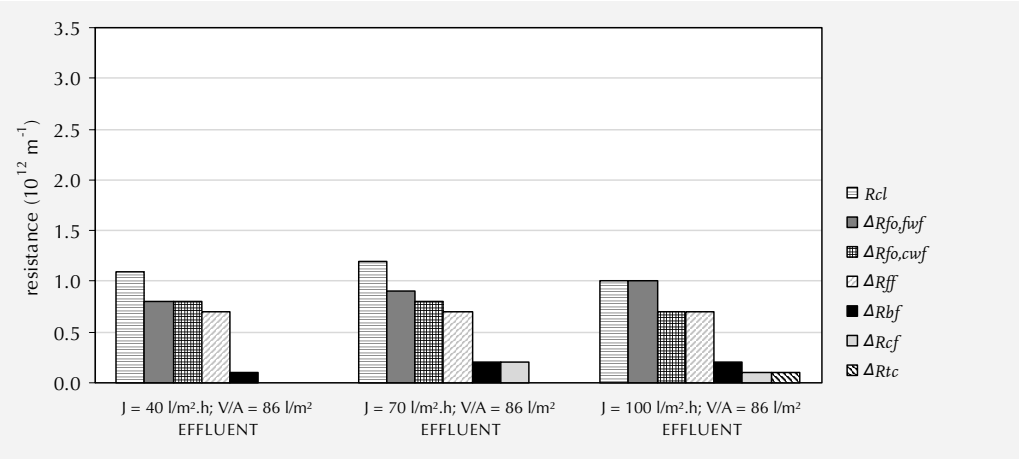


Figure 3-A.4 Graphical presentation of table 3-A.4; feedwater was effluent of wwtp Ede and membrane was PES/PVP 0.8 mm; measured at various fluxes, filtration was stopped at same filtered volume (V_{tot}/A_m)

3-A-5 Experiment 3-3: coagulated effluent of wwtp Ede with PES/PVP (0.8 mm) membranes

Table 3-A.5 Resistance increase found during experiments on dead-end ultrafiltration of coagulated effluent (wwtp Ede; PES/PVP (0.8 mm) membranes), date 1/6/'99 to 9/6/'99

Exp.	Feed	J (l/m².h)	V_{tot}/A_m (l/m²)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})	ΔR_{tc} (10^{12} m^{-1})
3-3	Effluent+	40	86	0.9	0.9	0.8	0.7	0.1	0.0	0.1
	Effluent+	60	86	0.9	1.0	0.9	0.8	0.0	0.0	-0.1

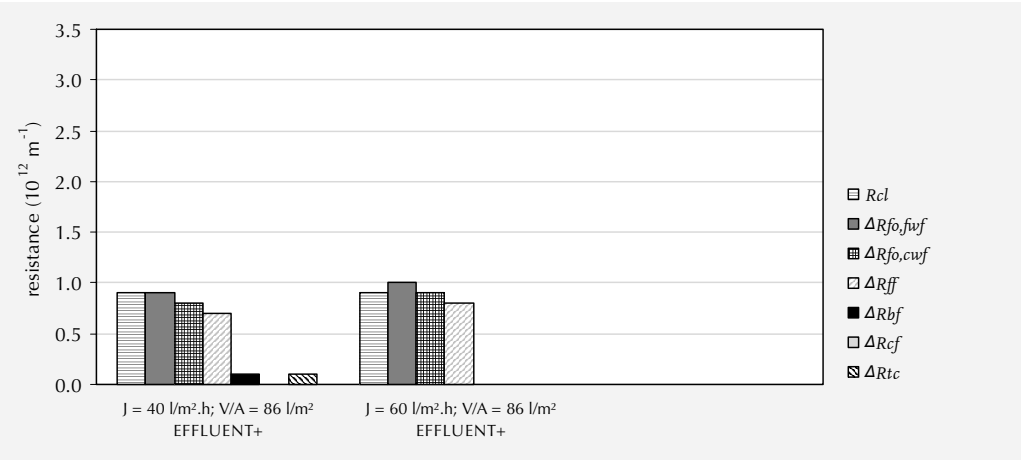


Figure 3-A.5 Graphical presentation of table 3-A.5; feedwater was coagulated effluent of wwtp Ede and membrane was PES/PVP 0.8 mm; measured at various fluxes

3-A-6 Experiment 3-4: filtrate of wwtp Ede with PES/PVP (0.8 mm) membranes

Table 3-A.6 Resistance increase found during experiments on dead-end ultrafiltration of filtrate (wwtp Ede; PES/PVP (0.8 mm) membranes), date 20/4/'99 to 27/4/'99

Exp.	Feed	J (l/m².h)	V_{tot}/A_m (l/m²)	R_{cl} (10^{12} m^{-1})	$\Delta R_{fo,fwf}$ (10^{12} m^{-1})	$\Delta R_{fo,cwf}$ (10^{12} m^{-1})	ΔR_{ff} (10^{12} m^{-1})	ΔR_{bf} (10^{12} m^{-1})	ΔR_{cf} (10^{12} m^{-1})	ΔR_{tc} (10^{12} m^{-1})
3-4	Effluent	40	180	1.2	3.1	2.5	2.1	0.3	0.2	0.0
	Effluent	70	210	1.0	2.7	2.0	1.8	0.8	0.5	0.1
	Effluent	100	142	0.9	1.7	1.0	0.8	0.1	0.1	0.0

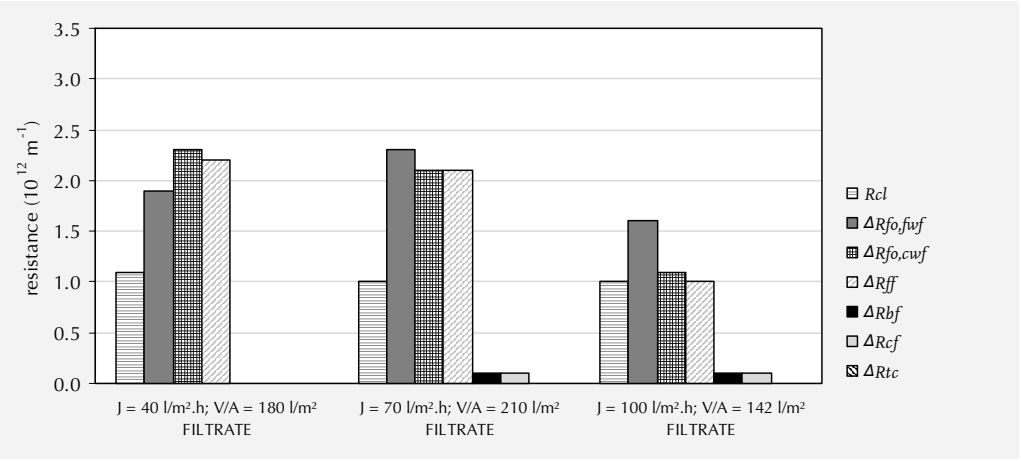


Figure 3-A.6 Graphical presentation of table 3-A.6; feedwater was filtrate of wwtp Ede and membrane was PES/PVP 0.8 mm; measured at various fluxes

Appendix 4- Specific Ultrafiltration Resistance

4-A Derivation of formula for the Specific Ultrafiltration Resistance (*SUR*)

1 – Darcy's law and dynamic viscosity

$$J = \frac{\Delta P}{\eta_T \cdot R_{tot}} \qquad \text{(eq. 4-A.1)}$$

where J = flux ($m^3/m^2.s$)
ΔP = pressure difference (N/m^2, Pa),
η_T = dynamic viscosity (N.s/m^2, Pa.s), related to feedwater temperature T (°C)
R_{tot} = total resistance over membrane (1/m)

$$\eta_T = \frac{497 \cdot 10^{-3}}{(T + 42.5)^{1.5}} \qquad \text{(eq. 4-A.2)}$$

2 – Flux definition (filtered volume divided by filtration time and membrane area)

$$J = \frac{dV}{dt} \cdot \frac{1}{A_{memb}} \qquad \text{(eq. 4-A.3)} \qquad \Rightarrow \qquad \frac{dV}{dt} \cdot \frac{1}{A_{memb}} = \frac{\Delta P}{\eta_T \cdot R_{tot}} \qquad \text{(eq. 4-A.3a)}$$

where V = filtered volume (m^3)
t = time (s)
A_{memb} = membrane area (m^2)

3 – Total membrane resistance R_{total} (sum of resistances)

$$R_{tot} = R_m + R_{cake} \qquad \text{(eq. 4-A.4)} \qquad \Rightarrow \qquad \frac{dV}{dt} \cdot \frac{1}{A_{memb}} = \frac{\Delta P}{\eta_T \cdot (R_m + R_{cake})} \qquad \text{(eq. 4-A.4a)}$$

$$\Rightarrow \qquad R_m + R_{cake} = \frac{\Delta P}{\eta_T} \cdot A_{memb} \cdot \frac{dt}{dV} \qquad \text{(eq. 4-A.4b)}$$

where R_m = membrane resistance (m^{-1})
R_{cake} = cake resistance (m^{-1})

4 – Cake resistance (product of specific cake resistance and cake thickness)

$$R_{cake} = \alpha_{av} \cdot c_v \cdot \frac{V}{A_{memb}} \qquad \text{(eq. 4-A.5)} \Rightarrow \qquad \frac{\Delta P}{\eta_T} \cdot A_{memb} \cdot \frac{dt}{dV} = R_m + \alpha_{av} \cdot c_v \cdot \frac{V}{A_{memb}} \qquad \text{(eq. 4-A.5a)}$$

$$\Rightarrow \qquad \frac{dt}{dV} = \frac{\eta_T \cdot R_m}{\Delta P \cdot A_{memb}} + \frac{\eta_T \cdot \alpha_{av} \cdot c_v}{\Delta P \cdot A_{memb}^{\,2}} \cdot V \qquad \text{(eq. 4-A.5b)}$$

where α_{av} = average specific cake resistance (m/kg)
c_v = concentration of solids in feedwater (kg/m^3)

5 – Integrating equation (4-A.5b) at constant pressure (ΔP) and constant temperature ($t_o = 0$; $t_t = t$; $V_o = 0$; $V_t = V$) gives equation (4-A.6)

$$t = \frac{\eta_T \cdot R_m}{\Delta P \cdot A_{memb}} \cdot V + \frac{\eta_T \cdot \alpha_{av} \cdot c_v}{\Delta P \cdot A_{memb}^{\ 2}} \cdot \frac{1}{2} \cdot V^2 \qquad \text{(eq. 4-A.6)}$$

$$\Rightarrow \qquad \frac{t}{V} = \frac{\eta_T \cdot R_m}{\Delta P \cdot A_{memb}} + \frac{\eta_T \cdot \alpha_{av} \cdot c_v}{2 \cdot \Delta P \cdot A_{memb}^{\ 2}} \cdot V \qquad \text{(eq. 4-A.6a)}$$

6 – Product of average specific cake resistance and solids concentration: Specific Ultrafiltration Resistance (SUR)

$$\frac{d(\frac{t}{V})}{d(V)} = \frac{\eta_T \cdot \alpha_{av} \cdot c_v}{2 \cdot \Delta P \cdot A_{memb}^{\ 2}} \qquad \text{(eq. 4-A.7a)}$$

$$\Rightarrow \qquad SUR = \alpha_{av} \cdot c_v = \frac{d(\frac{t}{V})}{d(V)} \cdot \frac{2 \cdot \Delta P \cdot A_{memb}^{\ 2}}{\eta_T} \qquad \text{(eq. 4-A.7)}$$

where SUR = Specific Ultrafiltration Resistance, resistance per meter filtrated feedwater (m^{-2})

7 – Modified Fouling Index (MFI) according to Schippers

Schippers (1982) defined the Modified Fouling Index for ΔP is 2.0 bar (200 kPa) and a temperature of 20 °C as the slope of the $t/V - V$ curve:

$$MFI = \frac{d(\frac{t}{V})}{d(V)} = \frac{\eta_T \cdot \alpha_{av} \cdot c_v}{2 \cdot \Delta P \cdot A_{memb}^{\ 2}} \qquad \text{(eq. 4-A.8)}$$

When the MFI is measured with different ΔP and/or temperature, the MFI can be corrected to the standard conditions as done in equation (A.8). The same is done in the MFI-UF described by Boerlage (2003).

$$MFI = MFI\text{-}UF = \frac{d(\frac{t}{V})}{d(V)} \cdot \frac{\eta_{20}}{\eta_T} \cdot \frac{\Delta P}{\Delta P_{200\ kPa}} \cdot \left(\frac{A_{memb}}{A_{0.45\ \mu m}} \right)^2 = \frac{\eta_{20} \cdot \alpha_{av} \cdot c_v}{2 \cdot \Delta P_{200\ kPa} \cdot A_{0.45\ \mu m}^{\ 2}} \qquad \text{(eq. 4-A.9)}$$

where
MFI = Modified Fouling Index (s/l^2)
$MFI\text{-}UF$ = Modified Fouling Index using Ultrafiltration membranes (s/l^2)
η_{20} = dynamic viscosity ($N.s/m^2$, $Pa.s$), related to a standard feedwater temperature of 20 °C
$\Delta P_{200\ kPa}$ = pressure difference of 200 kPa ($200 \cdot 10^3\ N/m^2$)
$A_{0.45\ \mu m}$ = membrane area of standard 0.45 μm microfilter, i.e. $13.8 \cdot 10^{-4}\ m^2$

8 – Filtration Index for effluent ultrafiltration

In Roorda and van der Graaf (2001) a new parameter for fouling prediction (MFI-UF_n) was introduced. This parameter was related to the Modified Fouling Index using Ultrafiltration membranes (MFI-UF) of Boerlage *et al.* (2002), but measured for a shorter period (14-30 minutes instead of 6-24 hours by Boerlage)

$$MFI-UF_n = \frac{d(\frac{t}{V})}{d(V)} \cdot \frac{\eta_{20}}{\eta_T} \cdot \frac{\Delta P}{\Delta P_{100\ kPa}} \cdot \left(\frac{A_{memb}}{A_{1\ m^2}} \right)^2 \qquad \text{(eq. 4-A.10)}$$

where $MFI\text{-}UF_n$ = normalised Modified Filtration Index using Ultrafiltration membranes at T = 20 °C
ΔP = 1 bar (100 kPa) and A_{memb} = 1 m²

The relationship between SUR and MFI-UF_n is presented in equation 4-A.11:

$$SUR = \alpha_{av} \cdot c_v = MFI-UF_n \cdot 10^6 \cdot \frac{2 \cdot \Delta P_{100\ kPa} \cdot A_{1\ m^2}^{\ 2}}{\eta_{20}} = MFI-UF_n \cdot 10^6 \cdot \frac{2 \cdot 10^5 \cdot 1^2 \cdot (20+42.5)^{1.5}}{497 \cdot 10^{-3}}$$

$$\Rightarrow \qquad SUR = MFI-UF_n \cdot 2.0 \cdot 10^{14} \qquad \text{(eq. 4-A.11)}$$

4-B Spreadsheet for analysing filtration data and for calculation of the *SUR*

Below the spreadsheet used for calculating the *SUR* is shown. On the left the filtration data are given, on the right the *SUR* is calculated.

Specific Ultrafiltration Resistance

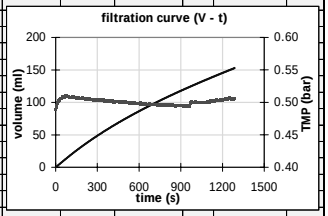
date:	22-2-2002			researcher:	Roorda	membrane code:	B-0410019														
filename:	020221_6.xls			location:	lab GZT	length	480	mm													
						int. diameter:	1.5	mm													
TMP:	0.501	±	0.004	bar	0.8%	surface area:	0.0023	m²													
J_n:	503	l/m².h.bar at 15°C																			
remarks:						feed:	effluent wwtp Berkel (21-2-02)														
sample from refrigerator (5°C)						average Temp:	17.0	°C													
TUDelft	**Roorda**																				
time:	*14:02 Feb. 21 2002*																				
A_m:	0.00226	m²																			
temperature:	17.0	°C																			
ref. temp.:	15.0	°C																			

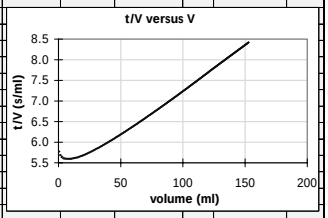
seconds	weight	flux	resistance	TMP		time	rel. time	volume	t/V	TMP	*SUR*
(s)	(g)	(l/m².h)	(m^{-1})	(bar)			(s)	(ml)	(s/ml)	(bar)	
1014296534	-0.02	7	0.00	0.267							
1014296537	0.25	124	1.07	0.398		21-02-02 14:02:17					
1014296541	0.26	5	31.05	0.429		21-02-02 14:02:21					
1014296544	0.52	119	1.25	0.451		21-02-02 14:02:24					
1014296548	1.08	256	0.60	0.463		21-02-02 14:02:28					
1014296551	1.65	263	0.59	0.470		21-02-02 14:02:31					
1014296555	2.24	270	0.59	0.479		21-02-02 14:02:35					
1014296558	2.85	280	0.58	0.490		21-02-02 14:02:38	0.0	0.0		0.490	
1014296562	3.45	276	0.59	0.493		21-02-02 14:02:42	3.5	0.6	5.78	0.493	
1014296565	4.07	285	0.58	0.496		21-02-02 14:02:45	6.9	1.2	5.69	0.496	
1014296568	4.68	280	0.59	0.498		21-02-02 14:02:48	10.4	1.8	5.69	0.498	
1014296572	5.31	289	0.57	0.499		21-02-02 14:02:52	13.9	2.5	5.64	0.499	
1014296575	5.92	280	0.59	0.501		21-02-02 14:02:55	17.3	3.1	5.65	0.501	
1014296579	6.56	293	0.57	0.503		21-02-02 14:02:59	20.8	3.7	5.61	0.503	
1014296582	7.17	280	0.60	0.504		21-02-02 14:03:02	24.3	4.3	5.62	0.504	
1014296586	7.8	289	0.58	0.505		21-02-02 14:03:06	27.7	5.0	5.60	0.505	
1014296589	8.42	285	0.59	0.506		21-02-02 14:03:09	31.2	5.6	5.60	0.506	
1014296593	9.04	285	0.59	0.506		21-02-02 14:03:13	34.7	6.2	5.60	0.506	
1014296596	9.66	285	0.59	0.506		21-02-02 14:03:16	38.1	6.8	5.60	0.506	
1014296600	10.28	285	0.59	0.507		21-02-02 14:03:20	41.6	7.4	5.60	0.507	
1014296603	10.9	285	0.59	0.508		21-02-02 14:03:23	45.1	8.1	5.60	0.508	
1014296607	11.52	284	0.60	0.509		21-02-02 14:03:27	48.5	8.7	5.60	0.509	
1014296610	12.13	280	0.60	0.508		21-02-02 14:03:30	52.0	9.3	5.60	0.508	
1014296613	12.76	289	0.59	0.510		21-02-02 14:03:33	55.5	9.9	5.60	0.510	
1014296617	13.37	280	0.60	0.509		21-02-02 14:03:37	58.9	10.5	5.60	0.509	
1014296620	13.98	280	0.60	0.508		21-02-02 14:03:40	62.4	11.1	5.61	0.508	
1014296624	14.59	280	0.60	0.509		21-02-02 14:03:44	65.9	11.7	5.61	0.509	
1014296627	15.2	280	0.60	0.509		21-02-02 14:03:47	69.3	12.4	5.61	0.509	
1014296631	15.81	280	0.61	0.511		21-02-02 14:03:51	72.8	13.0	5.62	0.511	
1014296634	16.42	280	0.61	0.511		21-02-02 14:03:54	76.3	13.6	5.62	0.511	
1014296638	17.02	276	0.61	0.509		21-02-02 14:03:58	79.7	14.2	5.63	0.509	
1014296641	17.63	279	0.61	0.509		21-02-02 14:04:01	83.2	14.8	5.63	0.509	
1014296645	18.24	280	0.60	0.509		21-02-02 14:04:05	86.7	15.4	5.63	0.509	
1014296648	18.83	271	0.62	0.509		21-02-02 14:04:08	90.1	16.0	5.64	0.509	
1014296652	19.43	276	0.61	0.509		21-02-02 14:04:12	93.6	16.6	5.65	0.509	
1014296655	20.02	271	0.62	0.508		21-02-02 14:04:15	97.1	17.2	5.65	0.508	
1014296659	20.61	271	0.62	0.508		21-02-02 14:04:19	100.5	17.8	5.66	0.508	
1014296662	21.2	270	0.63	0.508		21-02-02 14:04:22	104.0	18.4	5.67	0.508	
1014296665	21.79	271	0.62	0.508		21-02-02 14:04:25	107.5	18.9	5.67	0.508	

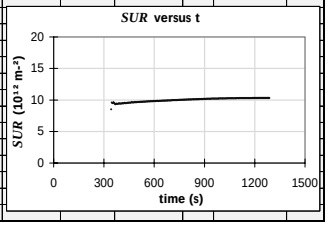
linearity between t/V and V_{total}:

	V_{total}	t/V
from:	54.2	6.27
to:	152.6	8.42

SUR	10.3	$\cdot 10^{12}$ m^{-2}			
SUR	10.0	±	0.3	$\cdot 10^{12}$ m^{-2}	2.9%







4-C Membrane resistance, fouling layer resistance, compressibility and the *SUR*

The Specific Ultrafiltration Resistance (*SUR*) is calculated according to equation 4.4, which is shown below. This equation is derived from Darcy's law.

$$SUR = \alpha_{av} \cdot c_v = \frac{d(\frac{t}{V})}{d(V)} \cdot \frac{2 \cdot \Delta P \cdot A_m{}^2}{\eta_T} \qquad \text{(eq. 4.4)}$$

In sludge filtration the resistance of the filter cake exceeds the resistance of the filter material by a factor more than 5 to 10. However, in ultrafiltration is the resistance of the filter material (the ultrafiltration membrane) in the same range as the resistance of the occurring fouling layer. The relationship between flux, resistance and TMP is therefore also determined by the initial resistance!

The relationship that has been derived in this dissertation for the *SUR* (see above eq. 4.4) does neglect the influence of the initial membrane resistance. The SUR that is found is comprised of a factor that is determined by the membrane itself, and by a factor that is determined by the developing fouling layer. The factor that is determined by the membrane is constant in measuring the *SUR* (at constant TMP) and therefore does not influence the outcome of a *SUR* measurement.

This is also the case in the results of experiments TMP-1, TMP-2 and TMP-3. The compressibility coefficient has also been calculated by correcting the *SUR* and the TMP for the membrane resistance. In figure 4-C.1, the results are presented for each experiment. In the upper left corner the trendline is presented for the results that neglected the membrane resistance, in the right corner is the trendline presented for the results that does not neglect the membrane resistance. As can be seen the compressibility factor are similar.

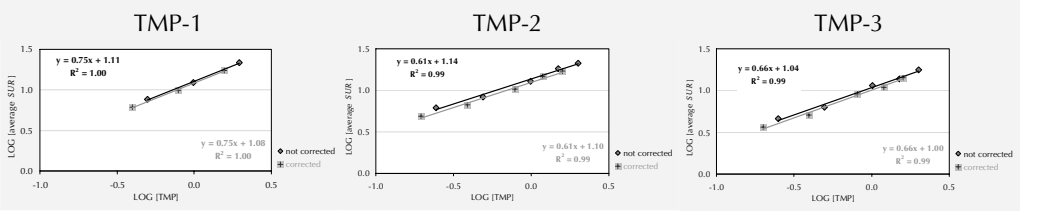


Figure 4-C.1 Compressibility coefficient calculated with and without neglecting the membrane resistance (see text for details)

Appendix 5 - Fractionation

5-A Particle distribution curves for effluent of wwtp Ede

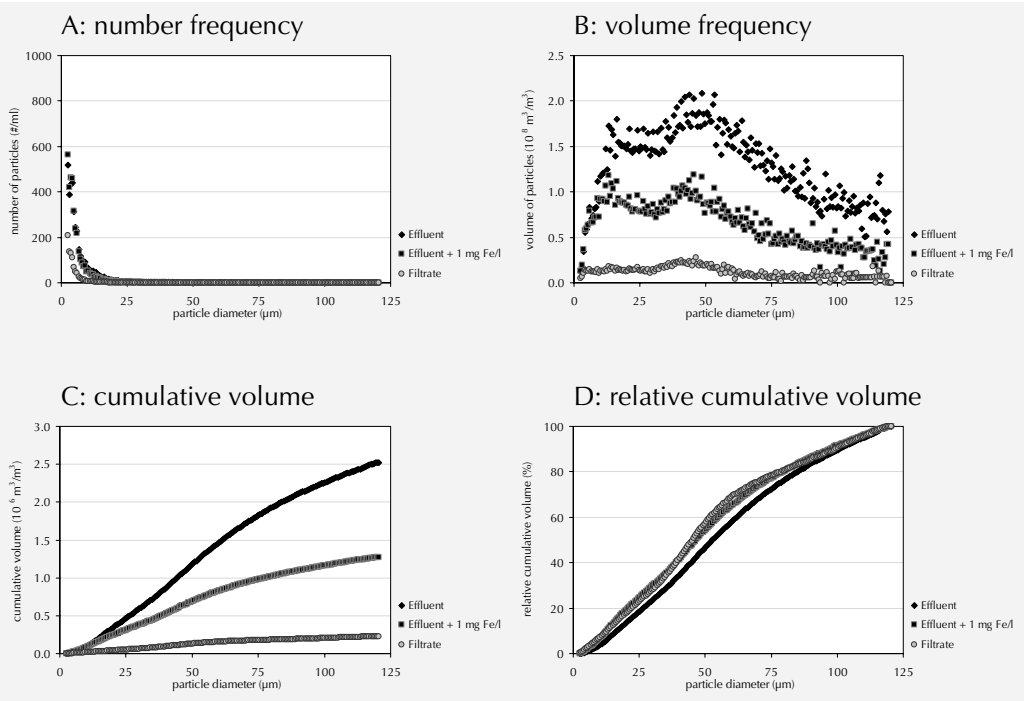


Figure 5-A.1 Particle distribution curves for effluent, effluent with 1 mg Al^{3+}/l and filtrate from wwtp Ede; A: number frequency, B: volume frequency, C: cumulative volume, D: relative cumulative volume.

5-B Changes in *SUR* for size fractions

Table 5-B.1 Percentage of the change in *SUR* for size fractions of raw and pre-treated wwtp-effluent

WWTP	**Berkel**			**HvHolland**	**Zaandam**
	Effluent	Effluent+[a]	Filtrate[b]	Effluent	Effluent
Feedwater fraction	% *SUR*	% *SUR*	% *SUR*	% *SUR*	% *SUR*
< 200 μm	100%	100%	100%	100%	100%
< 5.0 μm	103%	81%	104%	Nd[c]	97%
< 1.2 μm	80%	81%	90%	nd	92%
< 0.45 μm	76%	64%	82%	nd	81%
< 0.2 μm	65%	64%	70%	69%	77%
< 0.1 μm	17%	24%	29%	12%	nd

[a] Effluent + 1 mg Al^{3+}/l (PACl); [b] one layer sand filtration (10 m/h) without coagulation; [c] Not determined

Appendix 6 – Theoretical filtration curves

6-A Calculation of β-value

The characteristic form of the four filtration laws (Hermia, 1982) has been presented in §6.2 (equation 6.1). The β-values can be derived from a log-scale graph (equation 6.2), in which the slope of the curve is the β-value. Below, the procedure applied for the determination of the β-values is presented with its subsequent steps.

On lab-scale the filtration curve was measured for (pre-treated) effluent at constant TMP of 0.5 bar using an ultrafiltration membrane. The measurement usually lasted about 15 minutes. Every 3 to 5 seconds the filtered feedwater volume was measured as permeate mass, together with the feedwater pressure for determination of the TMP[1]. Permeate mass, feedwater pressure and time were stored on a personal computer for further analysis. For illustration, the calculation on a data set measured in the lab-scale unit with two pressure vessels is graphically presented for the subsequent steps below. The data set is measured on the 12th of April 2002 using raw effluent from wwtp Berkel.

Firstly, the relationship between time (t) and filtered volume (V) was drawn. In figure 6-A.1 an example of such a curve is presented. The black line shows the measured data, the dotted gray line shows the 3rd order polynomial that fitted the measured data[2]. The equation calculated by the spreadsheet program for this polynomial is given in the same curve together with the correlation coefficient R^2 (R^2=1 relates to a 100% correlation; R^2=0 relates to 0% correlation). The equation of the 3rd order polynomial was used for the subsequent steps in analyzing the filtration data.

[1] The Trans Membrane Pressure (TMP) was calculated by assuming that the pressure in the permeate tube is 0.0 bar; TMP is therefore equal to the pressure in the feedwater

[2] The intersection on t = 0 s and V = 0 m³ was used in experiments that had a constant TMP from their initial start; in some experiments the first 120 to 180 seconds were neglected because the TMP was still increasing, in this case the intersection was chosen by the spreadsheet program (*Microsoft® Excel 2000*); a verification was made in the second graph (fig. 6-A.2), which shows both dt/dV calculated from the raw data, as wel as dt/dV calculated from the 3rd order polynomial proposed by *Microsoft® Excel 2000*; in all experiments these values corresponded to each other (correlation coefficient R^2 is 1 with a deviation of less than 0.01%), showing that the polynomial could be used for further data analysis

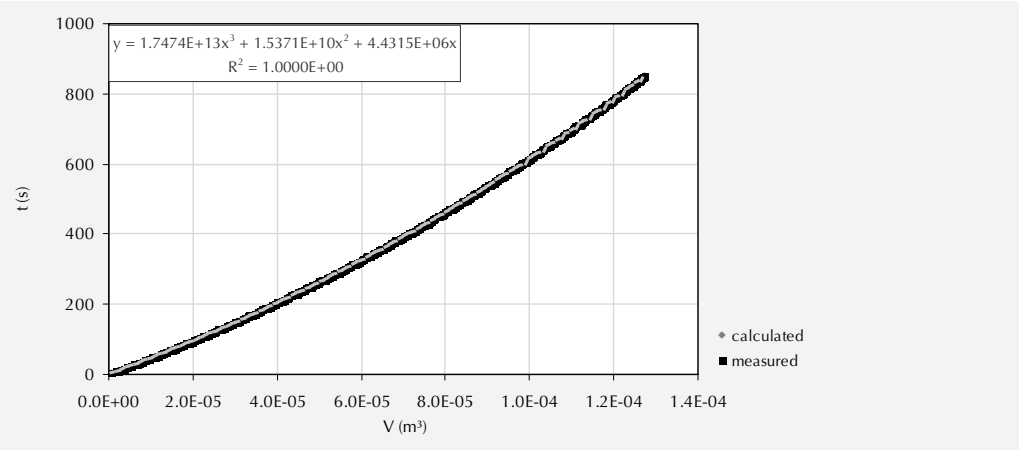


Figure 6-A.1 Example of filtration curve showing the relationship between time and volume, together with a 3rd order polynomial that fits the data correctly (correlation coefficient R^2=1)

The following step is the first derivative of polynomial. Figure 6-A.2 shows the measured first derivate (d*t*/d*V*) in black dots, the grey line shows the first derivate of the polynomial shown in figure 6-A.1. The black dots show the same curve as the calculated curve, verifying that the 3rd order polynomial accurately described the filtration data.

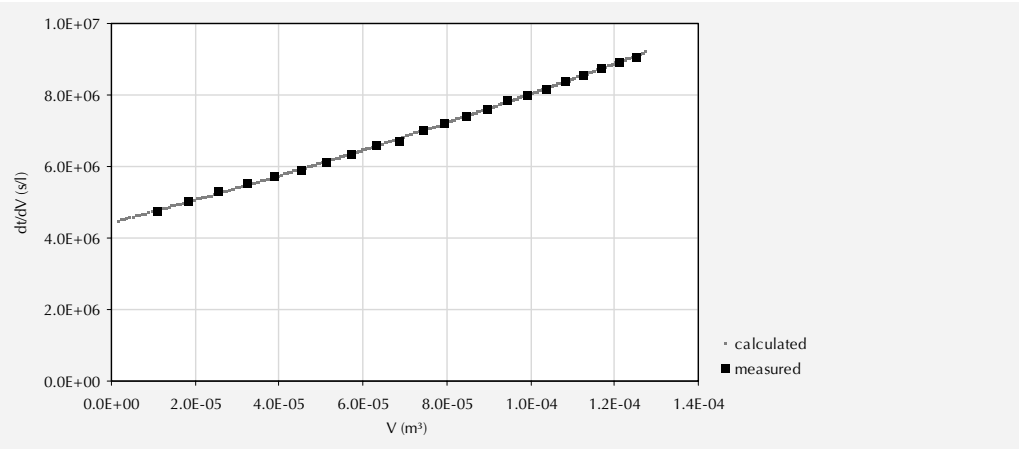


Figure 6-A.2 First derivate of the 3rd order polynomial (d*t*/d*V*) presented with the grey line; the black dots present the derivate calculated using the filtration data

Next, the second derivate of the polynomial (d^2t/dV^2) was calculated. In figure 6-A.3 the relationship between d^2t/dV^2 and V is presented graphically.

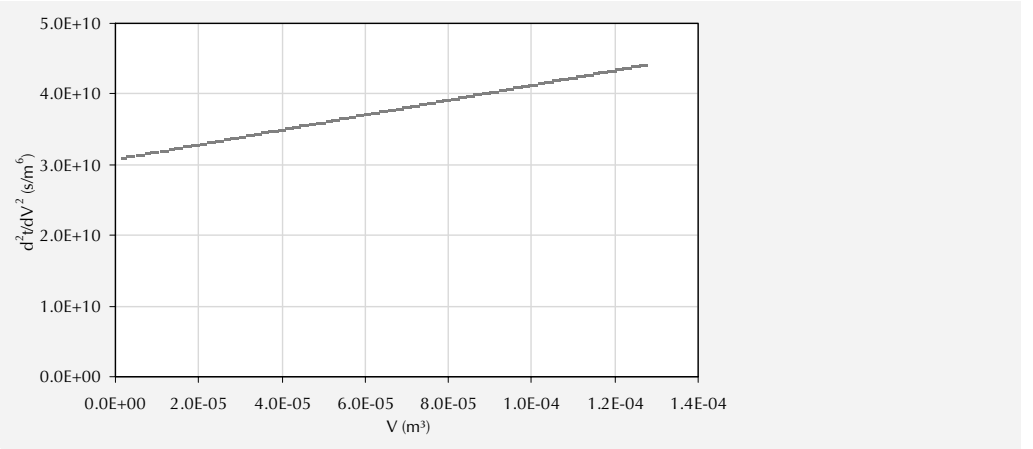


Figure 6-A.3 Second derivate of the 3rd order polynomial (d^2t/dV^2) presented with the grey line as a function of filtrated volume V

The relationship between the logarithm of the second derivate (d^2t/dV^2) and the first derivate (dt/dV) is presented below in figure 6-A.4. The slope of this curve presents the β-value, which is the characteristic for the four filtration laws. In figure 6-A.5 the β-value is presented for the filtration data discussed in this Appendix.

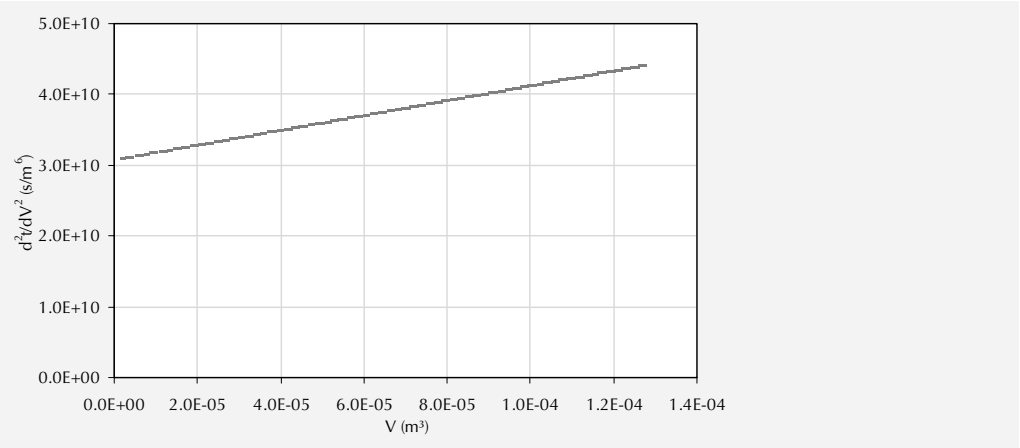


Figure 6-A.4 Relationship between the logarithm of the second derivate of the 3rd order polynomial (d^2t/dV^2) as a function of the logarithm of the first derivate (dt/dV); the slope of the curve presents the β-value that is a characteristic for the occurring filtration mechanism

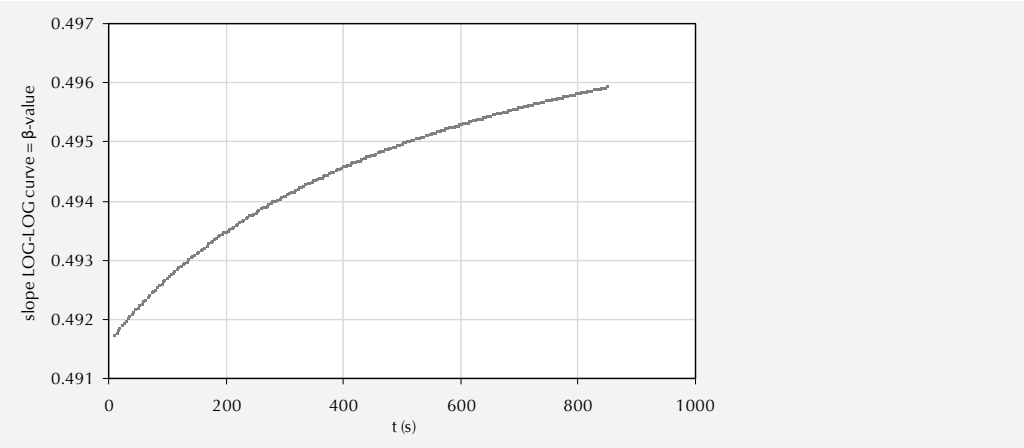


Figure 6-A.5 The β-value; a characteristic for the occurring filtration mechanism

Next to calculation of the β-values, the filtration laws were rewritten into a relation between t/V and t, and between t/V and V. Linearity of the first (t/V vs. t) indicates standard blocking, linearity of the latter (t/V vs. V) indicates cake filtration. In the left graph of figure 6-A.6 the curve for standard blocking is presented (for data used above: wwtp Berkel

12th April 2002). In the right graph (figure 6-A.6) the same is presented for the cake filtration law. For both curves the correlation coefficient R^2 was calculated, the highest R^2 indicated that the accompanying curve described the filtration mechanism the best. The example shown in figure 6-A.6 shows that the R^2 is for both curves about the same. This indicates that both filtration mechanisms could be applied to the filtration data.

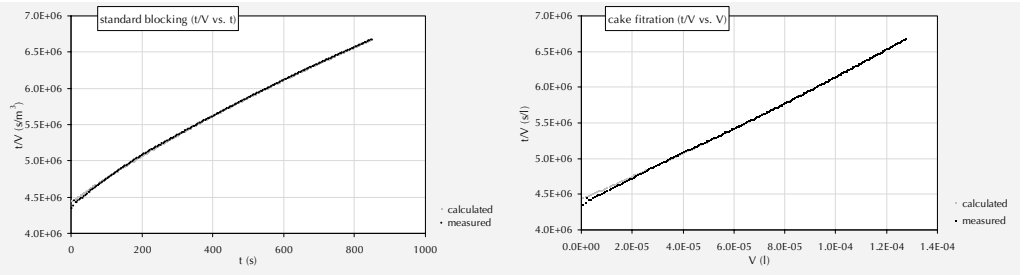


Figure 6-A.6 Linear relationship indicates filtration mechanism; in the left graph the relation between t/V and t is presented, linearity indicates standard blocking; in the right graph the relation between t/V and V is presented, linearity indicated cake filtration; a distinction is made by a comparison of the correlation coefficients; in these graphs from t = 160 s until the end of the filtration period R^2 is 0.9987 for the left graph and 0.9993 for the right graph, indicating that the filtration data can be described by both filtration theories

6-B Results of filtration mechanism analysis

The last column (Standard/Cake) represents the mechanism which filtration curve (see figure 6-A.6) had the R^2 closest to 1. As suggested in the previous Appendix 6-A, the R^2 for these two mechanisms is almost similar. This indicates that both filtration mechanisms fit the measured filtration data. Tables 6-B.1, 6-B.2, 6-B.4 en 6-B.5 are presented after each other, table 6-B.3 is the last table.

Table 6-B.1 Results of β-values calculated using (pre-treated) effluent of wwtp Tilburg-Noord; a 0.8 mm ultrafiltration membrane was used

Date	Exp.	Feed	*SUR* (10^{12} m^{-2})	β-value	Standard/Cake*
14-09-'00	E-1.1	Effluent	7.0	1.0	Standard
	E-1.2	Effluent	6.4	0.8 → 0.7	Standard
	E-1.3	Effluent	5.4	1.3 → 1.0	Standard
20-09-'00	E-1.4	Effluent	4.6	0.8	Standard
25-09-'00	E-1.5	Effluent	5.2	0.7	Standard
26-09-'00	E-1.6	Effluent	5.8	1.5 → 1.2	Standard
	E-1.7	Effluent	6.6	0.8	Standard
28-09-'00	E-1.8	Effluent	5.0	0.9	Standard
	E-1.9	Effluent	4.4	3.7 → 1.8	Standard
	E-1.10	Effluent	5.0	1.0	Standard
	E-1.11	Effluent	5.0	5.2 → 2.2	Standard
25-09-'00	E+-1.1	Effluent + 1 mg Al^{3+}/l	5.4	-0.2	Cake
26-09-'00	E+-1.2	Effluent + 1 mg Al^{3+}/l	5.0	5.0 → 1.7	Standard
28-09-'00	E+-1.3	Effluent + 1 mg Al^{3+}/l	6.2	-0.7 → -1.3	Cake
02-10-'00	E+-1.4	Effluent + 1 mg Al^{3+}/l	6.6	0.2	Cake
	E+-1.5	Effluent + 1 mg Al^{3+}/l	4.8	0.2	Cake
14-09-'00	F-1.1	Filtrate	3.6	1.2 → 1.0	Standard
20-09-'00	F-1.2	Filtrate	3.8	1.3 → 1.1	Standard
	F-1.3	Filtrate	4.2	1.7 → 1.3	Standard
25-09-'00	F-1.4	Filtrate	3.0	0.7	Standard
02-10-'00	F-1.5	Filtrate	3.8	4.5 → 2.5	Standard

* Based on linear relationship t/V vs. t (indicating standard blocking) or t / V vs. V (cake filtration)

Table 6-B.2 Results of β-values calculated using (pre-treated) effluent of wwtp Berkel; a 1.5 mm ultrafiltration membrane was used

Date	Exp.	Feed	SUR (10^{12} m^{-2})	β-value	Standard/Cake
17-10-'01	E-2.1a	Effluent (t < 1800 s)	13.7	0.5	Cake
	E-2.1b	Effluent (t > 1800 s)	13.7	0.1 → 0.4	Cake
27-10-'00	E-2.2a	Effluent (t < 1800 s)	8.9	0.3	Standard
	E-2.2b	Effluent (t > 1800 s)	10.5	0.1	Cake
18-10-'00	E+-2.1a	Effluent + 1 mg Al^{3+}/l (t < 1800 s)	6.6	-0.3	Cake
	E+-2.1b	Effluent + 1 mg Al^{3+}/l (t > 1800 s)	6.6	0.1	Cake
27-10-'00	E+-2.2a	Effluent + 1 mg Al^{3+}/l (t < 1800 s)	5.8	-0.5 → -1.0	Cake
	E+-2.2b	Effluent + 1 mg Al^{3+}/l (t > 1800 s)	5.6	-0.1	Cake

Table 6-B.4 Results of β-values calculated using (pre-treated) effluent of wwtp Berkel; a 0.8 mm ultrafiltration membrane was used

Date	Exp.	Feed	SUR (10^{12} m^{-2})	β-value	Standard/Cake
3-7-'02	E-4.1	Effluent	7.6	4 → 1.8	Standard
		Effluent <0.2 μm	6.0	3.9 → 1.5	Cake
		Effluent <0.1 μm	2.0	3.0 → 4.4	Cake
4-7-'02	E-4.2	Effluent	7.0	-0.3 → -0.4	Cake
		Effluent <0.2 μm	5.6	-0.5 → -1.1	Cake
		Effluent <0.1 μm	2.0	-3 → -4	Standard
5-7-'02	E-4.3	Effluent	7.0	-0.1 → -0.2	Cake
		Effluent <0.2 μm	5.4	-0.3	Cake
		Effluent <0.1 μm	1.6	-1.5 → -2.7	Standard

Table 6-B.5 Results of β-values calculated using (pre-treated) effluent of wwtp EMMtec; a 0.8 mm ultrafiltration membrane was used

Date	Exp.	Feed	SUR (10^{12} m^{-2})	β-value	Standard/Cake
6-03-'02	E-1	Effluent	7.6	0.1	Cake
11-03-'02	E-2	Effluent	12.9	0.5	Cake
6-03-'02	F-1	Filtrate	7.0	-0.2	Cake
11-03-'02	F-2	Filtraat	10.5	0.7	Standard
6-03-'02	F+-1	Filtrate + 2 mg Al^{3+}/l	5.4	-0.4 → -1.2	Cake
11-03-'02	F+-2	Filtraat + 2 mg Al^{3+}/l	6.8	0.6	Standard

Table 6-B.3 Results of β-values calculated using (pre-treated) effluent of wwtp Berkel; a 0.8 mm ultrafiltration membrane was used

Date	Exp.	Feed	*SUR* (10^{12} m^{-2})	β-value	Standard/Cake
15-03-'02	E-3.1	Effluent	13.1	0.6	Cake
		Effluent <5.0 μm	12.5	0.6	Cake
		Effluent <1.2 μm	11.5	0.4	Cake
		Effluent <0.45 μm	10.5	0.5	Cake
		Effluent <0.2 μm	10.5	0.6	Cake
18-03-'02	E-3.2	Effluent	13.1	0.6	Cake
		Effluent <5.0 μm	15.3	0.4	Cake
		Effluent <1.2 μm	12.9	0.5	Cake
		Effluent <0.45 μm	11.5	0.4	Cake
		Effluent <0.2 μm	11.3	0.4	Cake
12-04-'02	E-3.3	Effluent	11.9	0.5	Cake
		Effluent <0.2 μm	7.6	0.1	Cake
12-04-'02	E-3.4	Effluent	11.7	0.5	Cake
		Effluent <5.0 μm	9.7	0.1	Cake
		Effluent <1.2 μm	8.7	0.1	Cake
		Effluent <0.45 μm	8.7	0.2	Cake
		Effluent <0.2 μm	7.2	-0.2 → -0.7	Cake
19-04-'02	E-3.5	Effluent	10.3	-0.3	Cake
22-04-'02	E-3.6	Effluent	12.9	-0.5 → -2.0	Cake
		Effluent <1.2 μm	10.1	0	Cake
		Effluent <0.45 μm	8.9	0.3	Cake
19-04-'02	E+-3.1	Effluent + 1 mg Al^{3+}/l	8.0	-0.2 → -0.7	Cake
		Effluent + 1 mg Al^{3+}/l <5.0 μm	7.4	-0.4 → -1.7	Cake
		Effluent + 1 mg Al^{3+}/l <1.2 μm	6.2	-0.4 → -1.2	Cake
		Effluent + 1 mg Al^{3+}/l <0.45 μm	5.6	-0.4 → -0.8	Cake
22-04-'02	E+-3.2	Effluent + 1 mg Al^{3+}/l	11.3	-0.2 → -0.9	Cake
		Effluent + 1 mg Al^{3+}/l <1.2 μm	8.7	-0.1	Cake
		Effluent + 1 mg Al^{3+}/l <0.45 μm	6.8	-0.1	Cake