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**EUROMBRA**

Membrane bioreactor technology (MBR) with an EU perspective for advanced municipal wastewater treatment strategies for the 21st century.

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## D12 – Results of raw sewage challenge trials

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Membrane bioreactor technology (MBR) with an EU perspective for advanced wastewater treatment strategies for the 21<sup>st</sup> century

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**WP4**

**Process configuration**

**D12- Report**

**Results of raw sewage challenge trials**

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## Introduction

The aims of Work Package 4 are:

- correlate the impact of reactor geometry onto permeability;
- identify strategies for optimised nutrient elimination for MBR;
- characterize impact of peak loads on membrane performance and effluent quality;
- analyse the fouling composition and correlate the fouling and clogging tendency with sludge tested operational conditions (cooperation with WP5);
- formulate recommendations for reactor configuration, dimensioning and process control.

The Deliverable 12 is mentioned on the deliverables list but the content is not defined in the Work Package 4 description. Nevertheless, the main goal of Section 4.1 is to establish how long raw wastewater has to be in contact with sludge before reaching the membrane compartment in order to avoid severe fouling. This objective is based on the assumption known from practice that raw wastewater filtrated directly causes severe fouling.

The content of Deliverable 12 was therefore redirected for the documentation and analysis of the practical assumption that directly filtration of raw wastewater causes severe fouling. Having this goal in mind the following content was found relevant for this report:

1. Literature review: raw sewage characteristics related with MBR experience
2. Practical experiences (EUROMBRA/AMEDEUS)
3. Direct membrane filtration of raw sewage
4. Fouling rates comparison between raw sewage filtration and MBR filtration
5. Filtration characterization tests (Delft Filtration Characterisation method DFCm) on raw sewage
6. Filtration characterization tests during biological processes

The literature review and description of practical experiences from EUROMBRA/AMEDEUS MBR network partners aim to identify and characterize situations where a specific influent component had a detrimental effect on MBR performance.

Considering that membranes provide a physical barrier against contaminants and pollutants, the issue of using a single step process for wastewater treatment imposes by itself. DUT researched this possibility. The applied method and main results of raw sewage filtration are described in point 3.

To provide a base for comparison between raw sewage filtration and MBR filtration the fouling rates obtained with raw sewage and MBR activated sludge trials are described in point 4. Filtration characterization tests with the Delft Filtration Characterisation method DFCm using raw wastewater are described in point 5.

In order to gain insight about the consequences of reactor configuration filtration tests have been performed by DUT during the different steps of the biological process. Preliminary results are shown in point 6.

## **1- Literature review: raw sewage characteristics related with MBR experience**

The relation between feed water components and fouling has been suggested by the scientific community already many years ago. Considering that activated sludge is a complex and heterogeneous suspension constituted by feed water components, metabolites produced during the biological reactions and biomass, the hypothesis that many of these individual components (from flocculent solids to dissolved polymers) may contribute to membrane fouling seems to be logical [1, 2].

Fouling and its causes has been the most researched topic in recent years [3] but the link between raw sewage characteristics and full scale MBR operation (treating municipal wastewater) is not frequently mentioned. Situations describing problems in full scale MBR operation directly related to feed water characteristics are not abundant and there are even some authors that defend the opposite, that is, feed water characteristics do not affect MBR operation at all.

Some MBR network partner's experiences and a few literature cases, indicate that, in specific situations, raw sewage characteristics are detrimental to MBR operation. Experiences from MBR network partners, where decrease in full scale MBR's permeability was directly related to feed water quality, are described in detail in point 2.

In pilot plants treating sewage from a mixed sewer system it was found that strong varying influent flows influence the membranes performance even when compared to the effect of different pre-treatments and process temperatures [4].

In lab scale experiments, using a membrane aerated bio film reactor, the manipulation of culture conditions allowed the mineralization of perchloroethylene (highly chlorinated ethenes derived mainly from dry-cleaning and degreasing industries) [5]. Biomass was originally collected from a municipal wastewater treatment plant, exposed to perchloroethylene. Complete mineralization, however, was not achieved and COD removal rate was rather low (around 60%) for these type of reactors. The presence of the toxic compound affected the reactor performance.

The effect of a toxic component on the microbiological community in particular is also mentioned in other studies [6]. When exposed to a toxic environment, regardless the toxicity is of organic or inorganic nature, the microorganisms tend to form clusters to reduce the total surface area in contact with the environment. When exposed to a toxic environment microorganisms increase the production of EPS.

Nevertheless the experience in a full scale MBR, in operation since 2002 in Brescia, Italy, is that the efficiency of the MBR does not depend on the surrounding conditions because organic compounds and suspended solids are detained even if a toxic substance enters the MBR or if it suffers an organic, hydraulic or pH shock [7]. The decreases in permeability were then related to drops in temperature or malfunctions in the aeration system.

The quality of the feed water is mainly mentioned when the performance of the MBR is being tested. A simple division between pollutants can be considered: conventional pollutants (such as biochemical oxygen demand, total suspended solids, nitrogen, phosphorus and pathogens), non conventional pollutants (such as salts, metals and refractory organics) and emerging contaminants (such as pharmaceuticals, household cleaning products and endocrine-disrupting compounds) [8]. When the performance of an MBR treating municipal wastewater is being tested mostly conventional pollutants (though parameters like COD, BOD and nutrients) are measured.

Some specific cases may require measurements of other parameters as in the case of a full scale membrane bioreactor installed in Haidain community hospital [9]. The feed water and effluent

characteristics are provided in Table 1. The membranes were not chemically cleaned during six months, therefore no irreversible fouling was detected during this period, but operating flux was lower than 10 L/m<sup>2</sup>.h and suspended solids concentration was between 2.5 and 3.5 g/L.

Table 1- Feed and effluent wastewater characteristics[9].

Wastewater source Parameters	Hospital	
	Influent	Effluent
COD (mg/L)	48-277.5	<30
BOD <sub>5</sub> (mg/L)	20-55	<0.4
NH <sub>4</sub> <sup>+</sup> -N(mg/L)	10.1-23.7	<1
Turbidity (NTU)	6.1-27.9	<4
T(°C)	14-20	16-18
pH	6.2-7.1	6.2-7.1
Bacteria (number/L)	9.9×10 <sup>3</sup>	
<i>E.coli</i> (number/100 mL)	>1600	<23

In pilot scale, some trials have been performed to assess the effect of non conventional pollutants, such as salts (see point 2). Experiences were also done to study the removal of organic and inorganic micro pollutants [10] but fouling situations were not described.

The great amount of experience related with removal and effects of conventional, non conventional pollutants and emerging contaminants is provided by MBR's treating industrial wastewater. While municipal MBR have still a restricted application, MBR technology is already an attractive option for the treatment of industrial wastewater that have generally high pollutant loading but smaller plant size [11]. Experiments, with "real" influent, are described as follows.

Wastewater from an acrylonitrile-butadiene-styrene plant was treated with an MBR [12]. The wastewater has a high index of refractory biodegradation (total Kjeldahl nitrogen/COD ratio). The MBR achieved better COD and BOD removal than conventional biological treatment. Membrane cleaning (with water) was more frequent and chemical cleaning was required at lower hydraulic retention times (12 and 18 hours). Hydraulic retention times of 24 and 30 hours were also tested with less frequent membrane cleaning and no chemical cleaning was necessary.

A pilot unit with a membrane bioreactor (side stream ultrafiltration) coupled with reverse osmosis was used to treat tannery wastewater [13]. Feedwater and MBR permeate characteristics are provided in Table 2. Tannery wastewater is characterized by a high chemical and biological oxygen demand with high salt content. The influent was pre-treated with aeration, pH correction, coagulation/flocculation and phosphate addition. Biomass used for MBR inoculation was adapted to tannery effluents. In the MBR, COD concentration as well as sulphide and chrome were significantly reduced. Fouling situations were not described. The results lead to the design of a full scale MBR/RO plant.

Table 2- Feed and effluent wastewater characteristics[13].

Wastewater source Parameters	Tannery	
	Influent range	MBR Effluent range
COD (mg/L)	1704-4442	126-509
BOD <sub>5</sub> (mg/L)	650-1850	5-50
NH <sub>4</sub> <sup>+</sup> -N(mg/L)	5.4-295.2	0.056-47
S <sup>2-</sup> (mg/L)	0.6-52	0-0.4
Cr total(mg/L)	0.23-1.47	0.09-0.34

A combination of an up flow bed filter reactor with an MBR, at lab scale, enabled the treatment of swine wastewater containing high concentrations of organic solids and nitrogen [14]. Fouling situations were detected and chemical cleaning was performed.

A review on treatment processes regarding the removal of endocrine disrupting chemicals concluded that membrane bioreactors (which combine adsorption and biodegradation) are a good compromise for simultaneous carbon, nitrogen, phosphorus and endocrine disrupting chemicals removal [15]. Studies already done indicate that complex endocrine disrupting compounds are not removed to a satisfying level in a MBR [3]. A system combining MBR/Activated Carbon/ Nanofiltration was suggested to provide better COD removal for old leachate than any other conventional biotreatment scheme [16]. Landfill leachates include complex endocrine disrupting compounds so the set-up may be crucial for the removal of these substances. Specific effects of endocrine disrupting chemicals on membrane bioreactor operation are not yet known.

In a lab-scale MBR foul condensate, from a pulp mill, was used to test degradation of this specific wastewater, very rich in organic matter, especially methanol and total reduced sulphur compounds [17]. Biomass was acclimated and nitrogen and phosphorus were added to the bioreactor. High removal efficiencies of COD, methanol and total reduced sulphur were observed at 35, 45 and 55°C but, in general, the removal rate of contaminants decreased at higher temperatures. Fouling decreased at higher temperatures.

A lab-scale MBR was used to test textile wastewater treatment [18]. Textile wastewater is characterized by a low BOD/COD ratio, low nitrogen and phosphorus, and a high colour content (resulting from dyestuffs and accompanying chemicals). Biomass was adapted to textile wastewater. There was some decrease in the flux due to fouling, but during the 180 days of the test trial, no cleaning activities were performed. The system proved to be very resistant to changing loading rates and even at high loading rates COD removal occurred. Nevertheless, residual COD still prevails as well as colour components.

Surfactants are common in industrial effluents from cosmetic and detergent industries and laundry or car washing services. Conventional biological wastewater treatments can not achieve complete elimination of surfactants and their concentration in the medium cannot exceed certain values due to toxicity towards microorganisms and foaming in the aerated bioreactors. An ultrafiltration unit was coupled to an existing stirred tank reactor and a *Critrobacter braaktii* population inoculated into the industrial reactor (additional ammonium nitrate was added) [19]. The feed water and permeate characteristics are provided in Table 3. It is mentioned that membrane fouling affected the permeate flux.

Table 3- Feed and effluent wastewater characteristics[19].

Wastewater source	Cosmetic industry	
	Influent range	MBR Effluent range
Anionic surfactant (g/L)	0.13-0.47	0-0.04
COD <sub>soluble</sub> (g/L)	1.2-2.68	0.13-1.15
TSS (g/L)	0.13-0.37	0
MLVSS (mg/L)	0.1-0.27	-
pH	6.34-7.25	6.71-7.05
EC (dS m <sup>-1</sup> )	1.62-2.43	1.57-2.51

The dynamic behaviour of membrane bioreactors with negligible biomass growth, due to the presence of toxic substances, was modelled to simulate a pilot MBR in an industrial degreasing unit [20]. In the degreasing baths the most common toxic substances are free surfactants. Model results showed an improvement of specific substrate uptake rates with increased air supply, indicating a possible increase in the active cell population in the reactor due to the reduced toxicity.

## 2- Practical experiences (EUROMBRA/AMEDEUS)

The following MBR network partners communicated experiences where feed water characteristics influenced MBR operation and performance:

1. Cranfield University (CRAN)
2. Delft University of Technology (DUT)
3. Flemish Institute for Technological Research (VITO)
4. Aquafin (AQF)

Their specific knowledge is described as follows.

### 2.1 Cranfield University (CRAN)

In a pilot plant MBR trials tests were conducted to test the effect of saline wastewater shock loads on activated sludge characteristics and MBR performance [21]. The tests were performed with municipal wastewater and salinity was introduced by dosing NaCl solution directly into the bioreactor. Tests were done with two constant fluxes: 8 and 16 L m<sup>-2</sup> h<sup>-1</sup>.

After salt addition COD removal was immediately affected and took approximately one week to recover. Nitrification was also affected but in a lesser extent. When the salt was removed removal efficiencies were restored and permeability stabilized. These results show the inhibitory effect of salt shocking.

The carbohydrate and protein content of SMP and EPS increased rapidly at low salinities (0-1 g L<sup>-1</sup> NaCl) and less change was found at higher salinities (1-4 g L<sup>-1</sup> NaCl). The content of carbohydrate was generally higher than of protein, in both the SMP and EPS, with greater changes at lower flux values. A weak negative correlation was observed between permeability and SMP carbohydrate.

### 2.2 Delft University of Technology (DUT)

The Varsseveld MBR in the Netherlands started in 2004. After a few months of operation low permeability (around 300 L/(m<sup>2</sup>.h.bar)) was obtained requiring specific handling [22]. Decreases in permeability were confirmed by the Delft Filtration Characterization method. Filtration tests were performed with activated sludge collected from the pilot plant of Varsseveld MBR (scale down factor of 200 from the full-scale installation).

Visual inspection of the membranes revealed the presence of a substance, with slimy glue appearance, sticking together the membranes and therefore decreasing the available membrane surface [22]. Sludge with slimy appearance was also present in the compartment walls and membrane aerator tubes. The development of this substance was connected with the discharge of a polymeric substance by a local industry (cheese factory). A small perforation nearby a connection between the membrane module and cassette and the long period of anaerobic sludge conditions created the environment for the development and proliferation of the mentioned substance.

Batch tests were performed to assess the effect of the polymeric substance discharged by the cheese factory [23].

Permeability in Varsseveld MBR was restored with intensive cleaning and other maintenance procedures after the discharge of the polymeric substance ceased.

### **2.3 Flemish Institute for Technological Research (VITO)**

The Flemish Institute for Technological Research related the following experiences:  
in industrial wastewater from textile industry adsorbing dyestuffs affected membrane performance;  
in industrial wastewater from chemical industry lower values of sludge concentration decreased membrane performance.

### **2.4 Aquafin (AQF)**

The Schilde MBR in Belgium is in operation since 2003. Two fouling events were described as related to influent characteristics.

In May 2004 permeability decreased from 210 to 120 L/(m<sup>2</sup>.h.bar) within 15 days [24]. This occurred because of bad sludge quality and complete nitrification inhibition, possibly caused by a toxic cause. Microscopic sludge analyses showed partial deflocculation, increase in free swimming bacteria and decrease in protozoa, metazoan and ciliates in stalked condition. Permeability recovered autonomously by operating the system with its original settings.

In December 2006 foaming developed and persisted for 4 weeks [25]. The foaming development resulted in a significant decrease of permeability. This event happened after the implementation of butyric acid as carbon source. To this phenomenon two possible explanations were suggested:  
bad influent quality;  
switching from acetic acid to butyric acid caused a transitional phase in the microbial population which led to the foaming production (also from 26<sup>th</sup> December to the 8<sup>th</sup> January, the carbon source was dosed constantly causing probably carbon excess which could have increased foaming development).

Foaming disappeared on the second week of January followed by permeability recover.

### 3- Direct membrane filtration of raw sewage

In DUT intensive work was performed in Direct Membrane Filtration (DMF) [26, 27, 28, 29, 30]. The most relevant papers are presented in Annex I.

DMF is when wastewater is filtrated directly on a porous membrane. Membranes provide separation and not removal of wastewater constituents in a purely physical process so DMF has also been designated by Direct Membrane Separation. If the raw wastewater is filtrated directly on ultrafiltration membranes the process is more simply designated by Direct Ultrafiltration.

In this process the feedwater is taken directly from a collection system. Through simple mechanical pre-treatments such as screening large particles are removed before membrane filtration. The membrane separates the retentate from the filtrate.

The goal of the work developed in DUT was to provide a physical-chemical process based on membrane technology that could enable wastewater reuse also where biological systems were not suitable. As a single step process DMF guarantees only a partial treatment because many pollutants may not be removed but the water produced may have reuse options. If followed by further treatment DMF can be considered as a very advanced pre-treatment for the removal of particles, bacteria and turbidity.

For municipal wastewater submitted to DMF two applications are foreseen: main polishing step of wastewater (as an alternative to biological treatments) and partially concentrate the wastewater (extracting permeate and return the concentrate to the sewer) [28]. The advantages of DMF are: plants could be easily up-scaled and operation could be discontinuous and meet seasonal changes in water flow. DMF would be then suitable for small-scale reuse scheme where eventual permeate refinements would be determined according to reuse purpose.

The tests of DMF were performed in a crossflow filtration unit (see Figure 1) with hydrophilic tubular PVDF membranes of 5.2mm diameter (X-flow F 4385)[26]. The 1 meter module had five tubes placed vertically resulting in a total filtration surface of 0.073m<sup>2</sup>. Clean water (tap water quality), wastewater and cleaning solutions were fed with a centrifugal pump. Chemical feed to the inner side of the tubes was also “statically” possible from the top of the membrane module, and backflush with demineralised water was provided through an independent pump. The process parameters were calculated as follows from data automatically collected every 10 seconds:

- TMP: pressure meters were placed at the inlet of the membrane module, at the concentrate outlet and at the permeate outlet; TMP was calculated as the difference between the average pressure at the inner side and the permeate side of the membrane.
- Flux (J): the extracted permeate was weighed on a balance, therefore the flux was calculated as mass increase with time per membrane surface;
- Resistance (R): according to Darcy’s law

$$J = \frac{TMP}{\eta \cdot R}$$

resistance was calculated from J and TMP assuming  $\eta_{\text{solution}}$  equal to  $\eta_{\text{water}}$  at the measured temperature.

The crossflow ( $u_{cr}$ ) was calculated from flow measurements.

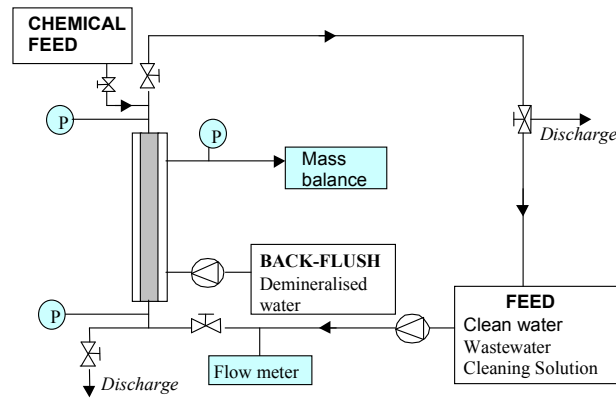


Figure 1: Schematic drawing of the filtration installation [26].

The experimental activities included filtration tests and physical-chemical analyses (to the feed water and permeate). A defined protocol was used to perform all the filterability tests [26].

As feed water raw sewage and effluent from primary clarifiers was used [26, 27, 29]. Samples were collected at the Wastewater Treatment Plant “De Grootte Lucht” in Vlaardingen, the Netherlands. Raw sewage was collected at the inlet, down stream of the rough screening and the effluent of primary clarifiers at the inlet of the aeration tank. Both wastewaters were then filtrated through a 0,56mm sieve. Short-term experiences where performed to evaluate the effect of  $u_{cr}$  and TMP on the filtration process and log-term experiments where performed to assess the possibility of continuous filtration. Batch filtration tests (with constant TMP or constant J at low fouling conditions) where performed to define process parameters for continuous filtration.

The main relevant conclusions were as follows:

- direct ultrafiltration of raw sewage and primary effluent results in the same fouling mechanism;
- changes in the feed water quality (and in the applied TMP and  $u_{cr}$ ) affect the characteristics of the forming cake layer;
- filtration of the primary effluent results in smaller resistance increase and higher permeate production;
- sustainable operation (when J, TMP and R change with time but do not inhibit continuous operation) is possible, for both wastewaters;
- fouling can not be avoided completely but can be controlled;
- After two hours the initial increase in reversibility becomes negligible (with exception of PACl).

In order to obtain a purely physical-chemical treatment of raw wastewater the addition of metallic coagulants and organic cationic polymer was tested [30]. For the applied conditions and dosages all coagulants produced similar effects. In the forming flocs only a limited fraction of COD and potential foulants was aggregated so the increase in filterability was between 20 to 35% maximum.

## 4- Fouling rates comparison between raw sewage filtration and MBR filtration

### 4.1 Raw sewage fouling rates

At DUT raw sewage trials have been performed as described in point 3.

The main goal of these experiments was not to assess fouling but to control it, through specific operational parameters, as a way to enable continuous treatment of raw wastewater. Operating conditions included imposed TMP or  $J$ , as the driving forces for permeation and fouling, and  $u_{cr}$  and backflush as “fouling removal agents”. Even if measuring fouling was not the main goal, fouling had to be dealt with in order to obtain the best strategy to minimize it. Fouling was characterized in terms of filterability and reversibility: filterability as the resistance increase over time that corresponds to fouling formation and reversibility as the possibility to remove fouling through hydraulic cleaning. Variations of TMP and  $u_{cr}$  affect the global resistance increase ( $\Delta R$ ) and the slope of  $R(t)$  [26].

Filterability tests were performed with raw wastewater. Results of filterability tests with primary clarifier effluent as wastewater are also shown to provide a base for comparison. The wastewaters were filtrated at the same conditions and tests were mainly performed at constant TMP [26]. An example of typical  $J$  and  $R$  development, at constant TMP, is shown in Figure 2.

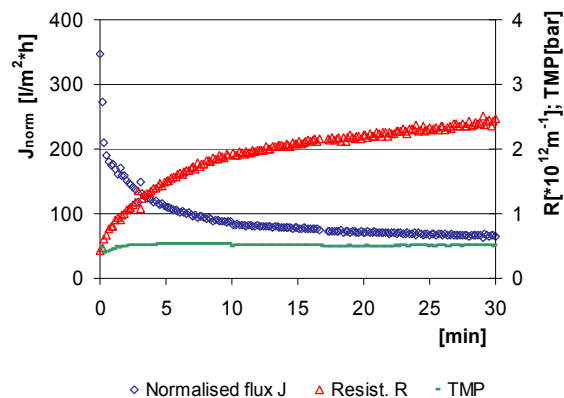


Figure 2: Typical  $J$  and  $R$  development during 30 minutes filtration at constant TMP [26].

During short-term tests the decreasing rate of  $J$  was very high in the first 5 to 10 minutes and decreased with time, therefore,  $R(t)$  increased more rapidly in the beginning of the test. This behaviour was consistent with the possibility of formation of a gel layer and eventually cake layer in the first minutes of the experience as a consequence of feed water quality and high fluxes applied. Operational conditions as well as production rates, during 30 m of filtration, are shown on Table 4.

Regarding the occurrence of fouling  $\Delta R$  was usually smaller during filtration of primary effluent but, along the whole test period, it was possible to obtain the same  $R(t)$  for both wastewaters while providing the same operational conditions.

Table 4- Production rates (L/m<sup>2</sup>.h) during 30 minutes of filtration [26].

TMP (bar)	0.3			0.5			1		
	2	1.5	1	2	1.5	1	2	1.5	1
Raw Sewage	94±25	83±18	70±9	102±3	97±2	81±10	110±9	98±9	79±4
Primary Effluent	168±11	140±20	91±18	167±2	129±10	97±19	143±10	117±2	96±11

The differences in filterability between the two wastewaters are reflected in the production rate. From table 4 it is visible that ultrafiltration of primary effluent, compared to raw sewage, results in an increase of 20 to 70% in the production rate. Also, increasing the applied TMP results in increasing production with raw sewage but has the opposite effect with primary effluent.

The tests described in paper [26] were repeated and extended. Experiments at constant TMP and constant J were performed with raw sewage aiming to obtain stable operation [27, 29]. With the same goal in mind, tests with constant J where preceded by a critical flux measurement. Imposed J was significantly below critical flux. The filtration cycle was set at 10 minutes filtration followed by 1 minute backflush in order to reduce fouling. Total duration of each test was variable but extended between 1 and 7 hours.

Tests with constant TMP showed that, with raw wastewater and starting from a clean membrane, fouling was irreversible during the first two or three cycles. After 30 minutes, resistance per cycle stabilized. This behaviour took place in short tests (1 h) and longer tests (7h). Operational conditions where TMP of 0.3 bar and  $u_{cr}$  of 2 ms<sup>-1</sup>. When the filtration cycle was varied the same results where obtained. After the first 30 to 50 minutes of filtration the additional resistance build-up during each cycle was reversible. When the filtration was performed without backflush the overall resistance reached higher values and did not stabilize.

Sustainable operations where obtained for constant TMP and constant J. Filtration tests at constant TMP showed a higher productivity.

In [30] filterability of raw wastewater was assessed by the total resistance increase produced during 30 minutes of filtration ( $\Delta R_{30}$ ). Values obtained for raw wastewater where in the range of 1.2 to 0.5 \* 10<sup>12</sup> m<sup>-1</sup>.

## 4.2 MBR fouling rates

DUT has developed the Delft Filtration Characterization method (DFCm) [31]. The main goal of this method is to provide a technique to characterize the activated sludge fouling properties. Operation conditions where defined to obtain fouling reactions while performing filterability measurements. In this case filterability is the resistance increase over volume that corresponds to fouling formation.

In DFCm activated sludge is filtrated at constant J,  $u_{cr}$  of 1 m/s, until TMP reaches approximately 0.7bar. The monitoring standard flux used is 80 L/m<sup>2</sup>.h and the total duration of the test is less than 30 minutes (25 minutes maximum). Tolls like backflush are not used because the goal is not to provide continuous filtration but to efficiently assess fouling characteristics so during the filtration test permeate is being extracted continuously. The filtration tests are performed in the Delft filtration Characterization installation (DFCi) described in point 5.

The DFCm has been applied during one year, on a weekly basis, in the full scale MBR of Heenvliet, The Netherlands (Waterboard Hollandse Delta - WHD) [32] and, through shorter campaigns, in the full scale MBR of Tilburg and pilot plants of Massbommel, Beverwijk and Hilversum, all of them in

The Netherlands [31, 33]. The result of each filtration test is a curve representing the increase in resistance *versus* the produced permeate volume by membrane surface. As a fouling indicator the additional resistance after 20 L/m<sup>2</sup> of permeate production is used ( $\Delta R_{20}$ ). This value corresponds to 15 minutes after the start-up of the filtration test. In Figure 3 some examples of the filtration curves obtained with the DFCm are shown as well as the values of  $\Delta R_{20}$  during one year of activated sludge monitoring in Heenvliet MBR.

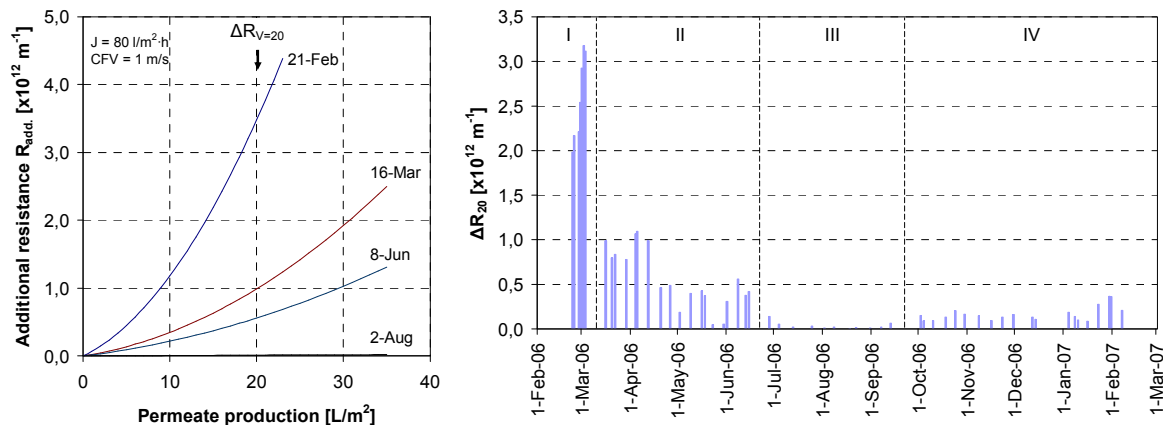


Figure 3: Examples of filtration curves (a) and  $\Delta R_{20}$  evolution (b) in Heenvliet MBR [32].

As can be seen from figure 3 (b) the filterability after the start-up of the MBR presented his worse values (period I). During March 2006 filterability began to improve, with a rather variable behaviour, but in general improving trend continued (period II). During the summer months filterability was the best of the whole year (period III). In October filterability got worse but could be still classified as a “good” filterability (period IV).

Results regarding filterability tests in the pilot plants of Massbommel, Beverwijk and Hilversum revealed that Hilversum showed the best filterability behaviour while Beverwijk was the worst of the three [31]. As far as Tilburg MBR is concerned the  $\Delta R_{20}$  was between  $0.2$  and  $0.4 \times 10^{12} \text{ m}^{-1}$ , which compared to the results of Heenvliet MBR, corresponds to the behaviour found in period IV.

#### 4.3 Comparison between raw sewage and MBR fouling rates

In order to allow comparison between raw sewage and MBR fouling rates the filtration curves obtained in February 2007 from Heenvliet MBR were used to estimate the  $\Delta R_{30}$  (resistance increase produced during 30 minutes of filtration). As mentioned before, in point 4.1, this value was used in raw sewage trials as a fouling indicator in terms of filterability. The chosen period for estimation of  $\Delta R_{30}$  corresponds to the period where the Heenvliet MBR sludge had “good” filterability, according to the DFCm.

The estimations of  $\Delta R_{30}$  for MBR sludge results in values between  $1.1$  to  $0.7 \times 10^{12} \text{ m}^{-1}$  so in the same order of magnitude as the values obtained for raw sewage trials ( $1.2$  to  $0.5 \times 10^{12} \text{ m}^{-1}$ ). These results do not reflect the behaviour in Table 4 where it is shown that the same method applied for raw sewage and primary effluent resulted in higher productivity for primary effluent.

The direct comparison between the DMF (applied in the raw sewage trials) and the DFCm (applied in MBR trails) must be considered carefully. The aim of DMF is to provide a single-step treatment using raw wastewater therefore the goal is not to assess fouling but to control it. Operational parameters were selected according to this objective so the optimal operational parameters where constant TMP of

0.3 bar, crossflow velocity of 1.5 m/s with a cycle of 10 m extracting permeate followed by 1 minute backflush. The productivity analyse of the tests was made within 30 minutes after the start-up in a total duration of 7 hours maximum. The aim of DFCm is to provide a technique to characterize the activated sludge fouling properties. Operation conditions are defined to obtain fouling reactions while performing filterability measurements, therefore standard measurements are performed using constant flux of 80 m<sup>2</sup>/L.h, crossflow velocity of 1 m/s with continuous extraction of permeate in a test that lasts less than 30 minutes. Because of all these differences fouling rates of DMF and DFCm trials can not be directly compared.

## 5- Filtration characterization tests (Delft Filtration Characterization method DFCm) on raw sewage

The DUT is planning to perform tests with raw sewage applying the Delft Filtration Characterization method DFCm [31]. Tests will be performed using the Delft Filtration Characterization installation (DFCi). The schematic drawing of the DFCi is shown in figure 4.

The heart of the filtration unit is a single tubular side stream X-Flow UF membrane with a length of 95 cm, a diameter of 8 mm and a nominal pore size 0.03  $\mu\text{m}$ . The measuring protocol consists of three steps: (a) clean water resistance determination, (b) sludge filtration and (c) membrane cleaning. The main step of the protocol is the sludge filtration: with a peristaltic pump an activated sludge sample is recirculated with a cross-flow velocity of 1.0 m/s, while a second peristaltic pump extracts permeate at a constant flux rate of 80 L/m<sup>2</sup>·h. During the experiment the transmembrane pressure TMP (Pa), sludge/permeate temperature T (°C) and flux J (m/s) are monitored. Permeate viscosity  $\eta$  (Pa·s) can be assumed equal to pure water. From these parameters the filtration resistance R (m<sup>-1</sup>) is calculated according to Darcy's law.

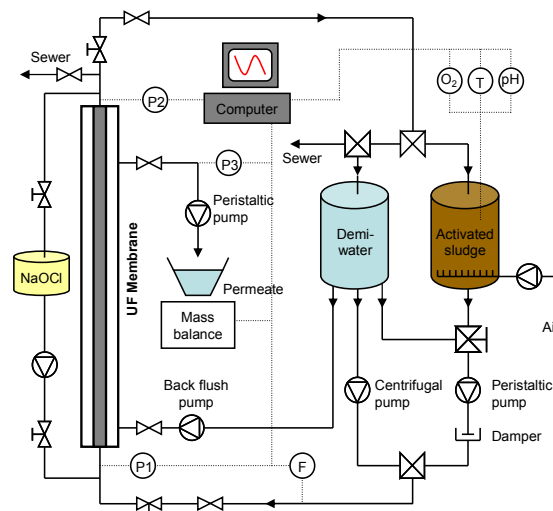


Figure 4- Schematic representation of the Delft Filtration Characterization installation (DFCi).

Filtration tests with raw sewage will be performed in parallel with filtration tests using activated sludge from membrane tanks of a full-scale MBR. The results will allow direct comparison between raw sewage and MBR activated sludge filterability.

Simultaneously, physical-chemical parameters will also be determined, namely, total suspended solids and soluble microbial products (proteins and polysaccharides). Total suspended solids will be measured according to standard methods, proteins according to Frølund et al. (1996) and polysaccharides according to Dubois et al. (1956).

## 6- Filtration characterization tests during biological processes

The DUT has performed filtration characterization tests during the biological process applying the Delft Filtration Characterization method DFCm. Samples were collected in different tanks of Heenvliet MBR, The Netherlands (Waterboard Hollandse Delta- WHD). The Heenvliet wastewater treatment plant was upgraded to membrane bioreactor through an innovative hybrid concept (the Heenvliet Hybrid MBR concept <sup>TM</sup> Witteveen+Bos and WSHD) (see WP8 Activity Report- Year 1).

Filtration tests followed a defined protocol [31] shortly described in point 5. Figure 5 shows the  $\Delta R_{20}$  (additional resistance after 20 L/m<sup>2</sup> of permeate production) obtained for filtration tests with activated sludge collected from the aerated tank and anoxic tank of Heenvliet MBR.

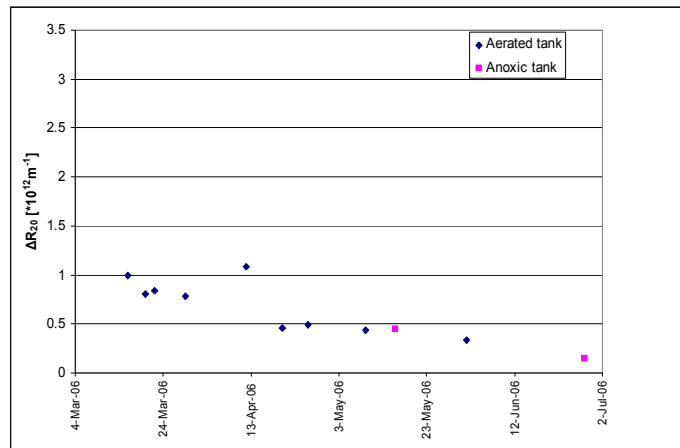


Figure 5-  $\Delta R_{20}$  from samples collected in the aerated and anoxic tank of Heenvliet MBR.

When comparing the results of figure 5 to the results shown in figure 3 (b) (obtained from filtration measurements in the membrane tank) for the same time period, the resemblance is evident. The filterability in the sampled tanks follows the same trend as the one obtained for the membrane tank.

To help clarify possible and expected differences between the different compartments of the Heenvliet MBR samples were collected from all the tanks on the same day and tested for filterability. The present layout of Heenvliet wwtp results from the upgrade of a wwtp with an activated sludge treatment [34] so samples from the carousel were also measured to provide comparison. Preliminary results from the filtration curves obtained for the different compartments of the MBR and carousel are shown in figure 6.

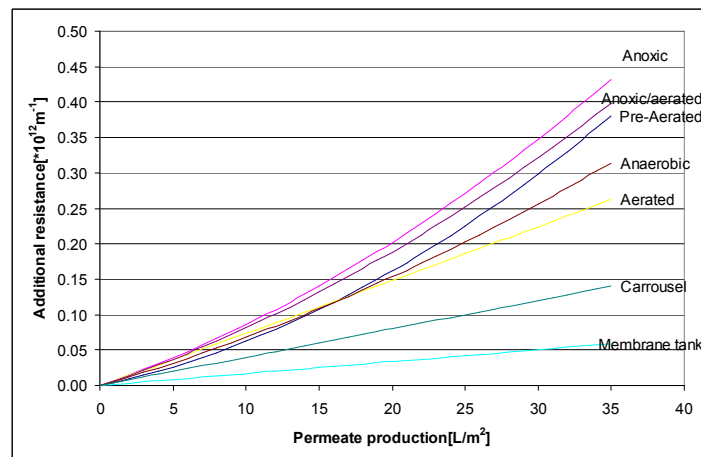


Figure 6- Filtration curves from samples collected in the different compartments of Heenvliet MBR.

Results show that activated sludge from the membrane tank has better filterability than activated sludge from conventional treatments and that along the different compartments of the hybrid MBR of Heenvliet activated sludge characteristics are changing until an improved sludge, in terms of filterability, is obtained in the membrane tank.

The adopted procedure, of sampling in the same day the different compartments of the MBR, will be repeated in future tests to extend results.

## Evaluation

From the literature review presented, mainly provided by studies with industrial wastewater, it is possible to conclude that effect of pollutants and contaminants on the biomass, and consequently on bioreactor performance is an accepted concept and is pushing the scientific community in search of other treatment solutions. Industrial MBRs are gaining acknowledgment and the MBR process is being tested for a wide variety of wastewaters. In a few cases MBR, or its optimization, seems to be the solution. Most of the industrial wastewater treatment solutions with MBR rely however on the set-up, biomass adaptation and/or specific microorganism species to obtain satisfactory results. The biomass adaptation to the wastewater quality seems an important factor mentioned in almost all the papers consulted. In general, recent experiments with industrial MBR's, make use of microorganisms capacity to degrade and utilize various toxic compounds and increase their rate of biodegradation by the use of acclimatized microorganisms and/or genetically improved bacteria.

Experiences from MBR network partners confirm the toxic effect of specific components on MBR performance. These situations are not very usual in full-scale installations but may occur due to the presence of uncommon substances. Salt intrusions, specific industrial wastewater components and changes in substrate were identified as specific foulant agents.

Direct membrane filtration (DMF) of raw municipal wastewater is possible even if in a limited scope. DMF assures a partial treatment, by the removal of particles, bacteria and turbidity. This process may be used as a main polishing step of wastewater (as an alternative to biological treatments) or to partially concentrate the wastewater (by extracting permeate and returning the concentrate to the sewer).

The application of DMF to raw sewage and primary effluent wastewater provided some results about the differences in filterability between these two wastewaters. Primary effluent wastewater has a better filterability than raw sewage.

The direct comparison between fouling rates provided by DMF and Delft Filtration Characterization method (DFCm) should not be done. The applied methods have different goals that led to the set up of different operational conditions. In order to provide comparable results DUT will perform raw sewage filterability tests using the DFCm.

Filtration tests (with the DFCm) are being performed in the different compartments of the full-scale Heenvliet MBR(WHD). The preliminary results obtained so far show that activated sludge from the membrane tank has better filterability than activated sludge from conventional activated sludge treatments (samples collected also in wwtp of Heenvliet). Along the different compartments of the hybrid MBR the activated sludge characteristics are changing until an improved sludge, in terms of filterability, is obtained in the membrane tank. These tests will be continued to obtain further insight about the influence of reactor configuration on MBR operation.

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## **Annex I**

### **Papers about Direct Membrane Filtration**

# ***Direct Ultrafiltration of Municipal Wastewater: comparison between filtration of Raw Sewage and Primary Clarifier Effluent***

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## **Abstract**

The increasing water demand encourages the development of non-conventional techniques for wastewater reclamation and reuse. In the treatment of municipal wastewater, direct membrane filtration is attractive because of the potential to produce high-quality water, enriched with valuable nutrients and organics, in one single step. This paper presents basic research into direct membrane filtration of municipal wastewater, focusing on filterability and fouling control. Primary clarifier effluent and raw sewage, pre-filtered on a 0.56mm sieve, were used in batch ultrafiltration experiments at constant TMP. Differences in filterability of the two feed-waters with TMP and crossflow velocity were explained as depending on the formation of a cake layer with different characteristics. Continuous filtration of both waters seems possible thanks to fouling reversibility with backflush.

**Keywords:** direct membrane filtration, crossflow, ultrafiltration, wastewater reuse, valuable in wastewater

## **Introduction**

In Western countries, treatment of wastewater of municipal origin is nowadays mostly accomplished through conventional biological processes. Coupled with disinfection and some other advanced processes, they have proved to be usually cost-effective and reliable in respect to discharge limits imposed by law. Nevertheless some specific conditions exist where biologically-based technologies are not efficiently applicable, for instance in case of extreme climates and discontinuous wastewater production during the year. In other cases, they can be not convenient for cost considerations or plant management problems related to small size.

Furthermore, in recent years water scarcity problems are increasing. Aim of wastewater treatment cannot be limited to merely achieve discharge limits anymore. In the Mediterranean countries, as well as in Australia and in the US, major attention is given to include recycling options within wastewater treatment schemes [1], in order to produce high quality effluent for different purposes. Treatment trains are becoming more specific as they are reuse-oriented [2], [3].

Within this scenario, alternatives to conventional treatments trains should be developed according to specific needs to fulfil. Membrane technology could be an answer to these needs, since different levels of purification can be achieved selecting the appropriate process [4].

## ***Direct Membrane Filtration***

Direct membrane filtration is intended to be a process where wastewater is filtrated directly on a porous membrane. Simplicity of design and maintenance (automation) and the high quality-water that can be produced would be the main advantages of such process. Filtrating raw wastewater, strong flux decline and fouling problems can be expected; therefore physical-chemical pre-treatments may be

added to enhance filtration rate and removal. In particular, sedimentation, coagulation, flotation or micro-sieving could be adequate.

In case of municipal wastewater, two different applications are foreseen, according to complete or partial water treatment. In the first case, direct membrane filtration would be the main polishing step of wastewater, as alternative to biological treatments. Given the characteristics of the produced permeate, further treatment would probably be necessary, for instance by more advanced membrane filtration. In the second case, direct membrane filtration can be used to partially concentrate the wastewater, extracting permeate and delivering the concentrate again to the sewer [5]. Water in combination with useful nutrients and organics for irrigation would be available, reducing the consumption of water from other sources and saving expensive artificial fertiliser [6].

Advantages can derive from the fact that membrane filtration is a purely physical process, with small use of chemicals involved. In particular:

Plants could be up-scaled, resized and adapted rapidly because of their modularity;

Operation could be discontinuous and meet seasonal changes in water flow.

Therefore, direct membrane filtration appears suitable for small-scale scheme for reuse, where eventual refinements would be operated on the permeate only to the extension required from the reuse purpose.

Since the actual applications of membranes in wastewater treatment are usually limited to post-biological treatments, mainly in MBRs and in the treatment of secondary effluent [7],[8], direct membrane filtration is a rather new concept. Few examples are found in literature about membrane processes where raw wastewater has been put in contact directly with membranes [9], [10], [11]. In other cases, pre-treated effluent of primary clarifier was filtrated on UF membranes [12], [13].

## **Theoretical background**

### ***Ultrafiltration***

Ultrafiltration (UF) is a membrane pressure-driven process for the separation of macromolecular solutions. Applied pressure (Trans Membrane Pressure, TMP) provides the driving potential to generate a flow through the membrane, which retains material larger than the pore size. The range of the pore size is about 1-100nm, thus suspended solids, colloids and macromolecules (for instance some proteins and polysaccharides [14]) can be retained. UF membranes constitute a complete barrier also for microbial species of health concern: Protozoan Cysts (*Giardia* and *Cryptosporidium*) and bacterial cells. Even viruses can be retained, according to the exclusion size of the membrane and eventual cake layer filtration that may occur [15].

Therefore, UF permeate is nominally particles-free and bacteria free. On the opposite, dissolved compounds and some macromolecules would be found in the permeate [7], [9].

### ***Fouling***

During the filtration process the retention of material operated by the membrane leads to accumulation on the membrane surface, resulting in concentration phenomena and eventually in the formation of a dense gel layer or a cake [16]. Material can deposit and be adsorbed within the membrane pores as well. Such phenomena are known as *fouling* and result in decline of flux with time.

The entity of fouling depends upon three main issues: operating conditions (TMP, hydraulic) feed water characteristics and membrane characteristics [17]. In particular, in [9] limiting TMP to constant operation was found at 0.4bar, and in [10] it is suggested that higher TMP would induce the formation of a cake-layer of higher density. Particle size distribution in the feed water is supposed to determine the major fouling mechanism. Cake layer filtration would be predominant when particles are larger than the pore size, while pore narrowing and pore blocking would be caused by components dimensionally comparable with the pore size [10], [11], [18], [19].

### ***Crossflow filtration***

In crossflow filtration, the feed water flows parallel to the membrane surface. Because of the pressure gradient across the membrane (TMP), part of the flow is extracted from the bulk solution as permeate, but most of the flow just passes along the membrane and is recirculated to the feed tank. Such mode of operation prevents excessive concentration of the transported material on the membrane surface and allows taking advantage of the hydraulic turbulence created by the flow. In fact a shear stress tangential to the membrane surface is created, which helps to reduce fouling phenomena.

Treating wastewater from a hotel building with tubular ceramic membranes, Kyu-Hong Ahn *et al.* [10] recognised the flux enhancement with increasing crossflow velocity. In Nieuwenhuijzen *et al.* [9] it is even suggested that in the ultrafiltration of municipal wastewater with tubular organic membranes, it would exist an optimal crossflow velocity above which no flux decreasing occurs.

### **Darcy's law**

Filtration through a porous mean can be described using the Darcy's law [19]:

$$J = \frac{TMP}{\eta \cdot R}$$

where J is the Flux, TMP is the Trans Membrane Pressure,  $\eta$  is the dynamic viscosity of the filtrating liquid and R is the total resistance to filtration.

In particular R is a general parameter that complies for the resistance to filtration offered by the membrane and by all the different kind of fouling mechanism that may occur (i.e. concentration polarisation, gel layer formation, pore blocking and adsorption). It is the so-called "Resistance in Series" model, described also in [14].

For water solutions the dynamic viscosity of the water is used. Since viscosity is dependent upon temperature, the generic Flux measured at the temperature  $T$ ,  $J_{(T)}$ , is corrected to a standard temperature of 15°C according to the following formula [20]:

$$J_{norm} = J_{(T)} \frac{(42,5 + 15)^{1,5}}{(42,5 + T)^{1,5}}$$

## **Experimental**

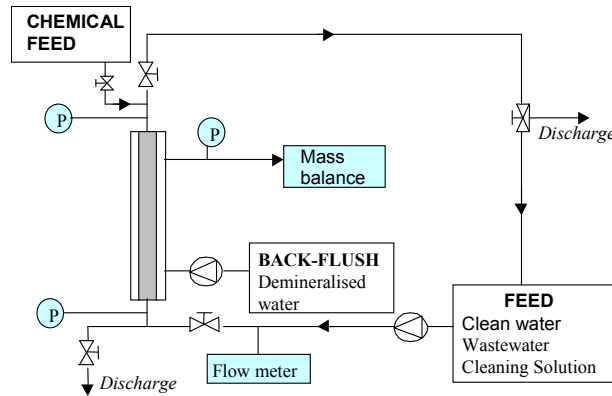
Experimental activities were carried out in two directions: filtration tests and physical-chemical analyses for water characterisation.

### **Filtration unit**

A crossflow filtration unit was built (see Figure 1), using hydrophilic tubular PVDF membranes of 5.2mm diameter (X-flow F 4385). 5 tubes were placed vertically in a 1m module resulting in a total filtration surface of 0.073m<sup>2</sup>. Clean water (tap water quality), wastewater and cleaning solutions are fed with a centrifugal pump. Chemical feed to the inner side of the tubes is also "statically" possible from the top of the membrane module, and backflush with demineralised water is provided with an independent pump. Process parameters are calculated as follows, on the basis of data automatically sampled every 10 seconds:

- TMP: pressure meters are placed at the inlet of the membrane module, at the concentrate outlet and at the permeate outlet; TMP is calculated as difference of the average pressure at the inner side and at the permeate side of the membrane.
- Flux (J): the extracted permeate is weighed on a balance, therefore the flux is calculated as mass increase with time per membrane surface;
- Resistance (R): according to Darcy's law, resistance is calculated from J and TMP assuming  $\eta_{solution}$  equal to  $\eta_{water}$  at the measured temperature.

The crossflow  $u_{cr}$  is calculated from flow measurements.



**Figure 1:** Schematic drawing of the installation rig.

### Water Analyses

Water analyses regarded feed water and permeate. As feed water, the effluent of primary clarifiers (from now on: “primary effluent”) and “raw sewage” were used. All the samples were taken at the Wastewater Treatment Plant (WWTP) De Groote Lucht, in Vlaardingen, NL. Raw sewage was collected down-stream the rough screening at the inlet of the WWTP, while primary effluent at the inlet of the aeration tank. Both waters were filtrated on a metallic 0,56mm sieve at sampling. Since three separate sewer systems deliver wastewater to the WWTP, as raw sewage a mixture of the three waters was used, according to the ratio of yearly average.

Feed water and produced permeate were analysed regarding Turbidity, Total Suspended Solids (TSS), Electro-Conductivity (EC), Chemical Oxygen Demand (COD), Phosphorous (total Phosphorous  $P_{tot}$  and orthophosphate  $PO_4^{3-}$ ) and Nitrogen compounds (total Nitrogen  $N_{tot}$  and Ammonium  $NH_4^+$ ). Measurements were performed on the feed water after the removal of suspended solids, i.e. after filtration on paper filter for TSS measurement (Schleicher & Schulle, 589<sup>2</sup>). Orthophosphates were measured after filtration over 0.45 $\mu$ m cellulose acetate filter (Sartorius). For the chemical analyses standard cuvette tests by Merck were used.

### Test Procedures

Every day about 100 litres of fresh sample were used, either raw sewage or primary clarifier effluent. In case more tests were performed on the same sample, at the end of each test the permeate was returned to the wastewater tank to avoid concentration phenomena.

Two kinds of experiments were performed: short-term and long-term tests. Nevertheless the testing procedure was made of the same following steps:

1. Measurement of membrane permeability with clean water at  $TMP=0.5bar$  and  $u_{cr}=1.5$  m/s (later referred as Clean Water Flux, CWF).
2. Regulation of  $TMP$  and  $u_{cr}$  with clean water.
3. Ultrafiltration of wastewater.
4. Cleaning.

In particular, the cleaning consisted in a rigid protocol:

- a) Membrane rinsing: 2 minutes of backflush and contemporary circulation of clean water at  $u_{cr} = 4$  m/s;
- b) Application of cleaning solution (Divos 109): 40 minutes of circulation and slow permeation;
- c) Rinsing of cleaning solution: 2 minutes of backflush and contemporary circulation of clean water at  $u_{cr} = 4$  m/s;
- d) Application of citric acid solution at  $pH=3$ ;
- e) Rinsing of citric acid: 2 minutes of backflush and contemporary circulation of clean water at  $u_{cr} = 4$  m/s;

### Short-term Experiments

Batch filtration tests at constant TMP were conducted for both the feed waters. Aim of the experiments was to evaluate the effect of  $u_{cr}$  and TMP on the filtration process, therefore different combinations of TMP and  $u_{cr}$  were tested: TMP values were 0.3, 0.5 and 1.0 bar;  $u_{cr}$  was 1.0, 1.5 and 2.0 m/s. During one day, 3 tests with duration of 30 minutes were performed on the same sample, where one parameter between TMP and  $u_{cr}$  was kept constant and the other was varied. In this way, four series of tests were performed, as represented in Table 1.

The same type of membrane was used all over the testing period, but between series B and C the membrane tubes were replaced because of damage.

**Table 1:** “Short Term” test scheme

Series	Feed water	Testing day	$u_{cr}$	TMP	Membrane
A	Raw Sewage	1	1.0 m/s	0.3-0.5-1.0 bar	1
		2	1.5 m/s	0.3-0.5-1.0 bar	
		3	2.0 m/s	0.3-0.5-1.0 bar	
B	Raw Sewage	4	1-1.5-2 m/s	0.3 bar	1
		5	1-1.5-2 m/s	0.5 bar	
		6	1-1.5-2 m/s	1.0 bar	
C	Primary Effluent	7	1.0 m/s	0.3-0.5-1.0 bar	2
		8	1.5 m/s	0.3-0.5-1.0 bar	
		9	2.0 m/s	0.3-0.5-1.0 bar	
D	Primary Effluent	10	1-1.5-2 m/s	0.3 bar	2
		11	1-1.5-2 m/s	0.5 bar	
		12	1-1.5-2 m/s	1.0 bar	

### Long-Term Experiments

As “long-term” experiments it is referred to continuous filtration tests where 10 minutes of filtration were alternated to 1 minute of backflush during 7-8 cycles. TMP was set at 0.3bar and  $u_{cr}$  at 2m/s. Aim of the experiments was to investigate if direct ultrafiltration can be operated continuously without chemical cleaning, thus to assess the fouling reversibility between successive filtration cycles. The test was executed twice on raw sewage and twice on primary effluent, during the series of tests indicated as A and C in Table 1.

## Results and Discussions

### Wastewater Characterisation

Analyses were performed about twenty times; results are reported in Table 2.

**Table 2:** Feed Wastewater analyses

		EC	Turbidity	TSS	COD	$N_{tot}^*$	$NH_4^+$	$P_{tot}^*$	$PO_4^{3-}$
		$\mu S/cm$	NTU	mg/l	mg/l	(mg/l-N)	(mg/l-N)	(mg/l-P)	(mg/l-P)
Raw	Average	1340	110.02	63	218	32.3	38.4	5.4	4.1
Sewage	St.dev.	197	37.44	41	43	5.2	12.9	0.9	1.0
Primary	Average	1042	54.62	46	135	23.8	29.9	5.0	3.6
Effluent	St.dev.	316	24.58	18	37	2.3	9.8	0.8	2.4

\*= on a smaller number of measurements

Difficulties were found for the analyses of total phosphate and total nitrogen, most probably because of interferences of the method with some compounds in the wastewater. The corresponding averages and standard deviations are calculated on few reliable measurements, thus they are not comparable

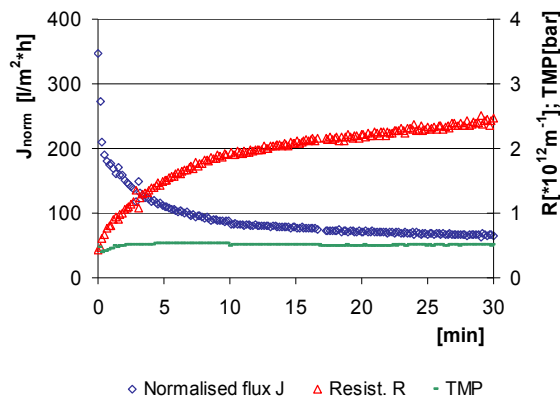
with the values indicated for  $\text{NH}_4^+$  and  $\text{PO}_4^{3-}$ . In general, in the same sample of feed water,  $[\text{P}_{\text{tot}}]$  exceeds  $[\text{PO}_4^{3-}]$  for less than 20% and  $[\text{N}_{\text{tot}}]$  exceeds  $[\text{NH}_4^+]$  for 4-15%.

### ***Filtration Characteristics at short-term tests***

Filtration characteristics are studied by plotting the flux  $J$  and the overall resistance  $R$  over time. Trends of  $J(t)$  and  $R(t)$  are similar for both the feed waters.

At  $\text{TMP}=0.3\text{-}1\text{bar}$ , rapid flux decline occurs. From values close to CWF, which means about  $700\text{l/m}^2\cdot\text{h}$  at  $\text{TMP}=1\text{bar}$  and  $230\text{l/m}^2\cdot\text{h}$  at  $\text{TMP}=0.3\text{bar}$ , during 30 minutes of filtration  $J(t)$  decreases to  $40\text{-}100\text{l/m}^2\cdot\text{h}$ . The decreasing rate is high in the first 5-10 minutes and decreases with time. Correspondingly,  $R(t)$  increases more rapidly in the first part of the test. Such behaviour can be explained assuming that the main fouling mechanism involved in the ultrafiltration of raw sewage and primary effluent is cake layer formation, as already assumed for UF and MF membranes during filtration of wastewater from a hotel building [10].

In the first minutes of filtration the accumulation of material on the membrane surface leads to gel layer formation and eventually to cake formation. This, as a consequence of the feed water quality and of the high fluxes generated at the starting of the test, when the membrane is clean. With time flux declines, the transport of material to the membrane decreases and the scouring effect of the crossflow velocity becomes more effective. Therefore, the deposition rate on the cake layer gets smaller. A typical development is plotted in Figure .



**Figure 2:** Typical  $J$  and  $R$  development during 30 minutes of filtration at constant TMP.

### ***Influence of applied $u_{\text{cr}}$ and TMP on cake layer formation***

Crossflow velocity and TMP are the main parameters suitable to control the filtration process. Their influence on cake layer formation can be followed indirectly observing the resistance increase with time,  $R(t)$ . During 30 minutes of filtration, the development of  $R(t)$  results in an overall resistance increase  $\Delta R$  and in a corresponding flux drop  $\Delta J$ .

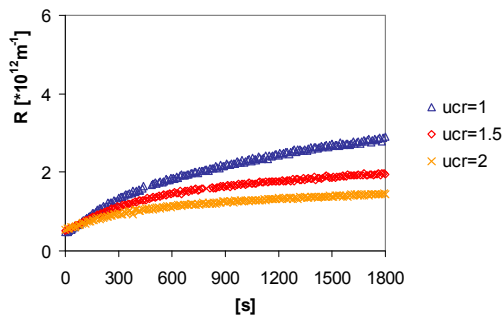
Comparing measurements from tests performed on the same sample, results agree with theoretical expectations. The crossflow velocity produces a scouring effect on the membrane walls, which helps reducing fouling phenomena. Therefore, increasing  $u_{\text{cr}}$  has always a positive effect, reducing the increase of resistance  $\Delta R$  and the corresponding  $\Delta J$ . On the opposite, increasing the applied TMP has a positive effect on the generated flux but also strong drawbacks. Indeed:

- From Darcy's law flux is directly proportional to applied TMP, therefore higher TMP results in higher starting flux  $J_{\text{start}}$ .
- High starting flux enhances the transfer of material towards the membrane, provoking severe fouling.

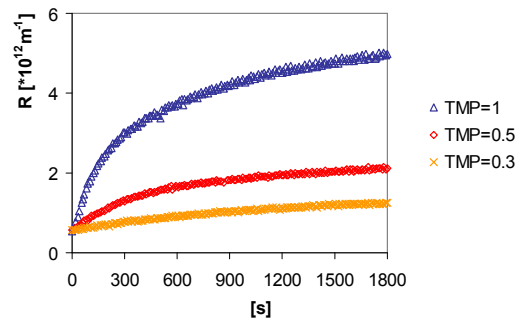
-When the cake layer is formed, it is also subject to the imposed TMP. Since the cake is a compressible layer [19], its density increases with TMP, resulting in higher resistance to filtration.

As a consequence, higher applied TMP produces higher resistance increase  $\Delta R$  and higher flux decline  $\Delta J$ . In Figure 3 and Figure 4 examples of the overall resistance increase when TMP and  $u_{cr}$  are varied filtrating the same sample are shown. It is visible that variations of TMP and  $u_{cr}$  affect the global resistance increase  $\Delta R$  as well as the slope of  $R(t)$ .

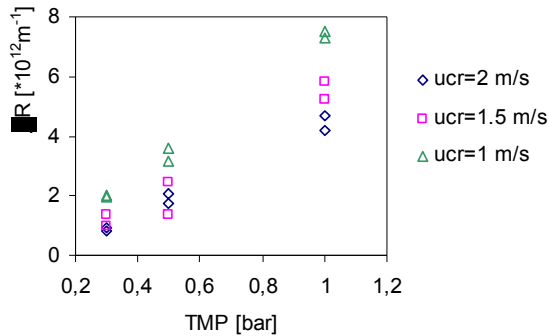
Comparing results from different samples shows that  $\Delta R$  depends upon changes in the membrane permeability and in the quality of feed water. During the testing period under discussion the membrane permeability was quiet constant, as explained in the next paragraphs. Despite changes in wastewater quality the influence of TMP and  $u_{cr}$  on the resistance increase was still comparable among different days (see Figure 5). Therefore, in the investigated ranges,  $u_{cr}$  and TMP strongly affect the cake layer formation and can be used to control fouling. In particular, filtration at TMP above 0.5bar is not recommended.



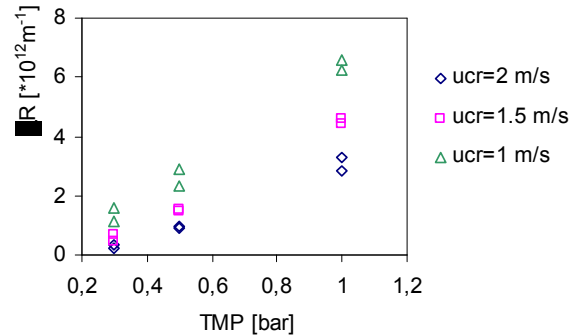
**Figure 3:** Resistance increase varying  $u_{cr}$  at TMP=0,5bar (primary effluent)



**Figure 4:** Resistance increase varying TMP at  $u_{cr}=1,5\text{m/s}$  (primary effluent)



a) raw sewage



b) primary effluent

**Figure 5:** Influence of TMP and  $u_{cr}$  on the resistance increase  $\Delta R$  filtrating different samples

### Comparison between raw sewage and primary clarifier effluent

Primary effluent and raw sewage were filtrated at the same conditions to evaluate if any difference in the filterability of the two waters exists. The two feed waters indeed differ at least for the content of particles, as the settleable fraction is removed in the clarifier. Particle size distribution has been reported to affect fouling ([10], [11], [12]), and in particular most of the interacting forces among particles in crossflow ultrafiltration of colloids are function of the particle size [20].

Considering the occurrence of fouling, from Figure it is visible that when the ratio of TMP and  $u_{cr}$  is varied, the trend of the overall resistance increase over time  $\Delta R$  is similar for the two feed waters.  $\Delta R$

is usually smaller during filtration of primary effluent, but during the testing period it was also observed that is possible that the same  $R(t)$  is recorded at the same applied conditions feeding raw sewage or primary effluent. Therefore within the changes in filterability with days, the filtration of primary effluent and raw sewage can result in the same resistance increase over time. Even if primary clarifier exhibits a slightly higher filterability, the same fouling mechanism occurs in the ultrafiltration of the two waters.

Looking at the permeate production, differences in the resistance increase reflect in remarkable differences. The production rate [ $l/m^2 \cdot h$ ], calculated as the ratio between the permeate volume produced during the 30 minutes of filtration and the membrane area, is used for comparison. In Table 3 calculated values of production rate are reported, as regarding to variations in TMP and  $u_{cr}$ .

**Table 3:** Production rates during 30 minutes of filtration of raw sewage and primary effluent.

Production rates ( $l/m^2 \cdot h$ )										
Series	TMP (bar)	0.3			0.5			1		
	$u_{cr}$ (m/s)	2	1.5	1	2	1.5	1	2	1.5	1
A,B	Raw Sewage	94±25	83±18	70±9	102±3	97±2	81±10	110±9	98±9	79±4
C,D	Primary Effluent	168±11	140±20	91±18	167±2	129±10	97±19	143±10	117±2	96±11

From Table 3 it is visible that during 30 minutes of filtration:

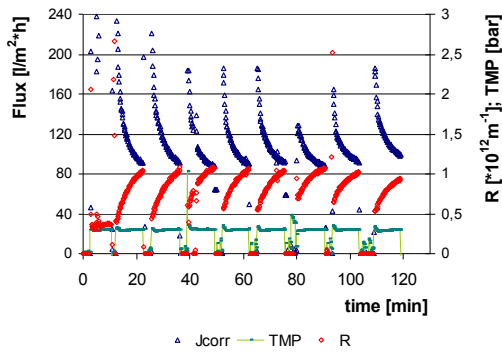
- ultrafiltration of primary effluent results in production rates 20-70% higher than during ultrafiltration of raw sewage;
- increasing the applied TMP production rates are increased in case of raw sewage and decreased in case of primary effluent. The mentioned difference of 70% indeed was measured at  $TMP=0.3bar$ .

This point represents the main difference in ultrafiltration of the two waters. To explain it, it is useful to recall that at the investigated TMP and  $u_{cr}$ , flux development with time is described by a rapid decline in the first five minutes and a slower decline in the residual time, where deposition/removal of material on the membrane surface approaches balanced conditions. Those conditions can be represented by the average flux over the last 5 minutes:  $J_{end}$ . During filtration of raw sewage, an increase of TMP corresponds to higher values  $J_{end}$ , but during filtration of primary effluent lower TMP may result in higher  $J_{end}$ . In particular, higher  $J_{end}$  at low TMP were measured for  $u_{cr}=1.5$  and  $2m/s$ , but not for  $u_{cr}=1m/s$ . It may be suggested that in case of raw sewage, a cake layer with characteristics of compactness and resistance to filtration above a certain degree is formed on the membrane surface at any conditions of TMP and  $u_{cr}$  investigated. On the opposite, in case of primary effluent, the building up of such dense cake layer is effectively tackled by the applied  $u_{cr}$ , especially at low TMP, resulting in a cake layer with less resistance to filtration, for instance because of higher porosity.

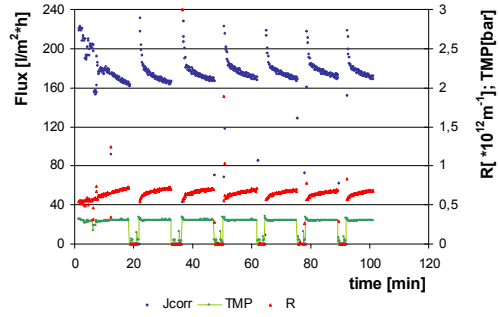
### *Filtration characteristics during long-term tests*

During long-term experiments filtration runs of 10 minutes were alternated to 1 minute of backflush with demineralised water.  $TMP=0,3bar$  and  $u_{cr}=2m/s$  were applied to both raw sewage and primary effluent. Both membrane 1 and 2 were characterised by rather high permeability ( $CWF \sim 650l/m^2 \cdot h \cdot bar$ ).

Average fluxes above  $120l/m^2 \cdot h$  and  $160l/m^2 \cdot h$  were measured respectively for raw sewage and primary effluent, confirming higher productivity in the filtration of primary effluent. The most interesting aspect is that for both feed waters, fouling reversibility was practically complete and sensible flux decline did not occur (see Figure 6). Therefore, continuous operations of direct ultrafiltration seem possible for both the wastewater and will be investigated in the future.



a) raw sewage

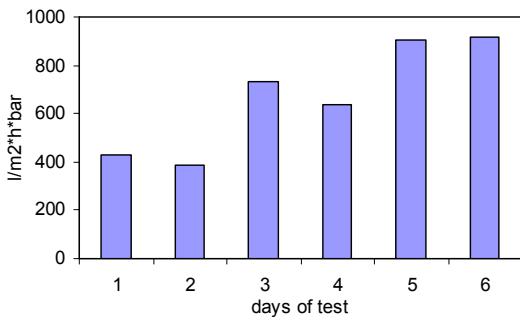


b) primary effluent

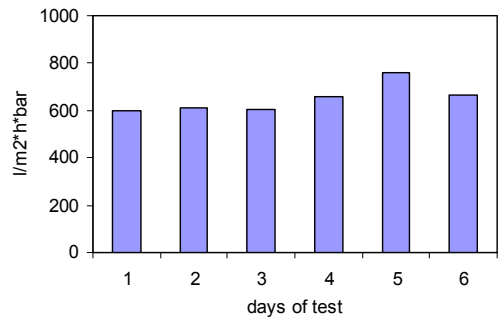
**Figure 6:** Filtration alternating 10 minutes of production to 1 minute of backflush at TMP=0,3bar, ucr=2m/s

### Membrane Cleaning

During the reported experimental activities, cleaning was intended as a tool to maintain the membrane in constant conditions. Therefore, chemical cleaning procedures were performed at the end of each experiment. When the membrane was not in use, a NaOCl solution (400ppm) was fed to the membrane from the feed-side, in order to avoid bio-fouling formation. Three cleaning products were tested: NaOCl (200ppm), Divos120CL and Divos109. The first two proved to be inadequate to maintain the membrane permeability at acceptable CWF during discontinuous testing periods. On the opposite, Divos109 showed satisfactory efficiency. In Figure 7 the permeability of membranes 1 and 2 during the testing period is shown as CWF measurements. Using Divos109 the permeability of membrane 1 was restored to 900l/m<sup>2</sup>\*h\*bar, when the nominal CWF of the clean membrane is 1000l/m<sup>2</sup>\*h\*bar. When membrane 1 was replaced with the new membrane 2, the permeability was rather constant for all the series of test with primary clarifier Effluent.



a) Series A,B: Membrane 1



b) Series C,D: Membrane 2

**Figure 7:** Membranes permeability over testing days: Clean Water Flux measurements at TMP~0,5bar

### Permeate Quality

The permeate quality is the starting point to recognise possible applications for direct ultrafiltration of municipal wastewater. Although reuse possibilities are not investigated in this paper (they will also be object of future work), results of the analyses on permeate are reported in Table 4.

The ultrafiltration membrane is a complete barrier for particles and particle-related load, while it is permeable for dissolved compounds. Therefore, suspended solids are not present in the permeate and turbidity is reduced below 1 NTU. COD and nutrients are reduced for the fraction associated to the removed particles: up to 40% in case of COD, 10% in case of Nitrogen compounds and 15% in case of Phosphorous compounds. Anyway, such figures are specific for the tested feed waters and can be

different for other wastewaters [21]. What must be remarked is that the total nitrogen concentration in the permeates is found to correspond mostly to the ammonia concentration in the influent and that the total phosphorous corresponds to orthophosphates in the influent (see Table 2).

**Table 4:** Analyses of produced permeate

		EC	Turbidity	TSS	COD	N <sub>tot</sub> *	NH <sub>4</sub> <sup>+</sup>	P <sub>tot</sub> *	PO <sub>4</sub> <sup>3-</sup>
		μS/cm	NTU	mg/l	mg/l	(mg/l-N)	(mg/l-N)	(mg/l-P)	(mg/l-P)
Raw	Average	1276	0.15	-	138	29.0	39.4	4.4	4.0
Sewage	St.dev.	207	0.13		26	3.0	11.6	0.6	0.8
Primary	Average	1036	0.10	-	78	28.1	30.3	4.1	3.4
Effluent	St.dev.	300	0.02		30	2.3	9.5	1.0	1.6

\*= on a smaller number of measurements

## Conclusions

In the investigated range of TMP and crossflow velocity, direct ultrafiltration of raw sewage and primary effluent results in the same fouling mechanism. The measured filtration characteristics are associated to the formation of cake layer on the membrane surface.

During 30 minutes of filtration, changes in the feed water quality and in the applied TMP and crossflow velocity affect the characteristics of the forming cake layer. At the tested conditions, the filtration of primary effluent results in smaller resistance increase and higher permeate production. Further research is needed to recognise the causes of the difference in filterability of the two feed waters and to optimise filtrating conditions in respect to productivity. In any case filtration at TMP above 0.5bar appears not recommendable.

During 1 hour alternating 10 minutes of production with 1 minutes of backflush, continuous operation at TMP=0.3bar and crossflow velocity=2m/s seem possible for both the feed waters. Further testing is needed to validate such preliminary results.

## Acknowledgments

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# Direct ultrafiltration of municipal wastewater: towards sustainable operations via low-fouling conditions

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## Abstract

Batch filtration tests are conducted on raw sewage of municipal origin and primary clarifier effluent, to investigate if stable operations are possible during crossflow direct ultrafiltration of raw wastewater. Experiments are run either at constant transmembrane pressure (TMP) and constant flux (J). To minimise fouling occurrence, so-called low-fouling conditions are applied: during constant TMP tests, TMP is fixed at low values (0.2-0.3 bar); during constant flux test, a sub-critical flux is imposed. For the investigated duration of some hours, the selected strategy appears successful, and stable filtration is obtained. Fouling cannot be avoided but can be controlled. At constant TMP = 0.3 bar and cross flow velocity ( $u_{cr}$ ) = 1.5 m/s, net-flux around 70 l/m<sup>2</sup>h are produced and the same is obtained at constant flux at  $u_{cr}$  = 2 m/s.

## Keywords

Critical flux; crossflow; direct ultrafiltration; fouling; fouling reversibility; raw municipal wastewater

## INTRODUCTION

Micro- and ultrafiltration applications in wastewater treatment are mainly limited to filtration of secondary effluent and membrane bioreactors (MBR); nevertheless, the increasing water demand encourages the development of other non-conventional techniques. Direct ultrafiltration is an interesting idea particularly appealing in respect to wastewater reclamation and reuse. Indeed, despite costs associated with membrane filtration, high quality water free of bacteria and particles can be produced in one single step.

Literature review shows that treatment of wastewaters from buildings with micro- and ultrafiltration membranes was studied to produce water for toilet flushing (Ahn *et al.*, 1998; Ahn and Song, 2000). Ramon *et al.* (2004), investigated ultra- and nanofiltration of low loaded greywater from a sport centre for non-potable on site reuse. Van Nieuwenhuijzen *et al.* (2000), suggested irrigation reuse for the ultrafiltrate of municipal wastewater. Analogous research were conducted on primary effluent (Abdessemed and Nezzal, 2002; Sethi and Juby, 2002).

All the reported examples show that the development of direct membrane filtration of wastewater is still at an experimental stage. This paper deals with crossflow ultrafiltration of municipal wastewater, aiming to move forward in the knowledge about the effect of operating conditions on fouling formation. Experiments with duration of some hours are conducted to identify what operating conditions would allow continuous operations. In regard to this point, the term *stable* operation is used to indicate that the monitoring parameters (J, TMP and resistance R) do not change with time, while *sustainable* operation is used when J, TMP and R change with time but not in a way to inhibit continuous operations. Obviously, in order to achieve sustainable operations, J, TMP and R must get stable within a certain range.

### *Theoretical background*

Filtration through a porous media is usually described by Darcy's law:

$$J = \frac{TMP}{\eta \cdot R}$$

where J is the permeate flux, TMP is the Trans Membrane Pressure,  $\eta$  is the dynamic viscosity of the filtrating liquid and R is the resistance to filtration. In particular, R is the sum of the resistance to filtration offered by the membrane itself and the additional resistance that may be induced by occurring fouling. In this paper, if not differently specified, R stands for the total resistance.

Filtration can be conducted in two different modes: at constant TMP or at constant flux. Apparently, in crossflow micro- and ultrafiltration fouling occurrence is deeply influenced by the operational mode. Using different synthetic wastewaters, Field *et al.* (1995) suggested that during constant flux operations fouling can be reduced by operating below a certain threshold value, called critical flux ( $J_{cr}$ ). Defrance and Jaffrin (1999) confirmed Field's suggestion stating that in a MBR constant flux mode is preferable to constant TMP mode as it avoids overfouling during the initial stage of filtration. Using Lactalbumin particles Vyas *et al.* (2002) reported that operations at constant  $J \sim J_{cr}$ , provoke more internal fouling whereas operations at constant TMP result in more superficial fouling. Lianfa Song (1998), showed that fouling is created only when TMP overcomes a certain critical pressure. Ravazzini *et al.* (2004), investigated the effect of TMP and crossflow velocity on fouling formation during ultrafiltration of raw sewage and primary effluent. However, fouling in crossflow system can be regarded as a problem of balance of forces on the particles in suspension (Fred Fu and Dempsey, 1998) or as a problem of reaching equilibrium in deposition and removal of particles on the fouling layer (Song, 1998).

## **MATERIAL AND METHODS**

### ***Filtration set-up***

Experiments are performed in crossflow on an ultrafiltration PVDF membrane with nominal pore size of 30 nm (X-flow F 4385). 5 tubes of 5.2 mm diameter are cast in a 1 m module, resulting in a total filtration surface of 0.073m<sup>2</sup>. Clean water, wastewater and cleaning solutions can be fed to the membrane module by a centrifugal pump: clean water consists of demineralised water added of MgSO<sub>4</sub> to enable flow measurement; wastewater is pre-filtered on a 0.56 mm sieve; the cleaning solution is DIVOS 109 (Johnson-Diversey). Backflush with demineralised water is also possible, provided by an independent pump.

Automatic data acquisition via computer allows a real-time calculation of TMP, J and R. TMP is calculated by pressure measurements and J by permeate mass measurements on a balance. Resistance (R) is calculated from J and TMP according to Darcy's law, assuming  $\eta_{feed}$  equal to  $\eta_{water}$  at the measured temperature.

During filtration calculated J is the extracted permeate flux, thus the gross-flux. during constant flux operations, this corresponds to the net-flux as well. During constant TMP operations, water consumption for the backflush must be taken into account and net-flux is calculated as produced permeate volume minus backflush volume per unit membrane surface. The backflush volume is calculated on the basis of the Darcy's law, where TMP is imposed by the backflush pump (around 0.75 bar) and R is assumed constant and equal to the value measured right after the backflush.

The crossflow velocity  $u_{cr}$  is calculated from flow measurements.

### Batch filtration tests

Raw sewage and primary clarifier effluent from a Dutch treatment plant (“De Groote Lucht”, Vlaardingen), are used as feed waters. Batch filtration tests are conducted either at constant TMP and constant J, following a standard procedure. At first, the membrane permeability is measured as Clean Water Flux (CWF) at TMP = 0.5 bar and  $u_{cr} = 2$  m/s, using demineralised water. Afterwards, the filtration experiment is performed using a 100 l sample. Finally, the membrane is cleaned by backflushing and rinsing with demineralised water and applying the cleaning solution for 40 minutes. If a second test is performed on the same sample, at the end of the first experiment the permeate is returned to the feed tank and the procedure is repeated. During the periods of no-use the membrane is stored in NaOCl 200ppm.

Three series of tests are performed, which are depicted in Table 1, Table 2 and Table 3. In the tables tests are numbered with letters; when two tests are performed on the same sample the letter is unchanged and a subscript is added, as for instance in the case of “m<sub>1</sub>” and “m<sub>2</sub>”.

During the first series, test at constant TMP are performed with raw sewage and primary effluent. The filtrating cycle is set to 10 minutes of filtration alternated to 1 minute of backflush, according to Van Nieuwenhuijzen *et al.* (2000). Low TMP (0.3 bar) and high crossflow velocity (2 m/s) are applied, in order to reduce fouling formation and increase the chances to observe stable operations, as in Ravazzini *et al.* (2004). As encouraging results are obtained, a longer experiment is carried on with raw sewage for 7 hours.

**Table 1.** First series of test at constant TMP.

Test	Feed water	TMP (bar)	$u_{cr}$ (m/s)	duration (h)	cycle	
					filtration time	backflush time
a	raw sewage	0.3	2	1	10 (min)	1 (min)
b	raw sewage	0.3	2	1	10 (min)	1 (min)
c	primary effluent	0.3	2	1.5	10 (min)	1 (min)
d	primary effluent	0.3	2	1.5	10 (min)	1 (min)
e	primary effluent	0.3	2	1.5	10 (min)	1 (min)
f	raw sewage	0.3	2	7	10 (min)	1 (min)

During the second series of test, primary effluent and raw sewage are filtrated at constant flux. To prevent fouling occurrence, the flux to impose is selected on the basis of a quick critical flux ( $J_{cr}$ ) measurement. Permeate is extracted at constant flux for 30 minutes (lately reduced to 20 minutes) and the development of resistance and TMP is observed. If it remains constant, the extracted flux is incremented of a certain step (10, 15, or 20 l/m<sup>2</sup>h) and measurements are repeated. The series continues until an increase in resistance and TMP shows that  $J_{cr}$  has been reached. Afterwards, the membrane is chemically cleaned (or not) and the flux is set 20 or 30 l/m<sup>2</sup>h below  $J_{cr}$  for a filtration test with duration of 2 to 4 hours.

**Table 2.** Series of test at constant flux.

Test	Feed water	flux (l/m <sup>2</sup> h)	$u_{cr}$ (m/s)	duration (h)
g	primary effluent	research $J_{cr}$	2	-
h	raw sewage	research $J_{cr}$	2	-
i <sub>1</sub>	primary effluent	research $J_{cr}$	2	-
i <sub>2</sub>	primary effluent	105	2	2
l <sub>1</sub>	raw sewage	research $J_{cr}$	2	-
l <sub>2</sub>	raw sewage	90	2	2
m <sub>1</sub>	raw sewage	research $J_{cr}$	2	-
m <sub>2</sub>	raw sewage	70	2	4.5
n	raw sewage	concentration experiment	2	4

During the experiments from “a” to “m<sub>2</sub>”, 100 l of wastewater are used to extract less than 20 l of permeate, in order to avoid concentration effects. On the opposite, during experiment “n”, it is investigated to what extension the feed water may be concentrated before operations turn from stable to unstable. Starting from 30 l of raw sewage, the flux is fixed to 100 l/m<sup>2</sup>h, in order to rapidly concentrate the wastewater; after two hours, further concentration is done at 40 l/m<sup>2</sup>h.

During the third series of test, raw sewage is filtered at constant TMP. Operational parameters such as TMP,  $u_{cr}$  and filtration cycle are varied in order to recognise “boundary” limits to sustainable operations (the ratio filtration time/ backflush time is kept constant). Each day two tests of 3 hours are performed on the same sample, modifying only one parameter per time.

**Table 3.** Second series of test at constant TMP, to optimise filtrating conditions.

Test	Feed water	TMP (bar)	$u_{cr}$ (m/s)	duration (h)	cycle	
					filtration time	backflush time
o <sub>1</sub>	raw sewage	0.2	2	3	3 (min)	18 (s)
o <sub>2</sub>	raw sewage	0.3	2	3	3 (min)	18 (s)
p <sub>1</sub>	raw sewage	0.2	1.5	3	3 (min)	18 (s)
p <sub>2</sub>	raw sewage	0.3	1.5	3	3 (min)	18 (s)
q <sub>1</sub>	raw sewage	0.3	2	3	1 (min)	5 (s)
q <sub>2</sub>	raw sewage	0.3	2	3	1 (min)	5 (s) <u>idle</u>

## RESULTS AND DISCUSSION

### Feasibility at constant TMP operations

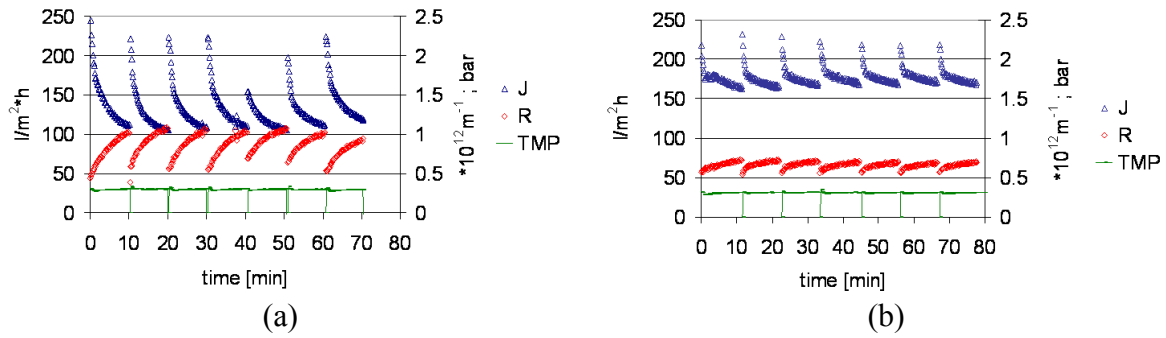
The aim is to recognise if stable ultrafiltration of raw wastewater is possible, by analysing fouling development with time. As stable operations it is meant that fouling is controlled and that resistance does not increase during filtration.

At the start of filtration, the membrane resistance is typically in the range  $0.38-0.54 \cdot 10^{12} \text{ m}^{-1}$ , slightly changing with use (the membrane resistance is maximum when the membrane is brand new). This corresponds to a clean water flux of 900-600 l/m<sup>2</sup>h\*bar.

During 1 h of test, feeding raw sewage and starting from a clean membrane, fouling is irreversible during the first 2-3 cycles. After about 30 minutes, resistance oscillations per cycle stabilise below  $1 \cdot 10^{12} \text{ m}^{-1}$  and the gross-flux typically declines from 200 to 90 l/m<sup>2</sup>h during each cycle (tests “a”, “b”).

When primary effluent is fed (“c”, “d”, “e”), resistance increases until  $0.8 \cdot 10^{12} \text{ m}^{-1}$  and gross-flux trend is between 200-150 l/m<sup>2</sup>h. Fouling appears usually reversible after the 2<sup>nd</sup> or 3<sup>rd</sup> run, except during test “e”(not shown). However, during that test the final value of resistance is about  $0.7 \cdot 10^{12} \text{ m}^{-1}$ , therefore lower than the value where it stabilises in the other cases.

The development of flux and resistance during test “a” and “d” is shown in Figure 1.



**Figure 1:** Development of resistance and flux during filtration of raw sewage (a) and primary effluent (b) at  $TMP = 0.3$  bar and  $u_{cr} = 2$  m/s.

Fouling occurs at the beginning of filtration and stabilises after a while. This behaviour suggests that first an increasing cake layer is forming and then a dynamic balance is reached in deposition and removal rates of fouling materials, because of the crossflow velocity and the backflush. Several aspects may contribute to this phenomenon:

- reduced mass transport towards the membrane with increasing resistance: as membrane fouls, flux diminishes and less material approaches the membrane;
- increased back-transport phenomenon's with increasing cake layer thickness;
- modification of size and characteristics of fouling material with time and recirculation;
- increase of temperature with recirculation: as feed water temperature increases viscosity decreases, thus flux increases and resistance apparently decreases (feed water temperature was found to rise almost linearly of  $0.5^{\circ}\text{C}$  per hour).

Independently from the actual mechanism at the basis of the results, operating at  $TMP = 0.3$  bar and  $u_{cr} = 2$  m/s allows sustainable direct ultrafiltration of raw sewage. The case of test "e" can be explained if it is supposed that the feed water of that day had a lower fouling attitude: the fouling layer build-up was so slow that it did not reach the point of equilibrium within test duration.

To test the stability of this supposed equilibrium, the same conditions are applied for 7 hours during test "f". Results confirm the observations of the short tests. Fouling stabilises rapidly, within 30 minutes. Average resistance is below  $0.7 \cdot 10^{12} \text{ m}^{-1}$ , and gross-flux oscillates between 125-105  $\text{l/m}^2\text{h}$ . With the applied scheme (10 minutes of filtration alternated to 1 minute of backflush), a net flux around 80  $\text{l/m}^2\text{h}$  is obtained. Measurements are plotted in Figure.

### Feasibility at constant flux operations

In this case, to prevent fouling, the feed water is filtrated at sub-critical flux ( $J < J_{cr}$ ).

$J_{cr}$  is found in the range 110-150  $\text{l/m}^2\text{h}$  for primary clarifier effluent and around 100  $\text{l/m}^2\text{h}$  for raw sewage. Measured values are shown in Table 4.

**Table 4:** Approximate values of critical flux for primary effluent and raw sewage at  $u_{cr}=2\text{m/s}$ .

Critical flux ( $\text{l/m}^2\text{h}$ ) at $u_{cr} = 2$ m/s					
primary effluent		raw sewage			
g	$i_1$	h	$i_1$	$m_1$	
$110 < J_{cr} < 150$	120	100	105	90	

After  $J_{cr}$  is found, 2 or 4 hours of filtration are performed at a flux 20-30  $\text{l/m}^2\text{h}$  below  $J_{cr}$ . When the membrane is chemically cleaned in between ("m<sub>2</sub>"), operations at sub-critical flux can be considered sustainable (see Figure 2). Little increase of resistance, from 0.6 to  $0.7 \cdot 10^{12} \text{ m}^{-1}$ , is observed during the first hour, but afterwards the filtration stabilises. Such

behaviour is in agreement with the findings of Defrance and Yaffrin (1999), who proposed a *weak* pragmatical concept of critical flux for the cases when the flux is set by a pump (our case): critical flux is the limit below which a stable filtration can be sustained for a long period with constant TMP. However, the resistance increase at constant flux resembles the trend observed for constant TMP operations, when it is supposed that during sustainable operations fouling occurs, but little, until an equilibrium point is reached.

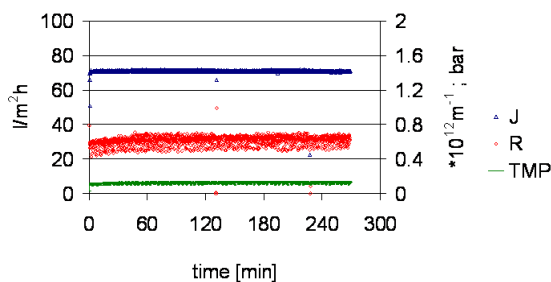
When the filtration at sub-critical flux is applied directly after that  $J_{cr}$  is found, thus without chemical cleaning in between, fouling slows down but does not stop.

It is concluded that operating in constant flux mode at sub-critical flux is a successful strategy to obtain stable operations. However, this is valid when starting from a clean membrane. Fouling can be defined as “instable” process, in the sense that when “uncontrolled” fouling is started because  $J > J_{cr}$  is imposed, stable conditions cannot be found at the same point anymore (as in Chen *et al.*, 1997).

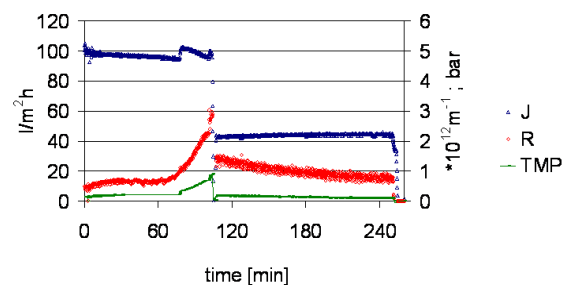
### Concentration at constant flux

Filtrating at 100 l/m<sup>2</sup>h, strong fouling occurs and resistance increases rapidly. Imposed J is clearly above  $J_{cr}$  and fouling becomes catastrophic with increasing concentration (see Figure 3). When flux is switched to 40 l/m<sup>2</sup>h, and without any chemical cleaning in between, filtration stabilises. Resistance even decreases, which is probably due the increase of temperature of the concentrated water (about 40° C at the end of the experiment). In facts the experiment is terminated when the wastewater is concentrated to 85%, because the feed water temperature is exceeding the limiting temperature for the membrane.

Apparently, even with a dirty membrane and a concentrated feed, it exists a  $J_{cr}$  below which stable operations are possible. In terms of process design, this means that concentrate of direct ultrafiltration may be treated with a cascade of concentration steps at decreasing flux. However, the issue of temperature increase with recirculation must be taken into account.



**Figure 2:** Sustainable filtration at sub-critical flux (70 l/m<sup>2</sup>h) and  $u_{cr} = 2$  m/s.



**Figure 3:** Concentration experiments with raw sewage until 85% water extraction.

### Optimisation at constant TMP operations

Beside operational stability, permeate production and energy savings are the other (conflicting) issues to be considered in the choice of the operating conditions during crossflow ultrafiltration. Therefore, a third series of test is performed to investigate the effect of TMP,  $u_{cr}$  and filtration cycle on fouling and productivity, as it is summarised in Table 5.

**Table 5: Objects of investigation during third test series.**

Parameter	TMP	$u_{cr}$	Filtration cycle
Values	0.2, 0.3 bar	1.5, 2 m/s	3'+18''; 1'+5''; 1'+5''idle
Effect	reducing TMP reduces beginning permeate production but may reduce fouling and flux decline	reducing $u_{cr}$ saves energy but may increase fouling	with time flux declines and fouling increases, reducing the filtration period may increase productivity and decrease fouling

*Fouling*

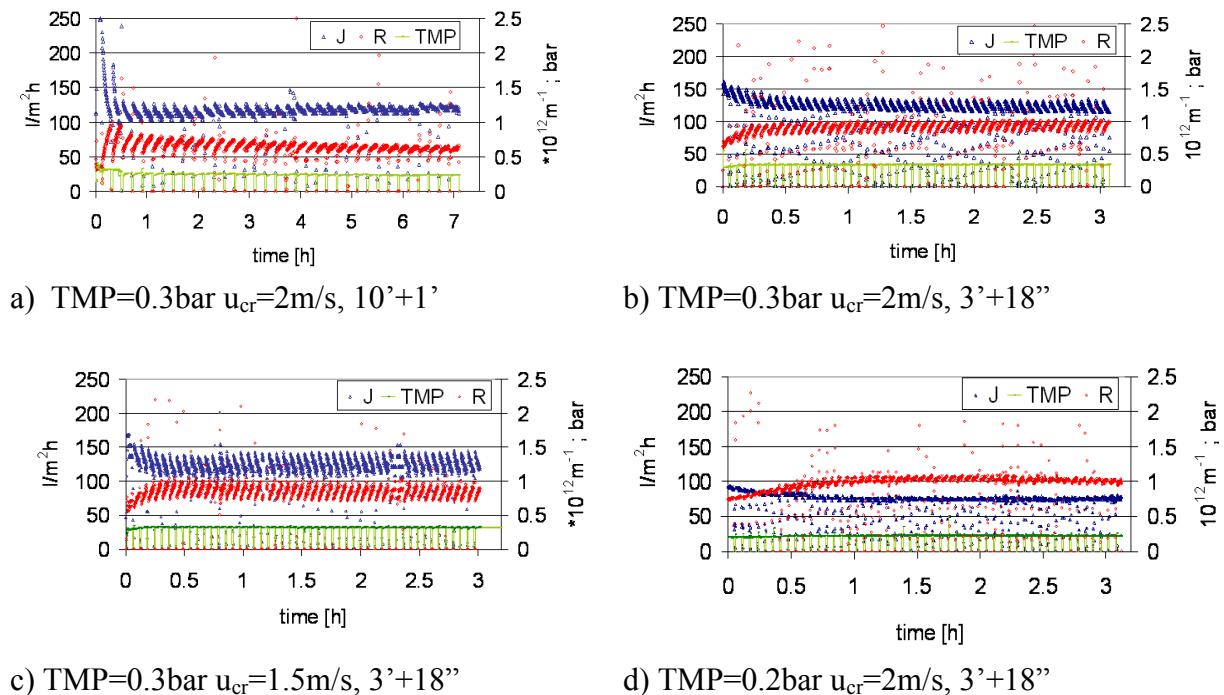
Relevant fouling occurs in any test. Starting from a membrane resistance around  $0.5 \cdot 10^{12} \text{ m}^{-1}$ , the overall resistance doubles until  $1 \cdot 10^{12} \text{ m}^{-1}$ . Usually, the resistance increase is slightly higher for the first of the two tests of the day (10-15%). As long as each day, during the first test, milder conditions are applied, this may indicate that modifications occur to the feed water because of recirculation, or because of temperature increase, or because of the removal of fouling particles that attach to the membrane during the first test and are washed out during the cleaning.

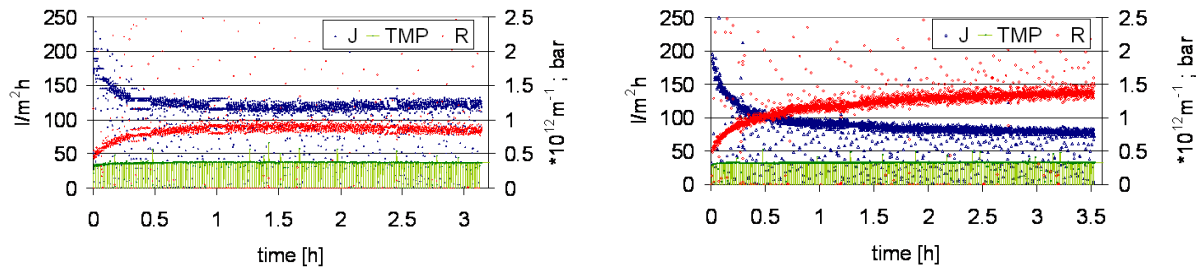
Apparently, fouling formation stops after 30-50 min of filtration. After this time the additional resistance build-up during each cycle is reversible, and the overall resistance stabilises around  $1 \cdot 10^{12} \text{ m}^{-1}$ . There are only two exceptions: a) during test “q<sub>1</sub>”, when the filtration time is reduced to 1 min and R stabilises around  $0.9 \cdot 10^{12} \text{ m}^{-1}$ ; and b) during the experiment “q<sub>2</sub>”, when in absence of backflush the overall resistance reaches the value of  $1.4 \cdot 10^{12} \text{ m}^{-1}$  and does not stabilise.

In regards to the feasibility of stable operations, it is concluded that:

- $u_{cr}$  can be reduced to 1.5 m/s;
- a smaller filtration time may help to reduce fouling;
- backflush is strictly necessary to control fouling.

Results are summarised in Figure 4.





e) TMP=0.3bar  $u_{cr}=2\text{m/s}$ ,  $1' + 5''$

f) TMP=0.3bar  $u_{cr}=2\text{m/s}$ ,  $1' + 5''$ (idle)

**Figure 4:** Comparison of filtration performance during constant TMP operations varying filtration cycle (a, b,e,f),  $u_{cr}$  (b,c) and TMP (b,d).

### Productivity

In Table 6, the measured gross-flux and the calculated net-flux per each experiment are presented. As gross-flux it is reported the flux measured after the filtration stabilises. In the calculation of average flux during backflush, resistance is assumed to be constant and equal to the values reported in the table.

The gross-flux is about  $75 \text{ l/m}^2\cdot\text{h}$  at  $\text{TMP} = 0.2 \text{ bar}$  and about  $125 \text{ l/m}^2\cdot\text{h}$  at  $\text{TMP} = 0.3 \text{ bar}$ . Therefore, operating at  $\text{TMP} = 0.3 \text{ bar}$  is more convenient in terms of productivity and does not affect stable operations. Decreasing crossflow velocity to  $1.5 \text{ m/s}$  and reducing the filtration interval, so as to backflush the membrane more frequently, did not result in any relevant variation of productivity.

**Table 6:** Summary of productivity data.

test	gross-flux ( $\text{l/m}^2\cdot\text{h}$ )	av.resistance after bf* ( $\cdot 10^{12} \text{ m}^{-1}$ )	average flux during bf* ( $\text{l/m}^2\cdot\text{h}$ )	production time (min)	backflush time (s)	net-flux ( $\text{l/m}^2\cdot\text{h}$ )
$o_1$	76	0.95	286	3	18	43
$o_2$	125	0.82	332	3	18	83
$p_1$	75	0.86	317	3	18	39
$p_2$	125	0.71	383	3	18	79
$q_1$	122	0.80	340	1	5	86
$q_2$	not stable			1	5 (idle)	not stable

\*bf = backflush

Eventually permeate saving might be realised reducing the ratio filtration time/ backflush time, which was not investigated. However, more testing is recommendable to generalise results in regards to changes of filterability of feed waters with days.

## CONCLUSIONS AND RECOMMENDATIONS

Batch filtration tests are conducted filtrating raw sewage and primary clarifier effluent of municipal origin on a ultrafiltration membrane. Experiments are run either at constant TMP and constant flux, at low-fouling conditions. Final goal is to recognise sustainable operating conditions for direct crossflow ultrafiltration. Even if further experimental confirmation is needed, some conclusions can be drawn on the basis of the measured results:

- primary effluent is slightly more filterable than raw sewage;
- fouling cannot be avoided completely;

- sustainable operations are possible for some hours either at constant TMP and constant flux, when low-fouling conditions are applied. In this case, operating in crossflow, the build up of the fouling layer stops after a while, supposedly when a balance is reached in removal/deposition of particles on the fouling layer;
- at constant TMP, the best operating conditions in respect to fouling, productivity and energy consumption are found to be  $TMP = 0.3$  bar and  $u_{cr} = 1.5$  m/s, alternating 1 min of production to 5 s of backflush. Net-flux around 70-80 l/m<sup>2</sup>h are obtained;
- at constant flux, operating at sub-critical flux leads to sustainable operations; at  $u_{cr} = 2$  m/s stable filtration is realised for 4 h at 70 l/m<sup>2</sup>h.
- fouling is an “instable” process: if the critical flux is reached and rapid fouling occurs, reducing the flux to sub-critical values helps to slow down the fouling process but is not sufficient to re-obtain stable operations. Therefore, starting the filtration from a clean membrane may help to obtain sustainable operations.

Further research will be conducted to extend results to longer durations. From an operational point of view, large attention will be given to the modifications occurring to feed water during recirculation, including the problem of temperature increase. From a theoretical point of view, the understanding and description of fouling build-up and stabilisation with time is regarded as a key feature for a future development of applications of direct ultrafiltration.

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## **Direct ultrafiltration of municipal wastewater: potential and opportunities for reuse**

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### **Abstract**

The novel concept of direct ultrafiltration for reclamation of municipal wastewater is introduced and discussed, on the basis of previous studies, actual research and existing similar processes. Advantages and disadvantages are highlighted in respect to opportunities and possible future applications. Great potential is recognised in the production of a clear bacteria-free filtrate in one single step, and in the flexibility typical of a purely physical membrane process. However, the knowledge about feasibility and cost is still limited and the process should be further developed.

### **Keywords:**

Direct membrane filtration, municipal wastewater, ultrafiltration, wastewater treatment cheme, wastewater reclamation, wastewater reuse.

### **INTRODUCTION**

Water sources are depleting, both in quality and quantity, leading to stricter discharge policy and increasing number of wastewater reuse projects all around the world (Bixio *et al.*, 2004).

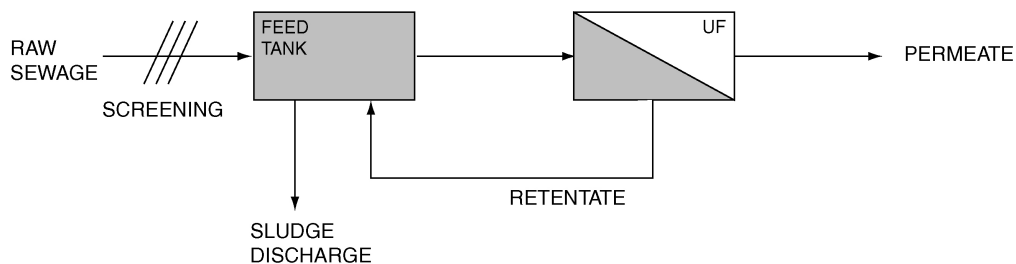
Because of its specific characteristics, membrane separation plays a significant role in these developments: membranes can provide both the rejection of harmful pathogens and clear filtrate suitable for reuse. As reported also by Wintgens *et al.* (2005), currently membranes are applied to the treatment of municipal wastewater mainly in two types of system. The first is a centralised scheme, where a double membrane system treats secondary or tertiary effluent from a conventional treatment plant, mostly using microfiltration (MF) or ultrafiltration (UF) followed by reverse osmosis (RO). The second is the application of membrane bioreactors (MBR), usually in local reuse systems. In both cases, a biological treatment precedes the application of membranes, aiming to remove COD (chemical oxygen demand), BOD (biological oxygen demand), and nutrients (Nitrogen and Phosphorous).

However, membrane separation is a purely physical process that may be considered as a stand-alone process as well. Separation and not “removal” of wastewater constituents would be realised, therefore this process has also been referred to as Direct Membrane Separation (DMS) (Ahn *et al.*, 2001). The typical limitations of biological processes (influence of temperature, feed stability and toxicity, start up period etc.) would be avoided.

In particular the concept of direct ultrafiltration of municipal wastewater is presented here. Given the absence of existing full-scale applications, advantages and disadvantages, opportunities and challenges for this novel concept are described in respect to similar applications.

## REVIEW ON DIRECT MEMBRANE SEPARATION (DMS) OF WASTEWATER

During DMS the feed wastewater is filtrated directly on a membrane, after simple mechanical pre-treatments (see Figure 1, where the example of direct UF is represented).



**Figure 1:** Schematic drawing of direct UF of raw sewage

High quality-water can be produced in one single treatment step. However, the quality of the permeate will depend on the feed water quality and the applied membranes. Strong fouling problems are likely to occur, as typically in membrane processes.

Literature review shows that the concept has often been related to reuse purposes, and in most cases the target quality of the product-water has been achieved. Different wastewater sources have been used: graywater, blackwater, manure wastewater and finally municipal wastewater, either raw or after primary sedimentation. Microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) have been applied. To the knowledge of the authors, the only full-scale application regards the treatment of pre-settled manure with MF or RO membrane, using VSEP (Vibratory Shear Enhanced Process) configuration. UF is used to concentrate the manure before digestion, and to produce a permeate that after chemical cleaning is recycled as irrigation, wash-down or cooling water; RO is used to produce drinking-water quality for the livestock and washwater for the barns (Johnson *et al.*, 2004).

All other researches have been limited to lab-scale or pilot scale for duration until one year. Ahn *et al.* (1998) and Ahn and Song (2000), treated pre-screened low loaded graywater from a resort and hotel complex with MF and UF tubular ceramic and hollow fibres membranes. They obtained an effluent satisfactory for Korean standard for secondary reuse (i.e. toilet flushing, landscape irrigation). During further research using MF hollow fibres on the effluent from septic tank of apartments and dormitory they studied the role of biological TOC removal in the equalisation and membrane tanks and decided to shift their attention to MBR systems (Ahn and Song, 1999; Ahn *et al.* 2001).

In California, on account of Orange County Water and Sanitation District, Sethi and Juby (2002) used a Memcor 6M10C pilot plant to test direct MF of primary clarifier effluent. The focus was on clarification and microbial removal, aiming to evaluate the suitability of the filtrate to direct ocean discharge (without chlorine disinfection) and further RO. The plant was stably operated during one year and results were defined “promising” for both applications.

In China, high strength wastewater from university apartments was filtrated on UF and MF ceramic and organic membranes (Hao *et al.*, 2005). The effluent was used to irrigate winter wheat in small laboratory test field. The membranes were able to remove 3-4 log of bacterial counts and 4-5 of coliforms, increasing the wheat production of 7.5% in weight in comparison to tap water enriched with fertilisers.

Other researches merely focused on the evaluation of obtainable permeate quality in respect to the reuse application. Abdessemed and Nezzal (2002) treated primary effluent by applying combinations

of coagulation, adsorption and UF in lab-scale experiments. Dosing 120 mg/l of FeCl<sub>3</sub> and 40 mg/l of PAC (Powdered Activated Carbon), they obtained a clarified filtrate with turbidity ~ 0 NTU and COD = 7 mg/l, which might be suitable for industrial applications. Ramon *et al.* (2004) tested UF and NF membranes for local reuse of graywater from a sport centre. They found that the permeate quality improves with the molecular weight cut-off (MWCO) of the membrane, and that only NF membranes would guarantee sufficient quality for the reuse applications, because of higher BOD removal.

In the Netherlands several studies focused on the feasibility of crossflow ultrafiltration of municipal wastewater either as a tool for advanced particle removal or for irrigation purposes (van Nieuwenhuijzen *et al.* 2000, Ravazzini *et al.* 2004).

In Table 1 an overview of the permeate quality achieved when MF and UF membranes were applied is given.

**Table 1: Permeate quality during MF and UF direct membrane separation of wastewater**

Ref.	Feed wastewater	Membr.	COD mg/L	BOD <sub>5</sub> mg/L	TOC mg/L	TSS mg/L	Turb. NTU	Total Coliform	Faecal Coliform	Coliphage
[8]	Primary effluent	MF		64.8		1.5	0.52	204 MPN /100mL	25MPN /100mL	838PFU /100mL
[13]	Primary effluent	UF	78			ND	<0.15			
[6]	Graywater (resort)	UF-MF	<15.6		<4.6		<0.7			
[11]	Graywater (sport centre)	UF	<80				<1.4			
[5]	Low-strength (hotel-shops)	MF	<20		<3.5	ND	0.2			
[4]	Low-strength (dormitory)	MF	11	4.1	4.8	0.2	0.01			
[9]	Low-strength (dormitory)	UF	67			ND	ND	<2000 ps/L		
[6]	Septic tank effluent*	MF	8.8	4.2	3.8	0.2	0.15			
[12]	Municipal raw	UF	210			ND	0.2			
[13]	Municipal raw	UF	138			ND	<0.3			

\*= biological removal in the tank

ND = not detected

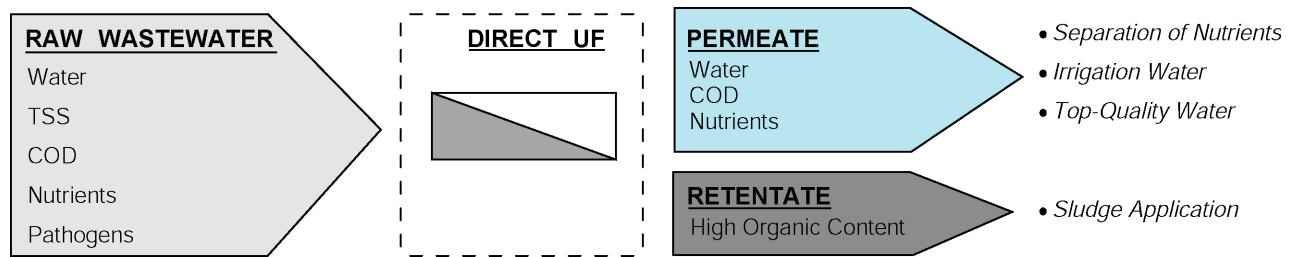
## DIRECT ULTRAFILTRATION OF MUNICIPAL WASTEWATER

In Figure 2, a concept-diagram of direct UF of municipal wastewater is presented. The raw wastewater is regarded as a mixture of valuable compounds (water and nutrients) with undesired elements (summarised as suspended solids “SS” and potentially infectious microorganisms, bacteria and viruses). The UF membrane realises the separation of the desired-undesired compounds by constituting a barrier to particulate, colloids and bacteria.

The permeate is water free of particles, microorganisms and bacteria, but rich in soluble COD, phosphorous (P) and nitrogen (N). Nutrients content may be interesting in regard to irrigation reuse, while the low turbidity and the absence of particles may enable the production of high-quality water. The concentrate contains the removed organic-rich particulate, microorganisms and bacteria, and may be treated as sludge. Given the absence of chemical addition, it may be considered for soil application.

Great advantages are recognised in respect to reuse applications:

- municipal wastewater is already a widely available water source;
- the process is purely physical/chemical, therefore operation can be discontinuous;
- high automation and remote control can be implemented.



**Figure 2:** direct ultrafiltration: separation of constituents and potential for reuse

## RESEARCH STUDIES

During our research we studied this concept feeding raw municipal wastewater and primary clarifier effluent to tubular PVDF membranes (vertically placed). Results were presented in Ravazzini *et al.* (2004, 2005) and can be summarised as follows:

- ultrafiltration of raw sewage and primary effluent on tubular organic membranes appears feasible both operating at constant TMP or at constant flux. To prevent fouling, low TMP (<0.3 bar) or flux below the critical flux were applied, at a crossflow velocity of 1.5-2 m/s (tests duration was between 3 and 7 hours);
- the difference in filterability of the two wastewaters is limited;
- during direct UF of raw sewage sustainable fluxes are in the order 70-80 l/m<sup>2</sup>h;
- permeate turbidity is in both cases steadily < 0.5 NTU;
- the nutrients content is practically unchanged by UF for both feed waters, while a large amount of COD is removed also when filtrating primary effluent (~40%).

Therefore it can be concluded that:

- direct UF is a very efficient pre-treatment when compared to primary sedimentation;
- the permeate from primary effluent has lower organic content then permeate from raw sewage but the same content of nutrients;

## APPLICATIONS AND REUSE POSSIBILITIES

Direct UF can be regarded either as end-of-pipe treatment for of municipal wastewater or as technology to generate reusable water from existing sewers. In this case, the concept of “sewer mining”, as reported in Butler *et al.* (1995), could be applied: water could be extracted on demand and the process-wastes returned to the sewer.

However, the quality of the product water greatly determines the type of application. The main characteristics of the filtrate are the following:

*Positive:* the permeate is clear (turbidity <1 NTU), particle-free, pathogens-free and rich in nutrients;

*Negative:* presence of unpleasant odours and organic components (eventually also undesirable micropollutants) which may affect stability during storage.

The following applications may be considered:

- *advanced pre-treatment:* in case of (temporarily) overloaded wastewater treatment plants (WWTPs), a reduction in the organic load can be achieved in a single step; the reject stream is treated as (anaerobic) sludge if the concentration factor is sufficient.
- *irrigation:* the presence of nutrients is valuable to increase crops productivity and realise saving on fertilizers. Blending with other water sources and confined agricultural practices can help to achieve water quality standards and protect water bodies from nutrients run-off.

- *ocean disposal*: the microbiological safety of the filtrate may enable a “sanitation” use of the process, where discharge of organic load is a secondary aspect (e.g. for small communities along the ocean coast, on ships, in emergency camps, etc.). Eventually, it can be considered also for the treatment of low loaded waters as in case of combined sewer overflow.
- *process water*: depending on the feed water quality, reuse can be considered directly or after some pre-treatment; in particular, the combination with coagulants or activated carbon (either granular or powdered) appears promising.
- *further membrane process*: direct UF permeate may represent a source for NF or RO; the dissolved organic content is high but the fraction of bacterial origin is lower than in membrane applications following biological treatment.
- *new treatment trains*: direct UF permeate is suitable for adsorptive and oxidising processes in absence of or prior to biological treatment; degradation or removal of dissolved organics (including refractory ones) could be enhanced and the biological treatment would operate on dissolved solids only.

## **COSTS**

Cost is a fundamental aspect in regard to process potential applicability. However, at least in the first stage of the development of a new technology, it should not be a strictly limiting factor. In some cases, ‘niche’ applications may result economical as a consequence of specific legislative-environmental circumstances. For instance, pre-settled manure wastewater is probably the most fouling wastewater listed in our review, but in this case direct UF has proved to be economical and is applied at full scale. Among the cited authors, only Ahn *et al.* (1998) presented calculations about energy consumption for tubular ceramic membranes. The minimum expenditure was found at 26 kW/m<sup>3</sup>, operating at 4 m/s and TMP = 0.12 bar with a MF membrane. Energy requirements for UF membranes, operating between 1 to 4 m/s at TMP between 0.12 to 0.366 bar, ranged between 53 to 125 kW/m<sup>3</sup>. At the actual Netherlands energy cost, this can be translated in 0.018 eurocents/m<sup>3</sup> for MF and 0.037-0.087 eurocents/m<sup>3</sup> for UF.

A preliminary estimation of the cost for the system we used was done, based on the operating conditions previously indicated ( $u_{cr}$  = 1.5-2 m/s, net-flux = 70-80 l/m<sup>2</sup>h). Calculations for a system of 100 m<sup>3</sup>/h indicate a total cost between 20 and 50 eurocents/m<sup>3</sup>. Such results must be considered only as a very rough estimation, as there is no real experience about flux decline and maintenance effort for a continuous system filtrating raw sewage. However, they indicate that the order of magnitude of cost of crossflow direct UF is encouraging. With time and developments on membranes and process control, this cost is expected to decrease.

## **PROSPECT**

In recent years Direct Membrane Separation (DMS) has regularly attracted attention for its interesting characteristics. In this paper, a literature review of available studies regarding the process has been conducted, focusing on MF and UF processes of wastewaters of household-municipal origin or similar. The aim was to discuss the potential of direct UF of municipal wastewater in regard to wastewater treatment and reuse. The characteristics of the process (flexibility) and of the product water appear suitable for the production of irrigation and process water, eventually on demand. Other applications may be for partial decontamination and “sanitation” of liquid wastes before discharge (e.g. to the ocean), or as advanced pre-treatment for existing WWTPs (e.g. for tourist areas with large oscillation in flow). However, the greatest challenge is still the further development of the process into a feasible option for wastewater treatment, as knowledge about operability and costs is still very little. When the feasibility is established, direct UF may even foster the development of new schemes for municipal wastewater treatment, for instance aiming to the removal of emerging pollutants.

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# Ultrafiltration of raw municipal wastewater: sustainable conditions for constant transmembrane pressure and constant flux operations

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## Abstract

The aim of this paper is to present investigation on sustainable operating conditions for direct ultrafiltration of wastewater of municipal origin. Batch filtration tests are conducted feeding raw sewage and primary clarifier effluent to tubular ultrafiltration membranes. Experiments are run at constant transmembrane pressure (TMP) and constant flux (J), at low-fouling conditions. During constant TMP tests, TMP is fixed below 0.4 bar and frequent backflushing is applied; during constant flux test, the flux is set below critical value. For the investigated duration of some hours, the selected strategies appear successful, and stable filtration is obtained. Fouling cannot be avoided but it can be controlled. Net- fluxes up 70-80 l/m<sup>2</sup>h and 60-70 l/m<sup>2</sup>h are measured at constant TMP and constant flux respectively, using crossflow velocities between 1 and 2 m/s.

## Keywords

Crossflow ultrafiltration; raw municipal wastewater; critical flux; fouling; fouling reversibility

## INTRODUCTION

In the treatment of municipal wastewater, more stringent discharge standards and increasing interest for water reuse encourage the development of new non-conventional techniques. Direct ultrafiltration is a recent idea and consists of filtrating raw wastewater directly on ultrafiltration membranes. In one single step, water free of bacteria and particles is produced. Literature review shows that the same concept was applied to treat wastewaters from buildings with micro- and ultrafiltration membranes, in order to reclaim water for toilet flushing (Ahn *et al.*, 1998; Ahn and Song, 2000). Ramon *et al.* (2004), investigated ultra- and nanofiltration of low loaded greywater from a sport centre for non-potable on site reuse. Van Nieuwenhuijzen *et al.* (2000), suggested irrigation reuse for the ultrafiltrate of municipal wastewater. Because fouling is the biggest threat to the development of the process, attempts are made not only with raw wastewater but also with partially treated wastewater: primary effluent has been used aiming to industrial reuse, ocean discharge or subsequent reclamation for reverse osmosis (Abdessemed and Nezzal, 2002; Sethi and Juby, 2002).

All the reported researches show that the development of direct membrane filtration of wastewater is still at an experimental stage. This paper deals with crossflow ultrafiltration of municipal wastewater, aiming to move forward in the knowledge about the effect of operating conditions on fouling formation. Experiments with duration of some hours are conducted to identify what operating conditions would allow continuous operations. In regard to this point, the term *stable* operation is used to indicate that the monitoring parameters (J, TMP and resistance R) do not change with time, while *sustainable* operation is used when J, TMP and

R change with time but not in a way to inhibit continuous operations. Obviously, in order to achieve sustainable operations, J, TMP and R must get stable within a certain range.

### *Influence of operational mode*

During crossflow micro- and ultrafiltration fouling occurrence is deeply influenced by the operational mode: at constant TMP or at constant flux. Using different synthetic wastewaters, Field *et al.* (1995) suggested that during constant flux operations fouling can be reduced by operating below a certain threshold value, called critical flux ( $J_{cr}$ ). Defrance and Jaffrin (1999) stated that in a MBR operations at constant flux avoid the overfouling during the initial stage of filtration mode that occurs at constant TMP. Vyas *et al.* (2002) reported that using lactalbumin suspensions operations at constant  $J \sim J_{cr}$  induce more internal fouling whereas operations at constant TMP result in more superficial fouling. Lianfa Song (1998) showed that fouling is created only when TMP overcomes a certain critical pressure. However, fouling in crossflow system is often regarded as a problem of reaching equilibrium in deposition and removal of particles on the fouling layer (Song, 1998), which depends upon feed water quality but also on operating conditions.

In a previous work we observed that during ultrafiltration of raw sewage and primary clarifier effluent there might exist an optimal range of applied TMP and crossflow velocity in respect to fouling formation (Ravazzini *et al.* 2004). Here we present the extension of that work to longer filtration times and constant flux operations. By operating at low TMP and sub-critical flux, we aim to obtain sustainable ultrafiltration of raw sewage.

## **MATERIAL AND METHODS**

### *Filtration set-up*

Crossflow ultrafiltration experiments are performed using tubular PVDF membranes with nominal pore size of 30 nm (X-flow F 4385). A 1 m module, filled with 5.2 mm diameter tubes, is connected to a recirculation loop fed by a centrifugal pump. Two different membrane modules are used, and a variable number of tubes are blocked with a rubber chord during the testing period. As a result, the total filtration surface in the reported experiments varies from 0.073 m<sup>2</sup> to 0.175 m<sup>2</sup> and 0.102 m<sup>2</sup>.

Different waters can be circulated: clean water, wastewater or cleaning solutions. Clean water consists of demineralised water added of MgSO<sub>4</sub> (to enable flow measurement); wastewater can be raw sewage or primary effluent, and is pre-filtered on a 0.56 mm sieve; the cleaning solution is DIVOS 109 (Johnson-Diversey). Cleaning operations include also backflush with demineralised water, which is provided by an independent pump.

TMP is calculated by pressure measurements, while J is calculated by permeate mass measurements (on a balance). Data acquisition is automatic and TMP, J and R are calculated in real-time. R is obtained by Darcy's law, which can be written as follows:

$$J = \frac{TMP}{\eta \cdot R}$$

where J is the permeate flux, TMP is the Trans Membrane Pressure,  $\eta$  is the dynamic viscosity of the filtrating liquid and R is the resistance to filtration. In particular, R is the sum of the resistance to filtration offered by the membrane itself ( $R_{mem}$ ) and the additional

resistance induced by occurring fouling. In this paper, if not differently specified,  $R$  stands for the total resistance and is calculated assuming  $\eta_{\text{feed}}$  equal to  $\eta_{\text{water}}$  at the measured temperature. During filtration the weighed extracted permeate mass corresponds to the gross-flux. During constant flux operations, this corresponds to the net-flux as well, whereas during constant TMP operations, water consumption for the backflush must be taken into account. The crossflow velocity  $u_{\text{cr}}$  is calculated from flow measurements and cross membrane section.

### Tests Procedures

Samples of 100-120 L of raw sewage and primary clarifier effluent are collected at a Dutch treatment plant (“De Groote Lucht”, Vlaardingen). Such a large quantity is necessary to avoid significant variation of suspended solids concentration during filtration.

Batch filtration tests are conducted with two different modes: at constant TMP and constant  $J$ . The procedure is standard. At first, the membrane permeability is measured as Clean Water Flux (CWF) at  $\text{TMP} = 0.5$  bar and  $u_{\text{cr}} = 2$  m/s, using demineralised water. Afterwards, the filtration experiment is performed. Finally, the membrane is cleaned by backflushing and rinsing with demineralised water and applying the cleaning solution for 40 minutes. If a second test is performed on the same sample, at the end of the first experiment permeate is returned to the feed tank and the procedure is repeated. During periods of no-use the membrane is stored in NaOCl 200ppm.

During tests at constant TMP, the TMP is varied between 0.2 and 0.4 bar, and  $u_{\text{cr}}$  between 1 and 2 m/s. Initially the filtrating cycle is set to 10 minutes of filtration alternated to 1 minute of backflush, but in the following it is also varied. In all the tests, the duration of the backflush period is approximately as 10% of the filtration period. Test duration ranges between 3 to 7 hours; a summary of operating conditions is reported in Table 1.

**Table 1:** Series of tests at constant TMP.

Feed water	TMP (bar)	$u_{\text{cr}}$ (m/s)	duration (h)	Cycle	
				filtration time	backflush time
raw sewage/ primary effluent	0.3	2	3-7	10 (min)	1 (min)
raw sewage	0.2-0.3-0.4	1-1.5-2	3	3 (min)	18 (s)
raw sewage	0.2-0.3	2	3	1 (min)	5 (s)
raw sewage	0.3	2	3	1 (min)	5 (s) <u>idle</u>

Tests at constant flux are executed at  $u_{\text{cr}} = 2$  m/s. A quick critical flux ( $J_{\text{cr}}$ ) measurement is used to select the operating flux. Permeate is extracted for minimum 20 minutes at constant flux, using a peristaltic pump. When the development of  $R$  and TMP remain constant, the extracted flux is incremented of 10, 15, or 20  $\text{l/m}^2\text{h}$  and a new observation takes place. The procedure is repeated until an increase in resistance and TMP shows that  $J_{\text{cr}}$  has been reached. Afterwards, the membrane is chemically cleaned (sometimes not) and the flux is set below  $J_{\text{cr}}$ , for a filtration test with duration between 2 and 6 hours.

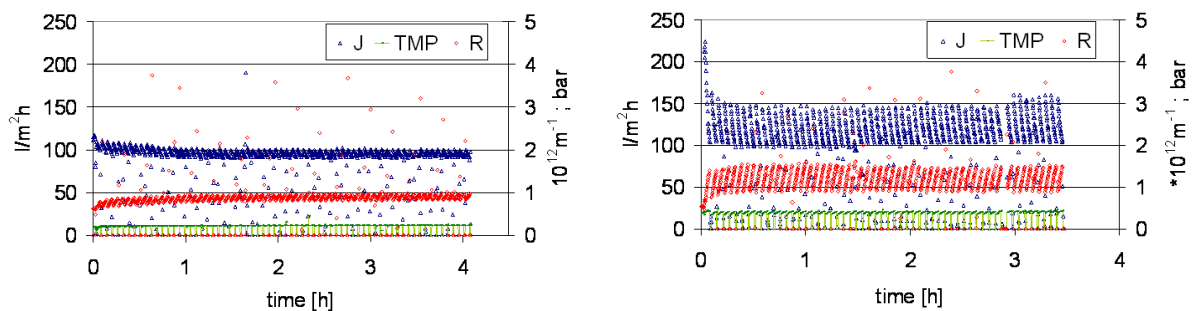
## RESULTS

### *Operations at constant TMP*

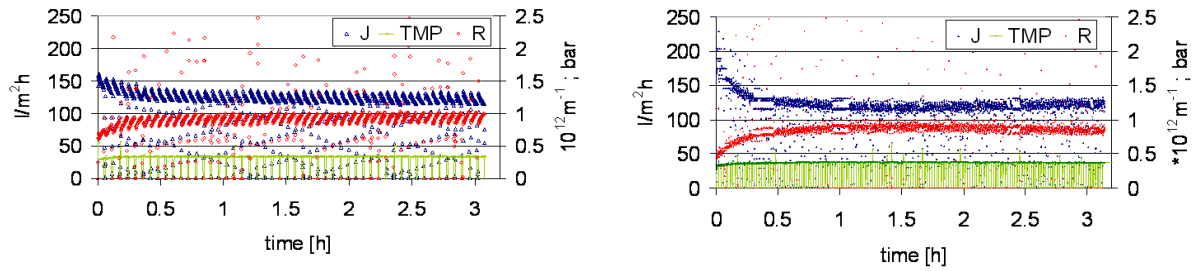
At the start of filtration, the membrane resistance  $R_{\text{mem}}$  is typically in the range  $0.38\text{-}0.54 \cdot 10^{12} \text{m}^{-1}$  for the first module and  $0.52\text{-}0.62 \cdot 10^{12} \text{m}^{-1}$  for the second.  $R_{\text{mem}}$  changes with use, being maximum when the membrane is brand new; however this seems to have negligible effect already after few minutes of filtration. Corresponding CWF are approximately in the range  $900\text{-}500 \text{l/m}^2\text{h} \cdot \text{bar}$  (measured flux at temperature between  $18^\circ\text{C}$  and  $22^\circ\text{C}$ ).

At the tested conditions, relevant fouling occurs in any test, but sustainable operations are possible with both feed waters. Apparently, fouling formation stops after 30-50 min of filtration:  $R$  increases until  $0.8\text{-}1.25 \cdot 10^{12} \text{m}^{-1}$ , then the additional resistance build-up during each cycle is reversible, and  $R$  stabilises. In regard to the effect of operational parameters on stability, it must be considered that the wastewater quality changes with days and may affect results. To limit this effect most of the times two tests of 3 hours are performed on the same sample, modifying only one parameter per time. Conclusions are drawn mainly on these direct comparisons, the most relevant observations being the following:

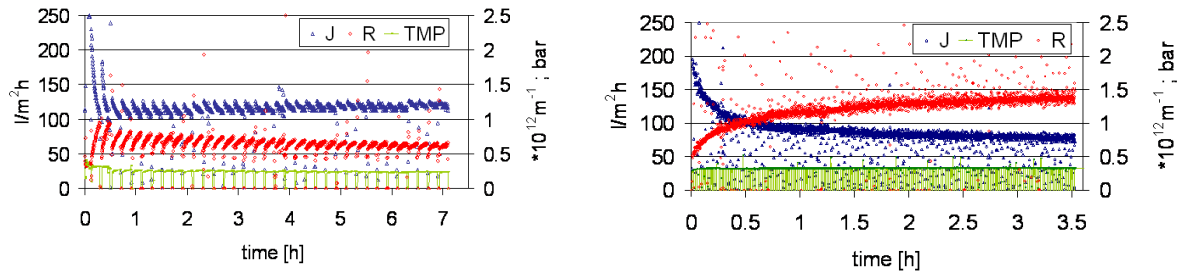
- average fluxes obtained with primary effluent are 20-30% higher than with raw sewage (not shown);
- increasing TMP increases fouling formation and productivity at any crossflow velocity (Figure 1);
- increasing crossflow velocity decreases fouling formation (not shown);
- reducing the filtration time may help to reduce fouling (Figure 2);
- the sustainability showed during 3h tests is confirmed with longer tests, up to 7h duration (Figure 3, left);
- backflushing is strictly necessary to control fouling (Figure 3, right).



**Figure 1:** effect of TMP increase during filtration at  $u_{\text{cr}}=1\text{m/s}$ , alternating 3' of filtration to 18'' of backflush: TMP=0.2bar (left) and TMP=0.4bar (right)



**Figure 2:** effect of increasing backflush frequency at TMP=0.3bar (left) and  $u_{cr}= 2$  m/s: filtration run of 3' (left) and 1'



**Figure 3:** filtration at TMP = 0.3 bar and  $u_{cr}= 2$  m/s: sustainability for 7h (left, 10' minutes filtration + 1' backflush) and unsustainable filtration without backflush ( right, 1' filtration + 5'' idle)

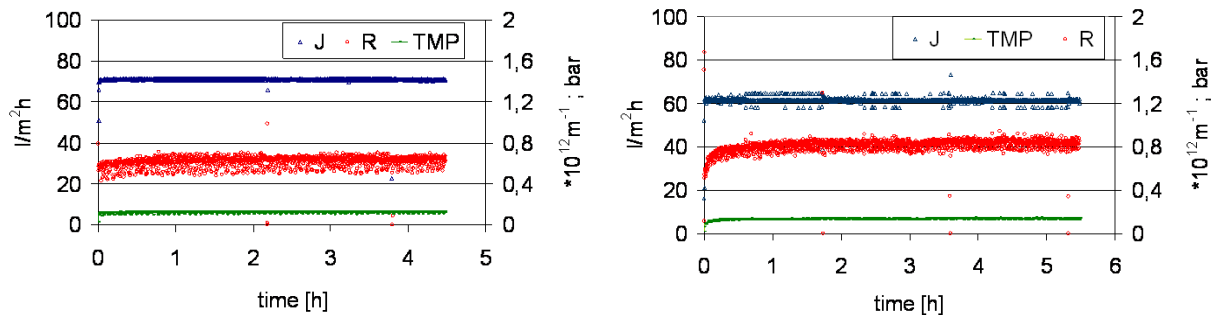
The permeate production varies with operating conditions; however the influence of wastewater changes with days in this case is not negligible. In general, during tests of 3 hours, the average gross-flux is about 75-90  $l/m^2 \cdot h$  at TMP = 0.2 bar and about 125  $l/m^2 \cdot h$  at TMP = 0.3 and 0.4 bar. Therefore, operating at TMP = 0.3 bar could be the optimal choice in terms of productivity and fouling formation. Since the backflush time have not been optimised, results cannot be properly extended to the net-production (when fouling is lower, at TMP = 0.2 bar, R during backflush is lower and therefore more backwash water is consumed). However, at the tested conditions, the best net productivity is about 80  $l/m^2 \cdot h$  at TMP = 0.3 bar,  $u_{cr} = 2$  m/s, using filtration run of 1 minute.

#### Operations at constant flux

The strategy to control fouling during constant flux operation, in absence of backflush, is to filtrate at sub-critical flux ( $J < J_{cr}$ ).  $J_{cr}$  is a function of  $u_{cr}$  and at  $u_{cr}= 2$  m/s, it is found to largely vary with feed water quality. Ranges are 90-150  $l/m^2 \cdot h$  for primary clarifier effluent and 40-100  $l/m^2 \cdot h$  for raw sewage. (Measured values were lower for the second module).

After  $J_{cr}$  is found, 2-6 hours of filtration are performed at a flux usually 20-30  $l/m^2 \cdot h$  below  $J_{cr}$ . When the filtration at sub-critical flux is applied without chemical cleaning in between, fouling slows down but does not stop (not shown). When the membrane is chemically cleaned in between, operations at sub-critical flux can be considered sustainable. A little increase of resistance is observed during the first hour, but afterwards the filtration stabilises (Figure, left) Such behaviour is in agreement with the findings of Defrance and Yaffrin (1999), who proposed a *weak* pragmatic concept of critical flux for the cases when the flux is set by a pump instead of being generated by a gradual increase of TMP :  $J_{cr}$  is the limit below which a stable filtration can be sustained for a long period with constant TMP. When J is set above  $J_{cr}$ ,

R does not stabilise during the test duration, while when J is set =  $J_{cr}$ , resistance increase is bigger but operation may stabilise again (Figure 4, right).



**Figure 4:** Direct ultrafiltration of raw sewage starting from a clean membrane and operating at sub-critical flux and (left) and at critical flux (right).

Also some concentration tests are conducted at constant flux, to investigate operation stability under these circumstances (not shown). Concentration limit is set to 85%, because the system in use presents some difficulties to handle some difficulties in handling the increase of temperature with recirculation after this point. During these tests J is varied above and below critical flux and the following is observed:

- concentration until 85% is possible and does not affect relevantly the fouling behaviour;
- the forming cake layer is not removed by  $u_{cr}$  when the extracted flux is reduced (from 60 to 20 l/m<sup>2</sup>h);
- reducing permeate extraction when filtration is not stable (from 100 to 40 l/m<sup>2</sup>h) may stabilise R.

## DISCUSSION AND CONCLUSIONS

Sustainable conditions are found for both the operational modes, at constant TMP or constant flux. However, fouling is not avoided but, more likely, only “limited”. The development of R with time suggests the formation of a cake layer which stabilises with time, when a dynamic balance is reached in the deposition and removal of fouling materials. The occurrence of equilibrium with time can result from the following:

- reduced mass transport towards the membrane with increasing resistance: as membrane fouls, flux diminishes and less material approaches the membrane;
- increased back-transport phenomenon’s with increasing cake layer thickness;
- modification of size and characteristics of fouling material with time and recirculation.

However, operating conditions play a fundamental role in regard to the existence of this dynamic equilibrium. Imposed TMP or flux are the driving forces for permeation and fouling, whereas crossflow velocity and backflush are “fouling removal” agents. Operations at  $TMP < 0.4$  bar with frequent backflushing and at  $J < J_{cr}$  seem effective strategies to obtain

sustainable operations. Especially the role of backflushing is emphasized in respect to fouling prevention.

A particularly positive aspect is that when fouling development shows an increasing trend, sometimes it is possible to obtain a new equilibrium operating on process parameters. However, this equilibrium would be found at lower flux values than before fouling occurrence (as in Chen *et al.*, 1997).

Direct ultrafiltration appears feasible for both primary effluent and raw sewage. In particular for raw sewage, net-fluxes up to 70-80 l/m<sup>2</sup>h and 60-70 l/m<sup>2</sup>h are obtained for 6-7h during filtration at constant TMP and constant flux respectively.

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