The Delft Filtration Characterisation method

Assessing membrane bioreactor activated sludge filterability

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Summary

Membrane bioreactors

The membrane bioreactor (MBR) process represents an innovative technology for the treatment of municipal wastewater. The basis for MBR technology is the well-known and widely applied activated sludge process, in which a concentrated suspension of micro-organisms is cultivated to biologically degrade the pollutants in the wastewater. After the biological treatment the biomass is separated from the purified water; in the conventional activated sludge process this is achieved by gravity settling, whereas in the MBR process this is achieved by a membrane filtration step (using microfiltration or ultrafiltration membranes). Superior effluent quality and reduced installation footprint are the main advantages of the MBR process. Nonetheless, the application of MBR technology is restrained by high operation and maintenance costs related to the prevention and removal of membrane *fouling*.

Membrane fouling

Fouling can be described as the deposition of feed water constituents on the surface or in the pores of the membrane during filtration or the decrease of performance that results from it. Fouling is a complex phenomenon that results from the interaction between three main factors: (1) the membrane characteristics, (2) the membrane operation and (3) the activated sludge properties. In addition to this the total fouling process can be subdivided in three components, based on the cleaning requirements to remove the fouling: (1) physically removable *reversible* fouling, (2) chemically removable *irreversible* fouling and (3) and non-removable *irrecoverable* fouling. The contribution of each of the components to the total fouling process depends on the time scale on which the process is considered.
Since a wide variety of research approaches are applied and each MBR plant has its unique combination of the three main factors affecting fouling, the explanations for fouling as discussed in literature are divergent and sometimes contradictory.

Methodology

Delft University has developed a method to characterise the *filterability* of activated sludge: the *Delft Filtration Characterisation method* (DFCm). The DFCm comprises a filtration unit with a single tubular sidestream ultrafiltration membrane and a well-defined measuring protocol. With the DFCm samples collected from any MBR plant can be filtrated under identical hydraulic circumstances and membrane starting conditions. In this way differences in the monitored filtration resistance increase can be attributed exclusively to differences in the activated sludge properties.

The conventional parameter to monitor and control the filtration process, the *permeability*, does not provide specific information about the role of each of the three main factors in the fouling process. The power of the DFCm is that it provides specific information about the potential of

an activated sludge sample to cause fouling (i.e. the filterability). This information can subsequently be used to allocate the limiting factor(s) in the filtration process in a full-scale MBR plant and to implement measures to improve/optimise the process.

The major limitation of the DFCm is that it does not necessarily provide information about irreversible fouling, which is the dominant fouling mechanism on the longer term. However, a relation between filterability and irreversible fouling can be assessed on the basis of an empirical comparison between DFCm measurements and the development of the permeability in the considered MBR plant.

DFCm output

In a short-term DFCm experiment the dominant fouling mechanism is reversible cake layer formation, i.e. the accumulation of substances on the membrane surface. A theoretical analysis on the basis of backtransport mechanisms indicates that only particles with a size smaller than approximately 5 μm are prone to accumulation on the membrane surface (for the standard measuring protocol: cross flow velocity = 1.0 m/s, flux = 80 $L/m^2 \cdot h$).

Total cake resistance ΔR [m^{-1}] can be expressed as a function of the specific permeate production V [L/m^2] with three constants that are related to the activated sludge characteristics: (1) the concentration of substances accumulating in the cake layer: c_i [g/L], (2) the specific cake resistance caused by the substances at a reference total cake resistance: α_R [m/kg] and (3) the compressibility coefficient of the accumulated substances: s [-]:

$$\Delta R = (\alpha_R \cdot c_i \cdot V)^{\frac{1}{1-s}} = (\alpha_R \cdot c_i)^{\frac{1}{1-s}} \cdot (V)^{\frac{1}{1-s}}$$

Every DFCm experiment provides a dataset from which the coefficients $\alpha_R \cdot c_i$ and s can be determined. A closer analysis indicates that in the relevant range of filtration resistance the compression of the cake layer plays a minor role; the total filtration resistance is predominantly determined by coefficient $\alpha_R \cdot c_i$.

To compare different DFCm experiments on the basis of a single value the ΔR_{20} value [$\cdot 10^{12}$ m^{-1}] was introduced, which represents the resistance increase after filtration of 20 L/m^2 permeate. Based on experience with numerous MBR plants a practical classification was made to qualify the filterability: for ΔR_{20} below 0.1 the filterability is good, for values between 0.1 and 1.0 it is moderate and when it exceeds 1.0 the filterability is indicated as poor.

Experimental results

The DFCm was applied as a tool to monitor the filterability over a period of several months in relation to the development of the permeability at the full-scale MBR plants of Varsseveld and Heenvliet. In addition the filterability of samples collected from MBR Heenvliet was manipulated in lab-scale experiments to create differences in filterability and to subsequently identify the activated sludge properties that play a role in the filtration process.

MBR Varsseveld
In the first few months of operation the performance of MBR Varsseveld was heavily influenced by the supply of wastewater from a local cheese factory. This wastewater appeared to contain a chemical polymer that was retained by the membranes and could not be degraded by the biomass. The severe fouling problems encountered in the full-scale installation were accompanied by extreme poor filterability of the activated sludge as measured with the DFCm. After the cheese factory was uncoupled from the sewer both the permeability and the filterability showed spectacular recovery. Hence, the DFCm experiments confirmed that the activated sludge quality was indeed a limiting factor in the filtration process. Due to the exceptional circumstances no clear conclusions could be drawn about which activated sludge characteristics were influencing its filterability.

MBR Heenvliet
In general the filterability at MBR Heenvliet was good throughout the measuring period, especially in the summer period. Nonetheless the permeability in the full-scale plant showed an unsatisfactory decreasing trend. Afterwards it appeared that the plant was subject to clogging and membrane integrity problems. In this case the DFCm demonstrated that filterability was not the limiting factor in the filtration process. The filterability showed a relation with the activated sludge temperature and the sludge volume index, but not with the Soluble Microbial Products (SMP) concentration which is often linked to fouling.

Stress experiments
Activated sludge samples collected from MBR Heenvliet were submitted to three different types of stress conditions: (1) prolonged low dissolved oxygen concentration, (2) high mechanic shear stress and (3) abrupt temperature decrease. The results demonstrate that for all three stress conditions the activated sludge deflocculates and its filterability deteriorates. This deflocculation is expressed by the release of SMP and sub-micron particles from the activated sludge matrix into the free water. The volume of sub-micron particles shows a closer relation with the activated sludge filterability than the SMP concentrations.

Conclusions
The DFCm has proven to be a useful tool to characterise and assess the filterability of activated sludge samples collected from full-scale MBR plants. Knowledge of the filterability can indicate whether a supposed permeability decrease should be attributed to poor activated sludge filterability or to inadequate operation of the filtration process.

Physical and chemical activated sludge analyses indicate that filterability is closely related to the volume of colloidal particles in the free water (i.e. particles up to 1 μm). The Soluble Microbial Products concentration, a parameter often linked to fouling, appears to be a weak indicator for filterability because the available methods do not distinguish soluble and colloidal SMP. Colloidal SMP are retained by the membrane and can cause fouling while soluble SMP can pass the membrane pores without contributing in the fouling process.

The concentration of colloidal particles in the activated sludge free water and therefore the filterability is closely related to the mechanisms of flocculation and deflocculation. In order to generate activated sludge with good filterability a balanced biological treatment process is required, preferably with a low loading rate. In case of (sudden) stress conditions supplementary measures could be applied to enhance the flocculation process (e.g. extension of the aerobic sludge retention time, dosage of a flocculation polymer).

Recommendations

The research described in this thesis has indicated that the colloidal fraction in the activated sludge free water plays an important role in the fouling process. More research is required to determine the exact particle size distribution in the colloidal range.

The mechanisms of flocculation and deflocculation play a crucial role in the fouling process. More research is required in order to optimise the activated sludge flocculation properties ones the activated sludge reaches the membrane area.

The current parameter used to control the MBR process is the *permeability*. The permeability can however be considered a weak parameter because it only provides information about the consequences and not about the causes of fouling.
Operation of the MBR filtration step should be more similar to the operation of the clarification step in the conventional activated sludge process. Whereas the clarification has its standardised parameter to characterise the settleability of the sludge (the *sludge volume index*), the MBR process also requires a standardised parameter to characterise filterability. Good filterability can be considered the starting point for a satisfactory filtration process. In addition the process operation should aim at maintaining a maximum membrane surface available for filtration. This so called *effective* membrane surface is continuously under pressure during operation, due to clogging and inadequate cleaning measures. When the effective membrane surface decreases the local fluxes in the membrane surface that is still available for filtration will increase. This is then also the case for the total fouling rate, irrespective of the activated sludge filterability. More sophisticated (local) flux measurements are required to understand the distribution of the total flow over the membrane surface.

Table of contents

1 Introduction

1.1 Background

Clean and fresh water is of vital importance to humanity. Evidently safe drinking water is of primary concern, but besides this water fulfils innumerable other important functions [I]. Through many of these functions the water is polluted, thereby becoming a threat to public health. Throughout history natural watercourses were used to get rid of our wastewater. Population growth and urbanisation however demonstrated that the natural drain off capacity has its limitations. In course of time the wastewater could not be carried off sufficiently anymore and people were compelled to use polluted sources for drinking water, with severe consequences [II]. It took until the second half of the 19th century before the relation between polluted water and public health was recognised. A few decades later, in the late 19th and early 20th century, the first large scale sewerage systems were constructed in Europe and the United States to carry wastewater away from urban areas. The contribution of this effort to public health is still apparent today, considering the situation in many developing countries in which sanitation facilities are still a privilege rather than self-evident [III].

I. The average daily water consumption of a Dutch citizen approximately consists of: 2 litres for direct consumption, 130 litres for domestic use (toilet, shower, washing, etc.) and 4000 litres through indirect use (production of food, clothes, industrial products, etc.). (Wikipedia, 2009)
II. In 1866 about 21.000 people in Amsterdam died from a cholera epidemic, directly caused by inadequate wastewater drain off facilities (Wikipedia, 2009).
III. Today still 2.6 billion people (40%) worldwide run continuous health risks due to poor sanitation facilities. Every day 7500 of these people die, from whom 5000 are children below the age of 5. (United Nations, 2009)

The construction of sewerage systems could be considered a blessing for public health, but at the same time it was a heavy burden for the environment. Pollutants, until then gradually discharged or degraded in cesspools, were now released directly and in higher concentrations into the surface water. Together with a strong growth of the population and industrial-, economical- and social development the modest self-purifying capacity of the environment was heavily overstressed. Aquatic ecosystems in densely populated areas were severely damaged [IV]. Besides the environmental aspect also several economical and social functions of the surface waters were threatened. When in addition also the groundwater, the primary source for drinking water, suffered quality deterioration it became clear that again drastic measures were necessary. As from the 1970s authorities (in Europe) commenced with industrial decontamination, legislation and the construction of wastewater treatment plants to improve the water quality. From this moment the situation improved considerably [V].

IV. A low point was reached in 1971, when as a result of industrial and municipal wastewater discharges the dissolved oxygen concentration in the Rhine had dropped so low that vast stretches of the river supported no form of life anymore. (Huisman et. el., 2001)

V. In 1970 the pollution discharged into surface water in the Netherlands amounted up to 45 million population equivalents (p.e.). The basic pollution load has now been reduced to 18 million p.e., mainly through industrial decontamination. After treatment the remaining 0.7 million p.e. are discharged into the surface water. (Dijk et al., 2001)

Although significant progress has been achieved in the recent decades, water pollution is still a current and not to be underestimated problem, also in Europe (VI). In addition the population, development and thereby the pressure on our water systems is still increasing (VII). Consciousness has grown that clean water is a valuable and vulnerable resource and that the current situation asks for efforts towards more sustainable consumption and management of our water sources.

In order to reach and preserve healthy water systems and sustainable sources for drinking water production the performance of wastewater treatment plants has to be upgraded. This concerns the degradation of nutrients (nitrogen and phosphorus) to very low concentrations, but also the removal of emerging pollutants such as heavy metals, medicines and pesticides. This requires advanced treatment technologies compared to the conventional methods that are now generally used. The ultimate goal, a healthy and sustainable (urban) water cycle, is not yet within reach. This stresses the importance of scientific research into advanced wastewater treatment technologies.

VI. 20% of all European surface waters are seriously threatened by pollution (European Commission 2009).

VII. 60% of the European cities overexploit their groundwater. 50% of the European wetlands have an endangered status due to groundwater overexploitation (European Commission, 2009).

Wastewater treatment and membrane bioreactors

The established method for the treatment of municipal wastewater is the widely applied and well-known *activated sludge process*. In this process a concentrated suspension of micro-organisms is cultivated which biologically degrades pollutants that are supplied with the incoming wastewater, primarily oxygen depleting substances and nutrients. After the biological treatment the micro-organisms are separated from the purified water (*effluent*) by gravity settling. The effluent is then discharged into the environment and the micro-organisms are carried back to new wastewater for repetition of the process.

In the innovative *Membrane BioReactor* (MBR) process the conventional settling process is replaced by a membrane filtration step. This membrane can be described as a fine sieve that allows water to flow through it (under influence of a pressure difference) while the relatively big micro-organisms are retained in the activated sludge suspension. The application of membrane filtration has two major advantages over sedimentation. In the first place the settling properties and thereby the biomass concentration are no longer a limiting factor in the process. This allows for a more compact reactor for the biological treatment. Together with the absence of the sedimentation tanks this results in a significant reduction of the installation footprint, which is economically beneficial for wastewater treatment plants in urbanised areas. The second important advantage of the MBR process is that the membrane filtration step provides a superior effluent quality compared to the settlers in the conventional activated sludge process.
In this way MBR technology can contribute in reaching the future stringent effluent quality discharges set by the Water Framework Directive [VII]. In addition MBR technology can produce high-quality effluent that has the potential to be used for specific reuse applications, such as irrigation and industrial process water.

VIII. **The Water Framework Directive** is the most substantial piece of water legislation ever produced by the European Commission, and will provide the major driver for achieving sustainable management of water in the European Union for many years to come (www.euwfd.com).

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1.2 Problem statement

Fouling

The most significant drawback of the MBR process is the inevitable problem of *membrane fouling*, which can be described as the accumulation and adsorption of feed water constituents on the membrane surface and in its pores. Due to fouling the water flowing through the membrane pores experiences a higher filtration resistance, i.e. the system performance decreases. The relatively high operation and maintenance costs associated to the prevention and removal of fouling are generally considered the primary constraint restricting the application of membrane technology. Especially in the last decade the MBR fouling phenomenon has become an extensively investigated topic. Significant progress has been achieved in understanding causes and mechanisms, but yet it has to be recognised that many questions still remain unanswered.

Research bottlenecks

The fact that MBR fouling is still insufficiently understood can only partly be attributed to the complexity of the (micro-scale) fundamentals of the process itself. An additional problem is that fouling is a difficult to investigate phenomenon. This can be explained on the basis of the three main factors that determine fouling: (1) the membrane properties, (2) the membrane operation and (3) the activated sludge properties.
Fouling is always the result of the interaction between these three factors. In practice each MBR plant, from lab- to pilot- to full-scale, has its own unique combination of these three parameters. In addition the activated sludge properties are highly heterogeneous and dynamic due to continuous fluctuations in the incoming wastewater concerning flow, composition and temperature. As a consequence it can be stated that each MBR plant has its own unique fouling process. Hence, the fouling processes of different MBR plants can not be compared unequivocally with each other. This explains the divergent and even contradictory explanations on the causes of MBR fouling that can be found in literature.

The established method to study problems such as membrane fouling is to simulate the filtration process on lab-scale. The well-defined and controllable circumstances that can be created in a lab-scale research approach are suitable (if not a prerequisite) for gaining fundamental knowledge that can subsequently be applied in practice. The MBR process however hardly lends itself for this approach. Several reasons can be mentioned for this:

- The hydraulic circumstances (and especially the spatial differences) prevailing in full-scale membrane modules are extremely difficult to “downscale” to lab-scale proportions.
- Lab-scale set-ups generally do not have access to real municipal wastewater. The homogeneous and stable (mostly synthetic) feed solutions that are often used are not

representative for the highly heterogeneous and dynamically alternating activated sludge suspensions as present in full-scale installations.

- MBR fouling consists of a short-term and long-term component. Especially the latter is difficult to investigate because it is a process manifesting on a time-scale of weeks or months and besides depends on the membrane cleaning measures. The gross of the lab-scale set-ups are not suitable to be operated on such a long term. Also the cleaning protocol as applied in full-scale installations can not be easily simulated in a lab-scale set-up.

1.3 Research approach

The problem statement indicates a controverse between scientific fouling research on the one hand and the actual practice of the filtration process in full-scale MBR plants on the other hand. The line of approach of the research described in this dissertation is based on overcoming this problem. To do so a method was developed that combines the accuracy of scientific lab-scale research with the representativeness of full-scale MBR installations. This so called *Delft Filtration Characterisation method* (*DFCm*) was designed and developed by Herman Evenblij (2006). The DFCm consists of a lab-scale filtration unit with a single tubular membrane and a measuring protocol. Key aspect of the DFCm is the possibility to measure the resistance increase created by any given activated sludge sample while filtrating with *similar membrane starting conditions* and under well-defined and *constant hydraulic circumstances*. In this way differences in filterability can be attributed exclusively to differences in activated sludge properties. The DFCm thus allows unequivocal comparison of the filterability quality of activated sludge samples collected under different circumstances or from different installations.

The representativeness of the research is formed by the strategy to only use “real” municipal wastewater activated sludge for the DFCm experiments. All activated sludge samples used in this research are collected from the first full-scale municipal MBR plants that have been put in operation in the Netherlands: MBR Varsseveld and MBR Heenvliet.

In the research described in this dissertation the DFCm is applied in two ways:

- As an *in-situ* tool to monitor activated sludge filterability at two full-scale MBR plants in relation to the permeability development and operational and external circumstances.
- As a *research* tool to assess changes in filterability and activated sludge properties of batch samples due to imposed stress conditions in lab-scale circumstances.

In its first application the DFCm acts as a tool that characterises the filterability parallel to the filtration process in the considered full-scale plant. The filterability as measured with the DFCm can be analysed in relation to the permeability in the full-scale installation. Drawback of this highly practical application of the DFCm is that the filterability that is actually measured is the sum of a multitude of operational parameters and external circumstances that can impossibly be all surveyed. This drawback can be partially overcome by imposing specific and well-defined stress circumstances upon activated sludge samples in lab-scale experiments. In this way the alternation of filterability and activated sludge properties that are created can be attributed exclusively to the imposed stress circumstances (when compared to an unaltered reference sample). This offers better possibilities for identifications of foulants that are involved.

1.4 Research objectives

The objective of the research described in this thesis is twofold. In the first place a next step has to be made in the development and assessment of the Delft Filtration Characterisation method. The second goal is to improve the understanding of the MBR filtration/fouling process by relating the DFCm filterability to the performance of the considered MBR plants and the properties of the examined activated sludge samples.

Delft Filtration Characterisation method assessment

The first steps concerning the validation and assessment of the DFCm were evidently made by its designer. Issues like reproducibility, sensitivity for changes in operational circumstances and sample maintenance were verified and described by Evenblij (2006). In this dissertation a next step in the assessment of the method is made by searching answers to the following questions:

i. What are the possibilities and the limitations of the DFCm?
ii. Is the filtration process as applied in the DFCm representative for the filtration process in practice?
iii. How should the DFCm be applied to yield optimal profit from its possibilities?
iv. What is the significance of the DFCm output and what information does it provide about the activated sludge properties and the occurring fouling mechanism(s)?

Fouling fundamentals

The second research goal is to improve the understanding of the fundamentals of the MBR fouling process. On the basis of DFCm results and the operational circumstances encountered in the considered full-scale MBR plants the following questions have to be answered:

v. What is the relation between filterability as characterised with the DFCm and the permeability development as occurring in full-scale plants?
vi. Which activated sludge properties are linked to its filterability and how?
vii. Can the properties and the filterability of the activated sludge collected from full-scale MBR plants be related to the process operation or to external circumstances?

1.5 Thesis outline

The remaining chapters of this thesis are structured as follows:

Chapter 2 *(Wastewater treatment and membrane bioreactors)* provides a general description of municipal wastewater treatment, membrane filtration and membrane bioreactor technology. The conventional activated sludge process and its main operational parameters are introduced. Subsequently the fundamentals and operational parameters of membrane filtration for water treatment processes are discussed. The chapter concludes with a description of the membrane bioreactor process, comprising of a general process description, an overview of operational parameters, the barriers and drivers related to the application of the process and a brief description of the development that MBR technology has experienced in the last decades.

Chapter 3 *(Fouling and filterability)* is dedicated to the phenomenon of membrane fouling. A general background and the theoretical mechanisms of membrane fouling in water treatment processes are addressed. Subsequently the analysis is specified to fouling in MBR, starting with an analysis of the factors that are involved. The concepts of reversible, irreversible and irrecoverable fouling are explained as well as the different fouling stages that can be distinguished. In conclusion a literature review of the three main factors affecting membrane fouling is presented.

Chapter 4 *(The Delft Filtration Characterisation method)* forms the descriptive part of the methods that are used in this research. First a historical background about the development of the DFCm is described. Subsequently the Filtration Characterisation unit and the measuring protocol that together form the DFCm are explained in detail. In conclusion the activated sludge analyses that accompany the DFCm measurements are described.

Chapter 5 *(Method assessment)* deals with the assessment of the Delft Filtration Characterisation method. The possibilities and limitations of the method are discussed and a comparison with several other activated sludge filtration characterisation methods is made. The raw output of the DFCm experiments is analysed in relation to the theory of cake filtration to discern the significance of the output. In conclusion the hydraulic regime in the membrane tube during a filtration characterisation experiment is assessed to verify a possible influence on the experimental results.

Chapter 6 *(Filtration Characterisation at MBR Varsseveld)* discusses the first measuring campaign, conducted at MBR Varsseveld. In this campaign a limited number of DFCm experiments were conducted with activated sludge collected from different compartments of the MBR. These experiments were accompanied by an extensive set of activated sludge quality analyses. The DFCm results are related to the specific circumstances and the permeability development in the full-scale MBR to verify a possible relation. In addition the DFCm results are related to the activated sludge properties to identify foulants.

Chapter 7 *(Filtration Characterisation at MBR Heenvliet)* addresses the second measuring campaign, conducted at MBR Heenvliet. The filterability and the activated sludge properties were monitored on a weekly basis for a period of approximately one year. As with the research campaign in Varsseveld the DFCm results are analysed in relation to the site specific circumstances, the development of the permeability and the activated sludge properties.

Chapter 8 *(Stress experiments)* provides an overview of several stress experiments that were conducted with activated sludge collected from MBR Heenvliet. In lab-scale circumstances the filterability and properties of the activated sludge samples were wilfully manipulated by imposing stress conditions upon them, in the form of long-term low dissolved oxygen concentrations, short-term high mechanic shear stress and an abrupt decrease of the temperature. Both the effect of the stress circumstances and the resilience of the activated sludge to recover are investigated in relation to several activated sludge properties.

Chapter 9 *(Discussion, conclusions and recommendations)* represents the final chapter of this thesis. The research questions posed in section 1.4 of this chapter are recalled and discussed. The final conclusions of the research are summarised and several recommendations for future research directions are proposed.

2 Wastewater treatment and membrane bioreactors

2.1 Introduction

In this chapter the basic principles of municipal wastewater treatment and membrane bioreactor (MBR) technology are discussed. In section 2.2 the background related to municipal wastewater treatment is briefly discussed. The basis for the MBR process is the well-known and widely applied activated sludge process, in which a concentrated suspension of micro-organisms biologically degrades the pollutants in the wastewater. The fundamentals of the conventional activated sludge process are discussed in section 2.3. In the MBR process the micro-organisms responsible for the treatment of the wastewater is not separated by means of sedimentation, but by membrane filtration. The fundamentals of membrane filtration for water treatment are discussed in section 2.4. Subsequently some specific characteristics of MBR technology and the differences compared to the conventional process are discussed in section 2.5. This chapter concludes with a short summary in section 2.6.

2.2 Background

Although the indispensability of wastewater treatment is nowadays widely recognised by policymakers and society it was only in 1970 when a legislative framework came into effect in the Netherlands: the *Wet Verontreiniging Oppervlaktewater* (*WVO*), Dutch for the Surface Water Pollution Act. The purpose of the WVO was to prevent and reduce the pollution of surface water with respect to the important functions that these waters fulfil in our society. The WVO did not deal with treatment methodology but set a framework for combating water pollution and allowed for enforcing stricter legislation. This resulted in the construction of wastewater treatment plants, enforcement of discharge permits, pollution levying and the execution of inspections. The responsibility for complying with the WVO was within the jurisdiction of the state (Ministry of Waterways and Public Works) and regional water quality administrators (mainly Provinces and Water Boards).

In the course of time the WVO has been modified several times, mainly for implementation of European directives. Currently the WVO is enforced by the most important piece of European water (quality) legislation: the *Water Framework Directive* (*WFD*). The WFD, which came into effect in December 2000, requires all waters (rivers, lakes, coastal waters and groundwater) within the European Union to have a “good status” by the year 2015. Since 2000 the WFD is gradually being transposed into the existing legislation of all EU members. With respect to municipal wastewater treatment the biggest impact of the WFD includes the ambition to reduce eutrophication of the European surface waters. Another attention point of the WFD is to suppress discharges and reduce the emission of heavy metals and priority hazardous substances such as pesticides.

2.3 The activated sludge process

The classical method for the treatment of municipal wastewater is the widely applied *activated sludge process*. The antecedents of the activated sludge process date from the late 19th century when several researchers studied the influence of aeration on the quality improvement of wastewater. Around 1913 Adern and Lockett (Manchester Sewage Works, England) observed that the “flocs of micro-organisms” that arose from aerating wastewater were capable of aerobic stabilisation of organic constituents. Besides this the flocs could be separated from the water relatively easily by sedimentation. Returning the settled flocs to new wastewater even led to an improvement and acceleration of the wastewater purification. Adern and Lockett named their discovery the *activated sludge process*, since the aeration of wastewater was accompanied by the production and activation of micro-organisms.

2.3.1 Process description

A schematic representation of the conventional activated sludge process as nowadays widely applied is presented in Figure 2.1. As a first step coarse material, sand, fat and settleable materials are removed from the wastewater by a pre-treatment step. This pre-treatment usually consists of screening, primary sedimentation and flotation. Subsequently the actual activated sludge process takes place, consisting of three basic components:

- A bioreactor in which the micro-organisms are aerated and kept in suspension.
- A secondary clarifier in which the activated sludge is separated from the treated water (*effluent*) by sedimentation.
- A recirculation system that returns the settled sludge back to the bioreactor.

The pollutants in the wastewater are used by the biomass as a source for metabolism and to multiply. As a consequence of this multiplication the biomass concentration is continuously increasing. To maintain a constant biomass concentration *excess* sludge (or *surplus* sludge) has to be removed periodically from the system.

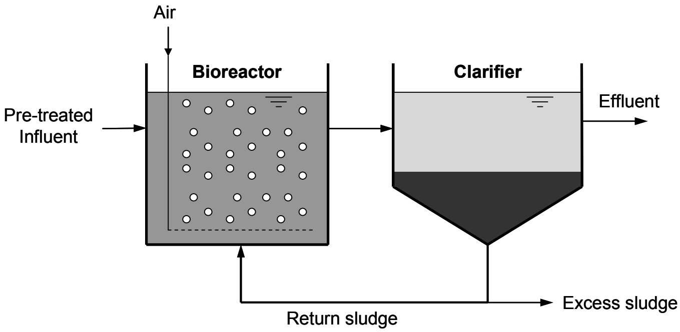


Figure 2.1: Schematic overview of the activated sludge process

Since the activated sludge process came into common use in the early 1920s it has evolved in many different ways, mainly with respect to the biological process. In the simplest configuration (continuously aerated bioreactor) the biomass is solely capable of removing oxygen depleting substances. By introducing aerobic, anaerobic and anoxic zones and internal recycle flows within the bioreactor the process can also incorporate biological removal of the nutrients nitrogen and phosphorus. In addition several chemicals can be added to enhance the treatment performance.

2.3.2 Effluent quality

The primary goal of a wastewater treatment plant is evidently to remove pollutants from the wastewater to a desired level. In the research described in this thesis the effluent quality plays a minor role; therefore it is only described briefly.

In general the treatment performance of a wastewater treatment plant is assessed on the basis of the removal of oxygen depleting substances (chemical oxygen demand, COD and biochemical oxygen demand, BOD), total suspended solids (SS), nitrogen (N_{tot}) and phosphorus (P_{tot}). In addition it is mentioned that the removal of heavy metals and micro-pollutants is gaining significance (Water Framework Directive, 2009).
The performance of a wastewater treatment plant is determined by its design, the influent properties (rate and composition) and the operational circumstances. Table 2.1 shows the average concentrations for the five main pollutants in Dutch municipal wastewater and the average treatment performance of the treatment plants. The numbers indicate that the removal of BOD, COD and SS is not the limiting factor for Dutch municipal wastewater treatment. With respect to eutrophication the performance of activated sludge systems is nowadays generally judged on the basis of nutrient removal. Currently the Dutch wastewater treatment plants do not meet the stringent WFD regulations with respect to nitrogen, phosphorus and suspended solids. This stresses the need for optimisation of the activated sludge process and the application of advanced technologies.

Table 2.1: Average Dutch municipal wwtp influent and effluent compositions and discharge standards (CBS Statline, 2007)

Component	Influent	Effluent	Removal efficiency	Discharge standards	WFD standards
COD (mg O_2/L)	471	38	91%	125	-
BOD (mg O_2/L)	196	4	98%	20	-
SS (mg/L)	223	10	95%	30	5
N_{tot} (mg N/L)	44	8	81%	10 (wwtp > 20.000 p.e.) 15 (wwtp < 20.000 p.e.)	2.2
P_{tot} (mg P/L)	7	2	79%	1 (wwtp > 100.000 p.e.) 2 (wwtp < 100.000 p.e.)	0.15

2.3.3 Process parameters

Sludge loading

A major process parameter determining the performance of an activated sludge system is the *Sludge Loading* or the *Food-to-Mass* ratio, which represents the ratio between the supply of organic contents in the influent wastewater and the available amount of biomass in the bioreactor to degrade it.

The most widely used parameter to express the amount of organic contents in the influent wastewater is the *Biochemical Oxygen Demand* (BOD, mg O_2/L). BOD represents the amount of dissolved oxygen consumed by micro-organisms to biochemically degrade the organic matter in the wastewater over a period of five days (Metcalf and Eddy, 2003). The amount of biomass can be determined and classified in several ways. In this dissertation the term *Mixed Liquor Suspended Solids* concentration (MLSS, g/L) is used. The MLSS concentration represents the dry weight of solids per litre resulting from combining recycled activated sludge with influent wastewater (see chapter 4.6 for determination).

From a treatment performance point of a view it is desirable to employ a sludge loading as low as possible; the lower the food supply per micro-organism is, the better the total degradation will be. Furthermore a low sludge loading results in a lower sludge growth which is beneficial for the solids retention time, the excess sludge production and the denitrification process. From an economical point of view a low sludge loading is unattractive since it increases the aeration need per micro-organism. Typical sludge loading values mentioned in literature vary between 0.04 and 1.0 gram BOD per gram biomass per day (Metcalf and Eddy, 2003). In the Netherlands the average sludge loading in conventional activated sludge plants is around 0.2 g BOD/g MLSS·day (CBS Statline, 2007)

In practice it is very difficult or impossible to influence the influent flow rate and composition. This implies that the sludge loading can not be controlled through the "*food*" supply, but only through the amount of "*mass*" available in the bioreactor. To reach the best treatment performance it is thus desirable to maximise the MLSS concentration in the bioreactor. The maximum MLSS concentration is however restricted by the secondary clarification step; when the MLSS concentration is too high the sludge can not settle sufficiently. Generally the MLSS concentration in conventional activated sludge systems is limited to values around 5 g/L (Van der Graaf, 1995). The average MLSS concentration in Dutch conventional activated sludge systems is 4.1 g/L (CBS Statline, 2007).

Solids retention time

The *Solids Retention Time* (*SRT*) or *Sludge Age* represents the average period of time during which the activated sludge remains in the system. The SRT is closely related to the sludge loading and forms an important parameter as it affects the treatment performance, the required aeration tank volume, the sludge production and mineralisation and the oxygen requirements (Metcalf and Eddy, 2003). The SRT is defined by the ratio between the total amount of activated sludge in the system (kg dry solids) and the excess sludge discharge (kg dry solids per day). As mentioned previously the continuous growth of activated sludge during the process has to be compensated by a proportional discharge of excess sludge to maintain a constant MLSS concentration in the system. The excess sludge production mainly depends on the sludge loading: the lower the sludge loading, the lower the excess sludge production. Obtaining a high sludge age thus requires a low sludge loading.

For BOD removal an SRT of 3 to 5 days is required, depending on the activated sludge temperature (Metcalf and Eddy, 2003). Denitrification also depends on SRT and temperature. The design SRT for nitrification is usually higher than for BOD removal because nitrifying bacteria grow relatively slow. Besides this a safety factor is usually applied to provide operational flexibility and the possibility to handle nitrogen peak loadings. Depending on temperature and sludge compounds the required SRT for nitrification can vary between 3 and 18 days (Metcalf and Eddy, 2003). Finally, a high SRT is required to achieve aerobic stabilisation of the sludge (in the range of 20 to 40 days, depending on temperature). Aerobic stabilisation of the sludge is necessary to minimise the sludge growth and to create a stabilised excess sludge (odourless, biologically stable).
The SRT of conventional activated sludge plants in the Netherlands varies between 13 and 32 days, with an average of 24 days (CBS statline, 2007).

Sludge Volume Index

Evidently the settling properties of activated sludge are crucial for good operation of conventional wastewater treatment plants. The most common parameter used to characterise activated sludge settling properties is the *Sludge Volume Index* (SVI, mL/g). The SVI represents the ratio between the volume of a sludge sample after a certain settling period and its MLSS concentration. The method to determine the SVI - an empirical parameter - is addressed in chapter 4.6. In general SVI values of 100 mL/g and lower indicate good settling properties, while SVI values above 150 mL/g represent bad settling properties. High SVI values are typically associated with the presence of filamentous bacteria (Parker et al., 2001).

Hydraulic retention time

The *Hydraulic Retention Time* (HRT) is defined as the ratio between the reactor volume (m^3) and the volumetric flow rate (m^3/h) of the plant and thus represents the period of time in which the wastewater remains in the system. An increase of the HRT is beneficial for the treatment performance of the system, but evidently requires a larger bioreactor. Typical HRT values applied in conventional activated sludge systems vary between 10 and 30 hours (Metcalf and Eddy, 2003).

2.4 Membrane filtration

In the membrane bioreactor process the conventional sedimentation step is replaced by a membrane filtration step. This section discusses several generalities considering membranes and the membrane filtration process for water treatment processes.

2.4.1 Process description

Since the nature, purpose and application of membranes are extremely diverse it is not easy to formulate a universal definition of a membrane. As a minimum all membranes have in common that they act as "*a selective barrier between two phases*" (Mulder, 1996). This is still a general depiction as it only provides information about one characteristic and for example not about structure, material or purpose.

Focused on water treatment applications, a membrane usually consists of a finely porous synthetic medium with the function to allow water to pass through it while constituents in the water are retained. The transport of water through the membrane can only take place under the influence of a *driving force*. Different driving forces exist (electrochemical potential gradient, temperature gradient, concentration gradient), but in water filtration usually a pressure gradient is used, referred to as the *Trans Membrane Pressure* (TMP). Depending on the height of the TMP and the filtration resistance water will flow from the *feed* side through the membrane to the *permeate* side with a certain flow rate called the *flux* (J).

The size of the retained constituents highly depends on the size of the membrane pores. All the particles bigger than the membrane pores are retained by the membrane. In case of a cake layer formation by the particles during filtration also particles smaller than the membrane pores can be retained (discussed more detailed in chapter 3.2).

Figure 2.2 schematically represents the filtration principle for an ultrafiltration membrane, which is the most common applied membrane type in MBR technology. Ultrafiltration membranes are designed to retain colloids, particulate material and bacteria, while water and dissolved substances can pass.

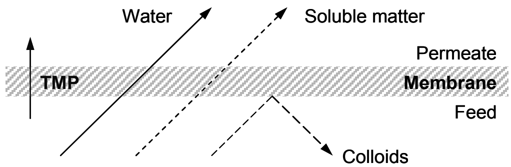


Figure 2.2: Principle of membrane filtration (ultrafiltration)

During filtration constituents in the feed water inevitably accumulate onto the membrane surface and into the pores, thereby hindering the filtration process. This process is called *fouling*. Fouling is a central topic in this dissertation; it is briefly considered in subsection 2.4.4 and discussed in detail in chapter 3.

2.4.2 Membrane characteristics

Material

In water filtration processes generally two kinds of membrane materials are used: *ceramics* and *polymerics*. Ceramic membranes are constructed from inorganic materials, such as aluminium oxides, titanium oxides, zirconia oxides and some glassy materials (Mulder, 1996). They offer several major advantages over polymerics; they are physically and chemically very stable and have a high durability. Nonetheless, in practice the advantages of ceramics are not (yet) counterbalanced by their only main disadvantage: they are expensive. In 1995 a cost difference of a factor 10 was reported in literature (Owen et al., 1995). Though cost reduction of ceramics has been achieved in recent years (Le-Clech et al., 2006), application remains very limited and restricted to specific circumstances. Examples mentioned in literature are ceramics used for the treatment of industrial waste and anaerobic biodegradation (Le-Clech et al., 2006).
In MBR technology practically all applied membranes are polymeric based. A rather wide variety of materials can be used; the most common ones are listed in Table 2.2.

Table 2.2: Polymeric membrane materials (Judd, 2006; Mulder, 1996)

Material	Abbreviation	Applications
Polyvinylidene difluoride	PVDF	MF, UF
Polyethylsulphone	PES	UF, RO
Polyethylene	PE	MF, UF
Polypropylene	PP	MF, UF

The membrane structure can be either *isotropic* or *anisotropic*. Isotropic membranes have a homogeneous structure, whereas anisotropic membranes consist of a thin top layer supported by a mechanically stronger underlying more porous layer (Baker, 2000). Membrane material can be either *hydrophobic* or *hydrophilic*. The constituents in the feed water are usually hydrophobic and therefore tend to accumulate on hydrophobic material. From a fouling point of view a hydrophilic membrane material is thus preferential. Several chemical techniques, like oxidation, plasma treatment and grafting are available to modify the hydrophobic character of a membrane surface (Judd, 2006). The relation between membrane characteristics and fouling properties is discussed more detailed in chapter 3.7.

Selectivity

On the basis of selectivity four types of membranes can be distinguished in water treatment processes: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO).

The selectivity of a membrane can be indicated by size or weight rejection. The molecular weight cut off (MWCO) of a membrane represents the molecular weight of solutes which are retained for 90% (Koros et al., 1996). Membranes can also be classified by their pore size, i.e. the size of constituents that can pass. Figure 2.3 schematically represents the MWCO and pore sizes of different membrane processes and the types of constituents that can be removed by it.

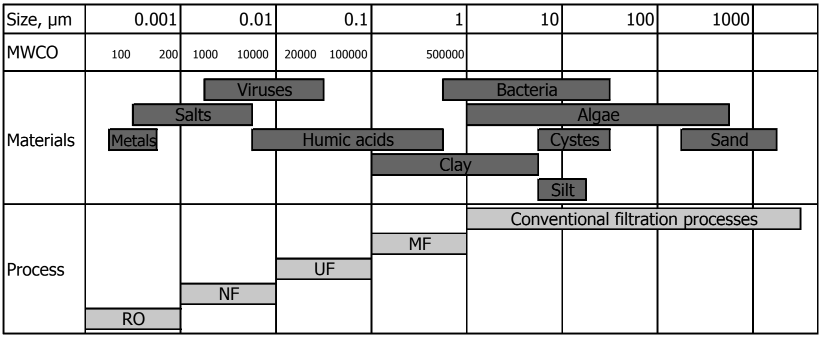


Figure 2.3: Classifications membrane separation processes (adapted from Van Dijk et al. 2001)

Smaller pores increase the hydraulic resistance of the membrane and thus imply a higher required TMP to maintain filtration. Table 2.3 outlines the range of pore sizes and the accompanying required operational TMP for the four different membrane applications (Mulder, 1996). It is mentioned that the pore size range related to each of the membrane process is not fixed; slight variations can be found in literature.

Table 2.3: Operating TMP and pore sizes for different membrane processes

Membrane process	Pore size [nm]	Pressure [bar]
Microfiltration	100 – 1000	0.1 – 2
Ultrafiltration	10 – 100	0.1 – 2
Nanofiltration	1 – 10	4 – 20
Reverse Osmosis	0.1 – 1	10 – 30

Configuration

Membrane configuration refers to the physical shape of the membranes and the way of fitting them in a full-scale installation. From an economic point of view it is desirable to maximise the ratio between the available surface (m^2) and the volume (m^3) occupied by the membranes. This so called *specific membrane surface* or *packing density* however conflicts with some operational aspects. The most significant one in this sense is the fact that a high packing density complicates the possibility to create turbulent circumstances near the membrane surface. As will be discussed in subsection 2.4.4 especially in MBR systems turbulent circumstances near the membrane surface are essential to maintain a good filtration performance.

Based on geometry six basic membrane shapes can be distinguished (Judd, 2006): flat sheet- (or plate-and-frame), hollow fibre-, tubular-, capillary tube-, pleated filter cartridge- and spiral-wound membranes. Each of these configurations has its own specific benefits and drawbacks. The major ones are indicated in Table 2.4.

Table 2.4: Membrane configurations (Judd, 2006; Baker, 2004)

Configuration	Cost	Turbulence promotion	Back flush	Packing density [m^2/m^3]
Flat sheet	High	Fair	No	100-300
Hollow fibre	Very low	Very poor	Yes	300-500
Tubular	Very high	Very good	Yes	150-300
Capillary tube	Low	Fair	Yes	1.500-5.000
Pleated filter cartridge	Very low	Very poor	No	500-1.500
Spiral-wound	Low	Poor	No	800-1.200

In MBR technology predominantly the first three configurations listed in Table 2.4 are applied (flat sheet, hollow fibre and tubular membranes); examples of these configurations are illustrated in Figure 2.4. Membrane configuration in MBR systems will be discussed more in detail in subsection 2.5.1.

Figure 2.4: Membrane configurations applied in MBR technology
tubular (X-flow), hollow fiber (Zenon) and flat sheet (Kubota)

2.4.3 Membrane operation

Constant TMP filtration and constant flux filtration

Permeate extraction can take place in two production modes: with *constant flux* or with *constant TMP*. In constant TMP operation the flux will decrease in time as a result of fouling, whereas in constant flux operation the TMP will increase in time. Since from an operational point of view it is more convenient to control the permeate production rather than the applied pressure, therefore large-scale MBR systems are generally operated in constant flux mode.

Crossflow and dead-end mode filtration

Two basic modes of membrane operation are employed in water treatment applications: *dead-end* filtration and *crossflow* filtration. In dead-end filtration the entire feed flow passes the membrane. This implies that all constituents in the feed flow that can not pass the membrane are retained on- or in it. In crossflow filtration a second flow is introduced, on the feed side and perpendicular to the membrane surface. In this way a shear force is created which prevents the deposition of particles on the membrane surface. Crossflow operation makes the membrane less sensitive to fouling, but requires an extra energy input component compared to dead-end filtration. In the MBR process the feed contains such a high amount of solids that dead-end filtration is not an option; in all MBR plants a tangential shear force has to be created to prevent rapid fouling.

Cleaning

Since fouling is inevitable periodical cleaning measures are required to maintain sufficient membrane filtration performance. Cleaning can be performed on the basis of time interval or a certain threshold pressure. Two general forms of membrane cleaning are distinguished: *physical* (or mechanical) cleaning and *chemical* cleaning.

Physical cleaning aims at creating shear circumstances along the membrane surface to remove accumulated particles. In chemical cleaning substances adsorbed to the membrane are removed by oxidation, usually through soaking with high or low pH chemicals. Physical cleaning has a number of advantages over chemical cleaning. It is usually a short process which is not suspected to be harmful to the membrane. In addition no chemicals are required and thus no chemical waste is produced. The main disadvantage of physical cleaning is that it is not as thorough as chemical cleaning; for the removal of more tenaciously adsorbed substances chemical cleaning is indispensible in the longer term.

2.4.4 Process and operational parameters

Flux

As discussed in subsection 2.4.1 the driving force for filtration is the trans membrane pressure (TMP), which represents the pressure difference between the feed and the permeate side of the membrane. Imposing a TMP induces a water flow through the membrane. The flow rate per membrane surface area unit is called the *flux*. The flux is a major operational parameter in membrane processes. Since the flow through the membrane can be considered laminar (Lojkine et al., 1992) the flux can be described according to Darcy's law:

$$J = \frac{Q}{A} = \frac{TMP}{\eta_p \cdot R_{total}} \tag{2-1}$$

With:

- J = flux, [m/s], in daily practice and in this thesis expressed as [$L/m^2 \cdot h$]
- Q = flow rate, [m^3/s],
- A = membrane surface, [m^2]
- TMP = transmembrane pressure, [Pa], in this thesis expressed as [Bar]
- η_p = dynamic viscosity of the permeate, [Pa·s]
- R_{total} = total resistance to filtration, [m^{-1}]

Equation 2-1 shows that the flux is inversely proportional to the permeate viscosity. This is explained by the fact that the more viscous or "thick" a fluid is, the higher the resistance will be against flowing through the narrow membrane pores. The viscosity of permeate can be assumed equal to pure water (Manem and Sanderson, 1996) and is therefore solely dependent on its temperature. In practice a temperature increase of 1 °C corresponds with a flux increase of approximately 3%. Several empirical relationships between temperature and pure water viscosity can be found in literature; in this dissertation the one as derived by Janssen and Warmoeskerken (1997) is used:

$$\eta_p = 10^{-3} \cdot \exp^{(0.580-2.520\theta+0.909\theta^2-0.264\theta^3)} \tag{2-2}$$

With:

- θ = empirical factor: 3.6610 · (T/(273.1+T)), [-]
- η_p = dynamic viscosity, [Pa·s]
- T = temperature, [°C]

Filtration resistance

Depending on its properties a membrane creates a certain amount of resistance ($R_{membrane}$) against filtration. In addition to the so called membrane resistance constituents in the feed water may accumulate to the membrane surface or in the pores during filtration. This process introduces an additional resistance factor called *fouling* ($R_{fouling}$). Total filtration resistance is the sum of the membrane resistance and the fouling resistance:

$$R_{total} = R_{membrane} + R_{fouling} \tag{2-3}$$

The challenge of membrane producers is evidently to create a membrane that combines a low membrane resistance with good “non-fouling” properties and good retention properties. Besides the size of the pores the membrane resistance depends on the tortuosity of the pores and the porosity and the thickness of the membrane. The membrane resistance can be calculated according to the Hagen-Poisseuille law which describes the flow through a porous medium:

$$R_{membrane} = \frac{8 \cdot \tau_p \cdot L}{\pi \cdot D^2} \tag{2-4}$$

With:
$R_{membrane}$ = membrane resistance, [m^{-1}]
D = pore diameter [m]
τ_p = pore tortuosity [-]
L = membrane thickness [m]

Understanding, modelling, characterising, predicting and preventing fouling is a major research theme. Chapter 3 is fully dedicated to the phenomenon of fouling.

Permeability

The permeability is the generally used parameter to express the performance of a membrane system. It is defined as the ratio between the flux and the TMP and can thus also expressed as the inverse product of permeate viscosity and total filtration resistance:

$$P = \frac{J}{TMP} = \frac{1}{\eta_p \cdot R_{total}} \tag{2-5}$$

With:
P = permeability, [m/s·Pa], usually expressed as [$L/m^2 \cdot h \cdot Bar$]
J = flux, [m/s]
TMP = transmembrane pressure, [Pa], usually expressed as [Bar]
η_p = dynamic viscosity, [Pa·s]
R_{total} = total resistance to filtration, [m^{-1}]

As shown in Equation 2-2 the water temperature affects the membrane permeability through the viscosity. To exclude the influence of the temperature on permeability in practice often the *corrected* permeability is used, which reflects the actual permeability multiplied by the quotient of the actual viscosity and a reference viscosity.

Crossflow

As mentioned in subsection 2.4.3 a shear force parallel to the membrane surface has to be created in the MBR process to prevent fouling. Two means of creating this shear force are applied in MBR technology:

- A controlled liquid flow tangential to the membrane surface
- Aeration of the liquid to induce motion in the fluid and recirculation

For energy efficiency reasons the crossflow in MBR systems is generally created by coarse bubble aeration (see section 2.5). The bubbles scouring the membrane surface induce local shear transients and liquid flow fluctuations, promoting the back transport of potential foulants. Contrary to sidestream systems, in immersed systems the shear force can not be regulated directly. The amount of shear energy induced in a two-phase flow system is usually expressed as the amount of air per hour (Nm^3/h) or per membrane surface (Nm^3/m^2). These parameters do however not provide a direct indication of the actual crossflow velocity. Determination of the liquid crossflow velocity in immersed MBR systems is extremely complex. Techniques used to do so are electromagnetic flow velocity meter (Sofia et al., 2004), particle image velocimetry (Cabassud et al., 2001) and constant temperature anemometry (Le-Clech et al., 2008). In practice these techniques are generally not available at full-scale plants.

2.5 Membrane Bioreactor technology

2.5.1 Process description

In membrane bioreactor technology the secondary clarification step, as applied in the conventional activated sludge process, is replaced by a membrane separation step. The crucial consequence of this replacement is that the settling properties of the activated sludge are no longer a limiting factor in the process. Whereas the MLSS concentration in the conventional activated sludge process is limited to about 5 g/L in the MBR process theoretically no upper limit exists. Nonetheless, for operational reasons (oxygen transfer efficiency, clogging prevention) in MBRs the biomass concentration is also limited. In MBR plants for municipal wastewater treatment the maximum applied MLSS concentration is generally 10 to 15 g/L. As a result of the higher MLSS concentration the bioreactor of an MBR can be designed more compact compared to a CAS system. Together with the absence of secondary clarifiers MBR technology thus has the benefit of a relatively small footprint.

System configuration

The membrane step can be implemented into an MBR system in two ways: *sidestream* or *immersed*.

In sidestream configuration the membranes are placed externally from the bioreactor, schematically presented in Figure 2.5. Activated sludge is recirculated through the membranes and permeate is extracted with inside-out filtration. In general tubular membranes are used in this configuration.

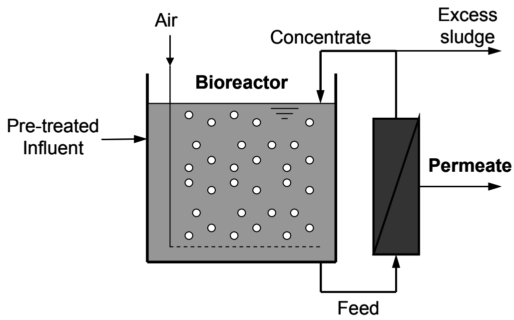


Figure 2.5: Sidestream MBR configuration

The first generation MBRs were designed according to the sidestream configuration. Fouling was traditionally prevented by applying a high liquid crossflow through the membrane tubes. Crossflow velocities mentioned in literature vary between 1 and 6 m/s (Evenblij, 2005).

Applying a high crossflow velocity makes the membranes less sensitive to fouling and thus allows for employing a relatively high TMP or flux. The main disadvantage of the traditional sidestream configuration is the high energy consumption required for the crossflow.

In the immersed or internal configuration the membrane modules are submerged directly into the activated sludge, either in a separate membrane tank or directly in the bioreactor. The immersed MBR configuration is schematically presented in Figure 2.6. Nowadays the membranes are more commonly placed in a separate membrane tank; in the first place this allows better control over the filtration process and besides this the wastewater undergoes superior biological treatment before reaching the membrane. The disadvantage of a separate membrane tank is the higher energy consumption for recirculation and aeration.

In the immersed MBR configuration usually hollow fibre- or flat sheet membranes are used. Permeate extraction takes place with outside-in filtration. Shear force along the membrane surface is created through scouring by coarse bubble aeration.

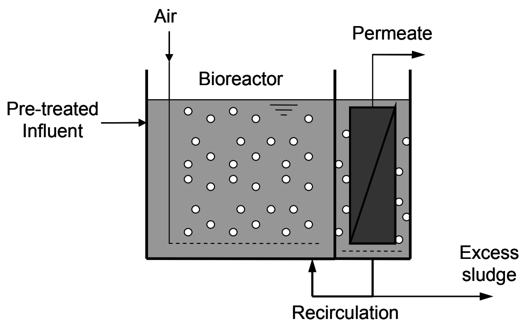


Figure 2.6: Immersed MBR configuration

Within immersed MBR systems two different membrane configurations are applied: *hollow fibre* and *flat sheet* membranes. Because the membrane channels in flat sheet membranes are well-defined compared to hollow fibre membranes the hydraulic circumstances are easier to control. This makes flat sheet membranes less sensitive to fouling and suitable for application of somewhat higher fluxes. On the other hand the packing density of flat sheet membranes is lower (Table 2.4) and they are estimated 20 to 25% more expensive compared to hollow fibres (Le-Clech et al., 2006).

The difference between sidestream and immersed systems as discussed here is based on the placement of the membrane modules. But actually the key difference between the two configurations is the means of creating shear force along the membrane surface: *liquid crossflow* versus *two phased air-liquid* configuration.

Some specific characteristics of the traditional sidestream and the immersed MBR configuration are summarised in Table 2.5 (Evenblij 2006; Judd 2006; Mulder 1996). As mentioned previously the traditional sidestream configuration with liquid crossflow is very energy consuming, about a factor 10 higher compared to immersed systems with coarse bubble aeration. For this reason in municipal wastewater treatment immersed MBR is by far the predominant configuration. In the three year period between 2003 and 2005 99% of the total installed membrane surface was immersed (Lesjean et al., 2007).

Table 2.5: Summary of sidestream and immersed MBR characteristics

Characteristic	Sidestream (liquid crossflow)	Immersed (coarse bubble aeration)
Membrane placement	Outside reactor	In bioreactor
TMP	2 – 6 Bar	0.2 – 0.5 Bar
Filtration type	Inside-out	Outside-in
Flux	50 – 100 L/m2·h	20 – 40 L/m2·h
Energy consumption	2 – 5 kWh/m3	0.2 – 0.5 kWh/m3
Crossflow speed	1 – 6 m/s	0.5 m/s

In a novel approach, introduced by NORIT (Futselaar et al., 2007), the concept of sidestream filtration is combined with bubble aeration. In the so called airlift system the activated sludge is circulated through tubular membranes by means of aeration, while a feed pump is only used to overcome the hydraulic losses. In this way the advantages of the classical sidestream configuration (easy accessibility of the membranes, relatively high flux) can be combined with a low energy consumption of 0.2 kWh/m^3 (Futselaar et al., 2007).

2.5.2 Additional remarks

Pre-treatment

Since membranes are sensitive to fouling and clogging MBR technology requires a more thorough pre-treatment compared to the CAS process. In the MBR process the commonly applied grid distance is between 1 and 3 mm, whereas in the CAS process this is about 5 mm. Since hollow fibre systems are more sensitive to fouling than flat sheet systems a lower grid distance (± 1 mm) is advised than in flat sheet systems (± 3 mm).

Effluent quality

Because the membrane forms an absolute barrier MBR technology is far superior compared to the CAS process when it comes to the removal of suspended solids, turbidity and bacteria. In addition it is mentioned that MBR plants are generally designed on the basis of a very low loading rate. This low loading rate is beneficial for the removal efficiency of oxygen depleting substances and nutrients. It has to be recognised that the loading rate is not necessarily associated to MBR technology. A supposed superior treatment efficiency of the MBR is thus not related to the means of sludge-water separation.

2.5.3 Drivers and barriers

Barriers

The most apparent barrier restraining the application of MBR technology for municipal wastewater treatment are undoubtedly the relatively high costs of the process compared to conventional treatment processes, both in terms of investment as well as in costs related to operation and maintenance.
Besides the cost aspect it can be considered that experience with MBR technology is relatively limited and it is still regarded a high-risk technology (Judd, 2006). As a result of this decision-makers might be reserved to invest in expensive MBR technology.

Drivers

Judd (2006) enumerates five main influences that promote the implementation of MBR technology:

- Legislation;
- State incentives to encourage improvements in wastewater technology and recycling;
- Local water scarcity;
- Decreasing investment costs;
- Increasing confidence in and acceptance of MBR technology;

Two supplementary drivers that can be mentioned are:

- The suitability of MBR technology to be retrofitted in an existing conventional wastewater treatment plant;
- Space scarcity.

The most important driver for MBR technology is undoubtedly upcoming more stringent legislation. The future WFD discharge standards require far-reaching nutrient removal. Contrary to the conventional activated sludge process MBR technology has the potential to meet the WFD requirements. In addition water is becoming a resource with increasing economical value and therefore water reuse applications become attractive. The high-quality effluent of an MBR plant can be suitable for reuse. References dealing with the application of MBR technology for reuse purposes are numerous.

2.5.4 MBR development

The first commercial application of MBR technology dates from the late 1960s, when Dorr-Oliver Inc. first combined an activated sludge bioreactor with a sidestream membrane filtration loop (Bemberis et al., 1971). MBR technology found applications in some niche areas like shipboard sewage, landfill leachate and highly loaded industrial effluents (Stephenson, 2000). However, broad application was restrained by the high costs for membranes and energy consumption, fouling problems and the low economical value of the effluent. Despite the drawbacks of the "first generation" MBRs the application increased steadily, predominantly in the USA and in Japan.

A new era in MBR technology commenced in 1989 when Yamamoto et al. (1989) introduced the idea to directly submerge the membranes into the biomass. Together with the application of modest fluxes the operational costs could be reduced drastically compared to the traditional sidestream systems. A gradual reduction of the membrane price promoted further growth of MBR technology in the mid 90s. Despite the challenges MBR technology is still facing its application has been increasing rapidly since the start of the new millennium. In the period from 2000 to 2005 the global MBR market doubled to €166 million and prognoses indicate an increase to €258 million in 2010 (Hanft, 2006). Atkinson (2006) even predicts a rise to €363 million by 2010.

Initially Europe stayed behind compared to Asia and the USA, with an MBR market size of €25.3 million in 1999. By 2004 the market size more than doubled to €57 million and this growth rate is expected to sustain over the coming years. The European growth of MBR technology is also illustrated by the number of installations that has become operational over the years; see Figure 2.7 (Lesjean and Huisjes, 2007).

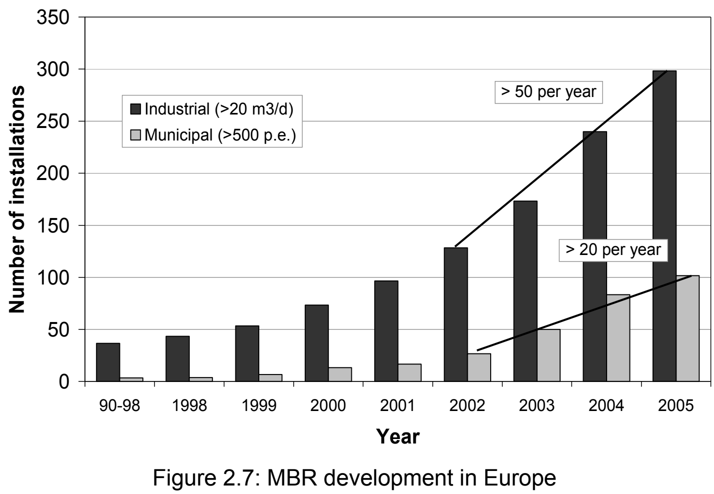


Figure 2.7: MBR development in Europe

2.6 Summary

The conventional method for the treatment of municipal wastewater is the activated sludge process. In the activated sludge process a concentrated suspension of micro-organisms is cultivated and used to biologically degrade pollutants in wastewater, primarily oxygen depleting substances and the nutrients nitrogen and phosphorus. After the treatment process the micro-organisms are separated from the treated water. In the conventional activated sludge process this is done by means of sedimentation.
In the innovative membrane bioreactor (MBR) process the activated sludge is not separated from the treated water by sedimentation but with a membrane filtration step. Superior effluent quality and a reduction of the installation footprint are the main advantages of the MBR process compared to the conventional activated sludge process.

The drawback of MBR technology is that it is relatively expensive compared to the conventional activated sludge process. These higher costs are predominantly associated to operation and maintenance measures required to prevent and remove fouling of the membranes that inevitably occurs during filtration. The phenomenon of fouling is discussed extensively in chapter 3. Despite the relative high costs MBR technology has experienced significant growth, mainly in the last decade. The most important driver for the application of MBR technology is legislation that sets higher standards to the effluent quality of wastewater treatment plants (predominantly through the European Water Framework Directive).

3 Fouling in membrane bioreactors

3.1 Introduction

This chapter addresses the fundamentals of the membrane fouling process in membrane bioreactor systems. During filtration constituents in the feed inevitably deposit on and into the membrane. This process, or the decrease of membrane performance that results from it, is called *fouling*. The operational and maintenance costs associated with the prevention and removal of fouling are generally considered to be the primary factor restraining the application of MBR technology for municipal wastewater treatment.

First in section 3.2 some general definitions of fouling and the different theoretical fouling mechanisms occurring in (waste)water membrane filtration processes are addressed. Subsequently the discussion is focused on fouling in MBR systems. In section 3.3 the main factors that are involved in MBR membrane fouling are structuralised. Section 3.4 discusses the different forms of MBR fouling with respect to rate and tenacity. The MBR fouling process can be subdivided in several stages, this issue is discussed in section 3.5. The subsequent sections provide a more detailed analysis of the three main factors that affect the fouling process: the membrane characteristics (3.6), the membrane operation (3.7) and the activated sludge characteristics (3.8). The chapter concludes with a summary and some concluding remarks in section 3.9.

3.2 Theoretical background

3.2.1 Definitions

In literature several definitions of membrane fouling can be found, with varying approach and comprehensiveness. Judd (2006) concisely defines it as "*processes leading to deterioration of flux due to surface or internal blockage of the membrane*". A slightly more comprehensive but comparable definition is formulated by the International Union of Pure and Applied Chemistry (IUPAC): "*fouling is the process resulting in the loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surface, at its pore openings, or within the pores*" (Koros et al., 1996). Metcalf and Eddy (2003) also formulate a concise definition, but contrary to Judd and Koros they do not include the deterioration of membrane performance in it. They describe fouling only as "*the potential deposition and accumulation of constituents in the feed stream on the membrane*".
The above mentioned definitions give a straightforward description of what the fouling process basically implies. In a different approach the terms reversibility and irreversibility are introduced. Van den Berg and Smolders (1990) state that fouling is a "*more-or-less irreversible*

and long-term process", while directly occurring and reversible flux decline is caused by concentration polarisation. Wiesner and Aptel (1996) state that fouling is "*a reduction in permeate flux that can not be reversed*", while Fane and Fell (1987) describe fouling as "*a flux decline which can not be reversed by simply altering the operating conditions*".
Although reversibility is closely related to fouling, in this dissertation the definition as proposed by the IUPAC is used. This implies that fouling is considered to be only the accumulation of particles to the membrane surface, leaving the removability of the accumulated particles out of consideration.

3.2.2 Fouling mechanisms

The definition of fouling as formulated by the IUPAC puts forward that it is a phenomenon that consists of several mechanisms. In crossflow filtration five theoretical mechanisms contributing to the total filtration resistance are distinguished (Berg and Smolders, 1990; Dijk et al., 2001), illustrated in Figure 3.1. The initial resistance is created by the membrane itself. The membrane resistance (**R_m**) can be defined as the resistance that is experienced by a clean membrane when demineralised water is filtrated.

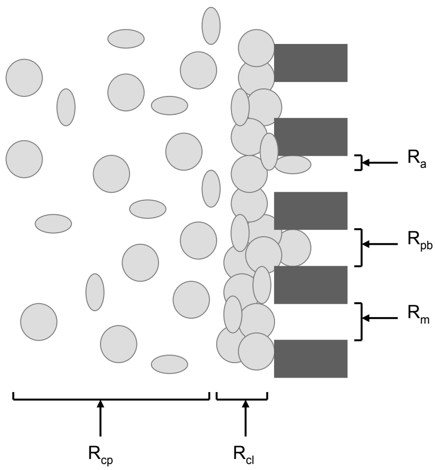


Figure 3.1: Schematic representation membrane filtration fouling mechanisms. (R_m = membrane resistance, R_{pb} = pore blocking, R_a = adsorption, R_{cl} = cake layer resistance, R_{cp} = concentration polarisation)

Subsequently the resistance increases due to the following theoretical fouling mechanism:

- **Pore blocking (R_{pb})**: the resistance caused by substances or particle(s) stuck in the membrane pore and thereby completely blocking it.

- **Adsorption (R_a)**: the resistance caused by particles or substances that are absorbed to the pore wall thereby narrowing the pore channel.
- **Cake layer resistance (R_{cl})**: the resistance caused by particulate material residing on the upstream face of the membrane surface during filtration. When the membrane is clean only particles with a size bigger than the membrane pores will be retained. In the course of a filtration cycle the thickness and density of the cake layer will increase and also particles smaller than the size of the pores can accumulate in the cake layer.
- **Concentration polarisation (R_{cp})**: the IUPAC defines concentration polarisation as: "*a concentration profile that has a higher level of solute nearest to the upstream membrane surface compared with the more-or-less well-mixed bulk fluid far from the membrane surface*" (Koros et al., 1996). The concept of concentration polarisation originates from RO applications. The retention of solutes by the membrane can lead to an osmotic pressure difference over the membrane. This results in convective transport of solvents from the permeate side to the feed side, requiring a higher TMP to maintain permeation. However, in membrane filtration processes with a higher molecular weight cut off, like MF and UF, the accumulation of material at the membrane surface is rather expressed by the formation of a cake layer. In crossflow MF and UF applications a common approach is to neglect the contribution of concentration polarisation to the fouling process (Evenblij, 2006; Ravazzini, 2008). In this dissertation concentration polarisation is also left out of consideration.

Together with the membrane resistance the fouling mechanisms cause the total resistance according to the resistance-in-series model:

$$R_{total} = R_m + R_{pb} + R_a + R_{cl} + R_{cp} \qquad (3\text{-}1)$$

Evidently the fouling mechanisms listed in equation 3-1 occur simultaneously during filtration. Significant progress has been booked in understanding the individual mechanisms, but this knowledge is predominantly based on synthetic, homogeneous and stable feed solutions. An important bottleneck in MBR fouling research is that these synthetic feed solutions can not be considered representative for the heterogeneous and dynamic nature of activated sludge.

3.2.3 Backtransport mechanisms

In the MBR filtration process two forces are imposed upon the particles near the membrane surface:

- A convective drag force towards the membrane surface due to the flux.
- A shear force parallel to the membrane surface due to the crossflow.

As will be discussed in chapter 4 the method used to characterise the filterability of activated sludge comprises a single tubular membrane. In a tubular membrane the backtransport mechanisms and their velocities are (Evenblij 2005; Belfort et al., 1994:

- **Brownian diffusion (V_{BD})**: the random movement of particles suspended in a fluid.

$$V_{BD} = 0.185\left(\frac{\gamma_0 \cdot k^2 \cdot T^2}{\eta_\mathrm{a} \cdot d_p{}^2 \cdot L} \cdot \frac{c_\mathrm{w}}{c_\mathrm{b}}\right)^{1/3} \tag{3-2}$$

- **Shear-induced diffusion (V_{SD})**: individual particles undergoing random displacements from the streamlines in a shear flow as they interact with and tumble over other particles.

$$V_{SD} = 0.072 \cdot \gamma_0 \cdot \left(\frac{d_p{}^4}{L} \cdot \frac{c_\mathrm{w}}{c_\mathrm{b}}\right)^{1/3} \tag{3-3}$$

- **Lateral migration (V_{LM})** (or inertial lift): sideward migration of particles which transports particles away from the membrane surface.

$$V_{LM} = 0.036 \cdot \left(\frac{\rho_{as} \cdot d_p{}^3 \cdot \gamma_0{}^2}{\eta_a}\right) \tag{3-4}$$

With:

γ_0 = shear rate near the membrane surface, [s^{-1}]
k = Boltzmann constant, [$1.38 \cdot 10^{-23}$ kg·m^2/s^2]
T = absolute temperature, [K]
η_a = apparent viscosity feed flow, [Pa·s]
d_p = particle diameter, [m]
L = length membrane tube, [m]
c_w = particle volume fraction at the membrane wall, [-]
c_b = particle volume fraction in the bulk, [-]
ρ_{as} = density activated sludge, [kg/m^3]

The three particle backtransport mechanisms occur simultaneously and the total backtransport velocity (V_{BT}) can be considered the sum of the three individual mechanisms:

$$V_{BT} = V_{BD} + V_{SD} + V_{LM} \tag{3-5}$$

The contribution of each of the individual mechanisms depends on the particle size and the operational circumstances. An analysis of the conditions in the filtration characterisation method applied in this research is discussed in chapter 5.6.

3.3 Factors affecting fouling in MBR

3.3.1 The "three-factor" approach

MBR fouling is a phenomenon in which a multitude of factors may play a role. Nonetheless, from a straightforward point of view the involved number of factors can be reduced to only three main ones, illustrated in the scheme in Figure 3.2. Simply stated, avoiding fouling problems requires the combination of a "good" membrane, "good" membrane operation and activated sludge with "good" properties. Evidently the scheme in Figure 3.2 does not comprehend the complexity of the process, but nonetheless the "three-factor" approach forms a good starting point to structure the problem of fouling and is widely accepted and used by several researchers (Chang et al., 2002; Lojkine et al., 1992; Le Clech et al., 2006; Evenblij, 2006).

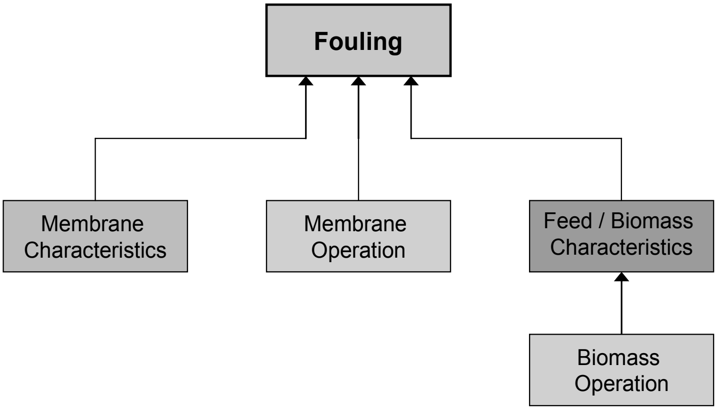


Figure 3.2: The three main factors affecting fouling in MBRs

The three main fouling factors mentioned in Figure 3.2 are generally assumed independent of each other. Some critical notes can be placed in this context. It can be expected that on the long term chemical cleaning measures (part of membrane operation) will influence the membrane properties in a negative way. Information about this matter in literature is poor, mainly because the experience concerning long-term membrane operation of full-scale MBR plants is still limited.

Furthermore some membrane operation aspects affect the characteristics of the activated sludge. Permeate extraction automatically implies an increase of the MLSS concentration in the membrane area, especially when the MBR is equipped with a separate membrane tank. It can be expected that the MLSS concentration in some way affects the fouling behaviour of the activated sludge. This complex issue will be discussed more in detail in subsection 3.8.2.

In addition the membrane operation could affect the biomass through the coarse bubble aeration, as a consequence of the input of oxygen and shear energy. On the other hand it can be expected that concerning the residence time the activated sludge properties are predominantly determined by the influent properties and the preceding biological treatment process in the bioreactor.

Evidently the operational circumstances and the biomass characteristics do not affect the actual (i.e. installed) membrane surface. However, the membrane surface can also be considered from an operational point of view (i.e. the membrane surface participating in the filtration process). Contrary to the installed membrane surface the so called *effective* membrane surface is closely related to the operational circumstances and the activated sludge properties. The significance of the operational parameter effective membrane surface is discussed more in detail in subsection 3.7.1 and chapter 5.3.

3.3.2 The "Judd" approach

A major critical note concerning the scheme as proposed in Figure 3.2 is that the diverse and complex phenomenon of fouling is represented as a single all-embracing parameter. Matters like mechanisms, the tenacity of the fouling and the time-scale on which the process takes place are left out of consideration in this approach. The disproportion concerning time-scale and the three parameters can for example be illustrated by comparing the factors membrane characteristics and biomass characteristics with each other: the membrane characteristics will change gradually during the membrane lifespan of several years, while the sludge characteristics represent a very dynamic parameter that can change on a time-scale of minutes. Figure 3.2 forms a good basis for structuring the fouling problem, but for good understanding a more extensive approach is required.

Judd (2006) proposes a somewhat more elaborate schematisation of the relation between fouling and several MBR parameters, represented in Figure 3.3. The three main factors pointed out in Figure 3.2 recur in a slightly different and extended way. Membrane operation and biomass operation are combined in the parameter "Operation", but the basic idea of fouling depending upon membrane (module) characteristics, membrane operation and biomass characteristics remains. The essential difference between the schemes in Figure 3.2 and Figure 3.3 is the addition of the phenomenon *clogging* and the distinction between *reversible* and *irreversible* fouling. The difference and potential relation between reversible and irreversible fouling form an important aspect of this dissertation and will be discussed more in detail in the next section 3.4.

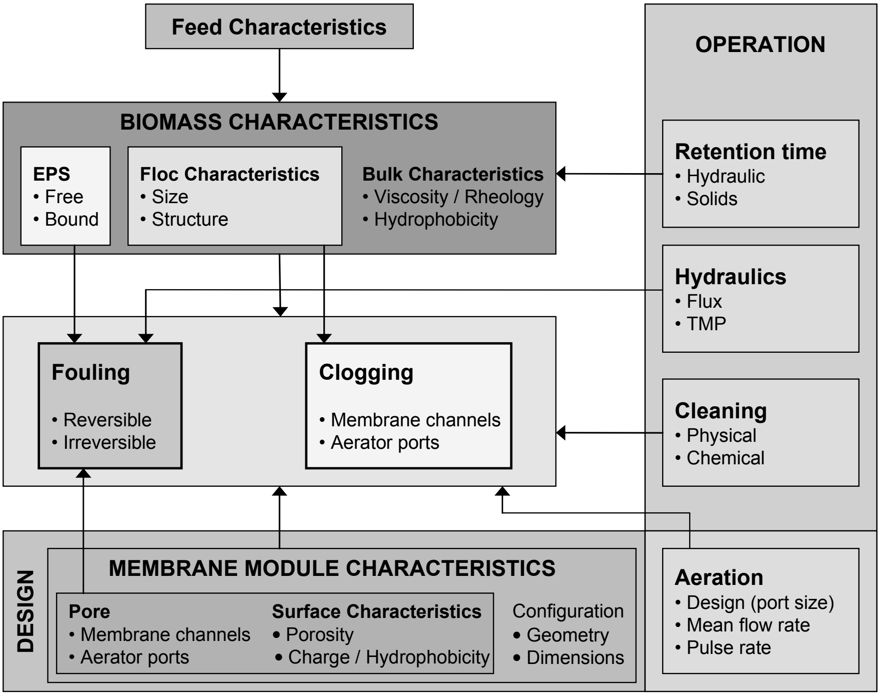


Figure 3.3: inter-relationships between MBR parameters and fouling (Judd 2006)

Clogging can be described as the accumulation of sludge particles in the membrane module near the membrane surface ("macro-scale"), whereas fouling is a process taking place right at the membrane surface or within the pores ("micro-scale"). Compared to fouling clogging is a simple and well-understood problem: as a result of poor hydrodynamic circumstances (usually caused by uneven distribution of aeration within the membrane channel/module) activated sludge flocs get the chance to coagulate and block up at the membrane surface, thereby making the membrane unavailable for permeation. Clogging and fouling are two different processes with the only obvious similarity that they both lead to a decrease of membrane performance. This similarity is important however, since it implies that it is not known a priori whether a decrease of membrane performance results from fouling or clogging. If for example a full-scale MBR plant suffers from a performance decrease it is tempting to impute this problem to fouling, meanwhile ignoring the possibility of clogging. Though clogging is much easier to avoid than fouling it still should be a major point of attention, especially since the current design trends (bigger plants, high module packing density) and preferred operation (high MLSS concentration, minimisation aeration rate) all promote clogging.

3.4 Reversible and irreversible fouling

3.4.1 Terminology

As put forward in Figure 3.3 the total fouling occurring in an MBR system can - or actually should - be subdivided in a *reversible* and *irreversible* component. In literature several different definitions and further subdivisions of reversible and irreversible fouling can be found. Terminology is confusing - or in fact incorrect - as irreversible fouling is actually reversible. To avoid misunderstandings the definitions of reversible and irreversible fouling as used in this dissertation are specified:

- **Reversible fouling** is fouling that can be removed by *physical* cleaning.
- **Irreversible fouling** is fouling that can be removed by *chemical* cleaning and not by physical cleaning.

To be complete a tertiary fouling component can be appointed, indicated as *irrecoverable* or *long-term irreversible* fouling:

- **Irrecoverable fouling** is fouling that can not be removed by any means of cleaning.

Ultimately the irrecoverable fouling determines the lifespan of a membrane (apart from other forms of damage). Since irrecoverable fouling is an inevitable and gradual process it can also be considered as depreciation of the membrane rather than a form of fouling.

Another term frequently used in the context of membrane fouling is *filterability*. Although closely related to each other fouling and filterability represent two very different concepts; fouling is a *process*, whereas filterability is a *characteristic*. The filterability is determined by the interaction between the membrane and the liquid to be filtrated. In the research described in this thesis all the membranes are of the type ultrafiltration and built of the material PVDF. Therefore in this research the filterability can be assumed a characteristic that is solely dependant on the activated sludge characteristics. Literature provides no universal definition for filterability. In this dissertation the following definition is used:

- **Filterability** is an activated sludge characteristic that reflects its *potential* to cause fouling, for reference operational circumstances and reference membrane characteristics.

3.4.2 Approach based on fouling mechanisms

The distinction between reversible and irreversible fouling is based on the cleaning measures required to remove it. However, the actual difference between the two obviously originates from

the tenacity with which the foulants are attached on or in the membrane and thus from the fouling mechanisms that are occurring.
The MBR process pre-eminently distinguishes itself from other water filtration processes by the high concentration of solids with divergent sizes in the feed flow (i.e. activated sludge). Due to the relatively high concentration of solids in the activated sludge that are larger than the membrane pores the dominant fouling mechanism on the short term is cake formation (Judd, 2006; Meng et al., 2009). Since cake fouling consists of substances "loosely" accumulated on the membrane surface it can be prevented and removed relatively easily by creating sufficiently high shear circumstances along the membrane surface. Focused on immersed configuration this is done by applying coarse bubble aeration. Subsequently cake fouling removal is enhanced by relaxation and/or back flushing of the membrane.

However, as discussed in subsection 3.2.2, simultaneously with cake fouling also other fouling mechanisms occur. Besides cake layer formation, substances will absorb more tenaciously to the membrane surface or within the pores. Theoretically back flushing can remove particles stuck in the pores, but on the long term mechanical measures are not sufficient to remove substances that are absorbed more tenaciously on the membrane surface or within the pores. As a consequence a chemical cleaning step is required to remove this irreversible part of the fouling. The considerations above indicate that reversible fouling is the result of cake layer formation, whereas irreversible fouling results from adsorption and pore blocking.

3.4.3 Fouling rates

Reversible, irreversible and irrecoverable fouling occur on different time scales. This is illustrated on the basis of Figure 3.4 and Table 3.1. In this figure the TMP development for constant flux operation of an MBR membrane during its total lifespan is schematically represented.

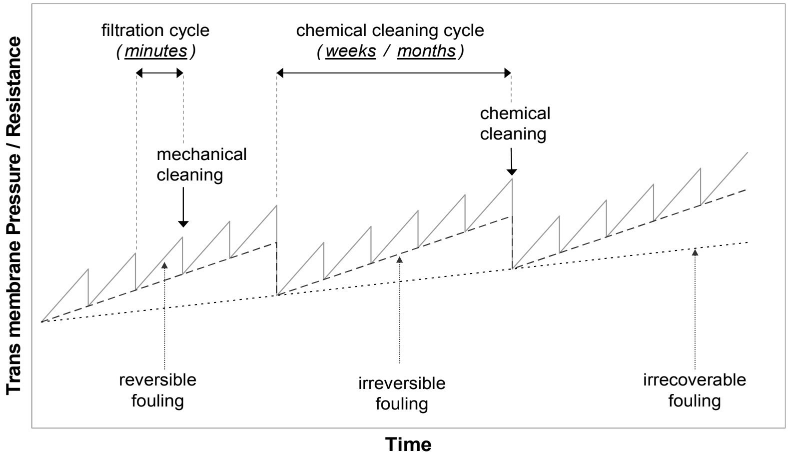


Figure 3.4: Different fouling rates during long-term MBR operation for constant flux operation (scale out of proportion, adapted from Judd 2006, Kraume 2007)

During filtration the TMP rise is predominantly caused by cake layer formation. After a short period –in the range of minutes– the *reversible* fouling is removed by a mechanical cleaning measures (relaxation, back flushing).

Table 3.1: Different fouling “forms” (adapted from Kraume (2007))

Fouling “form”	Fouling rate [mbar/min]	Time interval	Cleaning
Reversible fouling	0.1 – 1	10 minutes	Mechanical
Irreversible fouling	0.001 – 0.1	Weeks, months	Chemical
Long-term irreversible fouling	0.0001 – 0.001	Several years	Impossible

Since on a longer term mechanical cleaning is not capable of completely returning the TMP to its original value the TMP at the start of each filtration cycle will increase in time. This increase in time reflects the *irreversible* fouling rate. The filtration cycle is maintained until a certain pressure threshold (or permeability decline) is reached; at this point the membranes are cleaned chemically. In hollow fibre systems usually less-intensive maintenance cleanings are executed with a typical frequency of one to two weeks to remove *residual* fouling. High-intensive maintenance cleanings are used in both hollow fibre and flat-sheet system with a preferred frequency of once or twice per year. Ultimately also chemical cleaning measures can not prevent the gradual rise of the TMP over a period of years as a consequence of *irrecoverable* fouling.

Each form of fouling discussed above corresponds with a typical rate, time interval for cleaning and cleaning measure. An overview is given in Table 3.1. The irreversible fouling component is subdivided with respect to maintenance- and recovery cleanings. The fouling rate numbers presented in Table 3.1 indicate that irreversible fouling occurs with a rate that is about a factor 100 lower than reversible fouling.

3.5 Fouling stages

The TMP development as illustrated in Figure 3.4 is based on an idealised situation of steady and regular fouling rates and recoveries after the membrane cleaning measures. A steady fouling pattern is however not self-evident. Zhang et al. (2006) and Judd (2006) describe a fouling pattern according to three stages, schematically represented in Figure 3.5 and Figure 3.6.

In the initial conditioning stage fouling is the result of direct interaction between the membrane surface and constituents in the feed. The dominant fouling mechanism in this stage is pore blocking. Conditioning fouling has been reported to be independent of the flux (Zhang et al., 2006) and the tangential shear (Ognier et al., 2002). Ognier et al. report rapid irreversible fouling in the initial fouling stage, while Choi et al. (2005) found the contribution of conditioning fouling to total fouling to become negligible once filtration takes place.
The steady fouling stage commences when the membrane surface is mostly covered by foulants. This promotes further accumulation of particulate and colloidal material (Judd 2006). Pore blocking continues and a cake layer is formed. As long as the fouling is distributed more or less homogeneously over the total membrane surface the fouling can be indicated as steady fouling.
However, as a result of inhomogeneous distribution of the crossflow forces an irregular fouling distribution will inevitably occur. The permeability of fouled membrane areas will be lower compared to relatively clean areas, resulting in a higher flux in the clean areas. When this local flux exceeds the so called critical flux (further discussed in subsection 3.7.1) the fouling rate increases rapidly, indicated as a “TMP jump”.

The concept of different fouling stages indicates that the fouling rate is strongly depending on the effective membrane surface. This stresses the necessity of good membrane operation. Poor cleaning efficiency or (local) clogging due to poor aeration reduce the effective membrane surface and thus promote the development of irregular fouling.

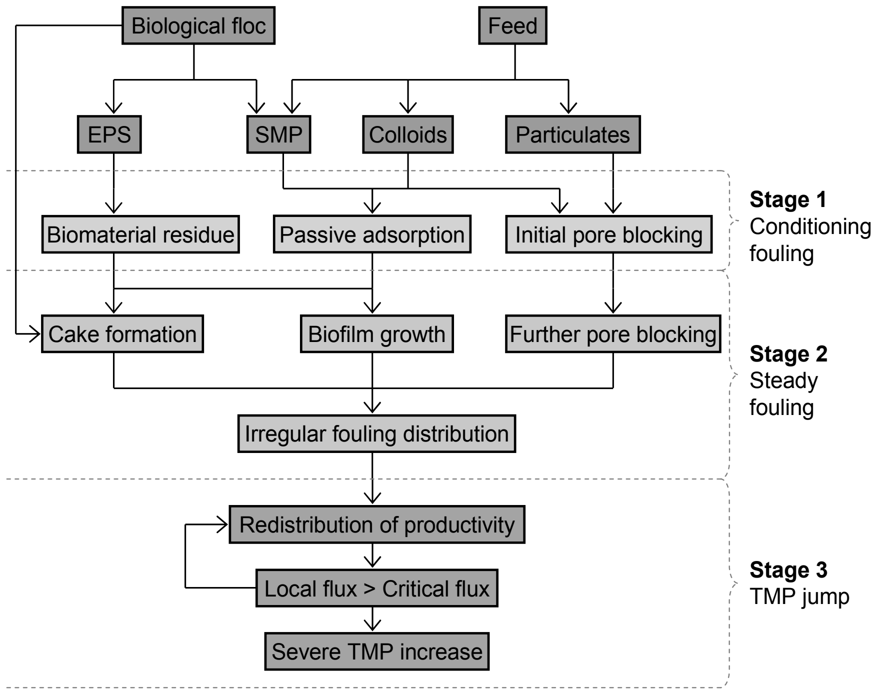


Figure 3.5: Fouling stages for MBR operated under constant flux circumstances

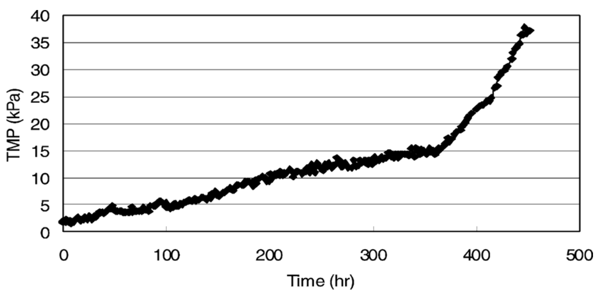


Figure 3.6: Fouling stages for MBR operated under subcritical constant flux circumstances

3.6 Membrane characteristics

Since the membranes involved in the research described in this dissertation all have comparable micro-scale characteristics this topic is only discussed briefly. Four membrane characteristics linked to fouling are considered: configuration, material, pore size, pore distribution and hydrophobicity.

3.6.1 Configuration

The configuration of the membrane modules affects the hydrodynamic circumstances near the membrane surface, but does not affect the filterability of the activated sludge. In other words, an activated sludge sample that shows poor filterability in a sidestream system will highly likely show similar behaviour in an immersed system. This hypothesis is confirmed by Le-Clech et al. (2005), who observed similar fouling propensity in short-term experiments with specific crossflow circumstances for both configurations. Differences in performance are all explained by differences in hydraulic circumstances and maintenance conditions rather than by differences in activated sludge properties.

3.6.2 Material

Since in MBR technology the application of ceramic membranes is (still) negligible compared to polymeric membranes only the latter are discussed here. As mentioned in chapter 2.4 several different polymeric-based membrane materials are available. A study by Yamato et al. (2006) demonstrated PE membranes to be more sensitive to irreversible fouling compared to PVDF membranes. For the research described in this dissertation the influence of membrane material on fouling is less interesting, since all considered MBR plants and the measuring tool are equipped with PVDF membranes.

3.6.3 Pore size and distribution

A membrane parameter susceptible to affect the fouling process is the pore size of the membrane. Since the effect of the pore size on fouling is closely related to the properties of the activated sludge the results reported in literature show opposite trends. It can be expected that a smaller pore size leads to rejection of smaller particles and the build-up of a cake layer with a higher resistance. On the other hand, larger pores are more suspicious to internal pore blocking and thus to irreversible fouling. In this research the influence of the pore size on fouling plays a minor role, since the membrane in the applied measuring tool and the considered MBR plants are all of the ultrafiltration type.

3.6.4 Hydrophobicity

As a consequence of hydrophobic interaction between feed constituents and the membrane material, fouling problems are expected to be more problematic with hydrophobic rather than hydrophilic membranes (Chang et al., 1999; Madaeni et al., 1999; Yu et al., 2005). Nonetheless, in practice membranes generally have a hydrophobic character. Besides, changes in membrane hydrophobicity are difficult to assess as they are often accompanied by changes in other important membrane characteristics such as pore size and morphology (Judd 2006). In the research described in this thesis the influence of membrane hydrophobicity on fouling is left out of consideration.

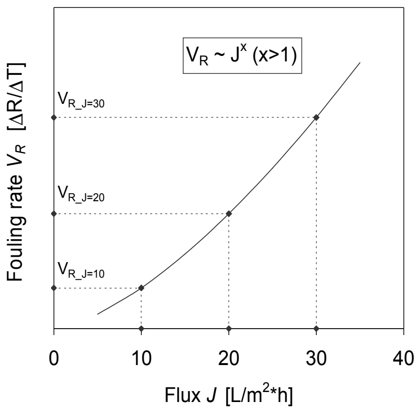
3.7 Membrane operation

MBRs are routinely operated under constant-flux conditions; hence the number of studies dealing with constant-TMP operation is limited. General outcome of studies comparing both operation modes is that constant-flux operation is the more favourable of the two since the relatively high initial flux at constant-TMP operation creates rapid fouling. On the other hand Le-Clech *et al.* (2006) suggest that constant-flux operation may result in irreversible fouling, as this operating mode tends to promote the mechanism of internal fouling of the membrane.

3.7.1 Flux

Evidently the flux is a main operational parameter affecting fouling. Because activated sludge has a high fouling potential MBR systems are usually operated under relatively low fluxes, preferably below the so called *critical flux*. Field et al. (1995), who introduced the critical flux concept, state that: "*The critical flux hypothesis for MF and UF processes is that on start-up there exists a flux below which a decline of flux with time does not occur; above it fouling is observed.*" After its introduction several slightly different definitions of the critical-flux concept have been proposed. In its most strict form the critical flux is equated to the clean water flux under similar operating conditions. However, an alternative weak form of the critical flux was formulated since clean water fluxes can not be achieved in practice for real feed waters due to adsorption of feed components to the membrane. In this approach the critical flux is the flux rapidly established and maintained after start-up of filtration, but not necessarily equal to the clean water flux. Nonetheless, several studies have demonstrated that in practice even the weak form of the critical flux can not be maintained (Brookes et al., 2004; Ognier et al., 2001; Wen et al., 2004). Feed constituents will inevitably and continuously absorb to the membrane surface for any flux, even at a zero flux, resulting in a constant increase of the fouling resistance in time.

In short-term filtration experiments Le-Clech et al. (2003), Gugliemi et al. (2007) and Geilvoet (2004) found fouling rates to increase exponentially with the flux. This exponential relationship implies that the filtration process is optimal for an evenly distributed flux over the membrane surface, see also Figure 3.7. In this figure the fouling rates for two hypothetic situations are represented: (1) even flux distribution and (2) uneven flux distribution. The average flux and total permeate production is similar for both cases, but the average fouling rate (V_R) for the situation with uneven distribution of the flux is higher. When parts of the membrane surface are not or less available for permeation the flux in the remaining membrane surface increases (to reach a similar total permeate flow rate) and the overall fouling rate will increase, irrespective of the activated sludge properties. The higher the distribution of the flux (ΔJ), the higher the fouling rate will be.



1. Even flux distribution

- $J = 20$
- $Q_1 = A * V_{J=20}$
- $V_{R1} \sim 20^x$

2. Uneven flux distribution

- $J = 10$ (50%) and 30 (50%)
- $Q_2 = 0{,}5A * V_{J=10} + 0{,}5A * V_{J=30}$
- $V_{R2} \sim (10^x + 30^x)/2$

$Q_1 = Q_2$
$X > 1 \rightarrow V_{R1} < V_{R2}$

Figure 3.7: Increasing fouling rate for uneven flux distribution

The analysis above stresses the importance of an even distribution of the permeate flow over the total membrane surface. This is however not self-evident in practice, especially when the scale (i.e. membrane surface) of the plant increases. The issues of irregular flux distribution and effective membrane surface are discussed more detailed chapter 5.3.

3.7.2 Crossflow

The balance between the flux and the degree of turbulence induced by the crossflow circumstances directly determines the extent of clogging and (short-term) fouling in an MBR system. As discussed in chapter 2.4 the turbulence in a sidestream system can be regulated by adjusting the crossflow velocity. A too high crossflow velocity should however be avoided, as it damages the floc structure and promotes the release of soluble microbial products from the activated sludge flocs into the free water (Tardieu et al., 1999; Wisniewski et al., 1998, see subsection 3.8.7). The degree of turbulence created by crossflow in tubular membranes is closely related to the diameter of the tube and the density and viscosity of the (activated sludge) liquid. This issue is discussed more extensively in chapter 5.6.

Choi et al. (2005) report a high CFV to be more effective in preventing fouling in a MF rather than in a UF membrane. This might be explained by the hypothesis that at a low flux the particles enter the relatively big pores of a MF membrane more easily, while at a higher CFV the particles do not deposit on or in either membrane. Considering the relatively low CFV applied in practice the use of UF membranes seems preferable.

In MBRs for municipal wastewater treatment crossflow circumstances are predominantly created by coarse bubble aeration. At the same air flow rate coarse bubbles create a higher turbulence and liquid crossflow velocity than fine bubbles (Sofia et al., 2003). Intuitively a higher aeration rate would result in higher turbulence and shear rates and thus decrease the fouling rate. Ueda et al. (1997) report an optimum aeration rate beyond which a further increase had no significant effect. As with liquid crossflow also a too intensive aeration can result in a damage of the flocs and subsequent deterioration of the activated sludge filterability (Ji et al., 2006; Park et al., 2005).

3.7.3 Cleaning

As discussed in chapter 2.4 membrane cleaning measures can be subdivided in physical and chemical cleaning. Physical cleaning techniques for MBR systems include relaxation and back flushing. Evidently the tenacity of the foulants determines to a large extent the efficiency of physical cleaning. As a consequence no universal optimum for the physical cleaning regime can be specified. Ng et al. (2005) report that relaxation allows filtration to be maintained for longer periods before cleaning is necessary. On the other hand relaxation has been considered to be economically unfeasible for large-scale MBR plants (Hong et al., 2002). Key parameters determining the efficiency of back flushing (besides the activated sludge properties) are frequency, duration, the ratio between these two and the intensity. Jiang et al. (2005) found less frequent, but longer back flushing (600 s filtration, 45 s flushing) to be more effective than more frequent and shorter back flushing (200 s filtration, 15 s flushing).

3.8 Activated sludge properties

The role of the activated sludge properties in the fouling process of MBR systems is an intensively investigated topic. Activated sludge characteristics can be classified in several different ways. A common way to classify activated sludge is on the basis of *physical characteristics* and *constituents* (Metcalf and Eddy, 2003). A crucial characteristic related to fouling is the size of the activated sludge constituents; this issue is discussed in subsection 3.8.1.
Physical characteristics of activated sludge that can be mentioned are: *suspended solids concentration, floc size distribution*, *temperature, viscosity, sludge volume index*, conductivity, colour, conductivity and pH. In subsections 3.8.2 to 3.8.6 the physical characteristics represented in italic are discussed with respect to their influence on the fouling process.
Activated sludge constituents that can be mentioned are: *extracellular polymeric substances, soluble microbial products,* biological oxygen demand and chemical oxygen demand, total organic carbon, nutrients (nitrogen and phosphorus), metals and priority substances. In MBR fouling research the attention is predominantly focused on the role of EPS and SMP. An analysis of the role of EPS and SMP and fouling is represented in subsections 3.8.7 and 3.8.8 respectively.

3.8.1 Size fractions

Evidently the *size* of the activated sludge constituents plays a major role in the fouling process. Judd (2006) broadly states that "*evidence suggests that it is the physical nature, and especially the size, of the foulant that has the greatest impact on its fouling propensity*". In addition to this citation it can be stated that in the MBR filtration process a certain size *range* with a lower and upper size limit can be marked off which is relevant with respect to the fouling process. From a straightforward point of view it can be argued that the smallest substances (in solution) can pass the membrane pores with the permeate without participating in the fouling process whereas on the other size spectrum the relatively big (suspended) material is restrained from reaching the membrane surface thanks to the crossflow backtransport mechanisms. This implies that fouling is caused by substances with a size approximately similar to the membrane pore size on the lower size spectrum and particles that are just not carried off by the backtransport forces on the upper size spectrum. An estimation of the upper critical particle size is addressed in chapter 5.6, but anticipating on this analysis it is already mentioned that in theory only particles up to several micrometers are subject to being involved in the fouling process (for typical MBR operational circumstances).

The conventional scheme to classify different size ranges of constituents in activated sludge is based on the distinction between three fractions: (1) suspended solids, (2) colloids and (3) solutes.
Different methods are used to obtain these three fractions. Colloids and solutes are generally separated from suspended solids by centrifugation, but sedimentation and paper filtration are

also applied (Evenblij 2005). Subsequently colloids and solutes are separated by dead-end filtration membrane cells, but the pore size of these membranes is not standardised. Since no standardised fractionation method exists, the size range allocation for the different fractions is divergent. In this research the classification as proposed by Metcalf and Eddy (2003) is used, who state that colloidal particles range from 0.01 to 1.0 µm. This results in the following classification for each of the three fractions:

- Suspended solids: > 1 µm
- Colloids: 0.01 – 1 µm
- Solutes: < 0.01 µm

Especially in the MBR filtration process a clear classifications is important, because the relevant size range of foulants covers the transition between different fractions. Evidently this leads to divergent results with respect to the relative contribution of each of the fractions to fouling. This is illustrated in Figure 3.8, which summarises the outcome of several of these studies, collected by Judd (2005). For example, in the one of the studies by Itonaga et al. (2004) the relative contribution of the soluble fraction to fouling was found to be approximately 60%. However, in this study the soluble fraction was obtained by dead-end filtration over a filter with a pore size of 0.5 µm, significantly bigger than the pore size of the MBR membrane used in this study (0.1 µm). In the study of Wisniewski et al. (1998) the soluble fraction was defined as the fraction after filtration over a 0.05 µm filter. Although the membrane used in this particular study was of the MF type with a nominal pore size of 0.2 µm the relatively big "soluble" substances are more prone to accumulate in the cake layer.

The above mentioned considerations emphasise that the results as presented in Figure 3.8 strongly depend on the used methodology (definition of the fractions, membrane type and membrane operation). In addition also the biological state of the activated sludge can be expected to differ and influence the results of the different studies.

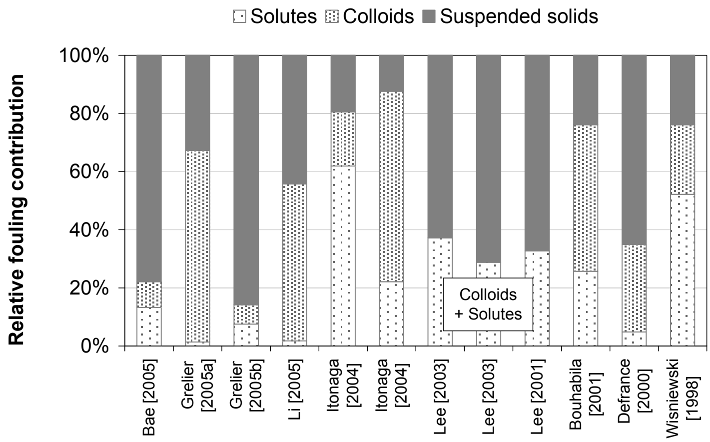


Figure 3.8: Relative contribution of different activated sludge fractions to MBR fouling reported in literature

As mentioned before, due to the crossflow backtransport mechanisms only the relatively small substances, in the size range of the membrane pores up to several micrometers, are involved in the fouling process. From this point of view it seems that especially colloidal material plays a crucial role in fouling. In this sense the significance of colloids is totally different from the conventional activated sludge process. In the conventional secondary clarification step colloids do not settle and leave the process with the effluent, but their share to the effluent quality is marginal compared to the solutes.

3.8.2 Suspended Solids concentration

Although at first sight a higher biomass concentration would seem an indicator for fouling propensity in practice the relationship between mixed liquor suspended solids (MLSS) concentration and fouling turns out to be way more complex. In literature an increase of the MLSS concentrations is reported to have a negative (Cicek et al., 1999; Chang and Kim, 2005), but also a positive (Defrance et al., 1999; Brookes et al., 2006) or insignificant (Le-Clech et al., 2003; Lesjean et al., 2005) impact on MBR performance. In other studies a threshold is reported above which the MLSS concentration promotes fouling (Lubbecke et al., 1995; Rosenberger et al., 2002). Above mentioned contradictory literature results at least seems to indicate that MLSS concentration itself is a poor parameter to indicate the fouling propensity of activated sludge. A presumable explanation for this is that the MLSS *concentration* itself does not provide any information about the *size* of the suspended solids. It can be expected that a if the activated sludge contains a high concentration of small suspended solids (up to several micrometer) the activated sludge has a higher potential to cause fouling compared to sludge that is constituted of suspended solids with a size of several tens of micrometers.

3.8.3 Particle Size Distribution

Since the size of an activated sludge floc is closely related to specific biological and operational circumstances the values mentioned in literature differ significantly. When other circumstances are left out of consideration, mean floc sizes mentioned in literature are 240 µm (Cabassud et al. 2004), 30 to 60 µm (Zhang et al., 1997), 25 µm (Bae et al., 2005) and 3.5 µm (Cicek et al., 1999). A main factor influencing the floc size is the shear stress experienced by the biomass. For example, Wisniewski and Grasmick (1998) report a mean floc size of 125 µm at a shear stress of 1.3 N/m^2 (in a tubular membrane module, 6 hours of recirculation). However, when the shear stress was increased to 72 N/m^2 the mean floc size decreased to 20 µm. The mean floc size itself provides no clear indication about the fouling propensity of an activated sludge. Given their relatively large size compared to the membrane pores sludge flocs can not block the membrane pores. Furthermore flocs are impeded from depositing on the membrane surface by drag forces and shear-induced diffusion (Judd, 2006). Nonetheless, floc size might affect the fouling process indirectly through the ratio between the concentrations of extracellular polymeric substances (subsection 3.8.6) and soluble microbial products (subsection 3.8.7).

3.8.4 Temperature

Activated sludge temperature affects the filtration process through its influence on the liquid viscosity (Darcy's law, see equation 2-1); a lower temperature results in a lower viscosity and a higher resistance to filtration. But besides this direct effect of the temperature several secondary influence factors can be mentioned, mostly related to the properties of the biomass:

- A low temperature promotes deflocculation of the activated sludge and results in the release of soluble microbial products (see subsection 3.8.7) into the activated sludge, which are widely linked to fouling.
- The activity of the biomass decreases at lower temperatures. As a result feed constituents that otherwise would have been degraded by the biomass can reach the membrane surface at lower temperatures and contribute to fouling.
- At lower temperatures the viscosity of the activated sludge liquor increases; this reduces the shear stress created by the coarse bubble aeration (in immersed systems).
- Particle back transport velocity near the membrane surface decreases at lower temperatures (Judd, 2006).

In addition it can be expected that the activated sludge temperature affects the properties of the membrane. At higher temperatures the membrane material will expand, resulting in a decrease of the size of the membrane pores. As discussed previously, the pore size is considered to be of importance with respect to fouling.

3.8.5 Sludge Volume Index

Although the Sludge Volume Index (SVI) is a parameter related to conventional activated sludge systems with a secondary clarification step it is also linked to fouling in MBR systems.
As discussed in chapter 2.3 poor settling properties (high SVI) are associated with the presence of filamentous bacteria. Several researchers found filamentous bulking to have an impact on MBR fouling (Chae *et al.,* 2006; Meng and Yang, 2007; Su et al., 2007; Sun et al., 2007). An overgrowth of filamentous bacteria is suspected to lead to an increase of the concentration of Extracellular Polymeric Substances (see section 3.8.7) and the viscosity (Meng et al., 2009). In addition the filamentous bacteria may enlace and fix the foulants on the membrane surface (Meng et al., 2009). As with other activated sludge properties research results are contradictory, considering the results reported by Fan et al. (2006) who state that the SVI itself is not a reliable parameter to predict fouling.

3.8.6 Viscosity

The viscosity of permeate can be assumed equal to clear water and is solely dependant on its temperature (see equation 2-2). The viscosity of activated sludge is predominantly determined by its MLSS concentration. The role of activated sludge viscosity in the MBR filtration process will be discussed extensively in chapter 5.6. For now it is mentioned that an increasing activated sludge viscosity has a negative impact on MBR fouling. In the first place the efficiency of the coarse bubble aeration reduces, resulting in lower shear rates and turbulence near the membrane surface. Besides this the oxygen mass transfer efficiency decreases, which is also detrimental for the activated sludge filterability properties (through the release of SMP, see subsection 3.8.7).

3.8.7 Extracellular polymeric substances

Activated sludge flocs contain a wide range of bacteria and protozoa. They are held together by so called extracellular polymeric substances (EPS). Flemming and Wingender (2001) describe EPS as "*the construction materials for microbial aggregates such as biofilms, flocs and sludge*". EPS has an essential function with respect to the survival of the biomass. Besides holding them together EPS forms a protective surrounding layer that protects the micro-organisms against harmful influences from the outside. In MBR fouling research EPS is often used as a generic term that encompasses a wide range of macromolecules. The characteristics of EPS are influenced by various factors, such as gas sparging, substrate composition, loading rate and solids retention time. Table 3.2 outlines the four main EPS components and the range of their relative content. The numbers presented in Table 3.2 emphasise that EPS has an extreme heterogeneous nature.

Table 3.2: Composition of EPS and range of relative component concentrations (Flemming and Wingender, 2001)

Component	Relative content in EPS
Polysaccharides	40 – 95%
Proteins	< 1 – 60%
Nucleic acids	< 1 – 10%
Lipids	< 1 – 40%

With respect to membrane fouling two basic forms of EPS are distinguished: *bound* (or *extracted*) *EPS* and *soluble EPS*. Bound EPS derives directly from the active cell, while soluble EPS is solubilised in the free water. Figure 3.1 shows a schematic representation of the mechanisms that affect the extracted and soluble EPS concentrations. Soluble EPS are released from the bacterial cell during cell lysis and diffuse through the cell wall into the free water. Soluble EPS can also be introduced with the influent. Especially soluble EPS (now generally termed *soluble microbial products*) are strongly linked to fouling and will be discussed in detail in section 3.8.7.

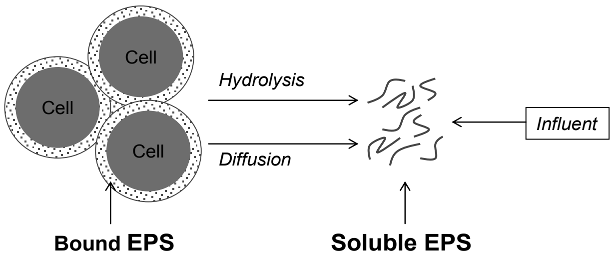


Figure 3.9: Schematic representation of bound and soluble EPS

The number of publications dealing with the role of (bound) EPS in the MBR filtration process is high and the results are diverse. An important issue in this matter is that no standard measuring protocol for the determination of the bound EPS concentration exists. A commonly used method to extract EPS is cation exchange resin (Jang et al., 2005; Gorner et al., 2003; Frølund et al., 1996), but also heating methods (Morgan et al., 1990) and centrifugation with formaldehyde (Zhang et al., 1999) are mentioned in literature.
Many researchers have linked EPS to fouling (Cho and Fane, 2002; Chang and Lee, 1999; Nagaoka et al., 1996; Nagaoka and Yamanishi, 1998; Rosenberger and Kraume, 2002). Especially the solids retention time is considered an important factor affecting the EPS concentration. Brookes et al. (2003) report decreasing EPS concentrations with increasing SRT; above an SRT of 30 days no further decrease was found. Cho et al. (2005) report bound EPS to have no significant effect on the specific resistance in the range below 20 and above 80 mg EPS/g MLSS, but to play an important role in between these two limits. Fawehinmi et al. (2004) report the resistance to increase linearly with increasing bound EPS concentration from 20 to 120 mg EPS/g MLSS. Gorner et al. (2003) report both EPS proteins and polysaccharides around the biological cell walls to be a key parameter in the flocculation process, thereby playing a major role in the fouling process.

An important critical note with respect to the EPS analyses in use is that they only provide information about the characteristic *concentration* and not about the *size*. It can be expected that EPS which are bound to sludge flocs of several tens of micrometers can not participate in the fouling process, whereas this could be the case for EPS bound to a single bacterial cell.

3.8.8 Soluble microbial products

Soluble microbial products (SMP) can be defined as *"soluble cellular components that are released during cell lysis, diffuse through the cell membrane, are introduced with the influent, are lost during synthesis or are excreted for some purpose"* (Laspidou et al., 2002; Li et al., 2005). Actually the term "soluble microbial products" is a misnomer, since SMP consist of both soluble and colloidal material (Judd, 2006).

Although SMP are now widely considered a crucial factor affecting MBR fouling, it was only in 2001 when SMP levels in MBR systems were reported for the first time (Chang, 2002). During filtration, SMP materials are thought to absorb onto the membrane surface, block membrane pores and form a gel structure on the membrane surface (Rosenberger et al., 2005). The influence of SMP on MBR fouling was demonstrated in several different ways. Ng et al. (2005) found a higher filtration resistance from the supernatant than from the biomass at a MLSS concentration of 4 g/L. Various fractionation experiments demonstrated higher SMP concentrations in the free water than in the permeate, which implies a retention and thus resistance increase as a result of SMP (Lesjean et al., 2005; Brookes et al., 2003; Evenblij and Graaf, 2004). Reported protein retention percentages are varying between 15% (Evenblij and Graaf, 2004) and 90% (Drews et al., 2005).

A logical strategy to reduce SMP fouling would be to minimise the production of EPS and SMP. The knowledge about the exact mechanisms that affect the production of EPS and their release into the free water as SMP is however limited. In the first place organisms excrete soluble organic material to establish concentration equilibrium across the cell membrane. Besides this the release of SMP into the free water is reported to be influenced by starvation of the biomass. Biomass starvation is influenced by various circumstances. Especially stress circumstances such as (sudden) temperature changes, osmotic pressure changes, deficit or surplus of nutrients, toxic influent shock and high shear circumstances can be mentioned (Kuo, 1993; Barker and Stuckey, 1999; Evenblij and Graaf, 2005).

A difficulty in understanding the role of SMP in MBR fouling is the diversity of methods to determine the SMP concentration of a given sample. In the first place there is no universal method for determining the total SMP concentration as it contains so many different constituents. A now often used simplification is to represent the SMP concentration as the sum of the protein- and polysaccharide concentrations (Rosenberger, 2002; Lesjean, 2005; Evenblij, 2005). Other analyses to characterise the SMP concentration are determination of the total organic carbon (TOC) level (Gao et al., 2004) and specific UV-absorbance (SUVA) measurements (Shin and Kang, 2003). Besides the issue of the multitude of constituents, three methods are used to separate the free water from the biomass: centrifugation, sedimentation and paper filtration. Evenblij (2004) found the latter one to be the most effective, but evidently different separation techniques can result in different results.

An important critical note with respect to the used methods to analyse SMP is that (as with the EPS analyses) they solely provide information about the characteristic *concentration* and not about the characteristic *size*. As discussed in subsection 3.8.1 the size of the foulants is considered a crucial parameter. In this context it is important to realise that SMP consist of both *colloidal* and *soluble* material. This implies that part of the SMP substances can pass the membrane without participating in the fouling process while other SMP substances are colloidal and are retained by the membrane. The ratio between “non-fouling” SMP and “fouling” SMP can however not be made on the basis of the colorimetric analyses. The issue of the concentration versus the size of SMP plays an important role in the research described in this thesis.

3.9 Summary and concluding remarks

In this thesis fouling is defined as "the process resulting in the loss of performance of a membrane due to the deposition of suspended or dissolved substances on its external surface, at its pore openings, or within the pores". MBR fouling is a complex phenomenon that is depending on three main factors: (1) the characteristics of the membrane, (2) the operation of the filtration process and (3) the properties of the activated sludge to be filtrated.

In the MBR filtration process three forms of fouling are distinguished: (1) reversible fouling, (2) irreversible fouling and (3) irrecoverable fouling. Although the distinction between these three forms of fouling is based on the cleaning measures required to remove it, the actual difference originates from the tenacity with which the foulants are attached on the membrane surface or in the pores.

3.9.1 Flux distribution and fouling rate

The research described in this thesis particularly focuses on the role of the activated sludge characteristics in the fouling process. Nonetheless, the characteristics and the operation of the membrane should not be neglected. A crucial parameter affecting the fouling process is evidently the flux. As discussed in subsection 3.7.1 several researchers demonstrate an exponential increase of the fouling rate with increasing flux. This implies that the fouling rate will increase in case of irregular distribution of the flux over the total membrane surface, irrespective of the activated sludge properties. In this context it is highly important to maintain a maximum effective membrane surface. This stresses the importance of the hydrodynamic circumstances (and their distribution) in the membrane modules.

3.9.2 Activated sludge properties and fouling

The role of the activated sludge properties in the fouling process is an extensively investigated topic, but despite of this no clear consensus has been reached about which activated sludge characteristic(s) is/are responsible for fouling. Three reasons for this can be mentioned:

- Fouling is not exclusively related to the activated sludge properties, but also to the membrane characteristics and the operational circumstances under which it is filtrated. Each MBR plant has its own unique combination of these three factors, which implies that the influence of the activated sludge properties in the fouling process can not be generalised.
- Fouling can be subdivided in a reversible, irreversible and irrecoverable component. It can be expected that these different forms of fouling are related to different activated

sludge characteristics. However, in various MBR fouling studies the distinction between different forms of fouling are not (yet) made.

- Activated sludge has a complex, heterogeneous and dynamic character. These properties complicate gaining scientific knowledge about its fouling behaviour. The synthetic, stable and homogeneous feed solutions on which fundamental knowledge about fouling is predominantly based are not representative for the nature of activated sludge in practice.

Various researchers report that fouling is predominantly determined by substances in the *free water phase* of the activated sludge. Especially soluble microbial products (SMP, see subsection 3.8.8) are suspected to play a crucial role in fouling. Drawback of the used methods to measure SMP is that they only provide information about the characteristic *concentration* and not about the *size*. The information about the influence of the size of the particles with respect to fouling is limited.

Despite the divergent and even contradictory research results obtained by various researchers some general statements about the influence of the activated sludge properties in the MBR fouling process can be posed:

- Fouling is depending on the concentration and especially on the size of the substances in the activated sludge. Bulk characteristics such as temperature, viscosity, sludge volume index, pH may all play a role, but their influence is indirect.
- In MBR systems relatively big particles are not involved in the fouling process, thanks to the crossflow operation. As a consequence it can be stated that the MLSS concentration itself is a poor parameter to indicate the fouling potential of activated sludge. The upper size limit of particles involved in fouling is depends on the ratio between the forces towards (flux) and away (crossflow) from the membrane. This issue is discussed more in detail in chapter 5.6.
- Also the lower size limit of substances involved in the fouling process can roughly be defined. Substances smaller than the membrane pores (and not accumulating in the cake layer) can pass the membrane without being involved in the fouling process. This implies that on the short term substances which are in solution can be assumed to not be involved in the fouling process (for UF and MF membranes).

4 The Delft Filtration Characterisation method

4.1 Introduction

The Delft Filtration Characterisation method (DFCm) is discussed in two parts. This chapter forms the descriptive part, whereas in chapter 5 several technical aspects of the method are assessed. Section 4.2 addresses the background of MBR fouling research at Delft University of Technology and the considerations that formed the basis for the development and the design of the DFCm. The DFCm consists of a Filtration Characterisation unit (discussed in section 4.3) and an accompanying well-defined measuring protocol (discussed in section 4.4). Section 4.5 addresses the raw output of a DFCm experiment. In section 4.6 the activated sludge analyses that accompany the DFCm experiments are discussed. This chapter concludes with a recapitulation, in section 4.7.

4.2 Background

In 2001 Delft University of Technology started a research project in cooperation with DHV Engineering Consultants. This research resulted in the PhD thesis: "*Filtration characteristics in membrane bioreactors*", by Herman Evenblij (2005). The broad objective at the start of the research was to improve the understanding of the MBR fouling process.

Practice proves that a wide variety of research methods and strategies can be employed to investigate MBR fouling. Since fouling is a complex and dynamic process, involves a multitude of factors and takes place on different time-scales it is practically impossible to cover all aspects of it in a single research program. Evenblij decided to focus the attention on the role of the activated sludge properties in the MBR filtration process. In order to do so a filtration apparatus and an accompanying measuring protocol were developed, later called the Delft Filtration Characterisation method (DFCm).

The primary feature of the DFCm is that it presents a method which allows filtration of different activated sludge samples with the same membrane (properties) and under identical hydraulic circumstances. In this way differences in filterability, as measured with the DFCm, can be attributed exclusively to differences in activated sludge properties. The DFCm thus offers the possibility to make an unequivocal comparison between the filterability of activated sludge samples collected from different MBR plants or under different circumstances.

A second feature of the DFCm is its aim to bridge the gap between lab-scale fouling research and the actual MBR filtration process in full-scale MBR plants in practice. As discussed in chapter 1.2 (Research bottlenecks) a common approach to investigate fouling is to simulate the filtration process on lab-scale and to use synthetic wastewater. This approach can be considered useful for gaining fundamental knowledge about the fouling process, but it lacks representativeness with respect to the complex, heterogeneous and dynamically changing activated sludge properties in full-scale MBR plants.

4.3 Filtration Characterisation unit

A schematic representation of the Filtration Characterisation unit is given in Figure 4.1. The final design of the unit is the result of an extensive trial-and-error process. Configurations were tested with different valves, pump types and membrane modules (Evenblij 2006). The main components of the Filtration Characterisation unit are:

- A single tubular vertically placed sidestream UF membrane,
- Equipment for controlled sludge recirculation and permeate extraction,
- A data acquisition system,
- Equipment for membrane cleaning.

It is emphasised that the Filtration Characterisation unit is not a lab-scale bioreactor aiming at simulating the MBR process. It is merely a tool to assess the momentaneous filterability of any given activated sludge sample, preferably but not necessarily originating from a pilot- or full-scale MBR. In order to exclude the role of biological processes on the activated sludge properties and thus on the filtration characterisation results the experiments have to be short and have to be executed as soon as possible after the activated sludge has been collected from the plant.

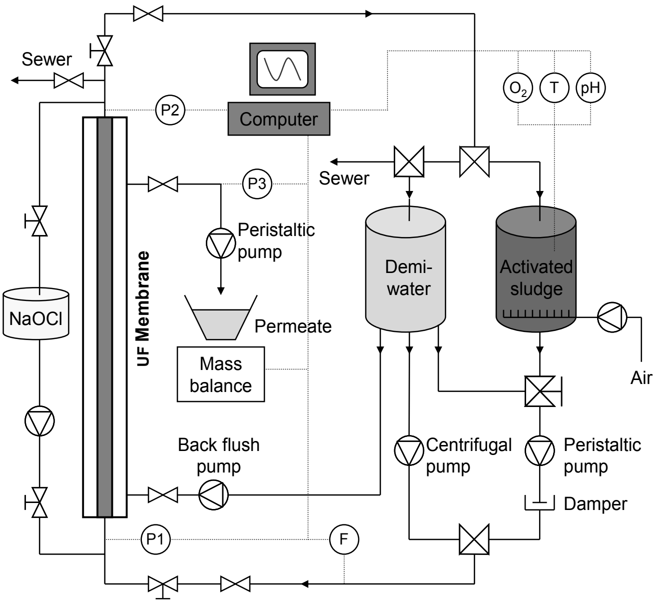


Figure 4.1: Schematic representation of the DFCm Filtration Characterisation unit

4.3.1 Membrane

The heart of the Filtration Characterisation unit is formed by a single tubular X-flow F5385 sidestream UF membrane with an internal diameter of 8 mm and a length of 95 cm. This equals a total membrane surface of 240 cm^2. The membrane is composed of the polymeric material polyvinylidene difluoride (PVDF) and has a nominal pore size of 0.03 µm. A detailed specification of the membrane can be found in Evenblij (2006). The membrane is glued with resin in a PVC tube with an internal diameter of 15 mm, schematically represented in Figure 4.2. The headers at the in- and outlet of the membrane module to which the feed and concentrate pressure sensors are connected also have an inner diameter of 8 mm, ensuring an undisturbed flow and the prevention of local energy losses.

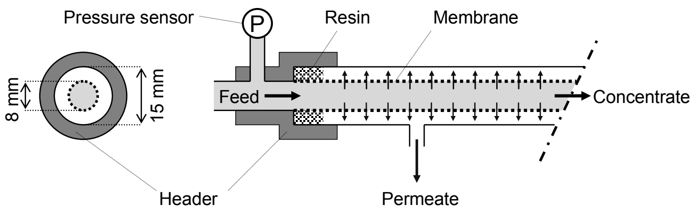


Figure 4.2: Membrane module FCu (Evenblij 2006)

4.3.2 Sludge recirculation and permeate extraction

An activated sludge sample of about 30 litres is recirculated through the membrane tube with a peristaltic displacement pump (Watson-Marlow 700 series). The pulse effect of the peristaltic pump is flattened out by a damper which is inserted at the upstream side of the pump. The pump speed can be adjusted accurately and the flow is measured with an electromagnetic flow meter (Krohne IFC 010K).
A second peristaltic pump (Watson-Marlow 520 series) is used to extract permeate by creating an under pressure at the permeate side of the membrane. The permeate mass production rate can also be adjusted by tuning the pump speed and is measured in time with a mass balance (Mettler Toledo).

4.3.3 Data collection and -processing

During a sludge filtration experiment several process- and sludge parameters are monitored. Using the software application Testpoint a program was designed that shows all relevant parameters online and logs them in an EXCEL file. For each time step of 12.5 seconds the following parameters are logged:

Activated sludge characteristics: During a filtration characterisation experiment the temperature [ºC] and pH [-] of the activated sludge are measured with standard electrodes (type WTW).

Crossflow velocity: The activated sludge flow rate Q [m^3/h] created by the peristaltic sludge pump is measured with the electromagnetic flow meter. Since the membrane diameter D [$8.0 \cdot 10^{-3}$ m] and thus the cross sections A_{cs} of the membrane [m^2] is known the crossflow velocity *CFV* can be calculated according to:

$$\mathrm{CFV} = \frac{Q}{A_{cs}} = \frac{Q}{0.25 \cdot \pi \cdot D^2} \quad [\mathrm{m/s}]$$

To obtain the standard *CFV* of 1.0 m/s a flow rate of 181.0 L/h is required.

Trans Membrane Pressure: The TMP represents the average pressure difference between the feed- and the permeate side of the membrane. To measure the TMP three pressure transmitters (type JandM CB3010) are installed at the feed- (P_1), concentrate- (P_2) and permeate (P_3) side of the membrane, see Figure 4.1. Because the permeate pressure sensor is placed in the vertical middle of the membrane the TMP can be calculated according to Equation 4-1:

$$\mathrm{TMP} = \frac{P_{feed} + P_{concentrate}}{2} - P_{permeate} \quad [\mathrm{Bar}] \qquad (4\text{-}1)$$

Permeate flux: With the mass balance the permeate production rate *dM/dt* [g/s] is monitored. The density ρ [g/m^3] of permeate is assumed to be equal to water (1000 kg/m^3). Since the available membrane surface A_m [m^2] is known the permeate flux J can be calculated according to Equation 4-2:

$$J=\frac{dM}{dt}\times\frac{3600}{A_m\times\rho}\ [L/\ m^2\cdot h] \tag{4-2}$$

Filtration resistance: Since the temperature of the activated sludge is known, the dynamic viscosity η of permeate can be calculated according to Equation 2-2. Subsequently, from the *TMP* [Pa] and the flux J [m/s] the total filtration resistance during filtration R_{tot} [m^{-1}] can be calculated from Darcy's law (see also Equation 2-1 in chapter 2.4.4):

$$R_{tot}=\frac{TMP}{\eta\cdot J}\ [m^{-1}] \tag{4-3}$$

4.3.4 Cleaning equipment

After each (activated sludge) filtration experiment the membrane in the Filtration Characterisation unit is cleaned. To do this a combination of three methods is available. Two centrifugal pumps can be used to perform either a forward flush or a back flush. Furthermore a small peristaltic pump is implemented that can be used to soak the feed side of the membrane with cleaning chemicals. The available cleaning measures are discussed more in detail in subsection 4.4.3 (Measuring protocol).

4.4 DFCm measuring protocol

In order to obtain unequivocal and mutually comparable results it is crucial that all filtration characterisation experiments are conducted according to the same procedure. For this reason a measuring protocol was formulated (Evenblij 2005), consisting of the following three steps:

- Membrane resistance determination,
- Activated sludge filtration,
- Membrane cleaning.

4.4.1 Membrane resistance determination

To be able to exclude the influence of the membrane properties on the filtration characterisation results the starting condition of the membrane has to be similar at the start of all experiments. In order to verify this the membrane resistance is determined prior to each sludge filtration experiment. Demineralised water is recirculated with a crossflow velocity of 1.0 m/s while permeate is extracted at a flux of 80 $L/m^2 \cdot h$. For a clean membrane the filtration resistance should be about $0.4 \cdot 10^{12}$ m^{-1} ($\pm$ 0.1 $\cdot 10^{12}$)[1]. In case of a too high membrane resistance at the start of an activated sludge filtration experiment supplementary cleaning measures are required. If the membrane resistance can not be restored below $0.5 \cdot 10^{12}$ m^{-1} the membrane has to be replaced.

4.4.2 Activated sludge filtration

The main step of the measuring protocol is the activated sludge filtration step. After the membrane resistance has been assessed an activated sludge sample of about 30 litres is recirculated with a flow rate of 181 L/h, corresponding with a crossflow velocity of 1.0 m/s in the membrane. Crossflow velocities used in practice in sidestream systems are in the range between 1 and 6 m/s. In immersed systems the liquid CFV is lower than 1 m/s, but in the DFCm membrane tube this value is required to exclude the possibility of clogging (Evenblij 2006).

Based on experiences with different types of activated sludge a standard value of the applied flux was chosen to be 80 $L/m^2 \cdot h$ (Evenblij 2006). Filtration lasts until a permeate production of at least 25 L/m^2 or the threshold TMP of 0.6 Bar is reached. The flux can be either increased or decreased respectively in subsequent filtration tests depending on extreme good or bad sludge filterability.
As a result of permeate extraction the MLSS concentration of the sludge sample increases during the experiment. A permeate production of 25 L/m^2 corresponds with a permeate volume of 0.6 litre. When commencing a DFCm experiment with a sludge sample of 30 litres the

[1] A filtration resistance of $0.4 \cdot 10^{12}$ m^{-1} corresponds with a TMP of 0.1 Bar (at T = 15ºC and J = 80 $L/m^2 \cdot h$)

concentration factor amounts up to only 2%. This concentration increase is assumed to be of no influence on the filtration characterisation results.

4.4.3 Membrane cleaning

After the activated sludge filtration step the membrane is cleaned for the next experiment. As briefly discussed in subsection 4.3.4, this can be done by a combination of three methods: (1) forward flush, (2) back flush and (3) chemical cleaning. After each cleaning method the membrane resistance is determined to verify whether the membrane is clean again.
With a centrifugal pump a forward flush with demineralised water can be executed. The pump creates a crossflow velocity that exceeds 5 m/s. A second centrifugal pump is connected to the permeate side of the membrane. With this a pump a back flush can be performed that creates a TMP of approximately -0.75 Bar. Finally the membrane can be soaked with chemicals. In this research only sodium hypochlorite (NaOCl) was used. The standard applied NaOCl concentration was 500 ppm. In case of tenacious fouling an increased concentration of 1500 ppm was used.

4.4.4 Summary measuring protocol

To conclude this section the measuring protocol is briefly summarised below:

Membrane Resistance determination

- Recirculation of demineralised water at CFV = 1.0 m/s,
- Permeate extraction at J = 80 $L/m^2 \cdot h$,
- Determination of the (constant) filtration resistance ($0.4 \cdot 10^{12}$ m^{-1} ± $0.1 \cdot 10^{12}$)
- Supplementary cleaning measures in case of a too high membrane resistance (>$0.5 \cdot 10^{12}$ m^{-1}).

Activated sludge Filtration

- Activated sludge recirculation at CFV = 1.0 m/s,
- Permeate extraction at standard flux of J =80 $L/m^2 \cdot h$,
- Permeate extraction until a permeate production of 25 L/m^2 or a TMP of 0.6 Bar is reached,
- Reconsideration of the flux in case of extreme bad or good filterability.

Membrane Cleaning

- Forward flush with demineralised water at CFV > 5 m/s,
- Back flush at TMP = ± -0.75 bar,
- Chemical cleaning by soaking with NaOCl (500 - 1500 ppm), at least 15 minutes.

4.5 DFCm output

This section briefly addresses the output and the conventional representation of the DFCm experiments. Based on the theory of cake layer filtration a new representation will be proposed in chapter 5.7.

4.5.1 Filtration curve

As discussed in subsection 4.3.3 several process parameters and activated sludge parameters are monitored during a filtration characterisation experiment. In the output all these parameters are plotted with the specific permeate production volume V [L/m^2]. During a filtration experiment the TMP and the filtration resistance R_{tot} will increase as a result of fouling; the other process parameters (J, CFV) and sludge parameters (temperature, pH) remain practically constant. In general filtration is maintained until a permeate production of 25 L/m^2, which results in a dataset of 90 values. In case of poor filterability the dataset is smaller because filtration has to be stopped with respect to the upper TMP limit of 0.6 Bar.

The main output of a DFCm experiment is a chart in which the value of the increase of the filtration resistance (ΔR) is plotted against the specific permeate production (V); an example for three experiments is represented in Figure 4.3. A closer analysis of the obtained DFCm curves shows that the trend of the filtration resistance can be fitted with high correlation factors to two types of functions, namely a quadratic (Q) and a power (P) function. The shape of the DFCm curve is discussed in detail in chapter 5.7.

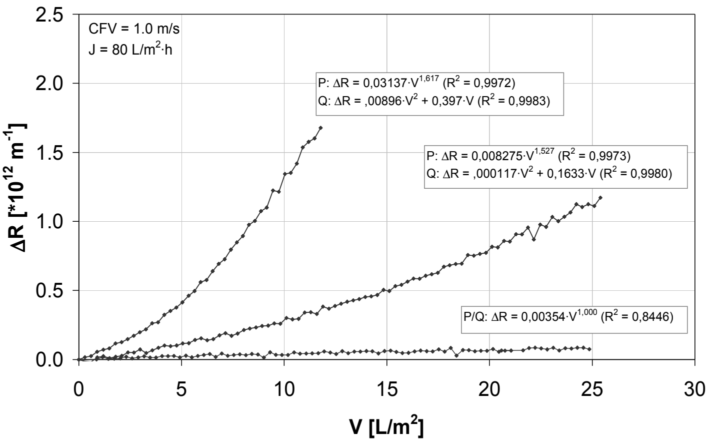


Figure 4.3: Example of DFCm output: filtration resistance with specific permeate production

4.5.2 ΔR_{20} value

Since the membrane resistance is practically similar for all experiments it is excluded in the comparison between different curves. Instead the results are compared based on the additional resistance that is caused by fouling. In order to be able discriminate different experiments it was decided to compare curves on the basis of the resistance increase after a permeate production of 20 L/m^2 (Evenblij 2006). For standard filtration circumstances (J = 80 L/m^2·h) this permeate volume is reached after 15 minutes of filtration. The resistance increase at this point is indicated as the so called ΔR_{20} value.

The ΔR_{20} value reflects the *filterability* of the sample. Based on the DFCm results gathered from various MBR plants and batch experiments (Geilvoet 2004, 2005) a practical classification was made to qualify the filterability, see Table 4.1. The representativeness and the significance of the ΔR_{20} value are discussed more in detail in chapter 5.7 based on the theory of cake layer filtration.

Table 4.1: ΔR_{20} and corresponding filterability qualification
(for the standard measuring protocol, CFV = 1.0 m/s, J = 80 L/m^2·h)

ΔR_{20} [·10^{12} m^{-1}]	**Qualification**
0 – 0.1	Good
0.1 – 1.0	Moderate
> 1.0	Poor

4.6 Activated sludge analyses

A major purpose of this research is to link the filterability of activated sludge to its properties. This section addresses the analyses that are performed in the measuring campaigns discussed later in this dissertation.

4.6.1 Temperature, pH and dissolved oxygen concentration

The temperature, pH and dissolved oxygen concentration of the activated sludge samples are monitored during filtration with standard electrodes (type WTW).

4.6.2 Mixed Liquor Suspended Solids

Since the *Mixed Liquor Suspended Solids* (MLSS) concentration is a prominent activated sludge characteristic it is analysed for all DFCm experiments. The MLSS concentration is determined according to the *NEN6621* standard.

A SchleicherandSchuell filter paper (D = 70 mm) is rinsed with demineralised water and subsequently dried and weighed. A known volume of sludge (50 or 100 ml) is filtrated over the paper in dead-end mode, after which the filter is dried at 105 ºC for a period of at least three hours. The MLSS concentration is represented by the ratio of the filter paper mass difference and the volume of the sample.

4.6.3 Sludge Volume Index

Although the *Sludge Volume Index* (SVI) is a parameter primarily associated with activated sludge settling properties it is also linked to membrane fouling (chapter 3.8). To verify this hypothesis several filtration characterisation experiments are accompanied by SVI determinations. The SVI, an empirical parameter, is generally determined according to the standard *NEN6624* method. An activated sludge sample of 1 litre is poured in a graduated cylinder and allowed to settle for 30 minutes. The SVI represents the ratio between the settled volume of sludge after this period and the corresponding MLSS concentration of the sample:

$$\mathrm{SVI} = \frac{\text{settled volume of sludge [L/L]}}{\text{MLSS [g/L]}} \tag{4-4}$$

The conventional method to determine the SVI is not suitable for activated sludge with high MLSS concentrations. Therefore the Diluted Sludge Volume (DSVI) was introduced (Jenkins, 1993). In this analysis the methodology is the same but the sample is diluted with effluent (or permeate). The value of the SVI equals the product of the DSVI and the dilution factor.

4.6.4 Soluble Microbial Products

As discussed in chapter 3.8.8 so called *Soluble Microbial Products* (SMP) are generally considered to be major foulants in the MBR filtration process. Since SMP represents a generic term for a wide range of substances it is practically impossible to determine the exact SMP concentration of a sample. A widely accepted simplification to express the total SMP concentration is the sum of the protein- and polysaccharide concentrations in the free water (Rosenberger, 2003). Different methods are available to determine SMP concentrations.
As a first step the free water has to be separated from the activated sludge. The generally applied methods to do so are paper filtration and centrifugation (in combination with paper filtration). Based on research by Evenblij and Van der Graaf (2004) it was decided to use paper filtration for the sludge-water separation (Schleicher and Schuell 589^2 filter paper with a pore size of 7 to 12 μm).

Proteins

The most common methods to determine protein concentrations are the ones as proposed by Lowry et al. (1951) and Bradford (1976). In general the method of Lowry is preferred, because the Bradford method does not allow for measuring macro-molecules with eight or more peptide bonds (Te Poele, 2005). Delft University uses the modified method described by Rosenberger (2003), which is based on the method of Lowry et al (1951). For the calibration Albumin Bovine, BSA, (Acros) fraction V, in a concentration range of 0-25 mg/L is used. The measured concentration can be calculated with the measured light extinction at 750 nm and the calibration curve.

Polysaccharides

The most commonly used method for the determination of polysaccharide concentration is the one described by Dubois (1956). In this method polymers are hydrolysed by concentrated sulphuric acid. Phenol is used as a reagent for colour development. Delft University uses the modified method described by Rosenberger (2003), which allows for measuring lower polysaccharide concentrations. For the calibration D(+) glucose (J.T. Baker), in a concentration range between 0-20 mg/L is used. The measured concentration can be calculated with the measured light extinction at 487 nm and the calibration curve.

4.6.5 Viscosity

The viscosity of a fluid can be described as a measure of its resistance to being deformed by shear stress. The *dynamic viscosity* of a fluid is defined as the ratio between the occurring shear stress and the velocity gradient perpendicular to the direction of shear (shear rate):

$$\eta = \frac{\tau_w}{\delta u / \delta z} = \frac{\tau_w}{\gamma_0} \tag{4-5}$$

With:
η = dynamic viscosity, [Pa·s],
τ_w = shear stress, [Pa],
$\gamma_0 = \delta u/\delta z$ = shear rate, [s^{-1}]

Pure water can be considered an incompressible Newtonian fluid, implying that the shear stress is linearly proportional to the velocity gradient. This implies that the viscosity of water is independent of the shear forces acting upon it. As discussed in chapter 2.4 the viscosity of water can be considered solely dependant on its temperature (Equation 2-2). Contrary to clean water, the rheological properties of activated sludge are non-Newtonian or pseudoplastic. This is explained by the bio-particulate structure of the activated sludge. Like polymer solutions, sludge flocks tend to flocculate in a large-scale network. By increasing the shear rate this network is disrupted, resulting in a decrease of viscosity. The main influence factor on activated sludge viscosity is the MLSS concentration (Rosenberger, 2002b; Judd, 2006). Other influence factors on viscosity that can be mentioned are the composition of the applied wastewater and the microbiological structure of the activated sludge (Rosenberger, 2002b).

Three equations are used to describe the rheology of activated sludge:

Bingham model: $$\tau_w = \tau_0 + k \cdot (\gamma_0) \tag{4-6}$$

Ostwald model: $$\tau_w = k \cdot (\gamma_0)^n \tag{4-7}$$

Herschel-Burkley model: $$\tau_w = \tau_0 + k \cdot (\gamma_0)^n \tag{4-8}$$

Two types of devices are used to measure activated sludge viscosity: *rotational* and *tubular* rheometers. The major disadvantage of rotational rheometers is their unsuitability for measuring at high shear rates (Rosenberger, 2002b). However, since rotational rheometers have better commercial availability and are more practical in use they are more commonly applied than tubular ones.

Delft University also uses a rotational rheometer, type Anton Paar. With the rheometer an aluminium cylinder is rotated in an activated sludge sample (100 ml) with several rotation speeds, creating different fixed shear rates (γ_0) in the range from 5 to 1000 s^{-1}. The rheometer measures the torque, from which the shear stress can be calculated. Subsequently the *apparent* viscosity (η_a) is calculated according to Equation 4-5. An example of the output of four random rheological experiments is represented in Figure 4.4.

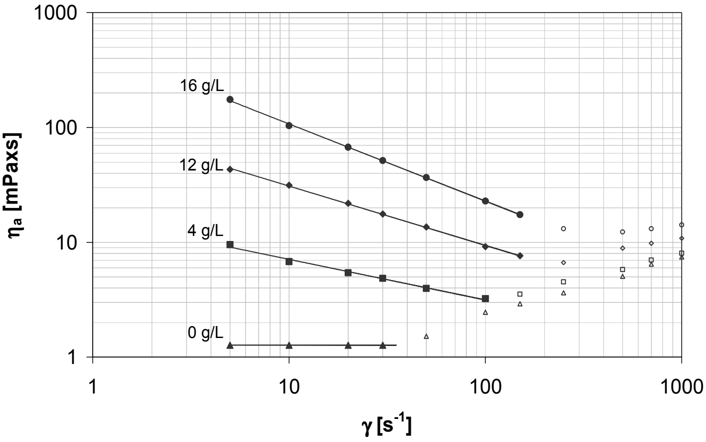


Figure 4.4: Example of raw output rheological measurement
(apparent viscosity with shear rate for 4 activated sludge samples)

The viscosity is highly depending on the MLSS concentration of the sample; a higher MLSS concentration corresponds with a higher viscosity. The open markers in Figure 4.4 confirm the unsuitability of the rheometer to measure at higher shear rates. Depending on the MLSS concentration as from a certain threshold shear rate the so called “slippage” effect occurs and the obtained data are not representative anymore. The activated sludge viscosity at higher shear rates and the rheological circumstances in the DFCm membrane element are discussed more in detail in chapter 5.5.

4.6.6 Particle Size measurements

As discussed in chapter 3.8 colloidal particles are linked to membrane fouling. Delft University uses a HIAC ChemShield MicroCount 100s particle counter which is capable of measuring particles with a diameter in the range between 0.4 and 5.0 µm. A schematic representation of the particle counting set-up is given in Figure 4.5. Since the amount of particles in activated sludge free water is above the measuring range of the particle counter the samples are diluted with demineralised water. The required energy level to generate a flow through the particle counter is created by the demi-water network in the lab and a weir. With a pinch valve the flow rate is adjusted to the required value of 100 mL/min.

As with the SMP analyses the free water is separated from the suspended material by paper filtration (Schleicher and Schuell 589^2 filter paper with a pore size of 7 to 12 µm). A peristaltic pump adds free water to the demi-water flow with a flow of 1 mL/min (dilution factor of 100) just prior to entering the particle counter. The potential influence of demi-water on the particles due to osmotic pressure difference is assumed negligible due to the short contact time. After passing the particle counter the sample is discharged to the sewer.

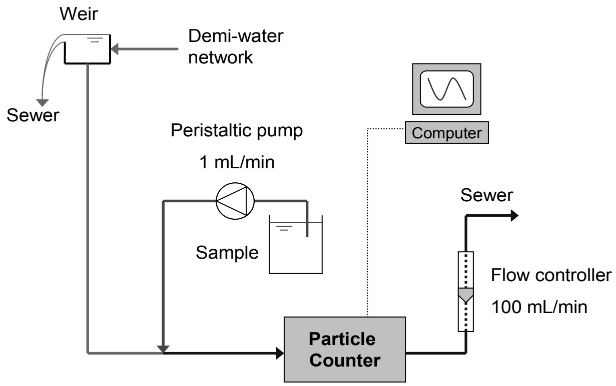


Figure 4.5: Scheme particle counter

A light scattering sensor counts the number of particles in the size range between 0.4 and 5.0 µm that pass the sensor every minute. These data are processed with the software application Particle Vision Online. The initial output consists of the number of particles per mL sample for different size range intervals; see Figure 4.6 for an example of a typical measurement. Note the wider range and thus higher number of counts for the size range between 1.5 and 5.0 µm. The results show that the number of counts decreases approximately exponentially with increasing particle diameter range. In advance of results discussed later in this dissertation, it is mentioned that this exponential distribution was found for all particle counting experiments.

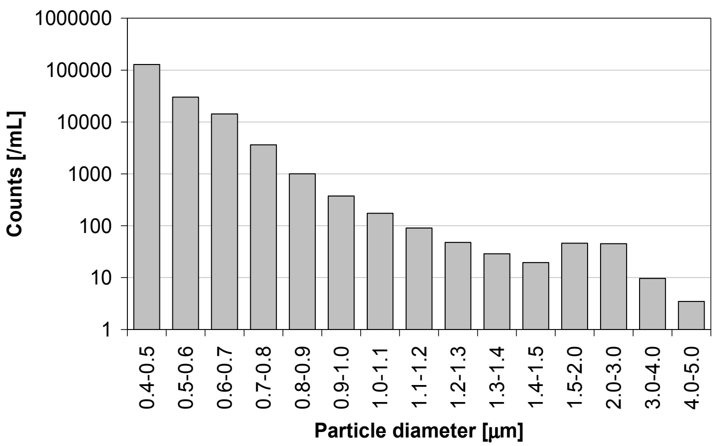


Figure 4.6: Typical example output particle counting experiment

The shape of the particles present in wastewater can be described as spherical, ellipsoids, rods and strings of various length and diameter, disk and random coils (Metcalf and Eddy, 2003). In this research a simplified approach is used by assuming all particles to be spherical and opaque. When the raw data are corrected for the demineralised water flow the volume distribution can be calculated. Figure 4.7 represents the cumulative volume distribution for the data originating from Figure 4.6.

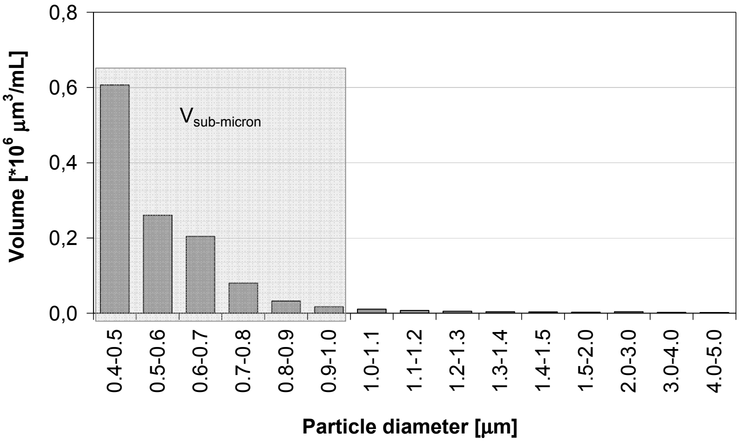


Figure 4.7: Volume distribution per range and determination $V_{sub\text{-}micron}$

Due to the relatively high number of particles with a diameter smaller than 1.0 µm the contribution of particles larger than 1.0 µm to the total volume in the size range between 0.4 and 5.0 µm becomes practically negligible (< 2%). Therefore only the particles smaller than 1.0 µm are considered in this research. In literature colloids are generally defined as particles in the size range between 0.01 and 1 µm (Metcalf and Eddy, 2003). The particle counter thus only measures a part of the total colloidal range. Nonetheless the measurement in the range between 0.4 µm and 1.0 µm is assumed to give a reflection of the total colloidal particle volume. In this thesis the volume of particles in the size range between 0.4 and 1.0 µm is denoted as the sub-micron particle volume ($V_{sub\text{-}micron}$).

Critical notes

It has to be recognised that the particle counting experiments as applied in this research hold several uncertainties. The results should therefore be considered with some reserve. Two major critical notes can be placed with respect to (1) the shape and nature of the particles and (2) the used dilution factor:

- *Shape and nature of the particles*

As discussed previously the particles are assumed spherical and opaque. Evidently in practice this is not the case. This implies that the recorded size of a particle depends on its position when it passes the sensor. In addition the ray of light can be refracted due to the transparent nature of the particles. The assumed spherical and opaque nature of the particles can thus lead to erroneous analyses.

- *Osmotic pressure*

  The osmotic pressure difference between the demi water and the sample might influence the size and properties of the particles. The effect is minimised by adding the sample to the demi water only seconds before the particle measurements. A possible effect of the osmotic pressure is left out of consideration in this study.
- *Dilution factor*

  In principle the particle counter is intended to analyse liquid contamination of high purity samples. Even at the applied dilution factor of 99% the amount of particles counted by the particle counter approaches or exceeds the concentration limit of 100,000 particles per mL. This problem could be overcome by further increasing the dilution factor. Additional tests at a higher dilution rate however demonstrated that the samples could not be mixed uniformly with the demi-water. Although at a dilution rate of 99% the concentration limit was sometimes exceeded the results for these operational conditions at least turn out to be stable and consistent.

The critical notes with respect to the particles size measurements can not be overcome. On the other hand, it is mentioned that all particle counting experiments are conducted according to the same protocol; this at least forms a solid basis for *mutual* and *relative* comparison between different samples.

4.7 Summary

Delft University of Technology has developed a method to characterise the filterability of MBR activated sludge. This so called *Delft Filtration Characterisation method* (DFCm) consists of a small scale Filtration Characterisation unit and a well-defined measuring protocol.

The heart of the Filtration Characterisation unit is a single tubular ultrafiltration membrane tube with a length of 1 m and an inner diameter of 8 mm (type X-flow, PVDF, nominal pore size 0.03 μm). In a DFCm experiment activated sludge samples (approximately 30 L) collected from full-scale or pilot-scale MBR plants are recirculated and filtrated under identical operational circumstances (CFV = 1.0 m/s, J = 80 $L/m^2 \cdot h$). The resistance increase during filtration is measured accurately. Thanks to the well-defined and identical hydraulic and operational circumstances in the membrane tube the differences in resistance increase occurring during filtration can be attributed exclusively to the properties of the activated sludge samples. In this research the ΔR_{20} value (resistance increase after filtration of 20 L/m^2) is used as the value to characterise the filterability of the activated sludge: an increasing ΔR_{20} value corresponds with a deteriorating filterability.

Parallel to the DFCm experiments several activated sludge characteristics are analysed to identify potential foulants. The activated sludge characteristics discussed in this thesis are: temperature, pH, mixed liquor suspended solids concentrations, sludge volume index, soluble microbial products concentrations in the free water phase, viscosity and the particle volume in the colloidal range.

This chapter formed the descriptive part of the DFCm. In the next chapter several aspects of the DFCm are assessed more in detail, including the possibilities and limitations of the method, a comparison with other filtration characterisation methods, the DFCm output in relation to the theory of cake layer filtration and the rheological regime in the membrane tube.

5 Method assessment

5.1 Introduction

In this chapter the Delft Filtration Characterisation method is assessed. Sections 5.2 and 5.3 present an exploration of the limitations and the possibilities of the DFCm. In section 5.4 the DFCm is compared with several other filtration characterisation methods for MBR activated sludge. Section 5.5 discusses the hydrodynamic circumstances in the DFCm membrane tube. Section 5.6 provides an analysis of the relevant size range of the foulants in a DFCm experiment. In section 5.7 the output of DFCm is discussed in relation to the theory of cake layer filtration. The chapter concludes with a recapitulation and some concluding remarks, in section 5.8. The topics discussed in this chapter are re-assessed in chapter 9 on the basis of the practical experience gained during the measurement campaigns that will be discussed in chapter 6 to 8.

5.2 DFCm limitations

Since fouling is a complex and comprehensive phenomenon it is practically impossible to incorporate all aspects that are involved in the process in a single research program. Evidently the DFCm does not form an exception. It is thus important to recognise the limitations of the DFCm in order to value the DFCm results. Two main aspects can be mentioned in which the DFCm filtration experiments differ from the MBR filtration process in practice. In the first place the DFCm is only suitable for short-term experiments. The consequences of this feature are discussed in section 5.2.1. Secondly the operational circumstances in the DFCm differ significantly from those in practice in full-scale MBR plants. This issue is discussed in section 5.2.2.

5.2.1 Reversible fouling

A DFCm experiment can be considered a reflection of a single filtration cycle (see Figure 3.4 in subsection 3.4.3). The total resistance increase during a filtration cycle consists of a reversible, irreversible and irrecoverable component. However, in a short-term DFCm experiment the irreversible and irrecoverable fouling rate are negligible compared to the reversible fouling rate. Although the irreversible fouling rate is partly an activated sludge characteristic it can not be investigated on the basis of DFCm experiments, because the irreversible fouling rate in an MBR is predominantly a reflection of the effectiveness of physical cleaning measures (relaxation, back flushing). Since these physical cleaning measures are not (yet) incorporated in the measuring protocol a DFCm experiment does thus not provide information about the irreversible fouling potential of the activated sludge.

5.2.2 Different filtration circumstances

The MBR fouling process depends on membrane characteristics, membrane operation and activated sludge properties. This is evidently also the case for the DFCm. Since the (starting) membrane conditions and the operational circumstances are well-defined and similar for all DFCm experiments the differences in filterability can be attributed exclusively to differences in the properties of the activated sludge mixed liquor. A major issue is whether the filterability as characterised with the DFCm is representative for the filterability in the considered full-scale MBR plant from which the activated sludge is sampled. This is discussed on the basis of (the differences in) membrane properties and process operation. In this context the process operation is assessed on the basis of the crossflow conditions and the value of the flux.

Membrane properties

The membrane properties of the membrane in the DFCm filtration unit are not necessarily comparable with the considered MBR plant. However, in the research described in this thesis by chance the membrane type and material in the DFCm (ultrafiltration, PVDF membrane material) are similar to the considered full-scale MBR plants (Varsseveld, see chapter 6.2 and Heenvliet, see chapter 7.2). As a consequence the micro-scale membrane properties of the DFCm membrane tube and of the membranes in the full-scale MBR plants can be considered similar.
The macro-scale differences in the membrane configuration (tubular membrane in the DFCm and hollow fibre and flat sheet in the considered full-scale plants) and the filtration mode (inside-out in the DFCm and outside-in in the full-scale plants) do have an effect on the hydraulic regime near the membrane surface, but they can not be considered to influence the fundamentals of the filtration process.

Process operation: air-liquid crossflow versus liquid crossflow

A major difference between the filtration regime in the DFCm and the considered full-scale MBR plants is the method to create crossflow circumstances. In the DFCm crossflow is created by liquid pumping while in the considered MBR plants this is achieved by coarse bubble aeration. In the case of coarse bubble aeration the shear forces are variable, whereas in sidestream liquid crossflow the shear force is more constant. Nonetheless, in the basis both means of crossflow aim for the same goal: the creation of a shear force along the membrane surface. The shear force may influence the fouling process, but it does not affect the potential of the activated sludge to cause fouling. It can thus be expected that an activated sludge sample that has poor filterability under coarse bubble aeration crossflow circumstances will also have a poor filterability under liquid crossflow circumstances (and vice versa).

The number of literature references dealing with the comparison between liquid crossflow and two-phase air-liquid crossflow MBR configurations is very limited. Le-Clech et al. (2005) report similar fouling behaviour in submerged and sidestream MBR configuration for specific liquid and air flow rates. Gunder and Krauth (1999) compared three pilot plants equipped with tubular-, plate- and hollow fibre membranes for a period of more than one year. The differences in fouling between the immersed and sidestream systems are attributed to differences in hydraulics rather than to differences in the activated sludge properties.

Process operation: low flux versus high flux

In practice submerged MBR systems are operated under relatively low fluxes to suppress fouling (below the critical flux, see chapter 3.7). In contrast to the situation practice in a DFCm experiment fouling is wilfully created by applying a much higher flux of 80 $L/m^2 \cdot h$. As a consequence of the relatively high flux in the DFCm the forces transporting constituents to the membrane surface are higher. Two mechanisms can be expected to occur in comparison with the application of a lower flux. In the first place bigger particles are transported to the membrane surface. Secondly smaller particles that could also have been involved in the fouling process at a lower flux are now transported to the membrane in a higher concentration.

Since fouling is predominantly attributed to fine particles it can be expected that particularly the higher concentrations of small particles attracted to the membrane is affecting the fouling rate. This implies that applying a higher flux accelerates the fouling process rather than results in a fundamental change of the occurring mechanisms.

5.3 DFCm possibilities

The parameter that is used in practice to manage and control the filtration process in an MBR system is the *permeability*. The DFCm is a method that allows characterisation of the *filterability* of a given activated sludge sample. This section discusses the shortcomings of the parameter permeability and the significance of the parameter filterability in the MBR process.

5.3.1 Permeability, a weak parameter

The conventional parameter to monitor the MBR filtration process is the *permeability*. The suitability of this parameter is however arguable, as it provides no specific information about which factors are determining it. The permeability is a function of all three main factors that are involved in the fouling process (membrane characteristics, membrane operation and activated sludge properties). The weakness of the parameter permeability is explained on the basis of the ways it can be expressed. These are (see Equation 2-5 in chapter 2.4 for denotation symbols):

$$P = \frac{J}{TMP} = \frac{1}{\eta_p \cdot R_{total}} = \frac{Q_{total}}{A_{membrane} \cdot TMP} \tag{5-1}$$

Flux-TMP ratio

By definition the permeability represents the ratio between the prevailing flux and TMP. In case of clean water filtration the *momentaneous* value of the permeability is independent of the imposed flux, because an increase of the flux is accompanied by a corresponding increase of the TMP according to Darcy's law (see Equation 2-1 in chapter 2.4). This is however not the case for the MBR filtration process, in which (cake layer) fouling plays an important role (see also section 5.7). This implies that the resistance caused by fouling is depending on the prevailing TMP. With increasing TMP the fouling layer is compressed and offers a higher resistance to filtration. This results in a decrease of the flux and in a lower value of the momentaneous permeability.

In addition the permeability *development in time* will evidently be affected by the height of the flux. With increasing flux the fouling rate increases and the permeability will show a more rapid decline. The absolute permeability value itself however provides no information about the applied flux. This can be considered an important shortcoming, because the flux (i.e. permeate production rate) is evidently a major operational parameter. The permeability development in time is an incomplete source of information about the fouling process when it is not considered in relation to the applied flux.

Membrane resistance and fouling resistance

The permeability is the inverse product of the permeate viscosity and the total filtration resistance. In practice the influence of the permeate viscosity (i.e. temperature) is overcome by using the temperature corrected permeability. However, the *total* resistance to filtration is the sum of the membrane resistance and the fouling resistance. On the basis of the permeability these two components can not be distinguished. The only way to overcome the problem mentioned above is to consider the actual permeability in close relation to the permeability directly after the last chemical cleaning procedure. The value at this moment reflects the membrane resistance (or indicates inadequate cleaning efficiency). The conclusion that stands is that the value of the permeability lacks representativeness when the contribution of the membrane resistance and the fouling resistance can not be distinguished.

Total and effective membrane surface

As discussed in chapter 3.7 the applied flux is a crucial parameter with respect to fouling. The average flux with which the activated sludge is filtrated equals the ratio between the total permeate production rate and the membrane surface that is available for permeation. A crucial misconception in this approach is the premise that the *total* membrane surface always equals the membrane surface that is actually available for permeation, indicated as the *effective* membrane surface. The effective membrane surface can be considered a membrane characteristic and is continuously under pressure during filtration. Three processes or events that reduce the effective membrane surface can be mentioned:

- Clogging.
- Irrecoverable fouling.
- Ineffective cleaning measures (with respect to both reversible and irreversible fouling).

From the three processes/events mentioned above clogging fully excludes the membrane from participating in the filtration process. A reduction of the effective membrane surface leads to an increase of the local fluxes in the membrane area that still is available for permeation. According to the theory of different fouling stages and irregular flux distribution (see chapter 3.7.1) this will inevitably accelerate the fouling process (i.e. the permeability decline), irrespective of the prevailing operational circumstances and activated sludge filterability. Irrecoverable fouling and ineffective cleaning measures also increase the local filtration resistance and thereby promote an irregular distribution of the flux and a higher fouling rate.

As discussed in chapter 3.3 clogging is the direct result of insufficient or irregular distribution of the hydraulic turbulent circumstances in the membrane modules. Clogging can be attributed to shortcomings in the design of the membrane modules and aeration system, inadequate operation or temporary failure of the operation. Clogging especially becomes a problem that has to be taken in account for bigger sized MBR plants in which the hydraulic circumstances are more difficult to control. On the basis of permeability measurements the occurrence of clogging can not be detected. This stresses the statement that permeability is not a suitable parameter for controlling and managing the filtration process. In order to detect (and prevent) clogging the process monitoring has to be extended by comprehending an analysis of the effective membrane surface. This issue is discussed more detailed in chapter 9.6.

5.3.2 Filterability, a key parameter

It can be stated that good filterability is one of three starting points for a satisfactory MBR filtration process, together with good membrane properties and good membrane operation. Although the filterability of activated sludge only partly determines the degree of fouling it is stressed that it still is an important parameter that deserves special attention. A better understanding of the filterability of activated sludge allows for optimisation of the filtration process. This is discussed in subsection 5.3.3.

5.3.3 DFCm potential in practice

Filtration process optimisation

As discussed in section 5.3.2 favourable activated sludge filterability properties can be considered one of three starting points for a satisfactory filtration process. A better understanding of the filterability and dynamic changes in this characteristic allows for optimising the process operation and anticipation to sudden changes in the filterability. Depending on the activated sludge filterability several operational parameters can be adjusted to minimise fouling problems (in case of bad filterability) or energy consumption (in case of good filterability):

- *Filtration cycle and physical cleaning.*
  As discussed in chapter 3.4.3 the current operation of an MBR filtration cycle is usually based on a fixed time interval for filtration and relaxation. Depending on the prevailing filterability the filtration time and relaxation time can be adjusted dynamically to optimise the filtration process with respect to energy consumption and fouling. Similar considerations count when the cleaning protocol also consists of a backflush (besides relaxation).
- *Membrane surface in use.*
  MBR plants are usually designed with separate membrane tanks that can be operated independent of each other. As discussed in chapter 3.7.1 the flux is a crucial parameter affecting the fouling process. In case of (temporary) poor filterability it might be advisable to minimise the prevailing fluxes in the system by maximising the membrane surface used to process the total flow. In this way the costs of extra energy

consumption might be counterbalanced by a reduction of the fouling rate (and the costs/maintenance due to fouling).

- *Coarse bubble aeration rate.*
  In case of good filterability the coarse bubble aeration rate in the membrane modules might be reduced to reduce the energy consumption. On the other hand it has to be kept in mind that the coarse bubble aeration rate is also linked with clogging. Reducing the aeration rate should thus not be solely based on the filterability properties of the activated sludge.

Filterability and permeability

Although the exact relation between irreversible fouling and filterability falls out of the scope of this research it can be expected that an indirect link between the two exists, since the irreversible fouling rate reflects the effectiveness with which the reversible fouling is removed by the physical cleaning measures. Intuitively a high reversible fouling rate interferes with a satisfactory recovery of the permeability after physical cleaning measures. When hypothesising that there is indeed a relationship between long-term permeability development and the filterability of activated sludge the potential of the DFCm in practice can be assessed. Figure 5.1 schematically illustrates four hypothetical combinations of DFCm results and the permeability development of the considered MBR plant. Roughly considered the DFCm filterability can be considered as *good* or *poor* and the permeability development of the considered full-scale plant as *satisfactory* or *unsatisfactory*.

The application of the DFCm lacks usefulness when the considered MBR plant is functioning properly. In theory a satisfactory development of the permeability in a full-scale installation could be accompanied by poor activated sludge filterability, when the operational circumstances are so excessive that they counteract the poor filterability. In practice this situation however seems unrealistic. In general a high and stable permeability can be expected to be accompanied by good activated sludge filtration properties.

DFCm filterability

Permeability development		Good	Poor
	Satisfactory	Desired situation	Poor filterability is overcome by excessive operational circumstances *(cost- and energy inefficient)*
	Unsatisfactory	Activated sludge filterability is **not** a limiting factor in the filtration process *(improve operation!)*	Activated sludge filterability is (at least partly) a limiting factor in the filtration process *(improve sludge quality!)*

Figure 5.1: Hypothetic combinations of DFCm filterability and permeability development (assuming a relation between filterability and permeability)

The application of the DFCm becomes interesting when the permeability development in the considered full-scale plant is not satisfactory. As discussed in section 5.3 a decrease in permeability can be the result of either poor operation or poor activated sludge properties. DFCm experiments can provide an explanation about the origin of the permeability development. When besides decreasing permeability also the filterability of the activated sludge is poor it can be concluded that *at least* the filterability is a limiting factor. It gets even more interesting when a decreasing trend in the permeability is accompanied by good activated sludge filterability. In this case the DFCm namely demonstrates that the activated sludge quality is not the limiting factor in the filtration process. This thus forms a strong indication that the operational circumstances are not sufficient to maintain the total membrane surface participating in the filtration process.

5.4 Comparison with other filtration characterisation methods

Since activated sludge filterability is an important factor with respect to economical operation of an MBR plant a multitude of methods have been developed aiming at characterising and understanding filterability. Nonetheless, no standard method exists to assess the filtration behaviour of activated sludge. This section describes several methods which are more or less comparable with the DFCm and can be applied to examine the filterability of activated sludge.

5.4.1 The Flux Step method

A well-known method to assess MBR fouling is the *flux-step method*, first described by Le-Clech et al. (2003). The flux-step method is based on the critical flux concept (see chapter 3.7). In the flux-step method the flux is increased and subsequently decreased stepwise. The highest flux for which the TMP stays constant is defined as the critical flux (in its weak form). A schematic representation of a flux-step method experiment is given in Figure 5.2.

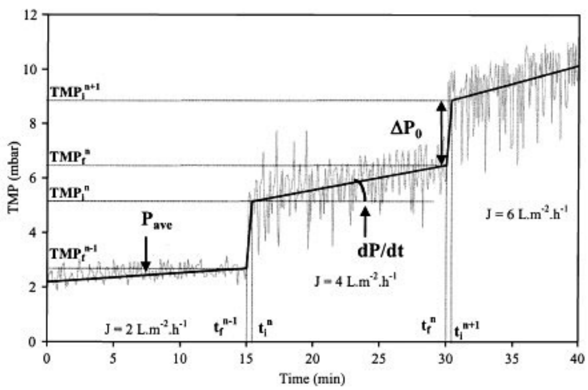


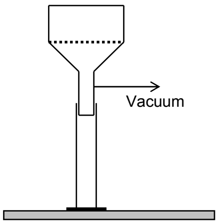
Figure 5.2: Schematic representation of the critical flux determination by the flux-step method (Le-Clech et al., 2003)

Although Le-Clech et al. propose an arbitrary chosen step height and duration of 2 $L/m^2{\cdot}h$ and 15 minutes respectively no standard protocol is defined for the flux-step method. Variables mentioned besides step height and duration are the initial state of the membrane, the feed characteristics and the system hydraulics, thus exactly the three main factors affecting fouling (see chapter 3.3.1, figure 3.2). The lack of a standard set-up and protocol is the main drawback of the flux-step method, since it allows no unequivocal comparison between different data. Nonetheless, the flux-step method provides a useful tool to assess the fouling potential of the activated sludge in an existing MBR plant.

A major finding in the flux-step method research is that long-term operation at a sub-critical flux (as determined in a short-term experiment) turned out to lead to a dramatic increase of fouling rate after a critical time period. From this observation it was concluded that the flux-step method is not suitable to predict long-term fouling behaviour in a real MBR system (Le-Clech et al., 2003).

5.4.2 The Time-to-Filter method

A very straightforward method to obtain some understanding about activated sludge filterability properties is the Time-to-Filter (TTF) method (Standard Methods, 1998). In the TTF an activated sludge sample is filtrated in dead-end mode over a filter paper using a vacuum pump providing constant pressure, see Figure 5.3. The time required to filtrate 100 mL permeate at a vacuum pressure of 51 kPa represents the TTF value.



- Filter paper (Whatman No. 1 or 2 or equivalent)
- Add 200 mL activated sludge to cylinder
- Vacuum: 51 kPa
- Start stopwatch until $V_{filtr.}$ = 100 mL

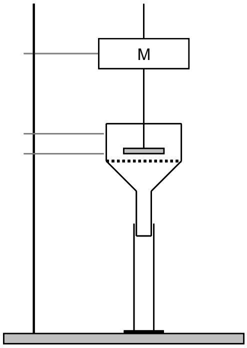
$$TTF = t_{V=100\ mL}\ (sec)$$

Figure 5.3: Schematic representation Time-to-Filter method

Evident advantages of the TTF method are that it is simple, low-tech, inexpensive and takes little time. However, the method does not comprehend the complexity of the MBR filtration process in practice. In the first place the TTF method is based on dead-end filtration, which implies that the MLSS concentration of the sample is strongly influencing the TTF value. Secondly the method is based on constant pressure filtration, whereas MBR systems are usually operated under constant flux filtration. Finally it is mentioned that the pore size of the filter paper used in the TTF differs from the pore size of membranes in MBR plants. This implies that the size of foulants determining the TTF value differs from the size of the foulants in practice in MBR systems.

5.4.3 The Sludge Filtration Index method

The Sludge Filtration Index (SFI) method (Raudies, 2008) is based on the Time-to-Filter method. For determining the SFI an activated sludge sample is filtrated over a filter paper by gravity (see Figure 5.4). To simulate crossflow circumstances a clamp blade agitator is rotating with a constant speed just above the filter paper. The SFI is represented by the ratio of the filtrate production in a certain period and the MLSS concentration of the sample.



- Filter paper (Schwarzband MN 85/70 Ø 150mm)
- Wing stirrer (D = 9-12 cm) placed 1 to 3 mm above filter paper
- Stirring rate:40 rounds/minute
- Add 500 mL activated sludge to cylinder
- Start timing at $V_{filtr.}$ = 100 mL
- Stop timing at $V_{filtr.}$ = 150 mL

$$\boxed{SFI = \frac{\Delta t}{MLSS}}$$

Figure 5.4: Schematic representation Sludge Filtration Index method

The advantages and disadvantages of the SFI method are comparable with those of the TTF method. Although the employed wing stirrer provides a better simulation of crossflow circumstances as occurring in full-scale MBR plants it can be expected that the influence of the MLSS concentration is more complex than the linear influence as presumed in the calculation of the SFI value.

5.4.4 The MBR-VITO Fouling Measurement method

The Belgian company VITO (Flemish Institute for Technological Research) developed a filtration characterisation method called the VITO Fouling Measurement method (VFMm) (Huyskens et al., 2008). The VFMm aims at characterising both the reversible and the irreversible fouling potential of MBR activated sludge. The VFMm bears several resemblances with the DFCm. Both methods consist of a comparable filtration set-up with a single inside-out tubular membrane (see Figure 5.5) and an accompanying well-defined measuring protocol. Furthermore both methods aim at characterising the fouling potential of activated sludge samples collected from an existing MBR plant.

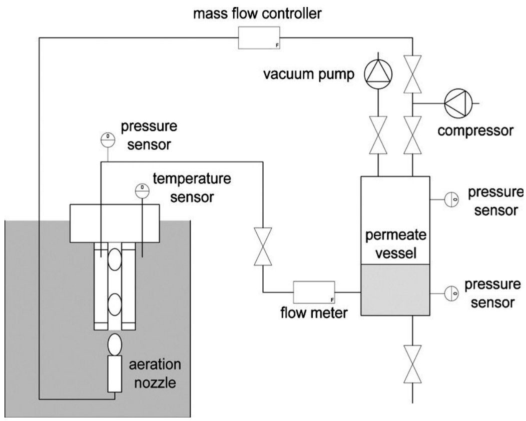


Figure 5.5: Schematic representation VITO Fouling Measurement set-up

An important difference between the VFMm and the DFCm is formed by the membrane operation. In the VFMm the crossflow is created by coarse bubble aeration, whereas in the DFCm this is done by fluid crossflow. In this sense the VFMm is thus more representative for the filtration mechanism in immersed MBRs. On the other hand it can be expected that it is more difficult to create constant and similar hydraulic conditions in the membrane tube with coarse bubble aeration. In addition it is mentioned that the VFM applies constant TMP filtration instead of constant flux filtration as in the DFCm. As discussed in chapter 3.7 these two different filtration modes are suspected to promote different fouling mechanisms. Since in practice MBRs are generally operated under constant-flux filtration in this sense the VFMm is thus less representative than the DFCm.

An interesting feature of the VFMm is that it aims at characterising the irreversible fouling potential of the activated sludge. In order to do so the measuring protocol is more extensive compared to DFCm. Whereas the DFCm restricts to a single filtration cycle, the VFMm implements multiple filtration cycles with mechanical cleaning steps (relaxation) in between them, see Figure 5.6 for an example of the VFMm output. The recovery of the permeability after the relaxation provides information about the activated sludge irreversible fouling propensity.

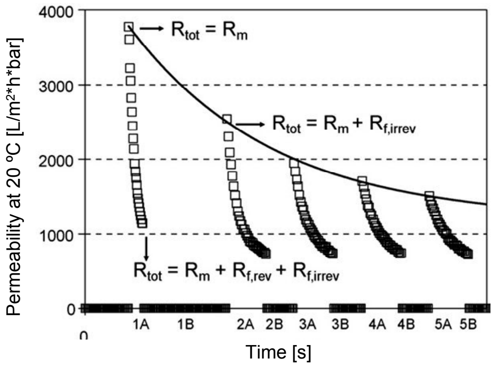


Figure 5.6: Example VFMm output

A critical note concerning the VFMm is that it aims at characterising the irreversible fouling rate in a much shorter period (hours) than it occurs in practice (weeks/months). As a consequence the irreversible fouling as characterised with the VFMm might not be representative for the irreversible fouling rate in practice.

5.5 Rheology in the membrane tube

With respect to the constant crossflow velocity of 1.0 m/s the hydraulic circumstances in the membrane tube can be considered similar for all DFCm experiments. However, the flow in the membrane tube should also be considered from a rheological point of view. Depending on the relative influence of inertial forces compared to viscous forces the flow in the membrane tube is either laminar or turbulent. Since both flow regimes result in different flow patterns in the membrane tube it can be expected that it also leads to different fouling behaviour. It is thus important to understand the rheological regime in the membrane tube.

The difficulty in determining the rheological regime in the membrane tube lies in the pseudoplastic behaviour of activated sludge. This implies that, contrary to pure water, the viscosity of the activated sludge is not known a-priori. Based on fluid mechanics theory (section 5.5.1) and DFCm measuring data first the flow regime in the membrane tube for pure water is analysed (section 5.5.2). Subsequently the conditions for activated-sludge flow are assessed (section 5.5.3). The findings are compared with some literature data (section 5.5.4) before ending with some concluding remarks (section 5.5.5).

5.5.1 Theoretical background

Reynolds number

The flow regime in a pipe is determined by the dimensionless Reynolds number (Re). The Reynolds number represents the ratio between inertial forces and viscous forces, in circular pipes (i.e. a tubular membrane tube) typically given by the following Equation:

$$\mathrm{Re} = \frac{\rho \cdot u \cdot D}{\eta} \tag{5-2}$$

With:

ρ = fluid density, [kg/m^3]

u = mean fluid velocity, [m/s]

D = hydraulic diameter, [m]

η = dynamic viscosity, [Pa·s]

For Reynolds numbers below 2300 the flow is laminar; when the Reynolds number exceeds 4000 the flow can be considered fully turbulent (Battjes, 1999). An exact value for the transition point can not be defined because the switch from laminar to turbulent circumstances is an instability phenomenon, sensitive for small immeasurable influences.

Viscosity

Equation 5-2 denotes the importance of the fluid viscosity with respect to the rheological regime. The fluid viscosity is defined by the ratio between the occurring shear stress and the velocity gradient perpendicular to the direction of shear (shear rate):

$$\eta = \frac{\tau_w}{\delta u / \delta z} = \frac{\tau_w}{\gamma_0} \tag{5-3}$$

With:

η = dynamic viscosity, [Pa·s]

τ_w = shear stress, [Pa]

γ_0 = $\delta u/\delta z$ = shear rate, [s^{-1}]

Water is considered an incompressible Newtonian fluid, implying that its viscosity is independent of the shear forces acting upon it. In contrast with this activated sludge is a non-Newtonian fluid; this implies that the apparent viscosity of activated sludge depends on the occurring shear forces (see also chapter 4.6.5).

Shear stress

A viscous fluid flowing through a pipe incurs a shear force parallel to the membrane surface in the opposite direction of the flow. The shear stress is the ratio between the shear force and the area parallel to the shear force and can be represented as a function of the occurring energy loss in the pipe or according to the Darcy-Weisbach Equation:

Energy loss: $$\tau_w = \rho \cdot g \cdot \tfrac{1}{4} \cdot D \cdot \frac{\Delta H}{\Delta L} \tag{5-4}$$

Darcy-Weisbach: $$\tau_w = \frac{\lambda}{8} \cdot \rho \cdot u^2 \tag{5-5}$$

With:

ρ = fluid density [kg/m^3],
g = gravity acceleration constant [m/s^2],
D = hydraulic diameter [m],
ΔH = energy loss [m],
ΔL = length membrane tube [m],
λ = Darcy-Weisbach friction factor [-],
u = mean fluid velocity [m/s].

Friction factor

The Darcy-Weisbach friction factor λ is a dimensionless number that describes the relationship between the mean fluid velocity and the pressure gradient according to:

$$\lambda = \frac{\left(-\frac{\partial p}{\partial z}\right) \cdot D}{\tfrac{1}{2} \cdot \rho \cdot u^2} \tag{5-6}$$

With:

$\delta p/\delta z$ = pressure drop gradient [Pa/m],
D = hydraulic diameter [m],
ρ = fluid density [kg/m^3],
u = mean fluid velocity [m/s].

The Darcy-Weisbach friction factor λ is related to the rheological circumstances in the pipe and can be derived from the Reynolds number according to the following empirical formulas (Battjes, 1999):

Laminar (Re <2300): $$\lambda = \frac{64}{\mathrm{Re}} \tag{5-7}$$

Turbulent (Re >4000): $$\frac{1}{\sqrt{\lambda}} \cong 1.8 \cdot \log\left(\frac{\mathrm{Re}}{7}\right) \tag{5-8}$$

Energy level

Equation 5-4 shows that the shear stress of a liquid flow in a pipe can be calculated from the occurring energy loss. The energy loss in the DFCm membrane tube can be characterised using the rigid column approach with the following presumptions:

- Energy is solely lost due to friction (and not due to local losses). In the DFCm filtration unit the headers to which the pressure sensors are connected have the same diameter as the membrane tube; therefore in this approach local losses between the feed and the concentrate pressure sensors are left out of consideration.
- The membrane tube is prismatic and the elasticity of the pipe is negligible; the cross section of the pipe does thus not change over the length of the pipe.
- The fluid is incompressible.
- The flow is uniform and stationary. Strictly taken this is not the case when permeate is extracted, because the ingoing feed flow does not equal the outgoing concentrate flow. However, when the standard protocol is applied (CFV = 1.0 m/s and J = 80 $L/m^2 \cdot h$), the permeate flow only accounts for approximately 1% of the feed flow. In this approach this difference is considered negligible.

When above mentioned presumptions are taken into account the energy loss in the membrane tube can be calculated according to the Darcy-Weisbach and Euler equations, valid in both laminar and turbulent circumstances:

Darcy-Weisbach: $$\Delta H = \lambda \cdot \frac{L}{D} \cdot \frac{u^2}{2g} \tag{5-9}$$

Euler: $$\Delta H = \Delta z + \frac{\Delta p}{\rho \cdot g} \tag{5-10}$$

With:
ΔH = energy loss [m],
λ = Darcy-Weisbach friction factor [-],
L = length pipe [m],
D = hydraulic diameter [m],
u = mean fluid velocity [m/s],
g = gravity acceleration constant [m/s^2].

Δz = height difference feed and concentrate sensor [m],
Δp = pressure difference feed and concentrate sensor [Pa],
ρ = fluid density [kg/m^3],

5.5.2 Analysis for water flow

Reynolds number

The theory discussed in section 5.5.1 is now applied to analyse the rheological circumstances in the membrane tube. In order to determine the rheological regime in the tube the viscosity of the fluid has to be known. In case of pure water flowing through the membrane tube the analysis is straightforward, because the viscosity is solely dependant on the water temperature. Nonetheless an analysis for pure water flow is useful for making a comparison with activated sludge flow.

Several DFCm experiments were conducted with demineralised water with a temperature of 16 °C. At this temperature the dynamic viscosity of water is 1.12 mPa·s (Equation 2-2). From Equation 5-2 the Reynolds number can be calculated for the standard crossflow velocity of 1.0 m/s:

$$\mathrm{Re} = \frac{\rho \cdot u \cdot D}{\eta} = \frac{1000 \cdot 1.0 \cdot 8E^{-3}}{1.12E^{-3}} = 7143$$

The results indicate a Reynolds number higher than 4000, and thus a turbulent hydraulic regime in the membrane tube.

Shear stress

Equation 5-2 demonstrates that the rheological regime in the membrane tube is highly dependant on the viscosity of the liquid flowing through the membrane. For activated sludge the apparent viscosity depends on the shear rate experienced by the flow. It is thus important to gain reliable information about the occurring shear rates in the membrane tube. For water the viscosity is known, which implies that the shear rate can be calculated both theoretically and on the basis of the DFCm pressure measurements. This comparison is useful to assess the reliability of the DFCm pressure measurements.

From a theoretical point of view the shear stress can be calculated from Equations 5-8 and 5-5. For the standard crossflow velocity of 1.0 m/s and a temperature of 16 °C this comes down to:

$$\frac{1}{\sqrt{\lambda}} = 1.8 \cdot \log\left(\frac{7143}{7}\right) \rightarrow \lambda = 0.0341$$

$$\tau_w = \frac{\lambda}{8} \cdot \rho \cdot u^2 = \frac{0.0341}{8} \cdot 1000 \cdot 1.0^2 = 4.3 \text{ Pa}$$

The shear stress in the membrane tube can also be derived from the pressure measurements (feed and concentrate pressure sensor) according to the Euler Equations. For the standard CFV of 1.0 m/s the measured pressure difference between the feed and the concentrate pressure sensor was 0.123 Bar. By implementing this pressure difference in Equations 5-10 and 5-5 the shear stress can be calculated:

$$\Delta H = \Delta z + \frac{\Delta p}{\rho \times g} = -1.0 + \frac{12300}{1000 \times 9.81} = 0.25 \text{ m}$$

$$\tau_w = \rho \cdot g \cdot 0.25 \cdot D \cdot \frac{\Delta H}{\Delta S} = 1000 \cdot 9.81 \cdot 0.25 \cdot 0.008 \cdot \frac{0.25}{1.00} = 4.7 \text{ Pa}$$

The theoretical and experimental results show a deviation of about 9%; this correspondence is considered reasonable. In order to form a more reliable comparison between theory and the pressure measurements more experiments were conducted, with six different crossflow velocities in the range from 0 to 1.5 m/s. Each crossflow velocity corresponds with a certain energy loss over the membrane tube. Figure 5.7 represents the theoretical energy loss (curve, according to the Darcy-Weisbach Equation) and the measured energy loss for the six different crossflow velocities (markers, according to the Euler Equations). The results show a high correlation between theory and the measured values. This confirms the representativeness of the pressure measurements. As can be expected the measured energy loss is somewhat higher than the theoretical energy loss, because in practice some local losses will inevitably occur.

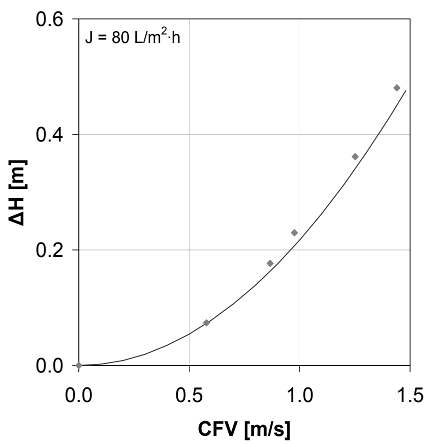


Figure 5.7: Energy loss with CFV in the membrane tube for demineralised water, theoretical (line) and measured (markers)

5.5.3 Analysis for activated sludge flow

Viscosity threshold

As discussed previously the rheological circumstances for activated sludge flow are more difficult to determine because the activated sludge viscosity is not known a-priori. From equation 5-2 the maximum viscosity η_{max} for which the circumstances are still turbulent can be calculated, assuming that the activated sludge density equals the density of pure water (1000 kg/m^3)[2]:

$$\mathrm{Re} = \frac{\rho \cdot u \cdot D}{\eta_{\max}} \rightarrow \eta_{\max} = \frac{1000 \cdot 1.0 \cdot 0.008}{4000} = 2 \text{ mPa·s}$$

In order to determine the apparent viscosity of the activated sludge first the occurring shear stress has to be known. Because with the apparent viscosity also the Reynolds number is not known a-priori the shear stress can not be calculated theoretically from the Darcy-Weisbach Equations.

Analysis based on energy loss

The shear stress can only be derived empirically from the measured pressure loss over the membrane tube. Since the viscosity of activated sludge increases with increasing MLSS concentration it can be expected that at a constant CFV also the shear stress increases with increasing MLSS concentration. Figure 5.8 represents a collection of 173 DFCm experiments, for which the measured shear stress is plotted against the MLSS concentration of the considered activated sludge samples (the values originate from the measured pressure loss and Equation 5-10). The value for pure water (4.7 Pa) is also included. Although the values are scattered the overall results confirm an increasing shear stress with increasing MLSS concentration.

[2] In practice the density of activated sludge is somewhat higher than of water, depending on the MLSS concentration, but in this analysis the difference is of negligible influence.

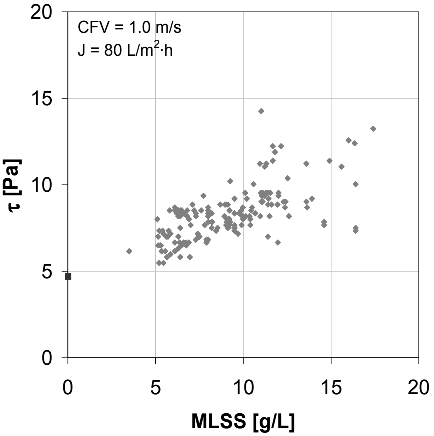


Figure 5.8: Measured shear stress with MLSS concentration

The dataset in Figure 5.8 indicates that an MLSS concentration of 10 g/L corresponds with a shear stress in the DFCm membrane tube in the order of magnitude of 8 Pa. From this value the Darcy-Weisbach friction factor can be estimated according to Equation 5-6:

$$\tau_w = \frac{\lambda}{8} \cdot \rho \cdot u^2 \rightarrow \lambda = \frac{8 \cdot 8}{1000 \cdot 1.0^2} = 0.064$$

Subsequently the accompanying Reynolds number can be determined in two ways, assuming a laminar flow (Equation 5-7) or a turbulent flow (Equation 5-8):

Laminar (Re < 2300): $\lambda = \frac{64}{\text{Re}} \rightarrow \text{Re} = 1000$

Turbulent (Re > 4000): $\frac{1}{\sqrt{\lambda}} \cong 1.8 \cdot \log\left(\frac{\text{Re}}{7}\right) \rightarrow \text{Re} = 1100$

Assuming turbulent circumstances leads to a Reynolds number lower than 4000; this starting assumption can thus be indicated as false. Based on this approach the results indicate that for an activated sludge sample with an MLSS concentration of 10 g/L the flow is laminar.

In order to have a fully turbulent regime in the DFCm membrane tube the Darcy-Weisbach friction factor has to be 0.041 at the maximum (calculated from Equation 5-8). This friction factor corresponds with a shear stress of 5.1 Pa. Comparing this value with the dataset represented in Figure 5.8 leads to the conclusion that the rheological regime in the membrane tube for activated sludge samples with an MLSS concentration higher than 5 g/L is laminar (at the standard crossflow velocity of 1.0 m/s).

Analysis based on viscosity measurements

The rheological regime in the DFCm membrane tube can also be analysed on the basis of the viscosity measurements. As discussed in chapter 4.6.5 the rheometer used in this research measures the torque at certain fixed shear rates. From the torque the shear stress and the apparent viscosity are calculated. Figure 5.9 originates from the same data as presented in Figure 4.4, but with the apparent viscosity plotted against the shear stress instead of the shear rate. The results demonstrate that the rheometer is not capable of measuring the apparent viscosity at the shear stress values that occur in the DFCm membrane tube.

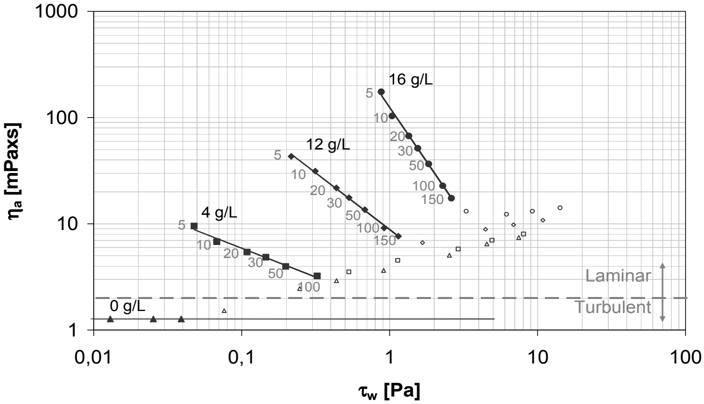


Figure 5.9: Apparent viscosity with shear stress and shear rate γ (tags)

A possible approach to estimate the apparent viscosity at higher shear stress values would be to extend the double logarithmic trend lines. However, simply extending the trend lines leads to an incorrect estimation because this would result in an activated sludge viscosity becoming lower than water with increasing shear stress. This is evidently impossible in practice. At a certain shear stress the apparent viscosity thus has to stabilise. Information about this phenomenon in literature is very limited, but it is confirmed by Khongnakorn et al. (2008). Figure 5.10 represents the output of a rheological measurement with an activated sludge sample (unknown MLSS concentration).

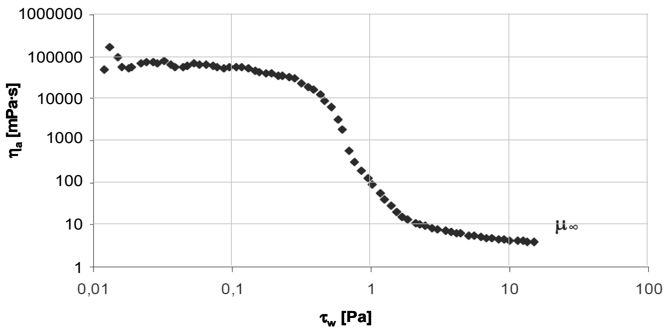


Figure 5.10: Apparent viscosity with shear stress (from Khongnakorn et al. 2008)

Khongnakorn et al. divide the evolution of apparent viscosity as a function of shear stress in three parts:

- In the first part the shear stress is so low that the activated sludge structure is not disrupted and the viscosity thus remains constant. In this range the activated sludge is in linear elastic state and behaves like a solid.
- In the second part the structure of the sludge is disrupted and the viscosity decreases logarithmically with increasing shear stress.
- Finally at higher shear stress values the viscosity tends towards a limit value and becomes practically constant.

The rheometer used in the research described in this dissertation (see chapter 4.6.5) is only capable of measuring in the second stage of the activated sludge viscosity development. The rheological measurements do thus not provide information about the apparent viscosity at the shear stresses that occur in the DFCm membrane tube and can thus not be used to elucidate the rheological regime.

5.5.4 Literature references

As mentioned previously the number of publications dealing with MBR activated sludge rheology is limited. Rheological investigations are mostly based on the conventional activated sludge process in which the MLSS concentrations are considerably lower compared to the MBR process. Since at lower MLSS concentrations activated sludge tends to exhibit Newtonian instead of pseudoplastic rheological behaviour the conditions are not comparable with MBR activated sludge. Nonetheless some papers deal with MBR activated sludge rheology. Findings reported in three different papers (Rosenberger et al, 2002; Laera et al., 2007; Khongnakorn et al., 2007) are applied to the circumstances in the DFCm membrane tube.

Rosenberger et al. (2002) and Laera et al. (2007) propose a relation based on the Ostwald model with apparent viscosity being dependant on the shear rate γ_0 and the MLSS concentration:

Rosenberger et al. (2002): $\eta_a = \exp(1.9 \cdot MLSS^{0.43}) \cdot (\gamma_0)^{-0.22 \cdot MLSS^{0.37}}$ (5-11)

Laera et al. (2007): $\eta_a = \exp(0.882 \cdot MLSS^{0.494}) \cdot (\gamma_0)^{-0.05 \cdot MLSS^{0.631}}$ (5-12)

The literature references are analysed on the basis of a presumed MLSS concentration of 10 g/L. For a fixed MLSS concentration the apparent viscosity η_a is only depends on the prevailing shear rate γ_0. Since the shear stress τ equals the product of the shear rate and the apparent viscosity (see Equation 5-2) the apparent viscosity can also be expressed as a function of the shear stress. Figure 5.11 represents the apparent viscosity with the prevailing shear stress for Equations 5-11 and 5-12, a repetition of the data as in Figure 5.10 and the average result for the measurements conducted with the rheometer (see chapter 4.6.5).

The apparent viscosity can also be estimated on the basis of the measured energy loss (i.e. shear stress) in the membrane tube. Figure 5.8 shows that at an MLSS concentration of 10 g/L the shear stress is approximately 8 Pa. Since the flow regime can be considered laminar the shear rate can be calculated according to Equations 5-2 and 5-7.

$$\eta_a \cdot \gamma_0 = \frac{64}{8 \cdot \mathrm{Re}} \cdot \rho \cdot u^2 = \frac{64 \cdot \eta_a}{8 \cdot \rho \cdot u \cdot D} \cdot \rho \cdot u^2 \rightarrow \gamma_0 = \frac{8}{D} \cdot u = \frac{8}{0.008} \cdot 1 = 1000 \text{ s}^{-1}$$

Subsequently the apparent viscosity can be calculated according to Equation 5-4:

$$\eta_a = \frac{\tau_w}{\gamma_0} = \frac{8}{1000} = 0.008 \text{ Pa·s} = 8 \text{ mPa·s}$$

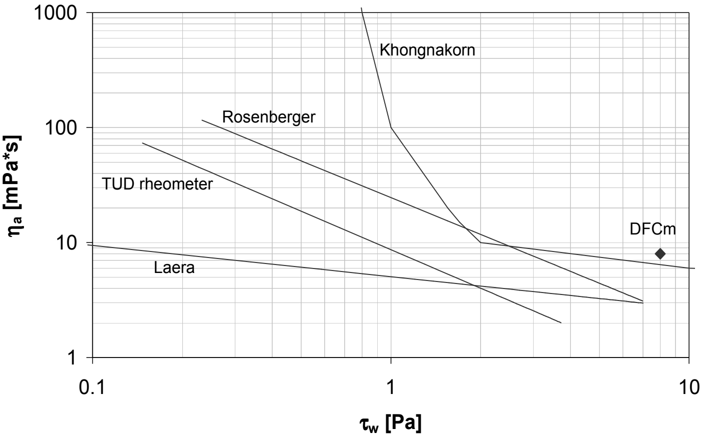


Figure 5.11: Apparent viscosity with shear stress for different literature references

Several critical notes can be placed with respect to the results represented in Figure 5.11:

- The measurements with the rheometer and the equations as proposed by Rosenberger and Laera can only be valid in a certain range. At some point the viscosity has to stabilise with increasing shear stress, as represented in Figure 5.10. Based on the results presented in Figure 5.10 this stabilisation is taking place at shear rates much lower than the prevailing shear stress of 8 Pa in the DFCm membrane tube. This implies that Equations 5-11 and 5-12 are not valid for the circumstances as occurring in the DFCm membrane tube.
- The MLSS concentration of the sample on which the data of Khongnakorn are based is not known. Concerning the steep trend of the apparent viscosity at relatively low shear stress values it can be expected that the MLSS concentration of this sample is higher than the reference value of 10 g/L.
- The relation between shear stress and the apparent viscosity is diverse for the different measurement campaigns. Apparently there is no universal relationship between shear rate/stress and apparent viscosity for activated sludge. The deviations might be explained by differences in the used methodologies (although all measurements were performed with rotational rheometers) and differences in the activated sludge properties (not verifiable).
- The DFCm is not a tool fitted for measuring the rheological properties of activated sludge. Although it can be expected that the energy loss in the membrane tube is predominantly caused by friction, the contribution of local losses might not be excluded. This reduces the accuracy and reliability of the apparent viscosity as determined on the basis of the energy loss over the membrane tube.

Above mentioned considerations indicate that it is complicated to draw accurate and reliable conclusions about the rheological regime in the membrane tube. Nonetheless, theory and experimental results indicate that the apparent viscosity in the membrane tube for standard circumstances is well above the critical value of 2 mPa·s. This indicates that the flow regime in the membrane tube is always laminar for activated sludge with an MLSS concentration of 10 g/L.

5.5.5 Concluding remarks on rheology

The rheological regime in the membrane tube was analysed for both (demineralised) water and activated sludge flowing through the DFCm membrane tube. In case of water flowing through the membrane the rheological circumstances are turbulent for the standard crossflow velocity of 1.0 m/s. The theoretical reasoning for this corresponds reasonably well with an analysis based on the measured pressure loss over the membrane tube.

With increasing MLSS concentration the viscosity of the activated sludge increases and at some point the flow has to become laminar. An analysis based on the theoretical Darcy-Weisbach Equations in combination with the measured pressure loss over the membrane tube indicates a laminar flow regime for activated sludge samples with an MLSS concentration above 5 g/L. Considering the actual MLSS concentrations in MBR systems the rheological circumstances for all DFCm experiments can thus be considered laminar. The rheological regime in the membrane tube could not be analysed on the basis of the measurements with the rheometer used in this research because the rheometer is not capable of measuring in the shear stress range that prevails in the DFCm membrane tube.

Based on the combination of theory, the measured pressure loss over the membrane tube and a comparison with literature results it can be concluded that the flow regime in the membrane tube is laminar for the MLSS concentration ranges of MBR activated sludge (MLSS > 5 g/L). The rheological regime is thus not a limiting factor with respect to unequivocal comparison between different DFCm experiments. It can be mentioned that with increasing MLSS concentration also the shear stress along the membrane surface gradually increases (at constant CFV). The influence of this increasing shear stress on cake layer formation can however be assumed negligible compared to the influence of the filterability properties of the considered activated sludge sample.

5.6 Analysis of foulants size range

The size of the particles in the activated sludge feed flow likely to deposit on the membrane surface during a DFCm filtration experiment can be estimated on the basis of the theory of backtransport velocity (discussed in chapter 3.2.3). The total backtransport velocity equals the sum of Brownian diffusion, shear-induced diffusion and lateral migration. When the drag velocity towards the membrane (due to permeation) exceeds the total backtransport velocity the particles are susceptible to accumulate on the membrane surface. Because the operational circumstances in the membrane tube are constant during the DFCm experiments also the size of the particles susceptible to be attracted to the membrane is constant. On the basis of several calculations and assumptions this particle size can be estimated using the equations for the three backtransport mechanisms (see chapter 3.2.3 for details):

- *Brownian diffusion:* $$V_{BD} = 0.185\left(\frac{\gamma_0 \cdot k^2 \cdot T^2}{\eta_a \cdot {d_p}^2 \cdot L} \cdot \frac{c_w}{c_b}\right)^{1/3}$$
- *Shear-induced diffusion*: $$V_{SD} = 0.072 \cdot \gamma_0 \cdot \left(\frac{{d_p}^4}{L} \cdot \frac{c_w}{c_b}\right)^{1/3}$$
- *Lateral migration*: $$V_{LM} = 0.036 \cdot \left(\frac{\rho_{as} \cdot {d_p}^3 \cdot {\gamma_0}^2}{\eta_a}\right)$$

Table 5.1 represents the input for the equations. Several input parameters are discussed more in detail:

- The flow in the DFCm membrane tube is laminar in the relevant MLSS range (see section 5.5). This implies that the shear rate γ_0 is constant for all experiments and equals a value of 1000 s^{-1} (for CFV = 1 m/s, based on Equations 5-1 and 5-6).
- The polarisation factor c_w/c_b, which represents the ratio between the particle fraction volume in the bulk and at the membrane surface, is difficult to measure. Based on a paper by Jiang et al. (2007) and Evenblij (2005) a value of 60 is assumed. Small mistakes in the assumption of the value do not have a big weight factor because the outcome of the equations is not sensitive for the value of c_w/c_b (due to the 1/3 power term).
- The exact value of the apparent viscosity γ_0 in the membrane tube is not known. A value of 5.0 mPa·s is estimated on the basis of the analysis of the hydrodynamics in the membrane tube that was discussed in section 5.5. It is mentioned that as with the c_w/c_b ratio a small deviation in the γ_0 value does not considerably affect the results.

Table 5.1: Input for the estimation of the critical particle size in a DFCm experiment

Parameter	**Symbol**	**Unity**	**Value**
Mixed liquor suspended solids	$MLSS$	g/L	10
Crossflow velocity	CFV	m/s	1.0
Shear rate	γ_0	s^{-1}	1000
Boltzmann constant	k	$kg{\cdot}m^2/s^2$	$1.38{\cdot}10^{-23}$
Temperature	T	K	288 (15º C)
Apparent viscosity	η_a	mPa·s	5.0
Length membrane tube	L	m	0.95
Polarisation factor	c_w/c_b	-	60
Density activated sludge	ρ_{AS}	kg/m^3	1000

Using the values listed in Table 5.1 as the input in the backtransport formulas allows estimation of the total backtransport velocity (V_{BT}) as a function of the critical particle size. Subsequently the critical particle size for the standard operational conditions (J = 80 $L/m^2{\cdot}h$) can be determined, see Figure 5.12. The results indicate that the critical particle size for standard operational circumstances is somewhat below 5 µm. In addition the conditions for other fluxes $L/m^2{\cdot}h$ are depicted. The results indicate that for instance at a flux of 20 $L/m^2{\cdot}h$ the critical particle diameter is below 2 µm.

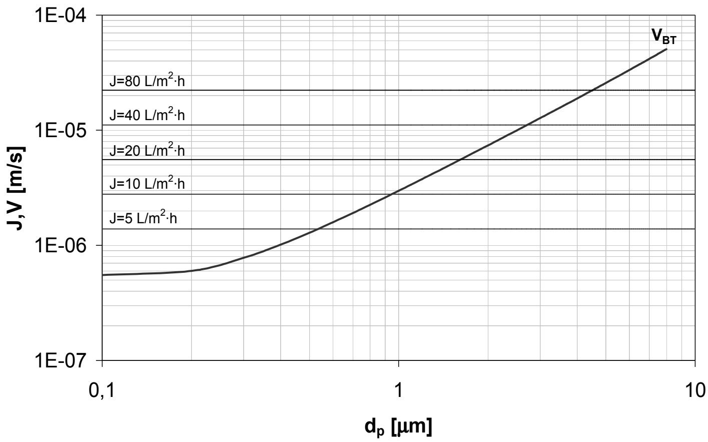


Figure 5.12: Backtransport velocity (V_{BT}) with critical particle size (for standard DFCm operational circumstances)

On the basis of the analysis represented in Figure 5.12 a conclusion can be drawn on the influence of the relatively high flux that is applied in the DFCm experiments compared to the considered full-scale MBR plants. It can be stated that the DFCm experiments would represent an incorrect characterisation of the filterability when the activated sludge contains a significant concentrations of particles with a size range between 2 and 5 μm. Particle size measurements however show that the volume (or concentration) of particles with a size between 2 and 5 μm is negligible compared the volume in the range between 0 and 2 μm (see Figures 4.7 and 4.8 in chapter 4.6). This indicates that the relatively high flux as applied in the DFCm is not an objection for characterisation of the filterability in general.

5.7 Analysis of DFCm output

As discussed in chapter 4.5.1 the main output of a DFCm experiment consists of a dataset that plots the increase of the filtration resistance against the specific permeate production. In this section the DFCm output is discussed more in detail in relation to the theory.

5.7.1 Fouling mechanism

As discussed in section 5.3 irreversible (long-term) fouling mechanisms can be assumed negligible in a (short-term) DFCm experiment. Directly after the start of filtration the membrane is still clean and the initial fouling mechanism will be pore blocking. Depending on the amount of substances accumulating on the membrane surface the dominant fouling mechanism will subsequently shift from pore blocking to cake layer formation. Considering the high concentration of particles in activated sludge and the high fouling rate that is pursued in a DFCm experiment the dominant fouling mechanism can expected to be cake layer fouling. This is confirmed by Jiang et al. (2003) who performed filtration tests in a set-up with comparable circumstances to the DFCm (sidestream X-flow membranes, fluxes of 52 and 72 L/m^2·h): they report pore blocking to be completed after only 8 seconds.

Studies on cake layer fouling are predominantly based on dead-end and constant-TMP circumstances (Roorda, 2004; Boerlage et al., 2003; Lee et al., 2003). The main difference between dead-end and crossflow filtration is that in crossflow filtration the resistance will not necessarily increase continuously during filtration. As a consequence of the back transport mechanisms created by the shear forces an equilibrium situation can set in between forces transporting constituents in the feed towards and away from the membrane (Judd 2006).
Evenblij (2005) presented the three possible development trends of the resistance when filtrating under constant-flux conditions, schematically illustrated in Figure 5.13. Apart from the exact shape of the curve three general trends are possible with respect to the fouling rate $\delta R/\delta V$: *constant* $\delta R/\delta V$ for a *linear* fouling cake layer, *decreasing* $\delta R/\delta V$ for an *equilibrium* cake layer and *increasing* $\delta R/\delta V$ for a *compressing* cake layer.

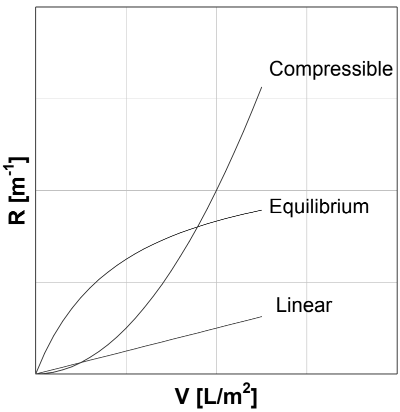


Figure 5.13: Hypothetic DFCm curves

5.7.2 Cake layer filtration theory

As discussed in chapter 3.2.2 cake layer fouling can be defined as resistance increase due to particulate material residing on the upstream face of the membrane surface during filtration. Theoretically the cake layer resistance can be represented by Equation 5-13 (Mulder, 1996):

$$R_{cake} = \alpha \cdot \left(c_{feed} \cdot \frac{V_{filtr}}{A_m} \right) \tag{5-13}$$

With:

R_{cake} = cake layer resistance, [m^{-1}]
α = specific cake resistance, [$m \cdot kg^{-1}$]
c_{feed} = solid concentration in feed water [$kg \cdot m^{-3}$]
V_{filtr} = filtrate volume, [m^3]
A_m = effective membrane surface, [m^2]

Within Equation 5-13 two components can be appointed that determine the total cake layer resistance. The second part of the Equation ($c_{feed} \cdot V_{filtr}/A_m$) represents the mass of substances accumulating in the cake layer and α represents the specific resistance caused by these substances.

Cake mass growth

The second part of Equation 5-13 ($c_{feed} \cdot V_{filtr}/A_m$) has the unity kg/m^2 and indicates the accumulated cake mass per membrane surface area. Thanks to the well controlled hydraulic circumstances in the DFCm membrane tube it can be assumed that the effective membrane surface A_m remains optimal and constant (240 cm^2) during a DFCm filtration experiment. The ratio between V_{filtr} [m^3] and A_m [m^2] thus equals the specific DFCm permeate production V [m^3/m^2 ~ L/m^2].

The parameter c_{feed} in Equation 5-13 represents the solids concentration in the feed water participating in the cake fouling process. In dead end filtration c_{feed} is equal to the total solid concentration in the feed water. This is however not the case for crossflow filtration. As a result of the shear force along the membrane surface only a part of the total solid concentration is involved in the fouling process.

The concentration of solids involved in the cake fouling process highly depends on the ratio between the forces transporting the foulants towards (J) and away (CFV) from the membrane surface. In a DFCm experiment both J and CFV are very constant; this implies that during filtration the same concentration of feed constituents are transported into the boundary- and cake layer. Together with the assumption that the activated sludge properties and composition do not alternate during a (short-term) experiment it can be assumed that c_{feed} is constant during a DFCm experiment. To stress the difference between the solid concentrations involved in the cake fouling process in dead-end and crossflow circumstances c_{feed} in Equation 5-13 is replaced by c_i, in which c_i thus represents the solid concentration that is *involved* in the cake formation.

Implementing above mentioned considerations in Equation 5-13 leads to the following equation for the cake layer resistance in a DFCm experiment:

$$R_{cake} = \alpha \cdot c_i \cdot V \qquad (5\text{-}14)$$

With:

R_{cake} = cake layer resistance, [m^{-1}]

α = specific cake resistance, [$m \cdot kg^{-1}$]

c_i = solid concentration involved in the fouling process [$kg \cdot m^{-3}$]

V = specific permeate production, [m^3/m^2 ~ L/m^2]

Equation 5-14 shows that in case of a constant specific cake resistance α the total cake resistance increases linearly with the specific permeate production. It also shows that, irrespective of the specific cake layer resistance an increase of the total cake layer filtration resistance can be combated by minimising the solid concentration accumulating on the membrane surface.

Cake compression

The initial mechanism resulting in cake layer fouling is the accumulation of solids on the membrane surface. The total cake layer resistance is subsequently determined by the specific resistance of these solids.

Theoretically the specific cake resistance α is depending on the density and the diameter of the particles in the cake layer and the cake layer porosity according to the Carman-Kozeny relation for spherical particles (Liew et al., 1995, Dijk et al., 2001):

$$\alpha = \frac{180}{\rho_p \cdot d_s^{\ 2}} \cdot \frac{(1-\varepsilon)^2}{\varepsilon^3} \tag{5-15}$$

With:
α = specific cake resistance, [$m \cdot kg^{-1}$]
ρ_p = density of the particles in the cake layer, [$kg \cdot m^{-3}$]
d_s = diameter of the particles in the cake layer, [m]
ε = cake layer porosity, [-]

The porosity can be defined as the measure of the void spaces in the cake layer and is expressed as a percentage between 0% and 100%.

From an empirical point of view the specific cake layer resistance can be considered in terms of compression and can be expressed as an empirical function of the pressure difference over the cake layer (Roorda, 2004; Boerlage et al., 2003; Lee et al., 2003; Matsumoto et al., 1999; Tiller and Yeh, 1987):

$$\alpha = \alpha_0 \cdot (\Delta P)^s \tag{5-16}$$

With:
α = specific cake resistance, [$m \cdot kg^{-1}$]
α_0 = specific cake resistance at reference pressure, [$m \cdot kg^{-1}$]
ΔP = pressure difference over the cake layer, [Bar]
s = compressibility coefficient, [-]

During a DFCm experiment the flux (J) and the permeate viscosity (η) are constant. Considering Darcy's law (equation 4-3) this implies that the pressure difference over the cake layer is directly proportional to the resistance difference over the cake layer ($\Delta R \sim \Delta TMP$). This implies that the specific cake resistance can also be expressed as a function of the resistance increase over the cake layer:

$$\alpha = \alpha_R \cdot (\Delta R)^s \tag{5-17}$$

With: α_R = specific cake resistance at reference resistance, [$m \cdot kg^{-1}$]

Both parameters α_R and s are related to the properties of the activated sludge. For constant-TMP filtration the specific cake resistance remains constant during filtration; the total filtration resistance will only increase as a consequence of cake mass growth. In constant-flux filtration the pressure difference over the cake layer and thus the specific cake resistance will increase during filtration. The compressibility coefficient s expresses the compression potential of the cake layer; when $s = 0$ no compression occurs and when $s = 1$ the compression is complete. In the absence of compression ($\alpha = \alpha_R$) the filtration resistance thus increases linearly with the specific permeate production.

5.7.3 Fitting cake layer filtration theory to DFCm output

As discussed in section 5.3 the dominant fouling mechanism in a short-term DFCm experiment is cake layer formation. This implies that the total resistance increase can be assumed to consist of cake fouling:

$$R_{cake} = \Delta R \tag{5-18}$$

Equation 5-17 shows that the specific cake resistance depends on the prevailing filtration resistance over the cake layer and the activated sludge properties α_R and s. The activated sludge properties can be assumed to remain constant during a DFCm experiment; this implies that the specific cake resistance is solely depending on the prevailing total cake resistance. Combining Equations 5-13, 5-17 and 5-18 results in the following relationship:

$$\Delta R = \alpha_R \cdot (\Delta R)^s \cdot c_i \cdot V \rightarrow \Delta R^{(1-s)} = \alpha_R \cdot c_i \cdot V \tag{5-19}$$

Subsequently Equation 5-19 can be rewritten to express the resistance increase ΔR as a function of the specific permeate production V:

$$\boxed{\Delta R = (\alpha_R \cdot c_i \cdot V)^{\frac{1}{1-s}} = (\alpha_R \cdot c_i)^{\frac{1}{1-s}} \cdot (V)^{\frac{1}{1-s}}} \tag{5-20}$$

The parameters α_R, c_i and s in Equation 5-19 all represent activated sludge properties and can be assumed to remain constant during a DFCm experiment. This implies that the resistance increases as a power function of the specific permeate production. As mentioned in chapter 4.5 the output of the DFCm experiments can be fitted with high correlation factors to a power function with two coefficients p and q:

$$\Delta R = p \cdot (V)^q \tag{5-21}$$

The course of the resistance increase as measured in the DFCm experiments thus corresponds with theory. Equation 5-20 holds three constant parameters that are related to the activated sludge properties (α_R, c_i and s). However, by extracting the logarithm of both terms the compressibility coefficient s can be isolated by placing it in front of the logarithmic term:

$$\log(\Delta R) = \left(\frac{1}{1-s}\right) \cdot \log(V) + \log\left((\alpha_R \cdot c_i)^{\frac{1}{1-s}}\right) \tag{5-22}$$

And for Equation 5-21 counts:

$$\log(\Delta R) = q \cdot \log(V) + \log(p) \tag{5-23}$$

Subsequently the compressibility coefficient s can be calculated from the coefficient q as obtained from the DFCm dataset:

$$\left(\frac{1}{1-s}\right) = q \rightarrow \boxed{s = \left(\frac{q-1}{q}\right)} \tag{5-24}$$

The product of the specific cake resistance α_R (at reference filtration resistance) and the concentration of particles c_i accumulating in the cake layer can be expressed as a function of p and s or p and q:

$$(\alpha_R \cdot c_i)^{\frac{1}{1-s}} = p \rightarrow \boxed{(\alpha_R \cdot c_i) = p^{1/q} = p^{1-s}} \tag{5-25}$$

Besides the fact that coefficient p is dependant on the compressibility coefficient s it has to be realised that its value furthermore only provides information about the combination of α_R and c_i, but not about the separate values. This is schematically illustrated for two hypothetic cake layers in Figure 5.14. In situation A the concentration of material in the cake layer is higher than in situation B, but since the specific resistance in situation B is higher (lower porosity) the value of $\alpha_R{\cdot}c_i$ can be similar for both situations.

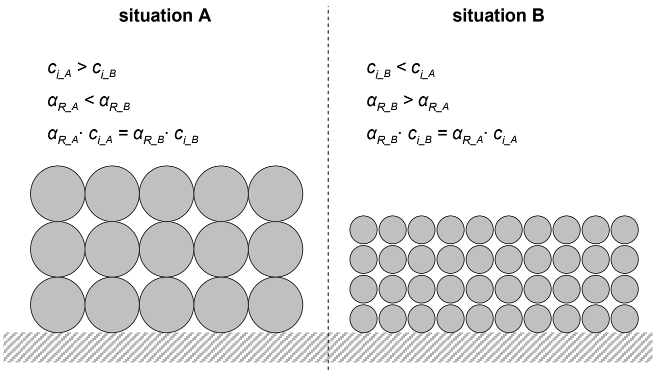


Figure 5.14: Hypothetic different cake layers with similar $\alpha_R \cdot c_i$ *values*

Logarithmic representation DFCm results

Considering the power relationship between the resistance increase ΔR and the specific permeate production V representing the DFCm results on a double logarithmic scale provides a superior insight into the significance of a DFCm dataset than a linear scale. Figure 5.15 represents an exact copy of Figure 4.3 in chapter 4.5, with the only difference that both the ΔR-axis and the V-axis are represented on a logarithmic scale. The renewed representation of the dataset allows a better understanding of the contribution of the compressibility coefficient s and the product $\alpha_R \cdot c_i$ to the total resistance increase.

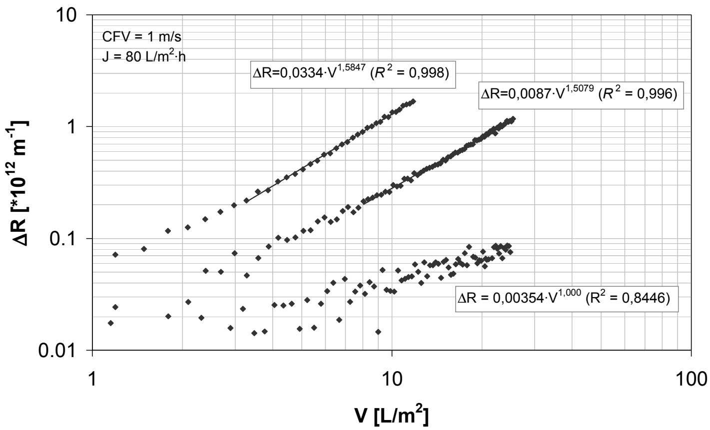


Figure 5.15: Example of DFCm output
(copy of figure 4.3, but with axes on double logarithmic scale)

Since the accuracy of each DFCm resistance measurement is in the range of 0.05 the correlation factor is low when the considered sample has a good filterability ($\Delta R_{20} < 0.1$). This implies that the compressibility coefficient can only be determined with reasonable reliability when the filterability of the sample is moderate to poor.

5.7.4 Re-assessment ΔR_{20} value

In order to discriminate between the filterability properties of different activated sludge samples in the initial stage of the DFCm research the ΔR_{20} value was introduced. As discussed in chapter 4.5 the ΔR_{20} value represents the resistance increase after a specific permeate production of 20 L/m^2. The value of ΔR_{20} corresponds with a certain filterability classification, see Table 5.2. In this section the significance of the ΔR_{20} value is re-assessed on the basis of the cake layer filtration theory.

Table 5.2: ΔR_{20} and corresponding filterability qualification

ΔR_{20} [$\cdot 10^{12}$ m^{-1}]	Qualification
0 – 0.1	Good
0.1 – 1.0	Moderate
> 1.0	Poor

An important critical note concerning the parameter ΔR_{20} is that it represents a single number while it is composed of two variables: (1) the product of the specific reference resistance and the accumulating concentration of particles in the cake layer and (2) the compressibility coefficient.
The question that comes up is whether ΔR_{20} is a representative parameter to characterise activated sludge filterability. Figure 5.16 represents the ΔR_{20} values for different combinations of $\alpha_R \cdot c_i$ and s (for a more comprehensible perception the ΔR_{20} axis is represented on a linear scale instead of a logarithmic scale). The 3-D surface forms a graphical representation of the cake layer filtration theory: in the absence of compression (s = 0) the ΔR_{20} value increases linearly with $\alpha_R \cdot c_i$ and in case of compression the (s > 0) the ΔR_{20} value increases according to an additional power term.

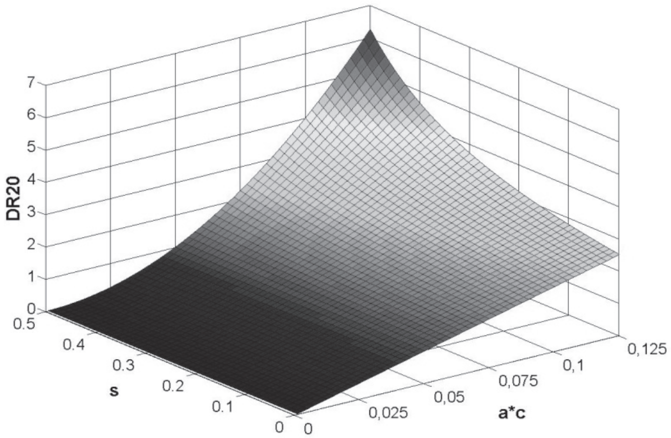


Figure 5.16: ΔR_{20} for different combinations of s and $\alpha_R \cdot c_i$

In the context of ΔR_{20} being composed of two coefficients it is important to recognise that the relevant range of ΔR_{20} only ranges from 0 to values somewhat higher than 1, see table 5.1. Values in the range much higher than 1 are less interesting, because in all cases the filterability can be qualified as poor. In the range between 0 and 1 the value of ΔR_{20} is predominantly determined by the value of $\alpha_R \cdot c_i$. In the lower range of ΔR_{20} the value of $\alpha_R \cdot c_i$ is automatically also low and this reduces the significance of the compressibility coefficient.

An interesting value in this analysis is $\Delta R_{20} = 1$, the (arbitrary) turning point for qualifying the filterability as poor (see Table 5.2). When ΔR_{20} equals 1 the power term in Equation 5-19 becomes insignificant and the value $\alpha_R \cdot c_i$ always equals 0.05 (because $\alpha_R \cdot c_i \cdot V = 1 \rightarrow \alpha_R \cdot c_i = 1/V = 1/20 = 0.05$). This implies that when the coefficient $\alpha_R \cdot c_i$ exceeds 0.05 the activated sludge filterability will always be poor, irrespective of the compressibility coefficient s. In order to be classified as “good” filterability ($\Delta R_{20} < 0.1$) the prerequisite is a low value for $\alpha_R \cdot c_i$. In the absence of compression the required value for $\alpha_R \cdot c_i$ is below 0.005. In case of compression the required value of $\alpha_R \cdot c_i$ slightly increases. For example, at a compressibility coefficient s = 0.2 the required value for $\alpha_R \cdot c_i$ is only 0.008. This increase is negligible in consideration of the range of $\alpha_R \cdot c_i$ values that were found in the measurement campaigns (as will be discussed in chapter 6 to 8).

The analysis above leads to the conclusion that the increase of the filtration resistance during a DFCm experiment can predominantly be attributed to the product of the specific reference cake resistance (α_R) and the concentration of substances accumulating in the cake layer (c_i). A possible relation between $\alpha_R \cdot c_i$ and s is discussed again in chapter 9.2.4 on the basis of the experimental results.

5.7.5 Recapitulation on DFCm output analysis

Considering the short duration of a DFCm experiment the contribution of the irreversible fouling rate can be assumed negligible compared to the reversible fouling rate. On the basis of the relatively high flux and high concentration of suspended material in the activated sludge liquor it can be assumed that the dominant fouling mechanism in a DFCm experiment is cake layer formation.

Theoretically the total cake layer resistance can be attributed to two components: (1) the concentration of substances accumulating in the cake layer and (2) the specific cake resistance caused by these substances. With respect to the constant operational circumstances and the assumption that the activated sludge properties do not change during an experiment the concentration of substances accumulating on the membrane increases linearly with the specific permeate production. According to theory during filtration the specific cake resistance will increase according to a power function due to cake compression. The datasets as obtained with the DFCm experiments correspond very well with this theory.

In the initial stage of the DFCm application it was decided to characterise the total resistance increase by the single value ΔR_{20}, the additional resistance after a specific permeate production of 20 L/m^2. It is important to mention that this value is composed of two components: (1) the product of the reference specific cake resistance and the concentration of accumulating in the cake layer and (2) the compressibility coefficient. A theoretical analysis however indicates that in the interesting range of ΔR_{20} (<1) the value is predominantly formed by the value of $\alpha_R \cdot c_i$ and not by the compressibility coefficient. This analysis will be verified for the results of the DFCm measuring campaigns (in chapter 6 to 8).

5.8 Summary and concluding remarks

The DFCm is a tool to characterise the *filterability* of an activated sludge sample collected from a (full-scale) MBR plant. Filterability is a parameter that partly determines the occurrence of fouling.

Together with favourable membrane properties and operational circumstances, good activated sludge filterability forms the starting point for a satisfactory MBR filtration process. The current parameter used to monitor and control the filtration process, the permeability, does however not provide information about the influence of each of these separate three factors in the filtration process. With the DFCm it is possible to elucidate the influence of the activated sludge properties in the filtration process. A better understanding of the activated sludge filterability forms a good basis for optimisation of the filtration process.

The DFCm measuring range is restricted to the potential of an activated sludge sample to cause *reversible* fouling. This is a drawback, because on a longer term the performance of an MBR plant is predominantly determined by the irreversible (and irrecoverable) fouling rate. Nonetheless, an indirect relationship between reversible and irreversible fouling can be expected, since the irreversible fouling rate is a reflection of the removal efficiency of reversible fouling by physical cleaning measures. The relation between filterability and irreversible fouling can be investigated on an empirical basis, by analysing the development of the filterability in relation to the permeability development in the considered MBR plant.

A rheological analysis of the flow in the DFCm membrane tube indicates that for the range of MLSS concentrations of MBR activated sludge the flow regime is always laminar. This implies that the rheological circumstances in the membrane tube do not affect the experimental results.

The dominant fouling mechanism in a DFCm experiment is cake layer formation. The cake layer resistance is formed by the concentration of substances accumulating on the membrane surface during filtration and the specific resistance created by these substances. The specific cake resistance is influenced by the prevailing pressure over the cake layer and the compression of the cake layer. However, theory indicates that in the relevant range the cake layer resistance is predominantly determined by the concentration of substances accumulating in the cake layer. This statement will be verified in chapter 9.2 on the basis of the experimental results that will be discussed in chapters 6 to 8.

6 Filtration characterisation at MBR Varsseveld

6.1 Introduction

The first of three measurement campaigns discussed in this thesis was performed at the wastewater treatment plant of Varsseveld in the eastern part of the Netherlands, in the period from January to October 2005. The study was performed in the framework of the STOWA-research program in which various aspects of MBR technology were investigated. Delft University was involved in the study on the relationship between the filterability and the quality of the activated sludge. The experiments described in this chapter were executed in close cooperation with Wetsus centre for sustainable water technology. In particular Maxime Remy is acknowledged for the analysis of the majority of the activated sludge characteristics discussed in this chapter.

Section 6.2 provides a general description of MBR Varsseveld. In section 6.3 the objectives and the approach of the research is specified. The research results are presented and described in three sections. Section 6.4 deals with the filtration characterisation results, section 6.4 with the relationship between the filterability as measured with the DFCm and the permeability development in the full scale installation and section 6.5 with the relationship between the filterability and several activated sludge characteristics. To conclude this chapter the results are summarised and the main conclusions of the measurement campaign are formulated.

6.2 MBR Varsseveld

In the context of upcoming stringent discharge legislation Dutch Waterboards are continuously exploring advanced wastewater treatment technologies. In the year 2000 a research program was launched at the wastewater treatment plant of Beverwijk to investigate the feasibility of MBR technology for municipal wastewater treatment in the Netherlands. Waterboard Hollands Noorderkwartier and the Dutch Foundation for Applied Water Research STOWA[3] commissioned DHV consulting engineers to carry out an extensive pilot-scale study. In a five year period seven MBR pilots with different configurations and membranes were tested. Based on the positive outcome of the Beverwijk study MBR technology was considered to be ready for application on a larger scale (Roest et al., 2002).

[3] The STOWA (Stichting Toegepast Onderzoek Waterbeheer) coordinates and commissions research on behalf of a large number of local water administrations. The bodies which contribute to the STOWA are the 26 Dutch Water Boards, the provinces and the Ministry of Transport, Public Works and Water Management (http://www.stowa.nl).

As a next step the first Dutch full-scale MBR was constructed at the wastewater treatment plant of Varsseveld (Waterboard Rijn & IJssel), with financial support of the STOWA, the Dutch Ministry of Economic Affairs and the European Union[4]. The wastewater treatment plant of Varsseveld discharges its effluent into ecologically sensitive surface water and for this reason an improvement of the effluent quality was required. DHV consulting engineers made the design for the MBR, which has been in operation since the end of 2004.

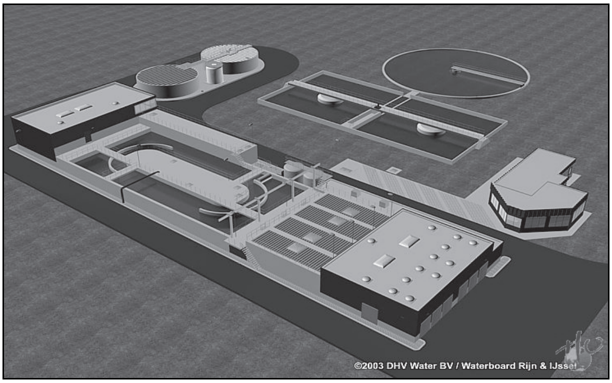


Figure 6.1: Aerial animation of wwtp Varsseveld
(Water Board Rijn and IJssel, DHV Water BV)

MBR Varsseveld is designed to treat a wastewater flow of 23,150 population equivalents or approximately 3,500 m^3/day during Dry Weather Flow (DWF) conditions. To cope with an increased flow during Rain Weather Flow (RWF) circumstances the maximum capacity is 755 m^3/h. The plant design is based on the future European Framework Water Directive discharge standards for nitrogen (< 5 mg N_{total}/L) and phosphorus (< 0.15 mg P_{total}/L). An aerial animation of the plant is represented in Figure 6.1. Table 6.1 shows a summary of the design data of MBR Varsseveld (Krzeminski, 2008).
As a pre-treatment step the influent passes a screen with a rod distance of 6 mm, an aerated sand and grease trap and a micro-sieve with a perforation of 0.8 mm respectively. The bioreactor is configured as a carrousel, with aerobic, anaerobic and anoxic zones and a design MLSS concentration of 10 g/L. The membrane part of the MBR consists of four separate membrane tanks or streets, each equipped with four cassettes Zenon ZW500d UF hollow fibre membranes with a nominal pore size of 0.035 μm. The total installed membrane surface is 20,160 m^2. To cope with the maximum flow during RWF conditions a net flux of 37.5 $L/m^2 \cdot h$ is required. Table 6.1 shows a summary of the design data of MBR Varsseveld (Krzeminski, 2008).

[4] The European Union's financial instrument LIFE supports environmental and nature conservation projects throughout the EU (http://ec.europa.eu/environment/life).

Table 6.1: Data MBR Varsseveld

Parameter	**Unit**	**Varsseveld**	
Start of operation	-	January 2005	
Water Board	-	Rijn&IJssel	
Function	-	Nitrogen and Phosporous removal	
Wastewater	-	municipal	
Biological Capacity	p.e.	23,150	
Hydraulic capacity (RWF)	m^3/h	755	
Hydraulic capacity (DWF)	m^3/h	250-300	
Hydraulic capacity (Average)	m^3/d	5000	
Process Configuration	-	Submerged	
Membrane type	-	Hollow fibre	
Membrane supplier	-	Zenon	
Product name	-	ZeeWeed 500d	
Membrane material	-	PVDF	
Bioreactor volume	m^3	4 tanks * 170 = 680	
Number of lanes	m^2	4 parallel tanks	
Total membrane area	m^2	20160	
Packing density	m^2/m^3	411	
Membrane pore size	µm	0,035	
Design Flux (net/bruto) RWF	$L/(m^2*h)$	37,5/45	
Average Flux (net/bruto) DWF	$L/(m^2*h)$	15-20	
Maximum Flux (net/bruto) RWF	$L/(m^2*h)$	50/60	
DWF : RWF	-	1:3 (250:755 m3/h)	
Design temperature	°C	7,5	
Sludge concentration	$kgMLSS/m^3$	10	
Cleaning	min	continuous mechanical + periodic chemical	
Cleaning	-	Mechanical and chemical	1 per week
Pre-treatment	-	grid removal (6 mm)+ sand trap + microsieve (0,8 mm)	
Aeration module - biology		plate diffusers (2x1 m)	
Aeration module - membrane		coarse: part of Zenon module (1-2 cm holes)	
Aeration intervals	sec	10/10 or 15/15	
Sludge retention time	days	35	

6.3 Research objectives and approach

6.3.1 Objective

The goal of the MBR Varsseveld project was to demonstrate that in the Netherlands MBR technology can be a feasible treatment method for municipal wastewater. This feasibility was defined as "reaching high effluent quality with a stable process and at reasonable costs". To verify the research goals a broad research program was formulated and executed in the first year of operation, covering all aspects of MBR technology and operation (STOWA, 2002). Delft University of Technology participated in the study *Activated sludge quality versus filterability*, together with Wetsus centre for sustainable water technology. The goal of this sub-study was to improve understanding of the complex interaction between activated sludge properties and its filterability to subsequently find indications for improvement of the biological process operation to reduce fouling.

6.3.2 Approach

In the period from January until October 2005 ten times samples were collected simultaneously from various compartments of the MBR, namely: (1) pre-treated influent, (2) predenitrification zone, (3) nitrification zone, (4) membrane tank and (5) permeate buffer.
The samples were transported to a laboratory of Wageningen University in Bennekom (30 minutes travel time) where all experiments were performed. Besides the filterability of the five samples, for the samples collected from the membrane tank also the filterability of its supernatant and its <0.45 µm fraction were characterised.

Parallel with the DFCm experiments an extensive sludge quality analysis was performed. The analyses consisted of fractionation in combination with the determination of various characteristics. The fractionations and analyses discussed in this chapter are summarised in Table 6.2 and Table 6.3.

Table 6.2: Fractionation activated sludge samples

Fraction	Method
Supernatant	Centrifugation (3000 rpm, 11 minutes) + paper filtration (S&S type ME 25/21 STL)
<0.45 µm (colloidal)	Filtration Kubota MF flat sheet membrane
<0.03 µm	Permeate Filtration Characterisation unit
<500, 100, 10, 1 kDa	Stirred UF cell, type Amicon model 8050

Table 6.3: Activated sludge quality analyses

Analysis	Unity	Reference to method
Activated sludge		
- Mixed Liquor Suspended Solids (MLSS)	[g/L]	Chapter 4.6.1
- Mixed Liquor Volatile Suspended Solids (MLVSS)	[g/L]	NEN 6621
- Bound exopolymers	[mg/g VSS]	Frølund 1996
- Particle Size Distribution (2-1000 μm)		Coulter LS230
Fractions		
- Soluble Microbial Products (SMP) proteins	[mg/L]	Chapter 4.6.4
- Soluble Microbial Products (SMP) polysaccharides	[mg/L]	Chapter 4.6.4
- Total Organic Carbon (TOC)	[mg/L]	NEN-EN 15936
- Biochemical Oxygen Demand (BOD)	[mg/L]	NEN-EN 1899-1

6.4 Filtration Characterisation results

The first observation concerning the DFCm experiments was the extreme poor filterability of the activated sludge samples in the first few months after the start-up of the MBR installation. As a consequence it was complicated to perform reliable DFCm experiments with the standard flux of 80 $L/m^2 \cdot h$. Therefore in this chapter the DFCm tests are predominantly analysed on the basis of DFCm experiments with a lower flux of 60 $L/m^2 \cdot h$.

6.4.1 Cheese factory wastewater

As will be discussed in section 6.5 severe problems were encountered with the permeability in the full-scale installation in this period. Soon it was suspected that the wastewater supply from a local cheese factory to the MBR was the malefactor. This wastewater appeared to contain the substance polyvinyl acetate (PVA), a rubbery synthetic polymer used to cover the cheeses with a protective plastic-like coating. Additional experiments with PVA wastewater in a pilot-scale simulation unit indicated that the PVA could not be degraded biologically and could not pass the membrane. As a result the PVA thus mounted up in the biological system and moreover appeared to form a sticky glue-like “curtain” on the outside of the membrane module, illustrated in Figure 6.2. To put a stop to the PVA problems the water board decided to uncouple the cheese factory from the sewer as from the beginning of May and to transport its wastewater to another (conventional) wwtp.

Figure 6.2: The effect of PVA wastewater on the membranes
(picture Water Board Rijn and IJssel)

A second important operational change was the start of the dosing of Iron (III) Chloride Sulphate ($FeClSO_4$) as from the beginning of June to enhance chemical phosphorus removal. The dosage was increased stepwise from 0.15 mol Fe^{3+}/mol P in June to 0.8 mol Fe^{3+}/mol P from October.

6.4.2 Membrane tank

Figure 6.3 and Table 6.4 represent the filtration characterisation results for the activated sludge samples collected from the membrane tanks for a flux of 60 L/m^2·h. In the period between January and the end of March the filterability of the activated sludge can be qualified as extremely poor (ΔR_{20} >> 1). Even at a relatively low flux of 60 L/m^2·h the intended permeate production of 20 L/m^2 could not be reached without exceeding the maximum allowable TMP of 0.6 Bar. The ΔR_{20} values represented in Table 6.4 are based on extrapolation of the trend lines in Figure 6.3.

The curve of February 14th forms a relative exception, but with a ΔR_{20} value higher than 1.0 the filterability can still be qualified as "very poor". After uncoupling the cheese factory from the sewer the filterability improves tremendously. At the first measurement on June 28th the ΔR_{20} decreases to 0.42, which corresponds with a "moderate" filterability. The improvement of the filterability seems to continue in the following period, given the result obtained in October. Nonetheless, with a ΔR_{20} value of 0.15 the filterability can still not be qualified as "good" (see Table 4.1 in chapter 4.5).

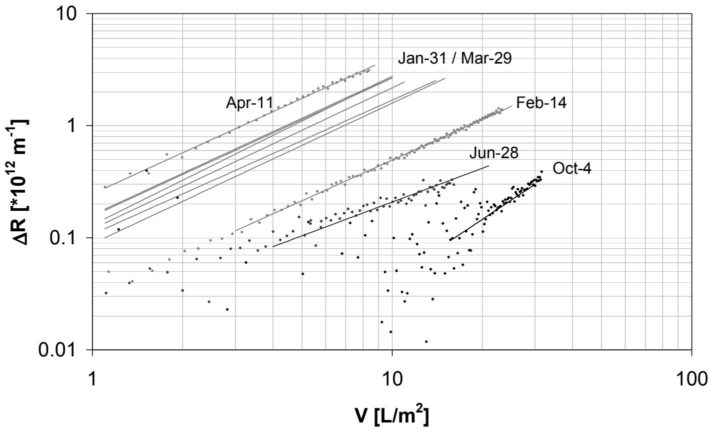


Figure 6.3: Filtration curves activated sludge membrane tank
(including DFCm dataset for Feb 14th, Apr 11th, Jun 28th, Oct 4th)

The most noteworthy DFCm results are highlighted in bold in Table 6.4. As also illustrated in Figure 6.3 in the period from January until April the compression factor *s* is more or less similar for all experiments. This implies that the differences in the ΔR_{20} values can be attributed to the differences in the coefficient $\alpha_R \cdot c_i$, i.e. the product of the specific reference cake layer resistance and the concentration of solids accumulating in the cake layer.

Table 6.4: DFCm results MBR Varsseveld measuring campaign (J = 60 L/m^2·h)

Date	$\alpha_R \cdot c_i$ [$\cdot 10^{-3}$ m^2]	s [-]	ΔR_{20} [$\cdot 10^{12}$ m^{-1}]
Jan-31	144.4	0.20	3.78
Feb-14	**55.6**	0.17	**1.14**
Feb-21	226.2	0.19	6.44
Feb-28	214.6	0.24	6.88
Mar-8	185.2	0.21	5.20
Mar-14	221.2	0.19	6.24
Mar-29	154.7	0.16	3.86
Apr-11	**316.3**	0.18	**9.44**
Jun-28	20.8	**0.00**	**0.42**
Oct-4	**17.0**	**0.44**	**0.15**

On June 28th the filtration properties of the activated sludge had improved drastically compared to the previous measurements; no compression was demonstrated (s = 0) and the resistance thus increases linearly with the specific permeate production. The improvement of the filterability results from the severe decrease of coefficient $\alpha_R \cdot c_i$ compared to the period from January until April. Remarkably the highest compressibility coefficient s was encountered for the sample with the best filterability, at October 4th. Nonetheless, ΔR_{20} is still relatively low because of the low value of $\alpha_R \cdot c_i$.

6.4.3 Flux sensitivity

In order to verify the sensitivity of the DFCm results for the value of the flux multiple filtration characterisation experiments were conducted with different fluxes. The obtained ΔR_{20} values for the different fluxes are plotted in Figure 6.4. It is mentioned that the values for J = 80 L/m^2·h in the period from January until April are derived from small DFCm datasets (since the upper TMP limit was reached rapidly) and can thus not be considered highly accurate. As can be expected ΔR_{20} rises with increasing flux. The results in Figure 6.4 emphasise the totally different filtration properties of the activated sludge before and after the uncoupling of the cheese factory. From January to March ΔR_{20} is very sensitive for changes in the low flux range (J < 60 L/m^2·h). For fluxes higher than 60 L/m^2·h the membrane seems to foul so quickly that the actual value of the flux is not of influence anymore.

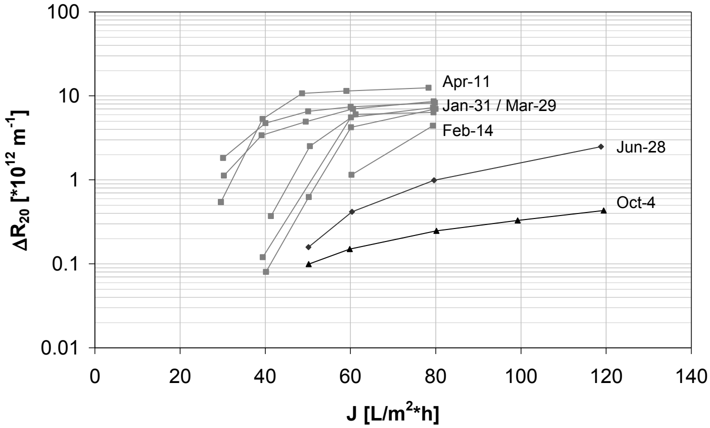


Figure 6.4: ΔR_{20} with applied flux (membrane tank)

The results illustrated in Figure 6.4 support the statement that activated sludge filterability is a parameter that is not dependant on the applied flux: the filterability as characterised with a high flux forms a representative reflection of the filterability at a lower flux.

6.4.4 Filterability per sampling point

To get an impression of the influence of the biological process on the filterability it was intended to collect samples from different compartments of the MBR. Unfortunately no samples could be collected from the predenitrification tank because of an impenetrable foam layer on top of the activated sludge. This reduces the number of sampling points to four: (1) pre-treated influent, activated sludge from the (2) nitrification zone and (3) the membrane tank and (4) permeate.

As can be expected the permeate samples did not cause an appreciable resistance increase and therefore they will be left out of consideration. Figure 6.5 represents the ΔR_{20} values for the pre-treated influent and the activated sludge samples collected from the denitrification- and membrane tank. In the period until the cheese factory was disconnected the pre-treated influent shows relatively better filtration properties than the activated sludge samples. Nonetheless the filterability of the influent can still be qualified as poor ($\Delta R_{20} \approx 1$).

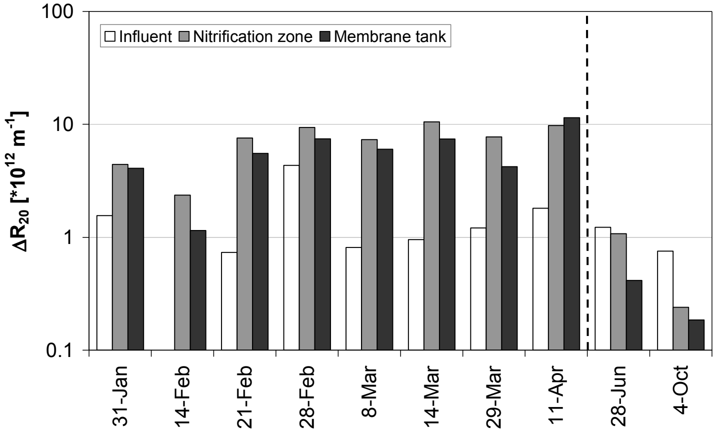


Figure 6.5: ΔR_{20} for different sampling points
(dashed line marks the uncoupling of the cheese factory)

Interestingly the PVA in the influent itself does not seem to cause extra fouling, considering the comparable ΔR_{20} values for the influent before and after uncoupling the cheese factory. Two possible explanations for this finding can be mentioned. The first explanation is that the PVA only causes problems from the moment it is brought in contact with the biomass. Another explanation might be that as a result of accumulation and the retention by the membrane the PVA concentrations in the activated sludge are higher than in the influent. The influence of the PVA on the activated sludge characteristics is discussed more in detail in section 6.6.

In general the filterability of the samples collected from the membrane tank is slightly better than of those from the nitrification zone. An explanation for this could be that the biological treatment process is still proceeding in the membrane tank, resulting in a lower concentration of potential foulants due to biological degradation or encapsulation within the activated sludge flocs. This issue is discussed more in detail in subsection 6.6.8.

6.4.5 Filterability of fractions

As discussed in section 6.3 it was intended to measure the filterability of the supernatant, the <0.45 µm fraction of the activated sludge samples collected from the membrane tank. However, since the filterability of the supernatant was very poor, it turned out to be practically impossible to filter a sufficient amount of supernatant with the flat sheet MF membrane to perform reliable DFCm experiments with it. Therefore the results obtained with this fraction are left out of consideration.

Figure 6.6 represents the filterability of the activated sludge samples collected from the membrane tank and its supernatant. The results clearly show the high fouling propensity of the supernatant. This indicates the free water constituents have a significant impact on the filterability. Interestingly most ΔR_{20} values for supernatant are comparable with the ones for the activated sludge; this supports the theory that relatively large sludge flocs are not involved in the fouling process. No clear trend between the filterability of activated sludge and its supernatant was demonstrated. The only weak noticeable trend is an improvement of both activated sludge and supernatant filterability after the cheese factory had been uncoupled from the sewer.

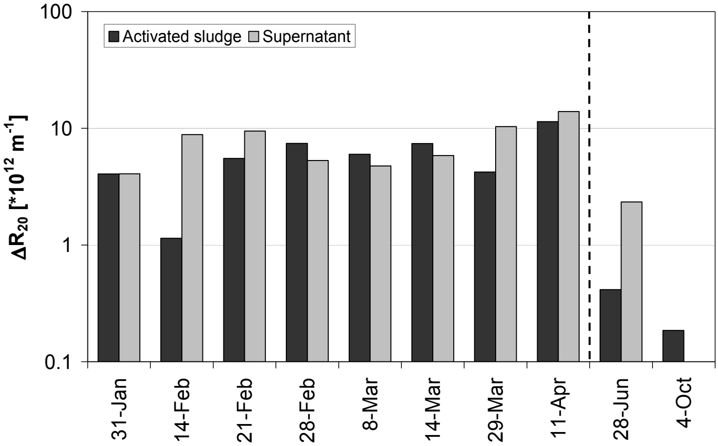


Figure 6.6: ΔR_{20} for activated sludge and its supernatant (membrane tank) (dashed line marks the uncoupling of the cheese factory)

6.5 Permeability

In this section the activated sludge filterability as characterised with the DFCm is analysed in relation to the development of the permeability in the full-scale MBR plant. First the development of the permeability throughout the total measuring period is addressed (section 6.5.1). Subsequently the permeability is analysed more detailed in the period that the samples were collected (section 6.5.2).

6.5.1 Permeability development full-scale plant

As briefly mentioned in section 6.4 the permeability of the full-scale installation was strongly influenced by the wastewater supply from a local cheese factory. Figure 6.7 represents the development of the permeability of one of the four membrane tanks (MT2) throughout the total measuring period, together with the activated sludge temperature and the applied flux. The results show a strong declining trend of the permeability in the first months of operation. An intensive chemical cleaning (IC) required after only a few months of operation, in the beginning of April, offered temporary recovery of the absolute permeability value but the decreasing trend persevered, even at very low fluxes. After uncoupling the cheese factory from the sewer the permeability recovers rapidly. From July the permeability stabilises and subsequently decreases slightly, but it has to be kept in mind that the applied flux in this period was relatively high.

Figure 6.7: Permeability development during measuring campaign

As from the beginning of June the dosage of Iron Chloride commenced. Unfortunately no samples were collected in between this moment and the stopping of the PVA wastewater supply. As a result only the combined effect of both events could be measured. Although a positive effect of the Iron Chloride on the filterability should not be excluded the recovery of the permeability is primarily attributed to the disappearance of the PVA in the wastewater. This is based on the considerable recovery of the permeability directly after the uncoupling of the cheese factory (and before the dosage of Iron Chloride).

6.5.2 Full-scale permeability and DFCm filterability

At first sight the permeability development seems to be related to the filtration characterisation results. To verify this three filtration curves are analysed more detailed in relation to permeability data. Three cases are discussed. First the situation on April 11th is analysed, the day that the worst filterability with the DFCm was measured. The second case is the situation around October 4th, the day that the best filterability was experienced. Finally the data of February 14th are analysed more in detail. On this day a relatively good filterability was measured compared to the other samples collected in this period.

April 11th

As illustrated in Figure 6.3 all activated sludge samples collected in the period from January to April showed a poor filterability quality. The absolute low point was experienced on April 11th, the last DFCm experiment before the cheese factory was uncoupled from the sewer[5]. Figure 6.8 represents the permeability development and the applied flux in one of the membrane tanks around the moment of sampling.

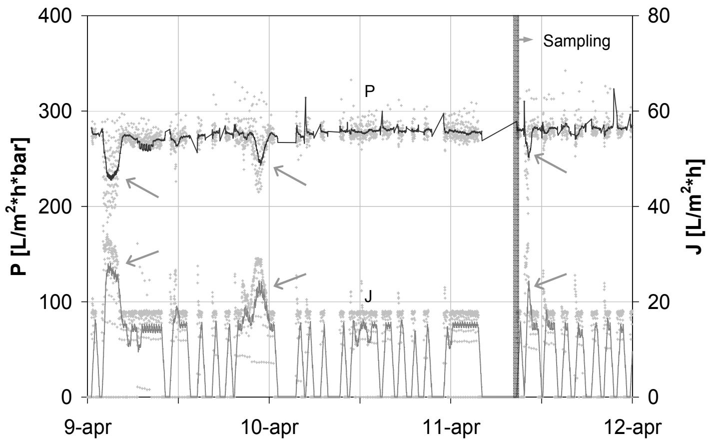


Figure 6.8: Permeability and applied flux around April 11th

[5] Actually, from hundreds of activated sludge samples tested with the DFCm over the years this sample has the doubtful honour to have the worst filterability ever measured…

The extreme poor filterability as measured with the DFCm was accompanied by severe permeability problems in the full-scale installation. In the first place at the time of sampling the permeability had dropped to worrying values below 300 L/m^2·h·bar, but moreover all four membrane tanks had to be put in operation at a flux of only 17 L/m^2·h to cope with DWF conditions. Higher fluxes immediately led to a sharp decline of the permeability, illustrated by the arrows in Figure 6.8. Since at this point the total functioning of the MBR was in danger an intensive chemical cleaning was performed, restoring the permeability to 500 L/m^2·h·bar. Clearly the IC was not a sustainable solution, considering the continuing decreasing trend of the permeability afterwards (see Figure 6.7).

October 4th

The two filtration characteristics gathered after the cheese factory was uncoupled from the sewer indicate a strong improvement of the activated sludge filterability (Figure 6.3). The DFCm results are again supported by the permeability data of the full-scale installation. Figure 6.9 represents the situation around October 4th. At the moment of sampling a capacity test was executed in which the maximum design flux of 37.5 L/m^2·h was imposed. The data clearly show that the permeability hardly suffers from the high flux. The permeability data thus support the statement that at the time of the capacity test the quality of the activated sludge was not a limiting factor in the filtration process.

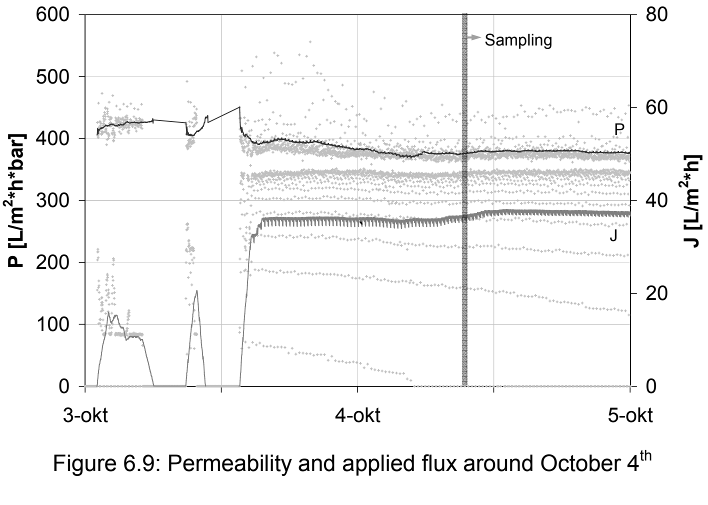


Figure 6.9: Permeability and applied flux around October 4th

February 14th

As illustrated in Figure 6.3 and Figure 6.4 the activated sludge sample collected on February 14th forms a relatively positive exception compared to the extreme poor filterability of the other samples collected in the first few months of operation. Figure 6.10 represents the full-scale permeability and flux data around the moment of sampling. As also indicated in Figure 6.7 the sample was collected during a capacity test in which a high flux of 37.5 L/m^2·h was imposed.

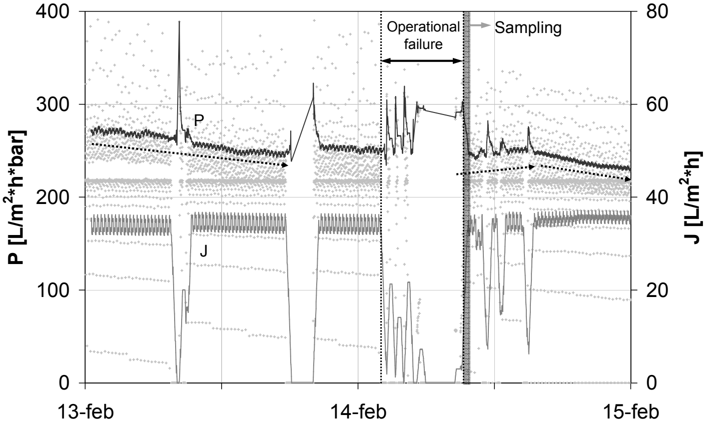


Figure 6.10: Permeability and applied flux around February 14th

Interestingly, by coincidence an operational failure occurred in the MBR plant just prior to sampling. As a result of this failure no permeate was extracted and no influent was supplied to the system for a period of about seven hours (this concerned all four membrane streets). The aeration in the bioreactor and the recirculation flows were however maintained. This thus implied an extension of the hydraulic retention time and extra time for the activated sludge to stabilise and degrade or encapsulate substances that could otherwise foul the membrane. After the permeate extraction was restored the permeability shows a slight increase, despite the fact that the applied flux was still 37.5 L/m^2·h. This indicates that at that very moment the filterability of the sludge was relatively good, which is confirmed by the DFCm experiment.

6.6 Activated sludge characteristics

As discussed in section 6.3 the DFCm experiments were accompanied by an extensive set of physical and chemical analyses. Unfortunately the majority of the research activities were planned and executed in the first few months after the start-up of the MBR. The major influence of the cheese factory on the filterability and the properties of the activated sludge could obviously not be foreseen. For planning and budget reasons the activated sludge properties were analysed only twice after the cheese factory had been uncoupled from the sewer. Evidently the activated sludge characteristics in the first few months of operation were unique and can not be considered representative for the MBR process in general. This reduces the value of the analyses in this period.

The analysis of the relation between filterability and activated sludge characteristics discussed in this section restricts to a comparison between the period before and after uncoupling the cheese factory. As explained in section 6.4 the ΔR_{20} values are obtained on the basis of experiments with a flux of 60 L/m^2·h.

6.6.1 Mixed Liquor (Volatile) Suspended Solids concentration

Figure 6.11 represents the ΔR_{20} values with the MLSS concentrations in the membrane tank and the nitrification zone. The gross of the samples have an MLSS concentration between 10 and 12 g/L. The results clearly refute a direct relationship between the activated sludge MLSS concentration and its filterability. Figure 6.12 represents the ΔR_{20} values with the organic fraction of the activated sludge (the ratio between the MLVSS concentration and the MLSS concentration). The results of June and October show a lower organic fraction of the activated sludge. This decrease is most likely not influenced by the PVA wastewater, but by the dosage of iron-chloride that commenced at the end of May.

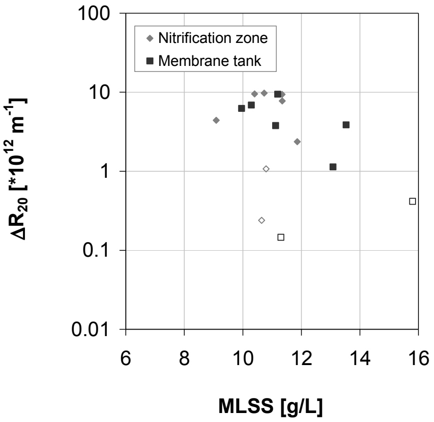


Figure 6.11: ΔR_{20} with MLSS concentrations
(open markers represent the results from June and October)

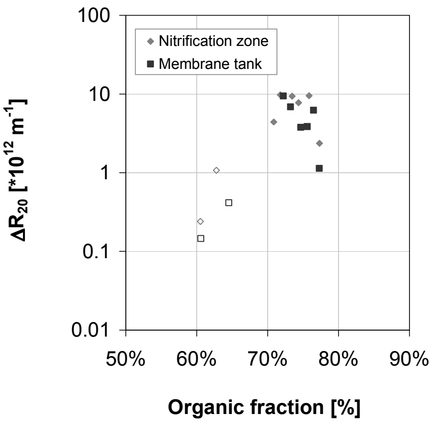


Figure 6.12: ΔR_{20} with organic fraction MLSS
(open markers represent the results from June and October)

The results in Figure 6.12 indicate that the properties of the activated sludge were affected by the Iron Chloride dosage. The improvement of the filterability is however predominantly associated to the cheese factory wastewater: on the basis of the available data the influence of the Iron Chloride dosing on the activated sludge filterability can not be verified.

6.6.2 Floc Size Distribution

The particle size distribution measurements in the size range from 2 to 1000 µm are analysed on the basis of the mean size of the activated sludge flocs. Figure 6.13 represents the ΔR_{20} values with the mean size of the activated sludge flocs (µm). In the period between January and April the mean particle size varied between 65 and 90 µm. Despite the relatively small range the results seem to indicate a relation with the filterability. This is in accordance with several literature results, which suggest a better filterability with larger flocs (see chapter 3.8.3). On the other hand in June and October the flocs were smaller while the filterability was better. Microscopic research showed that in June and October the flocs were smaller but less compact (STOWA 2006). Apparently the structure of the floc is a more important factor than its size. The results presented in Figure 6.13 at least refute a direct relationship between the floc size and the filterability. Unfortunately it can not be verified to what extent the uncoupling of the cheese factory or the dosage of iron chloride affected the floc size and –structure.

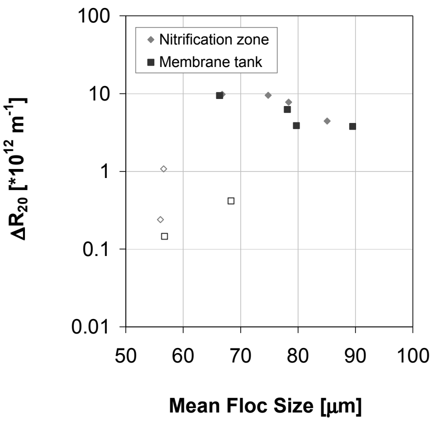


Figure 6.13: ΔR_{20} with mean floc size
(open markers represent the results from June and October)

6.6.3 Temperature

In Figure 6.14 the obtained ΔR_{20} values are plotted against the temperature of the activated sludge samples. The influence of the temperature on the filterability is insignificant compared to the influence of the cheese factory wastewater. With respect to the exceptional circumstances with the cheese factory wastewater a possible relationship between the filterability of activated sludge and its temperature can not be assessed.

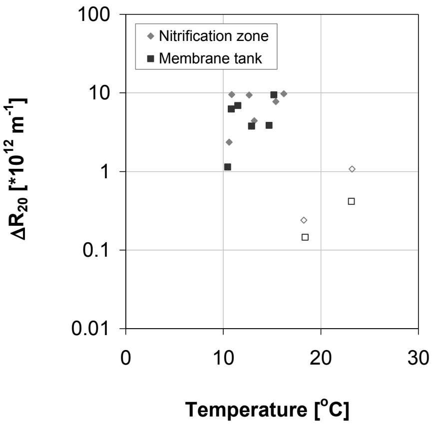


Figure 6.14: ΔR_{20} with activated sludge temperature
(open markers represent the results from June and October)

6.6.4 Extracellular Polymeric Substances

As discussed in chapter 3.8 several researchers suggest a relation between fouling and the extracellular polymeric substances bound to the activated sludge flocs. Figure 6.15 and Figure 6.16 represent ΔR_{20} values with the measured EPS concentrations (mg/g volatile suspended solids). For both proteins and polysaccharides the lowest values were measured in October, when the filterability was the best. The other results, including the ones from June, do not show a relation with the filterability. Moreover, the results obtained in June are comparable with several values measured in January to April (10-20 mg/g VSS for proteins and 20-30 mg/g VSS for polysaccharides), while the filterability was superior in June. This implies that the EPS concentration is not a strong indication for the filterability.

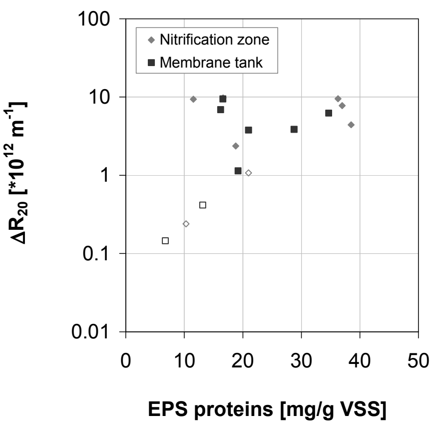


Figure 6.15: ΔR_{20} with EPS proteins
(open markers represent the results from June and October)

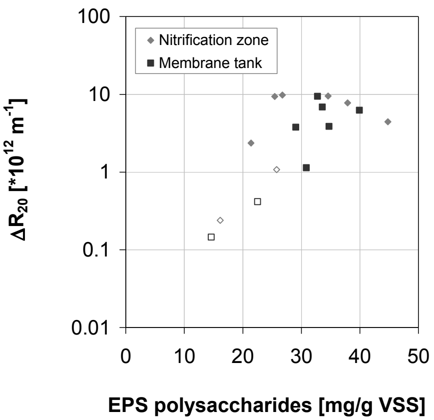


Figure 6.16: ΔR_{20} with EPS polysaccharides
(open markers represent the results from June and October)

6.6.5 Soluble Microbial Products

As discussed in chapter 3.8 the Soluble Microbial Products (SMP) concentration in the activated sludge free water is widely considered to be an important parameter affecting fouling and filterability. Though EPS and SMP are closely related to each other, in this research no clear relation between both concentrations was demonstrated (not represented graphically).

Figure 6.17 and Figure 6.18 represent the ΔR_{20} values with the protein- and polysaccharide concentrations in the supernatant. The relation between SMP and filterability is at most very weak. For both proteins and polysaccharides the lowest concentrations were indeed measured in October, when the ΔR_{20} values were the lowest. On the other hand the concentrations measured in June (20 mg/L for proteins and 40-60 mg/L for polysaccharides) is comparable with the ones in the period from January to April, while the filterability in both cases is totally different.

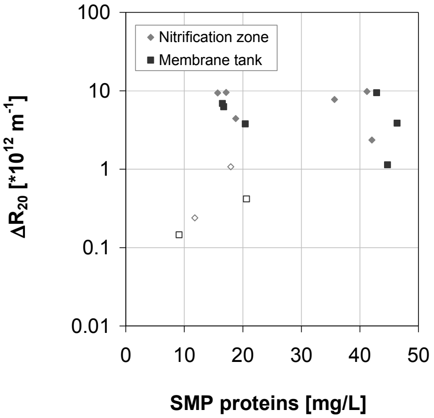


Figure 6.17: ΔR_{20} with SMP protein concentrations
(open markers represent the results from June and October)

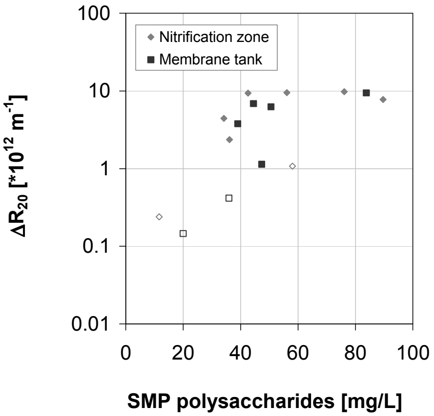


Figure 6.18: ΔR_{20} with SMP polysaccharide concentrations
(open markers represent the results from June and October)

6.6.6 Chemical Oxygen Demand and Total Organic Carbon

The relationship between filterability and the Chemical Oxygen Demand (COD) concentrations and the Total Organic Carbon (TOC) concentrations in the supernatant is comparable with the SMP concentrations. Again the lowest values were measured in June and October when the filterability was good (no TOC data available from the experiments in June). Although the amount of data after uncoupling the cheese factory is limited the results support the hypothesis that the filterability is determined by the concentration of substances in the activated sludge supernatant.

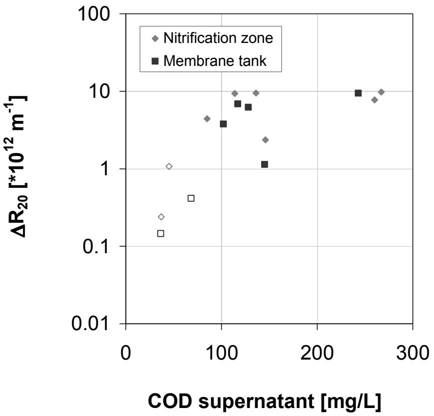


Figure 6.19: ΔR_{20} with COD supernatant
(open markers represent the results from June and October)

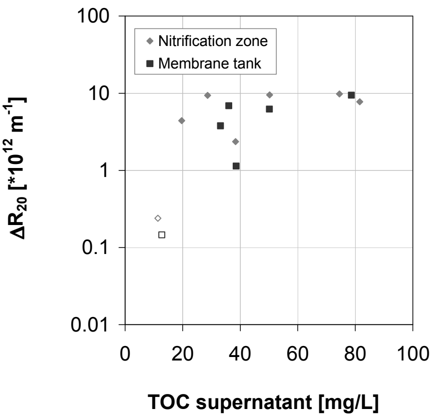


Figure 6.20: ΔR_{20} with TOC supernatant
(open markers represent the results from June and October)

6.6.7 Fractionation

As discussed in section 6.3 the activated sludge samples were fractionated to investigate the size range of several constituents in the supernatant. The supernatant of the activated sludge samples was fractionated with a MF membrane (<0.45 µm), a UF membrane (<0.03 µm) and with a stirred UF cell to separate MWCF fractions of 500, 100, 10 and 1 KDa respectively. Figure 6.21 and Figure 6.22 show the concentrations per fraction for proteins and polysaccharides (samples collected from the membrane tank). The results for COD and TOC show a similar trend (not graphically represented).

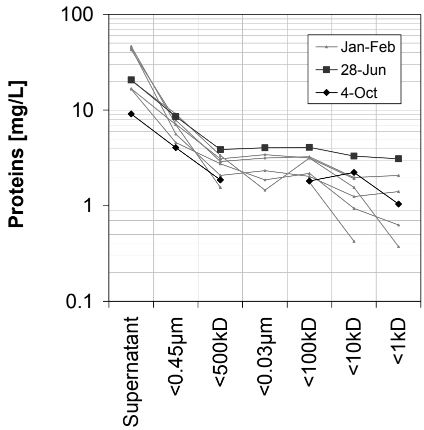


Figure 6.21: Fractionation results for proteins

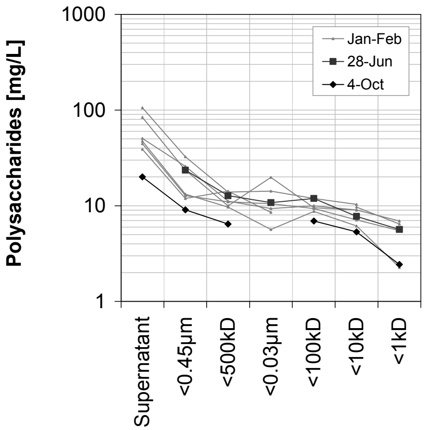


Figure 6.22: Fractionation results polysaccharides

The results demonstrate that the differences in the concentrations of substances in the supernatant before and after uncoupling the cheese factory are predominantly determined by substances larger than 0.45 µm. When analysing the fractionation results in relation to the ΔR_{20} values the results thus indicate that the filterability of activated sludge is linked with the concentration of substances in the supernatant that are more than ten times bigger than the membrane pores. On the other hand it has to be realised that the available information is limited and that the exact influence of the cheese factory on the particle size distribution in the supernatant is not clear.

6.6.8 Comparison nitrification zone and membrane tank

The filterability of the samples collected from the membrane tank was generally superior to those collected from the nitrification zone (see Figure 6.5). This is again illustrated somewhat differently in Figure 6.23, which plots the ΔR_{20} values from both samples against each other. The markers below the diagonal line indicate lower values and thus relatively better filterability of the activated sludge collected from the membrane tank. The improvement of the filterability is not supported by changing activated sludge properties. Figure 6.24 represents the SMP concentrations in the membrane tank versus the nitrification zone. In general the values are quite similar and even somewhat higher in the membrane tank. The better filterability in the membrane tank can thus not be explained on the basis of changes in SMP concentrations. Similar results were obtained for all other activated sludge properties; none of the other analyses could explain the alteration of the filterability.

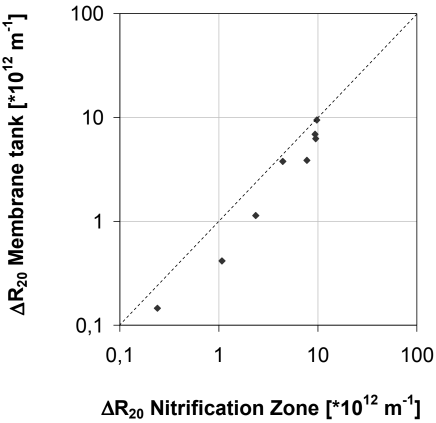


Figure 6.23: ΔR_{20}: membrane tank versus nitrification zone

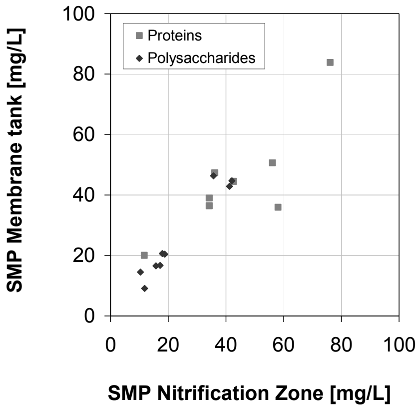


Figure 6.24: SMP concentrations: membrane tank versus nitrification zone

6.7 Summary and conclusions

An unavoidable aspect of conducting practical oriented research at a full-scale wastewater treatment plant is that the results are depending on a multitude of varying, unexpected and uncontrollable circumstances. Evidently these circumstances can not all be verified and this complicates the interpretation of the experimental results. For the Varsseveld measuring campaign the results were strongly influenced by the wastewater flow from a local cheese factory to the MBR in the first few months of operation. The Poly Vinyl Acetate (PVA) in the cheese factory wastewater turned out to be the cause of severe fouling problems, as it was retained by the membrane and could not be degraded biologically. After a few months of operation it was decided to uncouple the cheese factory wastewater flow from the sewer.

By chance the majority of the research activities were executed when the PVA wastewater was still discharged to the MBR plant. Evidently the activated sludge characteristics in this period were unique and not representative for the MBR process in general. As a consequence the value of the activated sludge property analyses in this period is questionable. On the other hand the problems with the PVA wastewater provided big differences in the activated sludge filterability compared to the situation after the cheese factory had been uncoupled. These variations were beneficial for analysing the DFCm experiments and assessing the relationship between the filterability and the development of the permeability in the full-scale installation.

6.7.1 Filtration Characterisation

As a consequence of the poor extreme poor filterability in the first few months after the start-up of the full-scale plant the DFCm experiments are not analysed on the basis of to the standard measuring protocol (80 $L/m^2{\cdot}h$) but on a lower flux (60 $L/m^2{\cdot}h$). Additional experiments with several different fluxes however demonstrate that the value of the flux is not affecting the filterability (a sample with poor filterability at a high flux has a correspondingly poor filterability at a lower flux and vice versa).

In the period before uncoupling the cheese factory the measurement frequency was approximately once every two weeks. In this period the filterability was stable and extremely poor (ΔR_{20} >> 1) with one exception. By chance one time the samples were collected just after an operational failure in the plant. During a period of approximately seven hours no influent was supplied to the plant and also no permeate was extracted. The internal recirculation and biological treatment however continued. The extension of the hydraulic retention time appeared to have a significant positive effect on the filterability, although the filterability could still be qualified as poor.

Unfortunately the filtration characterisation data gathered after uncoupling the cheese factory (May) is limited; only two isolated measurements were performed (June and October). The drawback of the isolated measurements is that they did not provide information about the development and the dynamics of the activated sludge filterability. The DFCm results however clearly show a spectacular improvement of the filterability. This confirms the proposition that the fouling problems in the full-scale plant had indeed to be attributed to the filterability properties of the activated sludge.

6.7.2 Activated sludge properties

As discussed in section 6.6 the activated sludge properties in the first few months of operation can not be considered representative for the MBR process in general. The data available after the cheese factory was uncoupled is very limited (only two sampling moments). Unfortunately these unforeseen circumstances severely diminish the value of the activated sludge quality analyses performed in this research campaign.

Comparing the activated sludge characteristics before and after uncoupling the cheese factory strongly supports the hypothesis that activated sludge filterability is related to the amount of substances in the free water with a size larger than 0.45 μm. From the two measurements in June and October the concentrations of SMP, COD and TOC in the supernatant are substantially lower than in the period from January to April. These conclusions have to be considered with some reserve considering the exceptional circumstances with respect to the cheese factory wastewater and the limited amount of information gathered after it was uncoupled from the sewer.

6.7.3 Conclusions

To conclude this chapter the main conclusions of the measurement campaign at MBR Varsseveld are summarised below:

- The DFCm has proven to be a useful tool to detect differences in the filterability of MBR activated sludge.
- The DFCm measurements demonstrate a clear relationship between the filterability of the activated sludge and the development of the membrane permeability in the full-scale installation. Poor filterability in the first few months is accompanied by severe permeability decrease. Stable permeability was encountered when the filterability was good. This finding supports the hypothesis that the filterability is related to the irreversible fouling rate.
- The DFCm experiments demonstrate that the activated sludge quality was a limiting factor in the filtration process at MBR Varsseveld.

- The encountered activated sludge properties in the first few months of operation were unique and not representative for the MBR process in general. As a consequence no clear conclusions can be drawn on which activated sludge properties are related to its filterability. The limited amount of data gathered after uncoupling the cheese factory however indicates that the improvement of the filterability is linked with the concentration of substances in the supernatant with a size larger than 0.45 µm.
- Due to the exceptional circumstances encountered during the most part of the measuring campaign no recommendations could be formulated on how to optimise the biological and operational process with respect to the prevention of fouling.
-

7 Filtration Characterisation at MBR Heenvliet

7.1 Introduction

In this chapter the measurement campaign performed at the MBR Heenvliet is discussed. Activated sludge samples were collected on a weekly basis for a period of more than one year and analysed for their filterability and several characteristics. In addition the filterability was analysed in relation to the development of the permeability.
Section 7.2 provides a general background about the MBR plant. In section 7.3 the objectives and the approach of the research campaign are addressed. In section 7.4 the filtration characterisation results are discussed. Subsequently the filtration characterisation results are analysed in relation the permeability data (section 7.5) and several activated sludge characteristics (section 7.6). The chapter concludes with a recapitulation of the research campaign and an enumeration of the main conclusions, in section 7.7.

7.2 Hybrid MBR Heenvliet

7.2.1 Background

The wastewater treatment plant of Heenvliet in the Netherlands (Figure 7.1), operated by Waterboard Hollandse Delta (WSHD), originally consisted of a conventional activated sludge system (type carrousel) with a design capacity of nearly 9.000 population equivalents. Because the wastewater treatment discharges its effluent into surface water with recreational functions additional chemical disinfection was applied in summertime by means of sodium hypochlorite dosage. When the sewage of the nearby village of Abbenbroek was also connected to wwtp Heenvliet the required capacity increased to 13.000 population equivalents (390 m^3/h). To increase the capacity and the efficiency of the existing plant it was upgraded with an MBR. In the new so called “hybrid” configuration the MBR and the conventional system are combined to treat the total wastewater flow.

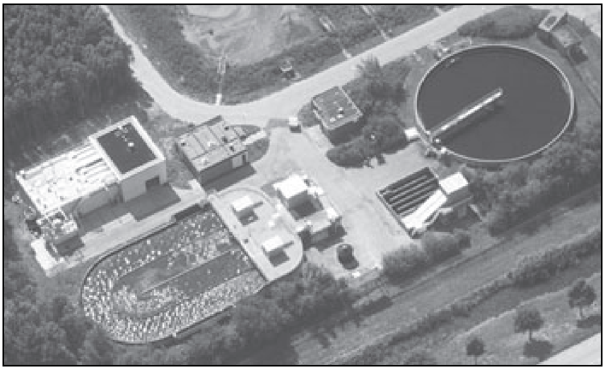


Figure 7.1: Aerial view of wwtp Heenvliet (WSHD)

7.2.2 Design

The MBR part of wwtp Heenvliet is designed to treat the dry weather flow (100 m^3/h, approximately 25% of the maximum hydraulic capacity of the total wwtp). As a pre-treatment step the influent passes a 6 mm screen selector before entering the carrousel. With an ultra-low design loading rate of 0.045 g COD/g MLSS·day the treatment plant aims at reaching a high removal efficiency for nitrogen and phosphorus.

In principle during DWF conditions the total influent flow is treated by the MBR; during storm weather events the extra flow is treated by the clarifier of the conventional system. The basic idea behind the hybrid MBR is thus the possibility to use the membrane surface to the maximum during DWF conditions. The hybrid MBR can be operated in two modes: parallel or in-series. In the parallel mode the MBR treats a fixed portion of the influent flow. In this way the MBR and the conventional system function totally independent of each other. During the period described in this chapter the plant was operated in the in-series mode, as schematically represented in Figure 7.2.

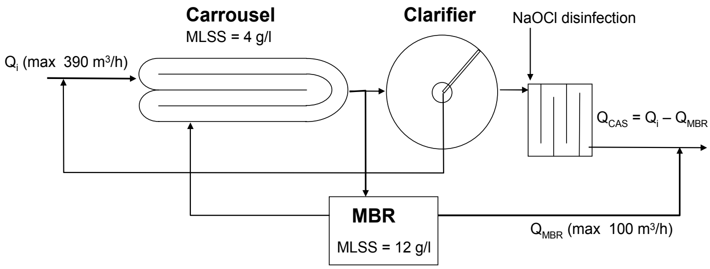


Figure 7.2: Schematic representation in-series operation of hybrid MBR Heenvliet

The in-series operation of the plant implies that the activated sludge that reaches the membrane tanks has undergone a double biological treatment: first in the carrousel and subsequently in the MBR. Table 7.1 (Krzeminski, 2008).

Table 7.1: Data MBR Heenvliet

Parameter	**Unit**	**Heenvliet**	
Start of operation	-	March 2006	
Water Board	-	Hollandse Delta	
Wastewater	-	municipal	
Biological Capacity	p.e.	3333	
Hydraulic capacity (RWF)	m^3/h	100	
Hydraulic capacity (DWF)	m^3/h	50-100	
Hydraulic capacity (Average)	m^3/d	1500	
Permeate production	m^3/h	100	
Process Configuration	-	Submerged	
Membrane type	-	Flat sheet	
Membrane supplier	-	Toray	
Membrane material	-	PVDF	
Bioreactor volume	m^3	2 tanks * 73 = 146	
Number of lanes	m^2	2 parallel tanks	
Total membrane area	m^2	4115	
Packagin density	m^2/m^3	28	
MF/UF	-	UF	
Membrane pore size	µm	0,08	
Design Flux (netto/brutto) RWF	$L/m^2 \cdot h$	24,3	
Average Flux (netto/brutto) DWF	$L/m^2 \cdot h$	12.2-24.3	
Maximum Flux (netto/brutto) RWF	$L/m^2 \cdot h$	45 or 56.3	
Peak flows	m^3/d	10000	
DWF : RWF	-	1:2.5 (38:100 m3/h)	
Sludge concentration	$kgMLSS/m^3$	10	
F/M ratio	gBOD/gMLSS*d	0,045	
Filtration	min	9	
Cleaning	min	1 (relaxation)	
Cleaning	-	Mechanical and chemical	1-4 per year
Aeration	$Nm^3/h*m^2$	0,3	
Aeration intervals	sec	20	
SRT	days	31	
HRT	hours	20 (Series)	

7.3 Research objectives and approach

The hybrid MBR at the wastewater treatment plant of Heenvliet serves as a demonstration project to gather experience with this relatively innovative configuration.

7.3.1 Objectives

The objectives for the research at hybrid MBR Heenvliet described in this dissertation are as follows:

- Verify the relation between activated sludge filterability as obtained with the DFCm and the development of the permeability in the MBR.
- Verify the relation between activated sludge filterability and several physico-chemical activated sludge properties. The emphasis again lies on the most mentioned parameter in literature: soluble microbial products.

7.3.2 Approach

During a period of more than one year activated sludge samples were collected from the MBR on a weekly basis. The aim of sampling on such a regular basis was to form a reliable indication of the development of activated sludge filterability throughout a long-term period. The sampling point was the aerated tank, where the activated sludge is aerated shortly just prior to entering one of the two the membrane tanks. The samples were transported to the laboratory of Delft University (approximately 45 minutes) where the analyses were performed. The physical-chemical analyses that were performed are listed in Table 7.2.

Table 7.2: Activated sludge quality analyses

Analysis	**Reference to method**
Activated sludge	
- Mixed Liquor Suspended Solids (MLSS)	Chapter 4.6.1
- Temperature (T)	-
- Sludge Volume Index (SVI)	Chapter 4.6.2
- Viscosity	Chapter 4.6.4
Free water	
- Soluble Microbial Products (SMP) proteins	Chapter 4.6.3
- Soluble Microbial Products (SMP) polysaccharides	Chapter 4.6.3
- Particle Size Distribution	Chapter 4.6.5
Permeate	
- Soluble Microbial Products (SMP) proteins	Chapter 4.6.3
- Soluble Microbial Products (SMP) polysaccharides	Chapter 4.6.3

7.4 Filtration Characterisation results

7.4.1 DFCm curves

In the first two months of the measuring campaign several different fluxes were tested in order to assess the reliability of the DFCm results and whether the standard measuring protocol could be applied. Initial results showed poor activated sludge filterability, but nonetheless the standard flux ($J = 80\ L/m^2 \cdot h$) could be applied. Figure 7.3 represents the results of four random DFCm experiments. In case of good filterability ($\Delta R_{20} < 0.1$) the compression coefficient s could not be determined with significant reliability. For the DFCm experiments that did show a significant resistance increase the compression factor was reasonably constant with a value of 0.24 on average.

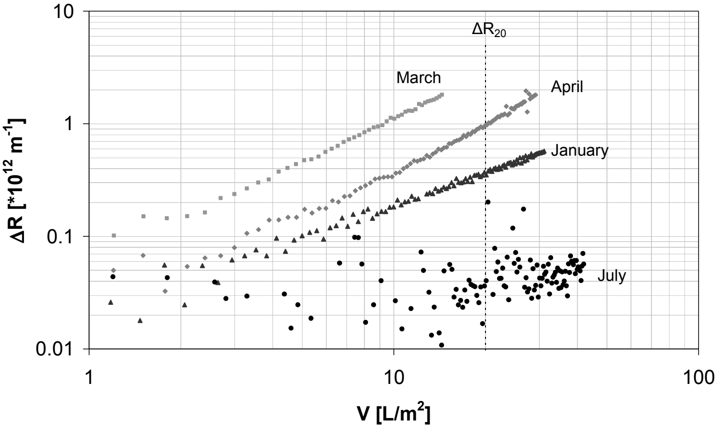


Figure 7.3: Output of four random DFCm experiments at MBR Heenvliet

7.4.2 Flux sensitivity

Several additional DFCm experiments with other fluxes besides the standard value of 80 L/m^2·h were conducted to verify the sensitivity of the results for the flux. Figure 7.4 shows the values for four random DFCm experiments. The results show consistent behaviour of the ΔR_{20} values for the different fluxes. The results support the hypothesis that the filterability quality of the activated sludge sample is not dependant on the applied flux (a sample with a poor filterability at a high flux also has a poor filterability at a lower flux, and vice versa).

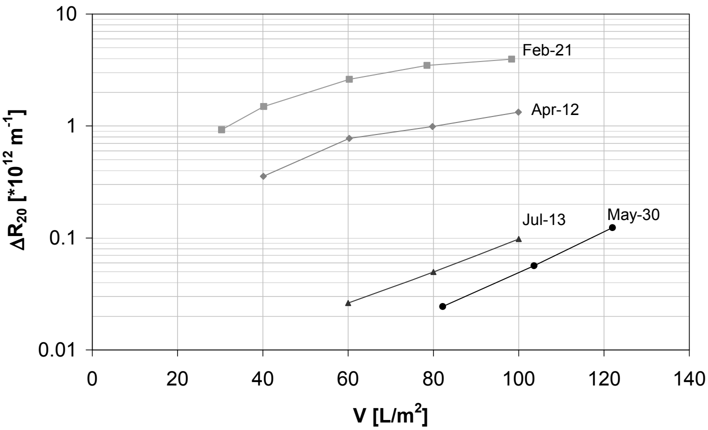


Figure 7.4: Sensitivity of ΔR_{20} for flux variations

7.4.3 ΔR_{20} evolution

During the campaign 58 DFCm experiments were performed according to the standard measuring protocol. When combining all the gathered DFCm curves the evolution of ΔR_{20} can be plotted during the total period, see Figure 7.5.
Based on the results a rough division can be made between four periods. In period I the MBR was still in preparation for the start-up. The membranes were not operational yet at this time. As a first step the required biomass concentration in the system had to be reached. To this aim additional activated sludge from the nearby wwtp of Rozenburg was added to the system. Subsequently the biomass concentration in the MBR was increased from about 4 to 10 g/L by supplying thickened activated sludge from the settling tank and by biological growth. Obviously the biomass was not in a stable situation in period I. This seems to be reflected by the DFCm results which show very poor filterability (ΔR_{20} >> 1).
As soon as the biomass concentration reached a constant level (approximately 10 g/L), at the start of period II, the ΔR_{20} values decrease considerably within a period of a few days (unfortunately no measurements were performed during these days). Apart from the fact that the data obtained in period II are rather unsteady the overall trend clearly shows a further improvement of the filterability. The lowest ΔR_{20} values were measured in period III; all activated sludge samples show excellent filterability with ΔR_{20} values of practically 0 m^{-1}. In period IV the ΔR_{20} values increase slightly, but the filterability can still be qualified as good to moderate.

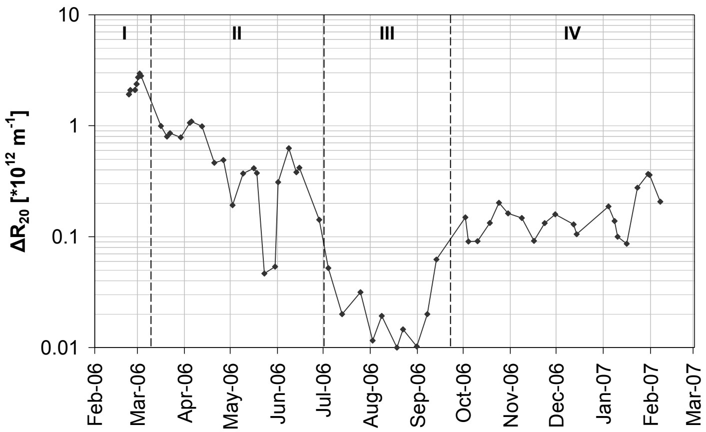


Figure 7.5: ΔR_{20} evolution during the measuring period
(standard measuring protocol, CFV = 1.0 m/s, J = 80 $l/m^2 \cdot h$)

The results gathered in period I and II demonstrate that activated sludge filterability is a dynamic parameter that can change considerably within a period of several days. On the other hand, the results gathered as from July also demonstrate that when the activated sludge experiences stable and probably favourable conditions the filterability is very constant. Although the DFCm experiments provide a momentaneous indication of the activated sludge filterability at the very moment of sampling there are no reasons to suspect differences in filterability throughout period III and IV. The constancy of the measured ΔR_{20} values also supports the reliability of the DFCm measurements.

7.5 Permeability

7.5.1 Permeability development

The permeability development in one of the two membrane tanks throughout the measuring period is represented in Figure 7.6, together with the generally applied flux and the activated sludge temperature (these data concern MT2, but both tanks show a comparable development). In the first few weeks after the start-up of the membrane filtration the permeability drops considerably and subsequently more or less stabilises at approximately 800 L/m^2·h·bar. After a decrease of the applied flux the permeability data are fragmentary, but the overall trend shows a recovery to values higher than 1000 L/m^2·h·bar.
However, as from the beginning of August a rather constant and continuous decreasing trend sets in. An acidic cleaning in September provides a modest recovery of the permeability, but the decreasing trend perseveres. At the end of January 2007 the permeability had dropped to about 200 L/m^2·h·bar. At this point another chemical cleaning was performed, this time with NaOCl.

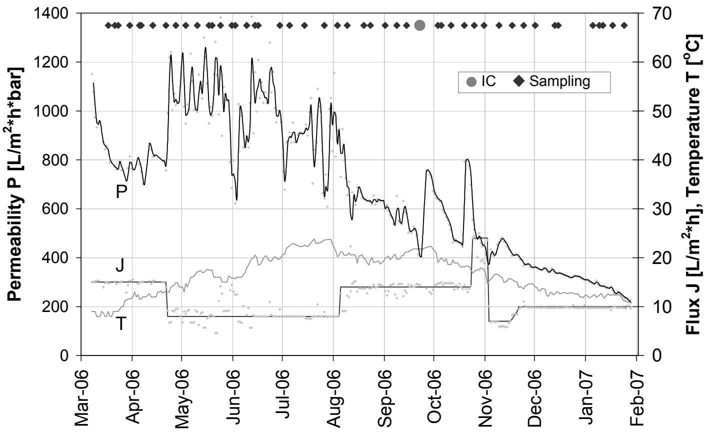


Figure 7.6: Permeability development (MT2)

7.5.2 Full-scale permeability and DFCm filterability

Evidently the DFCm results (Figure 7.5) do not match with the development of the permeability in the full-scale installation (Figure 7.6); while the permeability shows a continuous decrease the DFCm experiments indicate a good filterability. As discussed in chapter 5.3.3 two possibilities are available to explain this discrepancy. The first explanation would be that there simply is no relation between the short-term filterability (as measured with the DFCm) and the long-term permeability of the considered MBR plant. The second explanation would be that the origin of the permeability decrease was not related to the activated sludge properties but in shortcomings in the operation and/or the properties of the membranes. Additional study revealed that both the membrane operation and –properties were indeed far from optimal.

Membrane construction failures

From the second half of 2006 activated sludge particles were encountered in permeate of several modules. When the membranes were cleaned chemically at the beginning of 2007 constructional failures came to light. It appeared that for several membranes the glue connection between the membrane material and the frame had become unstuck.
Besides the obvious adverse effect these failures had on the permeate quality it could also have been detrimental to the filtration performance of the membrane, considering the accumulation of activated sludge that was noticed in the membrane headers. The exact consequence of the activated sludge accumulation in the membrane headers on the filtration performance can not be verified, but in theory a clogged header can totally exclude the membrane from participating in the filtration process.

Clogging

In the course of time doubts arose concerning the cleaning efficiency of the coarse bubble aeration. With a total installed capacity of 0.14 $Nm^3/m^2 \cdot h$ the aeration rate was already considered relatively low compared to reference values that were applied in the pilot-scale research in Beverwijk (STOWA 2002). But besides the low capacity there were also indications that the air was not distributed evenly over the total membrane surface:

- With respect to a possible future extension of the membrane surface the modules were not fully packed with membranes. The aerators were however installed over the full width of the membrane tank, resulting in a about at least 6% of the total supplied air not being functional.
- As a result of a lower resistance in the open area the air bubbles intended to scour the membranes were suspected to escape the membrane tank through this (lower resistance) open area. The more turbulent circumstances above the open area compared to the membrane area which were visually observed seemed to confirm this suspicion.
- Some operational problems were encountered with the aeration flush valves, resulting in temporarily uneven distribution of the aeration.

Based on above mentioned issues several measures were taken to improve the filtration process. An extra aeration pump was installed, aerators below the open area were cut off and a partition was made between the open area and the other aerators. As a result the potential aeration rate increased to approximately 0.3 $Nm^3/m^2 \cdot h$.
Based on the membrane construction failures that were encountered during the previously mentioned chemical cleaning it was decided to examine all the membranes. This event proved beyond any doubt that the created crossflow circumstances in the membrane tank had not been

sufficient, see Figure 7.7. About 25% of the membranes were covered with a thick black sludge layer that was expected to be highly impermeable.

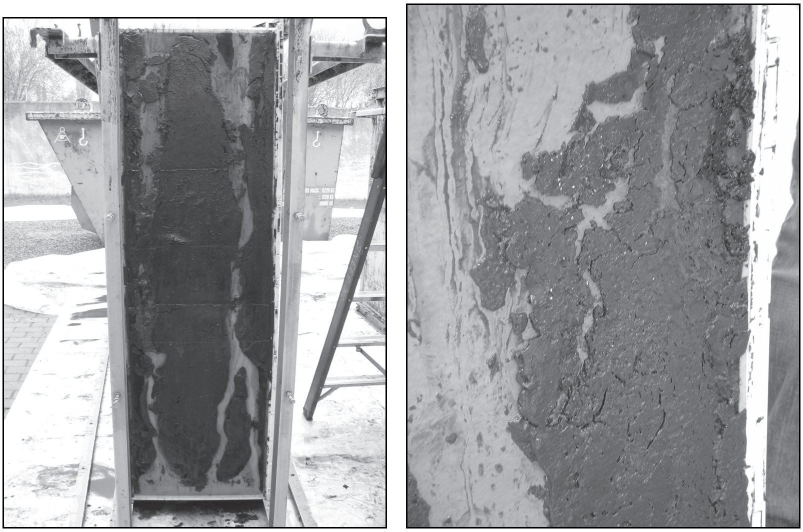


Figure 7.7: Clogged membranes MBR Heenvliet (WSHD)

The logical consequence of the clogging problems was that the total membrane surface available for permeation decreased. The total permeate flow had to be processed with a lower membrane surface and the flux in the membrane area that still was available for permeation increased correspondingly. As discussed in chapter 5.7 an irregular distribution of the flux inevitably accelerates the fouling process, irrespective of the activated sludge filterability. The observed clogging problems (in combination with the good filterability as measured with the DFCm) strongly support the suspicion that the decline in permeability could not be attributed to poor activated sludge filterability.

7.6 Activated sludge characteristics

This section discusses the relation between the activated sludge filterability and several physical-chemical activated sludge properties.

7.6.1 Mixed Liquor Suspended Solids concentration

Figure 7.8 represents the obtained ΔR_{20} values with the MLSS concentration of the activated sludge samples. As with the filterability a distinction can be made between different periods. In period *I*, when the biomass concentration was gradually increased from 4.6 to 11.4 g/L, the results show a similar gradual increase of ΔR_{20} from 1.9 to 2.9 ($\cdot 10^{12}$ m^{-1}). Presumably the explanation for the results in period *I* is related to the fact that the biomass had not reached stable biological conditions yet. The results seem to indicate an increase of fouling substances with increasing biomass. The results obtained in the other periods are in sharp contrast with the ones form period *I*. The overall results of period *II*, *III* and *IV* even suggest a better filterability with increasing MLSS concentration.

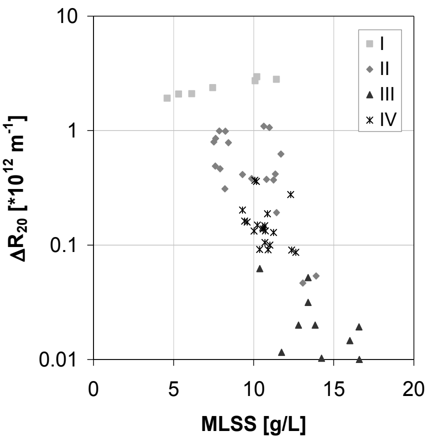


Figure 7.8: Filterability with MLSS concentrations

7.6.2 Activated sludge temperature

The excellent filterability that was measured in the summer months (Figure 7.5) suggests a seasonal or temperature effect on the DFCm results. The overall results indeed indicate a better filterability at higher temperatures, see Figure 7.9. On the other hand it has to be mentioned that the poor filterability measured in period I is more likely to be the effect of the unstable

biological circumstances that the biomass was experiencing rather than the low temperature. Also the improving trend of the filterability in period II is more likely to be related to the more stable biological conditions in the MBR rather than to the accompanying gradual temperature increase in this period.
Nonetheless a relationship between activated sludge filterability and temperature is plausible. Higher temperatures are beneficial for the biological treatment process. It can be expected that pollutants that can foul the membrane are degraded to a higher extent when the activated sludge temperature is higher. The relation between temperature and filterability is discussed more in detail in chapter 8.5.

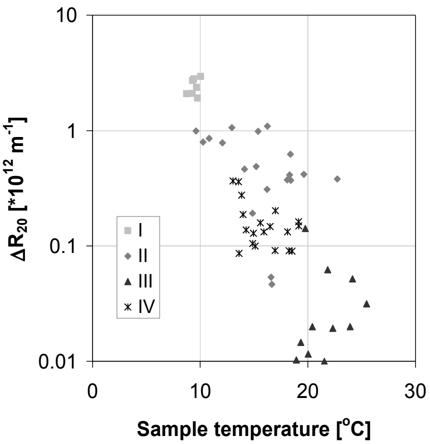


Figure 7.9: Filterability with activated sludge temperature

Evidently the activated sludge temperature itself is not the crucial parameter with respect to its filterability. Nonetheless, it can be expected that when the operational circumstances in the MBR are stable and favourable the temperature becomes a factor of influence.

7.6.3 Sludge Volume Index

The unstable biological circumstances at the start-up of the MBR also seem to be expressed in the SVI values. Although unfortunately no SVI data are available from period I the high SVI (171 mg/L) directly at the start of period II indicates poor activated sludge settling properties.
As soon as a stable biomass concentration was reached - as from the start of period II - the SVI starts to decrease. In a period of approximately two months the SVI decreases from 171 mg/L to 95 mg/L. From this moment the SVI stabilises at a value of approximately 100 mg/L. The decrease of the SVI is accompanied by a decrease of the ΔR_{20} values. As a consequence the overall results indicate a relationship between filterability and the SVI, illustrated in Figure 7.10.

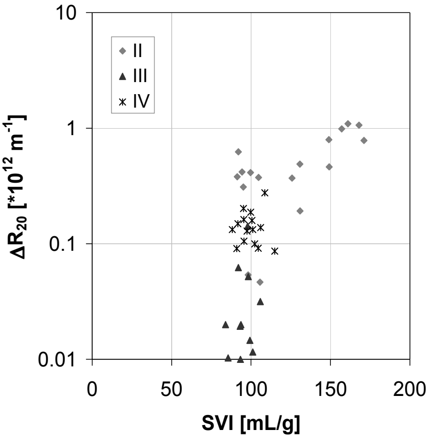


Figure 7.10: Filterability with Sludge Volume Index

7.6.4 Activated sludge viscosity

As discussed in chapter 3.9.3 the activated sludge viscosity is a parameter linked to fouling. Due to technical problems with the rheometer the viscosity measurements could not be performed throughout the whole period. However, the available results clearly show that there is no direct relation between filterability and viscosity (see Figure 7.11). The viscosity was found to be predominantly dependant on the MLSS concentration, illustrated in Figure 7.12. The influence of the temperature on the viscosity was found to be negligible compared to the MLSS concentration (not graphically represented).

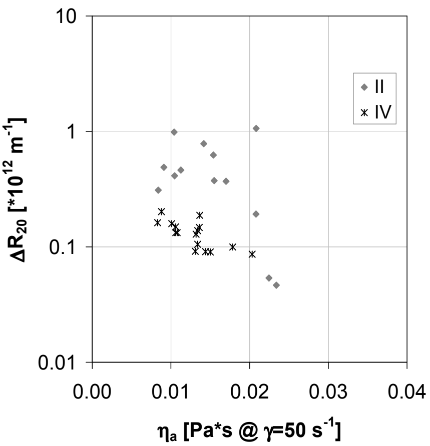


Figure 7.11: Filterability with apparent viscosity (at shear rate = 50 s^{-1})

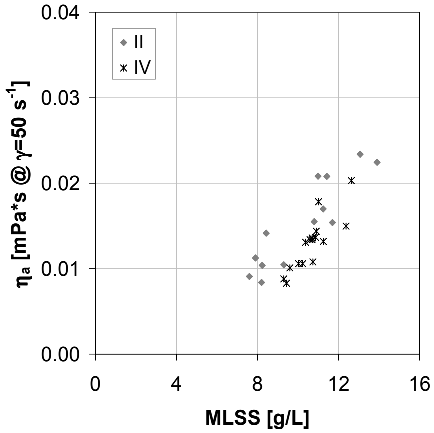


Figure 7.12: Apparent viscosity with MLSS concentration (at shear rate = 50 s^{-1})

7.6.5 Soluble Microbial Products concentration

The soluble microbial products concentrations fluctuated and no clear trend was observed throughout the whole measuring period. SMP protein concentrations varied between approximately 6 and 27 mg/L. Figure 7.13 represents the obtained ΔR_{20} values with the SMP protein concentrations.

The results do not confirm a correlation between SMP and filterability. The generally measured SMP concentrations between 10 and 20 mg/L are accompanied by a filterability ranging from good to very poor.

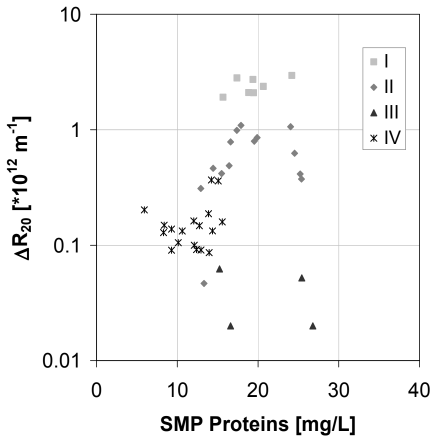


Figure 7.13: Filterability with SMP protein concentrations

The results for SMP polysaccharides are comparable with proteins. High concentrations are accompanied by poor filterability, but in the dominant concentration range the filterability again varies considerably.

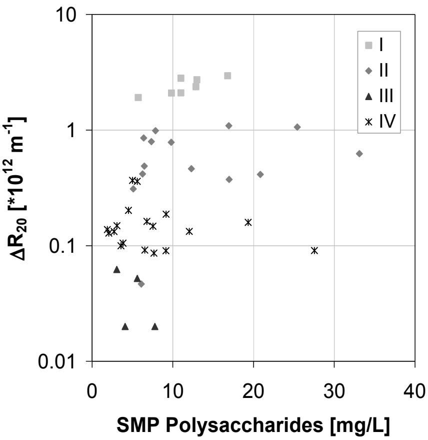


Figure 7.14: Filterability with SMP polysaccharide concentrations

7.6.6 Soluble Microbial Products retention

As illustrated in Figure 7.13 and Figure 7.14 the protein and polysaccharide concentrations in the free water do not show a clear relation with the activated sludge filterability. However, when the retention properties are considered proteins and polysaccharides do show different behaviour. Figure 7.15 and Figure 7.16 represent the SMP concentrations in the (DFCm) permeate versus the ones in the free water.

The results show that the amount of proteins that pass the membrane depends on the concentration in the free water. On average 60% of the proteins present in the free water breaks through the membrane. This indicates that the proteins in the free water have a size that is in the same range as the pores of the membrane. Although the protein concentrations could not be linked to filterability, theoretically the retention behaviour makes the proteins suspicious to being involved in irreversible fouling mechanisms like pore blocking.

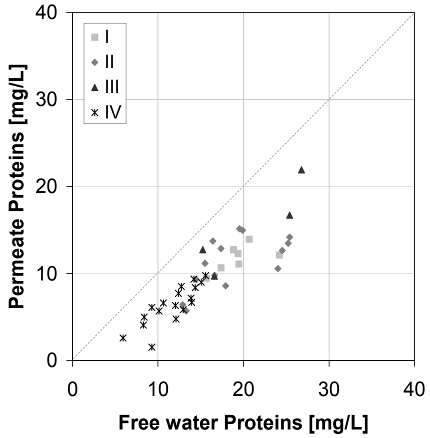


Figure 7.15: SMP protein concentrations: permeate versus free water

The polysaccharides show different breakthrough behaviour than the proteins. The polysaccharide concentrations in the (DFCm) permeate are more or less constant and relatively low (2.0 mg/L on average). The breakthrough appears to be independent of the concentrations in the free water.

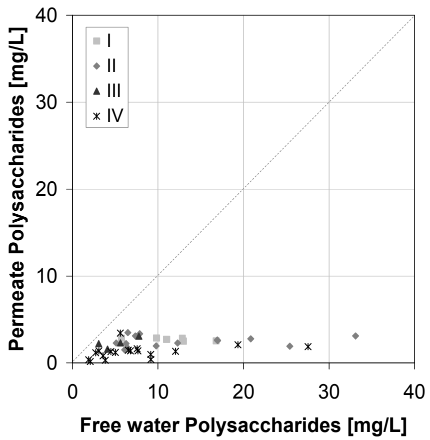


Figure 7.16: SMP polysaccharide concentrations: permeate versus free water

Similar retention behaviour of SMP proteins and polysaccharides was encountered for activated sludge sampled from several Dutch pilot MBR plants (Geilvoet, 2004).

7.6.7 Sub-micron particle volume

Particle size distribution (PSD) measurements were performed to investigate the influence of the colloidal fraction in the activated sludge free water on filterability. Unfortunately the particle counter was not yet operational in period I and II when the activated sludge still showed poor filterability. As a result only PSD data are available for samples with moderate to good filterability (ΔR_{20} < 0.5). Figure 7.17 represents the ΔR_{20} values with the sub-micron particle volume (defined by the total volume of particles with a diameter in the range between 0.4 and 1.0 µm). The lack of variation in the ΔR_{20} values makes it impossible to relate filterability to the colloidal particle volume in the free water. The PSD results mentioned here will be discussed again in chapter 9.4.4, together with the data obtained from stress experiments (chapter 8).

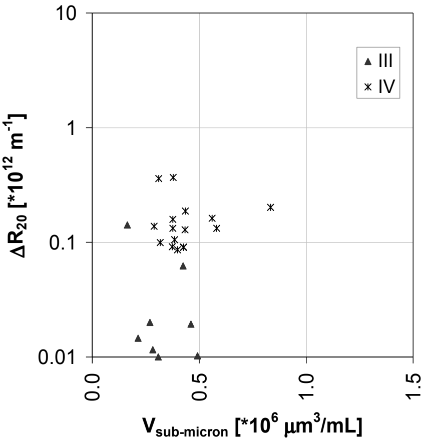


Figure 7.17: Filterability with sub-micron particle volume

7.7 Summary and conclusions

Activated sludge samples were collected on a weekly basis from the hybrid full-scale MBR in Heenvliet for a period of approximately one year (February 2006 until March 2007). The samples were analysed for their filterability and several activated sludge characteristics (mixed liquor suspended solids concentrations, temperature, sludge volume index, viscosity, soluble microbial products concentrations and sub-micron particle volume in the free water). In addition the filterability of the samples was analysed in relation to the development of the permeability throughout the measuring period.

7.7.1 Filterability

Based on the DFCm results a distinction could me made between four periods throughout the total measuring period. In the first period of approximately one week, just prior to the start-up of the MBR plant the filterability was extremely poor (ΔR_{20} >> 1). The poor filterability could more than likely be attributed to the unstable biological conditions in this period. The biomass concentration was increased from 4 to 10 g/L through biological growth and the addition of activated sludge from another nearby wastewater treatment plant. From period II, when stable MLSS concentrations were reached, the filterability showed an improving trend in the subsequent four months. In period III, during the summer months, the filterability was very good (ΔR_{20} < 0.05). From October the filterability showed a slight deterioration compared to the summer period, but it could still be qualified as reasonably good ($\Delta R_{20} \approx 0.15$).

The DFCm results show a consistent trend of the permeability throughout the measuring period. This indicates that the measurement frequency of one week was adequate for forming a reliable indication about the development of the filterability. Apart from the start-up phase the filterability throughout the measuring period can be qualified as good. A possible explanation for the good filterability is the thorough biological treatment before the activated sludge reaches the membranes. Due to the in-series operation of the wwtp the activated sludge undergoes a double biological treatment (in the carrousel of the conventional plant and in the MBR). This intensive biological process can be considered beneficial for the degradation of possible foulants and the flocculation of the activated sludge. A good flocculation process can be considered to lead to a reduction of the amount of fine material in the free water.

7.7.2 Activated sludge properties

As discussed in chapter 3.9 fouling in MBR is considered to be caused by fine particles in the free water of the activate sludge. In this sense the MLSS concentration can be considered an indirect parameter. In the start-up period of the MBR a clear relation was demonstrated between the MLSS concentration and the filterability. Concerning the unstable conditions in this period the MLSS concentration was an indication of the fine material in the free water. Contrary to the start-up phase in the remaining period the filterability was better at higher MLSS concentrations. An explanation for the better filterability at higher MLSS concentrations could be that at higher MLSS concentration fine material in the free water that can cause fouling are more easily absorbed into the activated sludge flocs.

On the other hand the relation between MLSS concentrations and filterability can also be a coincidence, since the highest MLSS concentrations were by chance accompanied by the highest activated sludge temperatures. The temperature is also a parameter that can be expected to indirectly affect the filterability. At higher temperatures the activity of the biomass increases. It can thus be expected that high temperatures enhance the degradations of substances that could otherwise reach the membrane and also that it improves the structure of the activated sludge with respect to filterability.

The SVI can also be considered a parameter indirectly related to fouling. During the start-up period poor filterability was accompanied by high SVI values. On the other hand the remaining measurements demonstrated that a low SVI is accompanied by varying filterability qualities, so the SVI itself is not a strong indicator for the filterability.

Two activated sludge properties were analysed that can be considered directly related to the activated sludge filterability: the concentration of soluble microbial products and the sub-micron particle volume (defined as the volume of particles with a diameter between 0.4 and 1.0 μm) in the free water. Despite being considered to have a significant impact on fouling, in this research no relation between SMP concentrations in the free water and filterability was demonstrated. Also the concentration of SMP that were retained by the membrane showed no relation with the filterability. The relation between filterability and SMP concentrations is discussed more in detail in chapter 9, when also the SMP analyses of the remaining measurement campaigns are incorporated.
Due to technical problems with the particle counter it could only be put in operation in the second half of the measurement campaign. In this period the filterability quality was stable and good; on this basis the relation between filterability and the sub-micron particle volume could not be verified.

7.7.3 Filterability and permeability

The high number and frequency of DFCm experiments performed at MBR Heenvliet form a good basis for comparing the development of the filterability with the development of the permeability in the full-scale plant. The DFCm measurements do not correspond with the development of the permeability. For the most part of the measurement campaign the filterability was good, whereas the permeability showed a continuous decreasing trend (with an unsatisfactory high rate). However, inspection of the membranes after the DFCm measurement campaign ended revealed that the membrane modules were subject to severe clogging problems. As a result of the clogging the effective membrane surface decreased with at least 25% and the flux in the membrane surface that still was available for permeation increased correspondingly. As discussed in chapter 3.6 and 3.10 an irregular distribution of the flux over the total membrane surface severely increases the fouling rate, irrespective of the activated sludge filterability properties.

Clogging and fouling could not be distinguished on the basis of the permeability development. The DFCm however demonstrated that the activated sludge had good filterability (and thus a low potential to cause fouling).

7.7.4 Conclusions

The main conclusions of the measurement campaign at the MBR of Heenvliet are summarised below:

- The relatively high number (58) and frequency (weekly) of DFCm measurements provided a consistent and reliable indication of the activated sludge filterability development throughout the total measuring period.
- DFCm experiments with different fluxes confirm the hypothesis that activated sludge filterability is independent of the applied flux (an activated sludge sample with poor filterability at a high flux also has poor filterability at a low flux and vice versa).
- The good filtration properties of the activated sludge that were generally measured throughout the measuring period can probably be attributed to the low loading rate and extensive biological treatment (both carrousel and MBR) before the activated sludge reaches the membrane modules.
- The visual observations strongly indicate that the decreasing trend of the permeability in the full-scale plant could not be attributed to poor activated sludge filterability, but to clogging problems. This implies that not the activated sludge filterability but the operational circumstances (i.e. turbulence creation) in the membrane modules were the limiting factor in the filtration process.
- The activated sludge filterability showed a clear relation with its temperature. The influence of the temperature is indirect and not crucial, but it can be expected that in case of stable and favourable operational conditions an increase of the temperature positively affects the activity of the biomass (mineralisation, flocculation) an thereby improves the filterability properties.
- The activated sludge filterability showed no relation with the soluble microbial products (SMP) concentrations in the free water or the concentrations retained by the membrane.

8 Stress experiments

8.1 Introduction

In this chapter several experiments are discussed in which the filterability and properties of activated sludge samples collected from MBR Heenvliet are manipulated in lab-scale circumstances. Imposing these so called stress conditions is favourable for effecting differences in filterability and the identification of activated sludge properties that are related to its filterability.
In section 8.2 the approach for the execution of the stress experiments is addressed. Subsequently three forms of stress are discussed: a prolonged low dissolved oxygen concentration (section 8.3), high mechanic stress circumstances (section 8.4) and a sudden decline of the activated sludge temperature (section 8.5). All imposed stress conditions are analysed on the basis of DFCm experiments and the analysis of several activated sludge properties. In section 8.6 the findings of the three stress experiments are summarised and discussed and the main conclusions of the stress experiments are formulated.

8.2 Approach

An important precondition to investigate the relationship between the filterability and the properties of MBR activated sludge is that variations in both characteristics do occur. A disadvantage of the measuring campaigns as performed in Varsseveld (chapter 6) and Heenvliet (chapter 7) is that these variations are not self-evident. This especially turned out to be the case for the measuring campaign at MBR Heenvliet where stable and good filtration properties were encountered over a long period (see Figure 7.5). In addition it can be mentioned that the variations in the activated sludge properties that do occur in a full-scale plant are the result of a combination between a multitude of operational parameters and influent characteristics that can impossibly be all surveyed.

A research approach to avoid the above mentioned problems is to manipulate the properties of activated sludge by imposing so called *stress* conditions upon it. In the context of this research stress can be described as any circumstance that adversely affects the filtration properties of the activated sludge.

Stress circumstances can be associated to either the process operation or to the properties of the incoming influent. Operational stress can have a biological or a mechanical background. Biological stress can be the result of any disruption of the activated sludge biological treatment process. Examples that can be mentioned are failing aerators, stirrers or recirculation pumps.

Mechanical stress can be the result of inappropriate recirculation pumps or too intensive coarse bubble aeration in the membrane area (Judd, 2006).

Examples of stress circumstances related to the influent properties are toxic shocks (chemicals, salts), a momentaneous extreme high organic loading rate or a sudden change in temperature (melt water). Evidently the properties of the incoming influent can not or hardly be influenced, both in terms of quality and quantity. Nonetheless a better understanding of these kinds of stress occasion is desirable, as it allows for a (temporary) adjustment of the process operation to prevent or minimise fouling problems.

On the basis of activated sludge sampled from MBR Heenvliet the following potential stress circumstances were investigated:

- Prolonged low dissolved oxygen concentration (biological stress)
- High shear stress circumstances (mechanic stress)
- Sudden temperature change (stress related to influent properties)

Each of these stress conditions were investigated in relation to reference samples that were aerated continuously. Evidently these stress occasions could not be created in the full-scale MBR plant. Therefore activated sludge samples were collected from the aerated tank and the stress conditions were simulated on a lab-scale basis. As discussed in chapter 1.2 a disadvantage of this lab-scale approach is that the created circumstances are not representative for the circumstances in practice in the considered full-scale MBR. This drawback can not be fully overcome. As soon as the activated sludge is samples it is taken away from its “familiar” biological process, which itself can be already considered as a stress circumstance. On the other hand the advantage of this lab-scale research approach is that the changes in filterability and activated sludge properties (compared to the reference sample) can be attributed exclusively to the imposed stress conditions.

8.3 Dissolved oxygen concentration stress

Aeration is a crucial parameter in the operation of an MBR plant as it is required for the biological treatment and membrane scouring. The use of aeration for membrane scouring falls out of the scope of this research, but the influence of oxygen supply on the microbial properties of activated sludge can be assessed with the DFCm. Aeration directly affects the dissolved oxygen (DO) concentration in the activated sludge. Lee et al. (2005) report the DO concentration to have an impact on fouling through the floc size distribution and the SMP concentrations. In addition Kang et al. (2003) link high DO levels with a better filterability due to larger particles resulting in a lower specific resistance of the cake layer.

In this section the sensitivity of activated sludge filterability and properties with respect to a cease of the oxygen supply are investigated. Not only is the effect of ceasing aeration studied, but also the behaviour when the aeration is resumed after a period of several days.

8.3.1 Approach and analyses

Three activated sludge samples (40 litres each) were collected from the aerated tank of MBR Heenvliet and transported to the Sanitary Engineering laboratory of Delft University. The following circumstances were imposed upon each of the samples:

- Sample #1: continuous aeration (72 hours)
- Sample #2: continuous stirring (72 hours)
- Sample #3: continuous stirring (92 hours), followed by continuous aeration

The following activated sludge properties were analysed regularly:

- Filterability, according to the standard DFCm measuring protocol
- MLSS concentration, [g/L]
- Temperature, [ºC]
- SMP proteins and polysaccharides, [mg/L]
- Sub-micron particle volume, [μm^3/mL]
- Dissolved oxygen concentration, [mg/L]

After each DFCm experiment the collected permeate was returned to the activated sludge sample to maintain a constant MLSS concentration. The experiments as described above were conducted several times. Since the results were comparable for all cases only one dataset is discussed here.

8.3.2 General activated sludge properties

Temperature

At the moment of sampling the activated sludge temperature was 18 ºC. This temperature was approximately similar to the ambient temperature in the lab. Since the aeration and stirring did not cause temperature alteration the activated sludge temperature was maintained at a constant level throughout the measuring period. The activated sludge temperature can thus be considered of no influence on the experimental results.

Mixed liquor suspended solids concentration

The MLSS concentration of the activated sludge was approximately 11 g/L at the moment of sampling. Throughout the measuring period the MLSS decreased slightly for all samples, but the differences were small (± 0.2 g/L). The influence of the MLSS concentration on the experimental results is assumed negligible.

Dissolved oxygen concentration

After arrival in the lab the dissolved oxygen concentration in the activated sludge was about 0.5 mg/L. As a result of aeration the dissolved oxygen concentration increased rapidly to values above 4 mg/L. In the stirred sample the oxygen concentration dropped below 0.1 mg/L in a period of about 2 hours and remained below this value.

8.3.3 DFCm results

The filterability of all three samples was characterised directly after arrival in the lab. Sample #1 was aerated for 10 minutes prior to the first DFCm experiment. The moment when each of the samples was filtrated for the first time was set as time point t = 0 (hours). All three samples showed similar and very good filterability with ΔR_{20} values of approximately 0.05 (*10^{12} m^{-1}). The comparable ΔR_{20} values for sample #1 and #2 indicate that the residence time without aeration during transport from the MBR plant to the lab (about 1 hour) had no significant effect on the filterability. The development of the ΔR_{20} values is represented in Figure 8.1.

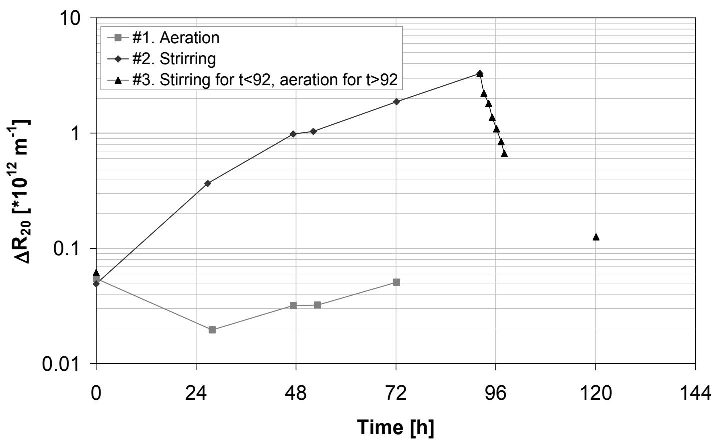


Figure 8.1: Filterability development for all three samples

Sample #1

The filterability of sample #1 remains constant and very good throughout the measuring period of three days (ΔR_{20} < 0.05). The good filterability implies that the value of the product of the specific reference cake resistance and the concentration of material accumulating in the cake layer is very low ($\alpha_R \cdot c_i$ < 0.003). The differences in the obtained ΔR_{20} values can be attributed to the lower accuracy in this measuring range rather than to changes in the filterability. As a consequence of the low ΔR_{20} values the compression coefficient could not be determined with significant reliability. A possible further improvement of the filterability as a result of the continuous aeration can not be confirmed since the starting filterability of the blank sample is already good.

Sample #2

The continuous anoxic circumstances imposed upon sample #2 clearly result in a deterioration of its filterability. The results of the DFCm experiments are summarised in Table 8.1. The deterioration of the filterability takes place gradually: after 27 hours ΔR_{20} increases to 0.37, which implies that the filterability can be qualified as moderate. Nonetheless the increasing trend of ΔR_{20} perseveres and on the long run the filterability becomes very poor. Remarkably the increase of ΔR_{20} is predominantly the result of an increase in the compressibility factor. This implies that the high resistance can be attributed to the high specific resistance of the cake layer and not to the amount of mass accumulating in the cake layer.

Table 8.1: DFCm results sample 2 (O_2 < 0.1 mg/L)

	$\alpha_R \cdot c_i$ ($\cdot 10^{-3}$)	s	ΔR_{20} ($\cdot 10^{12}$ m^{-1})
t = 0	2.5	-	0.05
t = 27 h	22.1	0.18	0.37
t = 47 h	49.5	0.40	0.98
t = 52 h	51.1	0.40	1.04
t = 72 h	71.2	0.42	1.88

Sample #3

After the first reference measurement (t = 0) sample #3 was filtrated for the second time after 92 hours of low dissolved oxygen concentration circumstances. Filterability is very poor with a ΔR_{20} value of 3.3. Since the imposed circumstances until this moment are similar to sample #2 it is not surprising that this value corresponds very well with the ΔR_{20} trend of this sample (indicated with the dotted line in Figure 8.2). After 92 hours of anoxic circumstances the compressibility factor is about 0.4. With respect to the similar numbers that were found for sample #2 (see Table 8.2) it seems that the compressibility factor stabilises at this value.

Table 8.2: DFCm results sample 3 (O_2 > 4 mg/L for t > 92 h)

	$\alpha_R \cdot c_i$ ($\cdot 10^{-3}$)	s	ΔR_{20} ($\cdot 10^{12}$ m^{-1})
t = 0	3.1	-	0.05
t = 92 h	101.2	0.41	3.30
t = 93 h	85.3	0.33	2.22
t = 94 h	75.8	0.30	1.81
t = 95 h	63.0	0.26	1.37
t = 96 h	53.3	0.24	1.09
t = 97 h	43.9	0.24	0.84
t = 98 h	36.0	0.20	0.67
t = 120 h	6.3	-	0.13

As soon as the aeration commences at t = 92 h the dissolved oxygen concentration instantly increases to values higher than 4 mg/L. In addition also the filterability improves rapidly. In a period of about six hours the ΔR_{20} value drops from 3.3 to 0.7. After a full day of aeration ΔR_{20} recovers to a value of 0.1 and the activated sludge filterability can thus be qualified as good again. The improvement of the filterability can be attributed to a decrease of both $\alpha_R \cdot c_i$ and s.

Remarkably the rate of filterability deterioration as a result of low dissolved oxygen concentration circumstances is totally different from the rate of improvement after the start of the aeration. The average increase of ΔR_{20} in the four days without aeration is $0.035 \cdot 10^{12}$ m^{-1}/hour, while the decrease of ΔR_{20} in the first few hours after resumption of the aeration is 0.44 $\cdot 10^{12}$ m^{-1}/hour, approximately a factor twelve higher. Apparently after three days without oxygen supply the activated sludge still has a strong potential to recover from these stress circumstances.

8.3.4 Soluble microbial products

Sample #1

As mentioned previously the filterability of sample #1 is constant and excellent throughout the measuring period. Also the SMP concentrations are quite stable. The protein concentrations vary between 11.6 and 13.7 mg/L and the polysaccharide concentrations are in the range between 3.5 to 8.0 mg/L (not presented graphically). As will be discussed later in this section these values and variations are relatively low.

Sample #2

As discussed in section 8.3.3 the continuous low dissolved oxygen concentration imposed upon sample #2 results in a gradual deterioration of the filterability. Figure 8.2 represents this development, together with the obtained SMP concentrations in this period. The polysaccharide concentration clearly shows an increasing trend, from 3.5 to 18.1 mg/L in the three day period.

The increase of the protein concentrations is less manifest. In the first 52 hours only a modest (but steady) increase from 15.0 to 18.7 is measured. In the last measurement (t = 72 h) the protein concentrations shows a decrease again, but the reliability of this measurement is questionable. In general the increase of the SMP concentrations indicate that as a result of anoxic circumstances bound EPS is released from the activated sludge flocs into the free water.

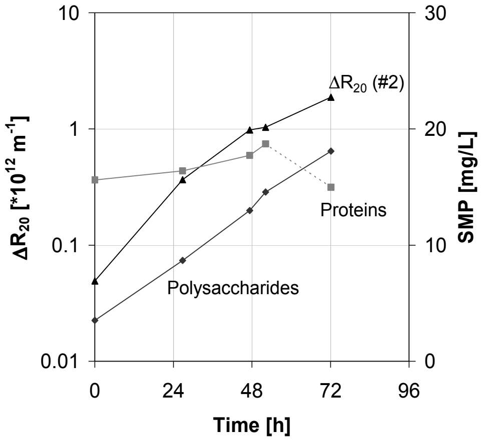


Figure 8.2: Filterability and SMP concentration development for sample #2 (low dissolved oxygen concentrations)

Sample #3

After 92 hours of low dissolved oxygen circumstances the ΔR_{20} of sample #3 increases to 3.5. As soon as the aeration was started the dissolved oxygen concentration instantly increased to values above 4 mg/L and the filterability recovered rapidly. Figure 8.3 represents the development of ΔR_{20} and the SMP concentrations after the aeration commenced (note the time scale difference between Figure 8.2 and Figure 8.3). The rapid improvement of the filterability is accompanied by a just as rapid decrease of the SMP concentrations; within a period of less than 6 hours the protein concentrations drop from 27 to 16 mg/L and polysaccharide concentrations from 18 to 10 mg/L. Apparently the SMP released from the activated sludge flocs into the free water due to the low dissolved oxygen concentrations are encapsulated (again) by the activated sludge flocs.

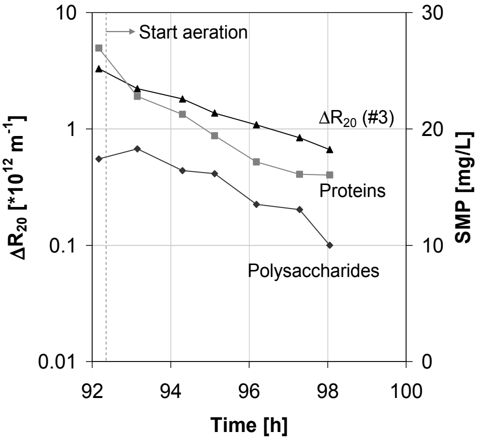


Figure 8.3: Filterability and SMP concentration development for sample #3
(92 hours stirring followed by aeration)

Filterability and SMP concentrations

Figure 8.4 shows a compilation of the SMP concentrations and the obtained ΔR_{20} values for all three samples. As mentioned previously the SMP concentrations in sample #1 are relatively low. The overall results demonstrate a relation between the SMP concentration and the filterability; increasing SMP concentrations are accompanied by higher ΔR_{20} values.

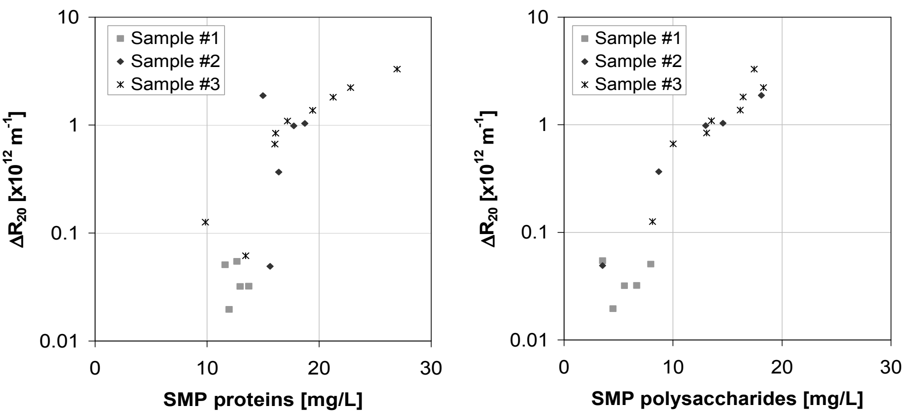


Figure 8.4: Filterability with SMP concentrations for all three samples

8.3.5 Sub-micron particle volume

As with the SMP concentration also the volume of colloidal particles in the free water can be expected to increase as a result of the stress conditions imposed upon the activated sludge sample. As discussed in chapter 4.6.5 the results of the particle size distribution measurement in the free water phase of the activated sludge are compared on the basis of the total volume of particles in the size range between 0.4 and 1.0 μm. Although this size range does not encompass the whole colloidal range the value of $V_{sub\text{-}micron}$ is considered to form a reflection of the colloidal particle volume.

Sample #1

As discussed in section 8.3.3 the filterability of the continuously aerated sample is excellent and remains constant. In addition also the sub-micron particle volume is very constant. The six particle counting experiments are in the range between 0.17 and $0.44 \cdot 10^6$ μm^3/mL. As will be discussed later in this section these values are very low.

Sample #2

The development of ΔR_{20} and the sub-micron particle volume of sample #2 are graphically presented in Figure 8.5. Initially the development of the filterability corresponds with the volume of sub-micron particles in the free water. As a result of the continuous anoxic circumstances the activated sludge seems to release colloidal particles into the free water. After 48 hours the sub-micron particle volume seems to stabilise at $1.5 \cdot 10^6$ μm^3/mL while the filterability continues to deteriorate. On the other hand, this assumed stabilisation is only based on one measurement (t = 72 h) and should thus be considered with some reserve.

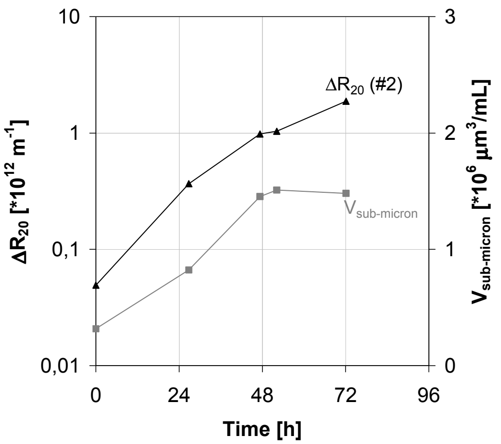


Figure 8.5: Filterability and $V_{sub\text{-}micron}$ development for sample #2

Sample #3

Figure 8.6 represents the ΔR_{20} and $V_{sub\text{-}micron}$ development for sample #3. The rate of improvement of the filterability is accompanied by a comparable rate of decrease in sub-micron particle volume. In a period of less than six hours the sub-micron particle volume decreases from 1.7 to $0.7 \cdot 10^6$ μg^3/mL, approximately a factor of 2.5. Apparently particles that were released in the free water are encapsulated again by the activated sludge matrix under the influence of the aerated circumstances.

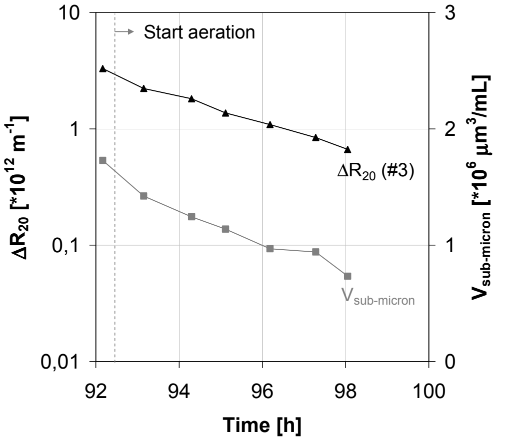


Figure 8.6: Filterability and $V_{sub\text{-}micron}$ development for sample #3

Filterability and sub-micron particle volume

Plotting the ΔR_{20} values against the sub-micron particle volumes for all three samples results in the dataset as represented in Figure 8.7. As mentioned earlier the sub-micron particle volume in sample #1 is very low compared to the other two samples. The results clearly indicate a correlation between the filterability and the sub-micron particle volume.

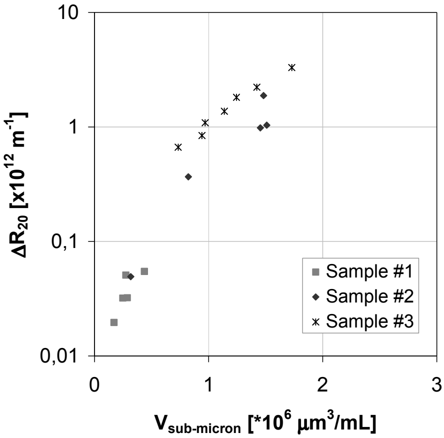


Figure 8.7: Filterability with sub-micron particle volume (all 3 samples)

8.3.6 Summary and concluding remarks

Stress circumstances were imposed upon an MBR activated sludge sample (MLSS = 11 g/L, T = 18 ºC) by means of ceasing the oxygen supply for several days. Although an aeration cease of several days is not likely to occur in practice in a full-scale MBR plant the experiments have been useful for understanding the influence of the DO concentration on activated sludge filterability. The DFCm experiments clearly demonstrate a deterioration of the activated sludge filterability in case of long-term low DO concentrations. In a period of almost four days without aeration the ΔR_{20} value increases from 0.05 to $3.3 \cdot 10^{12}$ m^{-1}. In a continuously aerated reference sample the filterability remained stable and very good, proving that the deterioration of filterability could indeed be attributed to the low dissolved oxygen concentration.

Besides a deterioration of the filterability both the SMP concentrations and the colloidal particle volume in the free water increase during the anoxic circumstances. This indicates that during these stress conditions substances are released from the activated sludge flocs into the free water. This leads to the conclusion that as a consequence of the stress conditions the activated sludge is deflocculating.

After four days without oxygen supply the aeration was resumed. The activated sludge shows a strong ability to recover from the stress circumstances. The filterability improves rapidly, with a rate of about 12 times higher compared to the deterioration rate for the anoxic circumstances. The dynamic improvement of the filterability is accompanied by a just as dynamic decrease of the SMP concentrations and the sub-micron particle volume. The experimental results indicate that the activated sludge (re)flocculates again in the newly imposed aerated circumstances. The experiments have demonstrated that the mechanisms of flocculation and deflocculation play an important role in the filtration process.

8.4 Mechanical stress

It can be expected that the filterability of activated sludge is adversely affected when it is experiencing high mechanical shear stress. In practice these circumstances can occur in an MBR plant when inappropriate (recirculation) pumps are installed or when the coarse bubble aeration in the membrane zone is too intensive (Judd, 2006). Several researchers report a relation between high shear stress and fouling problems due to the break-up of the sludge flocs and the accompanying release of SMP into the free water (source source source). In the experiments described in this section high shear stress circumstances were simulated by means of short term recirculation of an activated sludge sample with a centrifugal pump. In the first place the supposed deterioration of the filterability is verified in combination with several activated sludge properties. Subsequently the activated sludge is submitted to tranquil circumstances (continuous aeration) to assess its ability to recover from the imposed stress circumstances.

8.4.1 Approach and analyses

Two activated sludge samples (40 litres) were collected from the aerated tank of MBR Heenvliet and transported to the laboratory of Delft University. After arrival the samples were aerated and a blank measurement was performed to determine the starting filterability and -properties. The reference sample was continuously aerated. The other sample was recirculated with a centrifugal pump (DAB single impeller, type K30/70) for a period of 5 minutes. Unfortunately the exact shear stress induced by the pump on the activated sludge could not be calculated. The capacity of the pump at minimal head loss is 8 m^3/h. This implies that in a period of 5 minutes each activated sludge particle passes the pump 16 times on average. This high recirculation can not be considered representative in comparison with a full-scale plant, but nonetheless this stress experiment can be considered a reflection of the effect of high shear stress on activated sludge

After the imposed high shear circumstances the sample was aerated to investigate the recovery from the stress circumstances. Approximately every 45 minutes the following characteristics were monitored or analysed:

- Filterability (ΔR_{20}), according to the standard DFCm measuring protocol (J=80 L/m^2·h)
- Temperature, [ºC]
- SMP proteins and polysaccharides in the free water, [mg/L]
- Sub-micron particle volume, [μm^3/mL]
- Dissolved oxygen concentration, [mg/L]

After each DFCm experiment the collected permeate was returned to the activated sludge sample to maintain a constant MLSS concentration. The experiments as described here were conducted several times. The results for all trials were similar; therefore only one set of experiments is discussed in this section.

8.4.2 General activated sludge properties

Reference sample

Both the filterability and all the measured characteristics of the reference sample did not show significant changes due to the continuous aeration in the measuring period of 5 hours. It can thus be assumed that the alternation of the other sample can be exclusively attributed to the imposed high shear circumstances.

Temperature

At the time of arrival at the laboratory the activated sludge temperature was 17.2 ºC, which was approximately similar to the ambient temperature. As a result of the energy input by the centrifugal pump the sample temperature increased to 19.1 ºC. Subsequently the temperature gradually decreased to 17.5 ºC in a four hour period. The influence of the temperature on the experimental results is considered negligible compared to the imposed high shear circumstances.

Mixed liquor suspended solids concentration

The MLSS concentration was determined at t = 0 and was 9.3 g/L. The MLSS concentration is assumed to remain constant and to have no influence on the experimental results.

Dissolved oxygen concentration

As a consequence of the continuous aeration the dissolved oxygen concentration was always higher than 4 mg/L and is assumed to have to influence on the results.

8.4.3 Filtration Characterisation

The results of the DFCm experiments are represented in Table 8.3 and Figure 8.8. The filterability of the blank sample at t = 0 was good, with a ΔR_{20} value of 0.10 ($\cdot 10^{12}$ m^{-1}). The recirculation with the centrifugal pump has a devastating effect on the filterability; ΔR_{20} increases to 3.9. From this moment the activated sludge is aerated and the filterability recovers rapidly. Within two hours of aeration ΔR_{20} decreases from 3.9 to 0.6. An analysis of the DFCm curves indicates that the decrease of ΔR_{20} can be attributed to a decrease in both the coefficients $\alpha_R \cdot c_i$ and s.

Table 8.3: DFCm results shear stress experiments

Sample #	t (h)	$\alpha_R \cdot c_i$ ($\cdot 10^{-3}$)	s	ΔR_{20} ($\cdot 10^{12}$ m^{-1})
1 (blank)	0:00	4.8	-	0.10
2	0:36	117.2	0.41	3.91
3	1:02	91.4	0.33	2.59
4	1:41	62.9	0.30	1.42
5	2:36	33.5	0.26	0.57
6	3:50	31.2	0.24	0.52
7	4:35	22.1	0.24	0.38

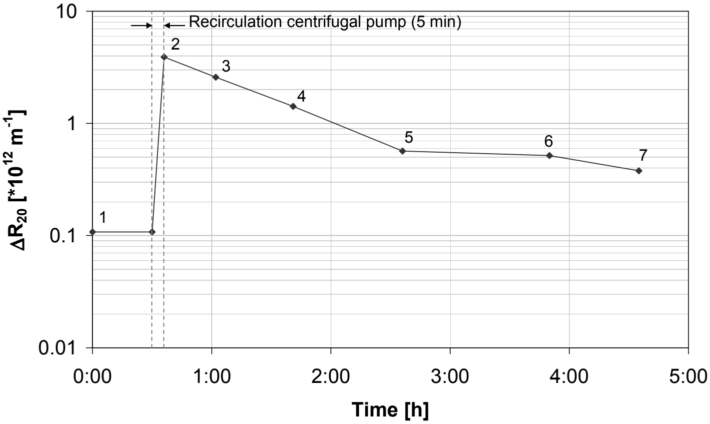


Figure 8.8: Filterability development in time after short-term high shear stress circumstances

8.4.4 Soluble microbial products

The high shear forces imposed upon the activated sludge sample with the centrifugal pump result in destruction of the flocs. It can thus be expected that bound EPS is released into the free water and results in an increase of the SMP concentration. This is confirmed by the SMP measurements, see Figure 8.9. Compared to the strong deterioration of the filterability the increase of the SMP concentrations is relatively modest, both protein- (from 11 to 19 mg/L) and polysaccharide concentrations (from 5 to 9 mg/L) almost double.

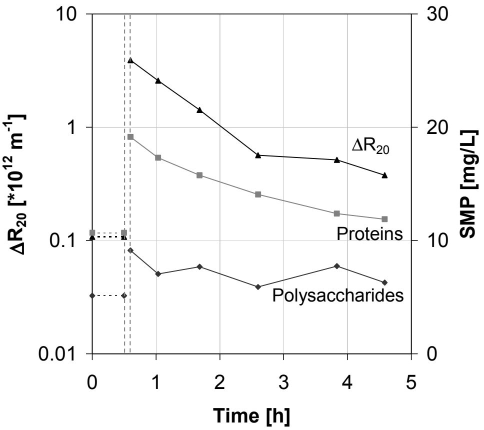


Figure 8.9: Filterability and SMP concentration development

As a consequence of the subsequent aerated circumstances the SMP concentrations decrease again. For proteins this decrease is more consistent than for the polysaccharides. All the polysaccharide measurements are in the range between 5 and 10 mg/L. These modest variations do not correspond with the strong filterability differences that were measured.

8.4.5 Sub-micron particle volume

The supposed destruction of the activated sludge flocs due to the high shear circumstances is clearly supported by the particle size measurements. In the free water of the starting sample the volume of particles with a diameter in the range between 0.4 and 1.0 μm is $0.5 \cdot 10^6$ μm^3/mL. After the recirculation with the centrifugal pump the volume of sub-micron particles increases with approximately a factor four to $2.1 \cdot 10^6$ μm^3/mL. As a consequence of the subsequent tranquil aerated circumstances the sub-micron particle volume decreases again, see Figure 8.10. This indicates that the particles in the free water are encapsulated again in the activated sludge flocs. Within four hours of aeration the sub-micron particle volume is reduced to its approximate original value again ($0.6 \cdot 10^6$ μm^3/mL). The activated sludge thus shows the ability to (re)flocculate.

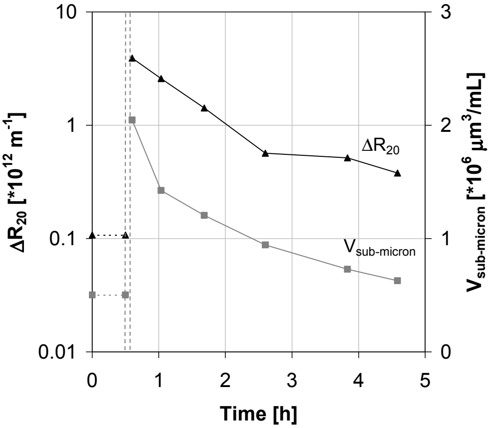


Figure 8.10: Filterability and sub-micron particle volume development

8.4.6 Summary and concluding remarks

Activated sludge samples collected from the aerated tank of MBR Heenvliet were submitted to a short period of high mechanical stress with a centrifugal pump. As can be expected the filterability of the activated sludge is adversely affected by the high shear stress. The strong deterioration of the filterability is accompanied by an increase of both the SMP concentrations and the sub-micron particle volume in the activated sludge free water. Both results show that the structure of the activated sludge is damaged and that the flocs are broken up into smaller substances that end up in the free water.
After the high shear stress the activated sludge was submitted to tranquil operational conditions by means of continuous aeration. As a consequence the filterability improves rapidly, accompanied by an analogous decrease of the SMP concentrations and the sub-micron particle volume in the free water.

The straightforward high shear stress experiments have demonstrated that the mechanisms of deflocculation and flocculation play an important role with respect to activated sludge filterability. On the basis of this experiment both the SMP concentrations and the sub-micron particle volume in the free water seem to provide information about the flocculation status of the activated sludge.

8.5 Temperature shock

Temperature is an important parameter in MBR filtration as it affects the process in two ways: through the water viscosity (Darcy's law, see equation 2-1.) and through the microbial activity of the biomass (see chapter 3.9.4). Especially the interaction between temperature, biological activity and fouling is complex.
The optimal temperatures for bacterial activity are in the range between 25 and 35 ºC. It can thus be expected that activated sludge experiences a sudden temperature drop as a stress situation. In the experiments described in this section a sudden temperature change was simulated by using a thermostat. Subsequently the influence of this change on the filterability and the activated sludge properties was investigated.

An extensive set of experiments has been conducted to verify the influence of temperature on the filterability (discussed in Benschop 2008). The results are not as consistent and reproducible as the stress experiments discussed in sections 8.3 and 8.4. Therefore four datasets are discussed.

8.5.1 Approach and analyses

For each of the four measurement series two activated sludge samples (40 litres each) were collected from the aerated tank of MBR Heenvliet. The samples were transported to the lab and aerated. In the first sample the temperature was lowered three steps (each ΔT step approximately 5 ºC) with a thermostat. Each ΔT step is reached in a period of approximately 30 minutes. Subsequently at each temperature the filterability and several activated sludge properties were analysed. The second sample served as a reference sample and was only aerated to assess the (possible) alternation of the activated sludge in time. Each set of the experiments was conducted within a time period of approximately 4 hours. The following activated sludge properties were analysed:

- Filterability, according to the standard DFCm measuring protocol
- MLSS concentration, [g/L]
- Temperature [ºC]
- SMP protein and polysaccharide concentrations in the free water and in the permeate, [mg/L]
- Sub-micron particle volume, [μm^3/mL]
- Dissolved oxygen concentration, [mg/L]
- Apparent activated sludge viscosity, [Pa·s]

8.5.2 General activated sludge properties

Mixed liquor suspended solids concentration

The MLSS concentration of the samples is between 9.0 and 11.6 g/L, with an average of 10.2 g/L. These modest variations in the MLSS concentration are assumed to be of negligible influence on the experimental results.

Dissolved oxygen concentration

The samples were aerated directly after arrival in the lab. As a consequence the dissolved oxygen concentration is always above 4 mg/L for all samples and can thus assumed to be of no influence on the results.

Activated sludge viscosity

Since the viscosity of water depends on the temperature it can be expected that it is an important factor in these temperature experiments. Nonetheless, rheological experiments demonstrated that the (apparent) viscosity change due to temperature differences is negligible compared to the influence of the MLSS concentration.

8.5.3 DFCm results

Figure 8.11 represents the DFCm results for four measurement series. The first experiment was conducted at the sludge temperature as it was at the moment of sampling (in between 12 and 15 ºC). Subsequently the temperature was lowered with the thermostat in three steps. The open markers in the figure represent the DFCm experiments with the reference activated sludge samples.

Two types of results were obtained, depending on the filterability of the starting sample. In case of a poor starting filterability (ΔR_{20} > 1.0) the effect of temperature on the filterability is minimal. This is the case for samples #3 and #4, with ΔR_{20} values of 1.1 and 2.0 ($\cdot 10^{12}$ m^{-1}) respectively for the starting sample. When the filterability of the starting sample is better a temperature effect is noticed. Samples #1 and #2 clearly show a decreasing filterability at lower temperatures.

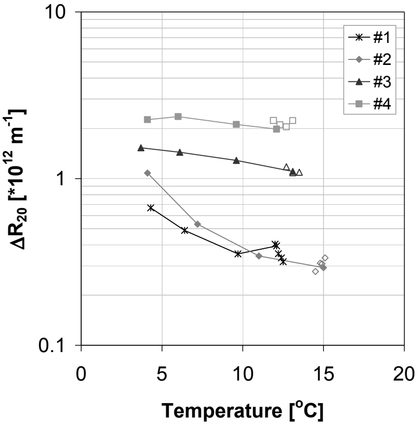


Figure 8.11: Filterability with temperature for 4 samples
(open markers represent the values for the reference sample)

The results also show that for the reference samples the temperature and the filterability are constant in time (not represented graphically). This indicates that the alternation of the filterability of the samples from which the temperature is lowered can be attributed exclusively to the temperature changes. The results for the two reference samples with a poor starting filterability are remarkable. It appears that the filterability is not improving despite to the continuous aerated circumstances (approximately 3 hours) that are imposed upon the activated sludge. This indicates that the filterability of an activated sludge sample with a poor filterability can not be improved by simply aerating it. This issue is discussed again in section 8.6, in relation to the dissolved oxygen concentration stress experiments as discussed in section 8.3.

8.5.4 Soluble microbial products

As a consequence of the stress circumstances experienced by the activated sludge due to the temperature alternation it can be expected that the biomass releases EPS into the free water as SMP. This is however not clearly confirmed by the experimental results.

Proteins

In all four experiments the temperature alternation appears to have no effect on the SMP protein concentrations (not represented graphically). This implies that for samples #1 and #2 at different protein concentrations variable ΔR_{20} values are obtained, as illustrated in Figure 8.12. For samples #3 and #4 both the filterability and the protein concentrations are constant. The conclusion on the basis of these experiments is that the deterioration of the filterability due to the induced temperature decrease can not be linked with the protein concentrations in the free water. On the other hand the overall results do indicate a relation between filterability and SMP proteins, since the concentrations in samples #3 and #4 are significantly higher compared to samples #1 and #2.

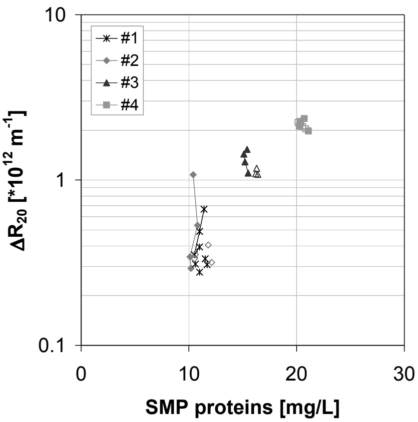


Figure 8.12: Filterability with SMP protein concentrations
(open markers represent the values for the reference sample)

Polysaccharides

The polysaccharide concentrations in the free water have values that are totally independent from the filterability. In Figure 8.13 the SMP polysaccharide concentrations are plotted against the obtained ΔR_{20} values for the four samples. The poor and constant filterability of sample #4 (both the reference and the alternated sample) is accompanied by polysaccharide concentrations varying between 13 and 28 mg/L. For sample #3 both the SMP polysaccharide concentrations and the filterability are constant, but the combination of relatively low polysaccharide concentrations (7.5 mg/L) and a poor filterability (ΔR_{20} > 1.0) does not correspond with the other samples. For samples #1 and #2 a relation between filterability and temperature was demonstrated (see Figure 8.12), but again this relation is not accompanied by analogous change of the polysaccharide concentrations in the free water. In addition to the inconsequent relation for each of the four separate samples also the overall results support the statement that activated sludge filterability can not be clearly linked with SMP polysaccharide concentrations.

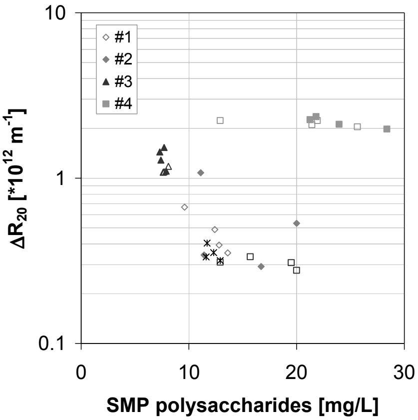


Figure 8.13: Filterability with SMP polysaccharide concentrations
(open markers represent the values for the reference sample)

SMP retention

In order to gain understanding of the retention by the DFCm membrane the SMP concentrations in the free water are compared with the ones in the permeate. The results are comparable with the retention behaviour as found during the measurement campaign in Heenvliet (see chapter 7.6.6). For polysaccharides the concentration in the permeate is very low and constant (2 mg/L), irrespective of the concentration in the free water.

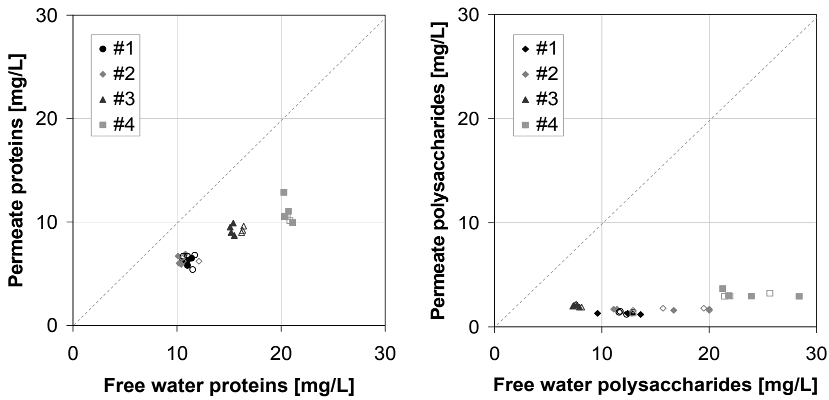


Figure 8.14: SMP concentrations permeate versus free water
(open markers represent the values for the reference sample)

The proteins show a different retention behaviour, with the concentration in the permeate being dependant upon the concentration in the free water. On average 55% percent of the proteins present in the free water pass the membrane. The retention behaviour of the proteins and polysaccharides is discussed more in detail in section 8.6.2 and chapter 9.4.3.

8.5.5 Sub-micron particle volume

Contrary to the SMP concentrations the sub-micron particle volume in the free water does show a clear link with the activated sludge filterability, see Figure 8.15.

In sample #1 and #2 the increase of ΔR_{20} at lower temperatures is accompanied by an analogous increase of the sub-micron particle volume. Sample #3 shows a modest variation of the sub-micron particle volume at more or less constant ΔR_{20} values, but when including the data of sample #4 and reviewing the overall results the relation between sub-micron particle volume and filterability is manifest. This relation was not demonstrated with respect to the SMP concentrations in the free water and thus seems to demonstrate that the sub-micron particle volume in the free water provides a better indication of the fouling potential than the SMP concentrations.

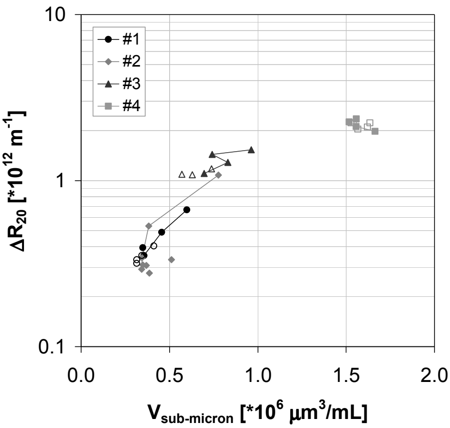


Figure 8.15: Filterability with sub-micron particle volume
(open markers represent the values for the reference sample)

As with the SMP concentrations also the sub-micron particle volume in the reference samples stays more or less constant. This again demonstrates that for the samples with a poor starting filterability (#3 and #4) the sludge properties (and filterability) could not be improved by simply aerating the sample for several hours.

8.5.6 Summary and concluding remarks

Stress circumstances were imposed upon several activated sludge samples (collected from MBR Heenvliet) by abruptly decreasing their temperature with a thermostat. Depending upon the filterability of the starting samples two types of results were obtained. In case of a relatively good starting filterability ($\Delta R_{20} \approx 0.2$) the temperature drop leads to a considerable decrease of the filterability. In case of a poor starting filterability ($\Delta R_{20} > 1$) the filterability remains more or less constant. These findings indicate that in case of poor starting filterability the contribution of the temperature stress becomes insignificant. A remarkable result with respect to the reference samples is that the filterability could not improved by aerating the sample for several hours. This conflicts with results obtained in other stress experiments (see section 8.3 and 8.4). This issue is discussed more in detail in section 8.6.

The imposed temperature shock appears to have no effect on the SMP protein concentrations, whereas the filterability does deteriorate (for the two samples with a good starting filterability, see Figure 8.12). The SMP polysaccharide concentrations do not show any relation at all with the filterability (see Figure 8.13). In contrast with the SMP concentrations the relation between filterability and the volume of sub-micron particles in the free water is evident. In the two samples with a poor starting filterability both the filterability and the sub-micron particle volume is already high and remains constant. But for the two samples that were affected by the temperature shock the deterioration of the filterability is accompanied by an increase of the sub-micron particle volume. Considering the overall results of the four experiments clearly demonstrates a poorer filterability with increasing sub-micron particle volume in the free water. This leads to the conclusion that the sub-micron particle volume in the free water seems a better indicator for the activated sludge filterability than the SMP concentration.

8.6 Summary and conclusions

Activated sludge samples were collected from the aerated tank of MBR Heenvliet. The samples were submitted to three different types of stress conditions that could also occur in a full-scale MBR plant: (1) prolonged low dissolved oxygen (DO) concentrations, (2) mechanic shear stress and (3) abrupt temperature decrease. Subsequently the alternation of the activated sludge filterability and properties were verified in relation to a continuously aerated reference sample.

8.6.1 Filterability

The DFCm results indicate that for the high mechanical stress and low dissolved oxygen concentration stress experiments the filterability of the activated sludge samples could be easily manipulated. For the temperature stress experiments the filterability deterioration was less manifest and consistent. Two explanations for this can be mentioned:

- For the DO stress and mechanical stress experiments the starting filterability was always good. This was (by chance) not the case for the samples collected for the temperature stress experiments. The results indicate that when the starting filterability is already poor the subsequent imposed stress circumstances do not cause significant additional deterioration of the filterability.
- The imposed stress upon the samples in the temperature experiments can be considered less extreme than the DO and mechanical stress experiments.

In the case of the DO and shear stress experiments the degree of stress imposed upon the activated sludge to force filterability deterioration can be considered extreme. After a full day of DO stress the filterability of the sludge could still be qualified as modest. Only after several days without oxygen supply the filterability became poor. These circumstances are not likely to occur in practice in a full-scale MBR plant. In the shear stress experiments filterability deterioration was only clearly demonstrated when the activated sludge was recirculated with a centrifugal pump several times in a short time. This can also not be considered representative for the situation in practice. The DO and mechanic stress occasions can be regarded as an exaggeration of stress circumstances as they may occur in practice and can thus not be considered fully representative. Nonetheless the experiments still provide information about the mechanisms that are associated to these stress occasions. It is unclear to what extent the “resilience” of activated sludge against quality deterioration could be attributed to its good starting filterability.

In addition to the resistance to quality deterioration the activated sludge showed a strong potential to recover from the stress as soon as mild circumstances were put into operation, in the form of continuous aeration. Filterability deterioration due to DO stress caused in a period of several days was counterbalanced in a period of only several hours (deterioration rate ΔR_{20} of $0.035 \cdot 10^{12}$ m^{-1}/h versus a recovery rate of $0.44 \cdot 10^{12}$ m^{-1}/h, difference of a factor 12). Also in the mechanic stress experiments the activated sludge recovers rapidly from the stress circumstances, considering the average ΔR_{20} decline of $0.88 \cdot 10^{12}$ m^{-1}/h during the first four hours of aeration.

The temperature shock experiments can be considered reasonably representative for circumstances that can occur in practice in a full-scale MBR plant. The filterability alternations created in these experiments were smaller compared to the other two stress experiments. Besides this the temperature effect was only demonstrated when the filterability of the starting sample was good. In case of a poor starting filterability the filterability could not be manipulated. Remarkably the filterability of the reference samples with a poor filterability could not be improved by continuous aeration. These activated sludge samples thus shows different behaviour compared to the DO and mechanic shear stress experiments.
The poor filterability of the reference sample has to be the result of a stress occasion experienced by the activated sludge in the full-scale installation. Contrary to the stress circumstances described in this chapter the filterability could not be improved by simply aerating the sample. Unfortunately the reason(s) for the poor filterability of the reference samples could not be retrieved.

8.6.2 Activated sludge properties

Although the three imposed stress circumstances had a different character they all seemed to have a similar effect on the activated sludge properties. In all cases the structure of the activated sludge flocs appears to be damaged, resulting in a release of fine particles in the free water. This supposition is explained on the basis of the SMP concentrations and the sub-micron particle volume in the free water.

Figures 8.16, 8.17 and 8.18 represent a compilation of the SMP concentrations and the sub-micron particle volume with the obtained ΔR_{20} values for all three stress experiments. For all three parameters the overall results indicate a worse filterability at higher concentrations, although the best correlation was found for the sub-micron particle volume. An explanation for the SMP concentration being a weaker indicator for the fouling potential of the activated sludge is likely to be the size of the SMP substances, at least for the proteins. Protein analyses of the DFCm permeate showed that more than half of the proteins present in the free water pass the membrane and end up in the permeate. These particles are thus not participating in the fouling process. The polysaccharides showed different retention behaviour; almost all polysaccharides are retained by the membrane. Despite of this the polysaccharides showed a weaker relation with the filterability than the proteins. The relation between filterability, SMP concentrations and sub-micron particle volume is discussed again in detail in chapter 9.4.

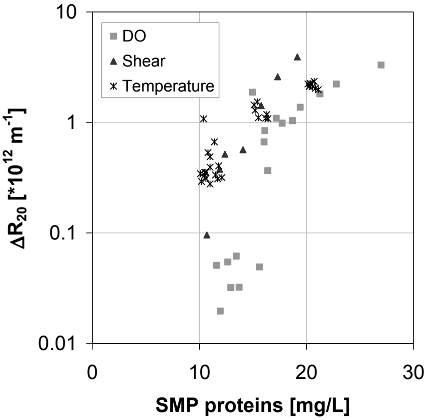


Figure 8.16: Compilation ΔR_{20} with SMP proteins

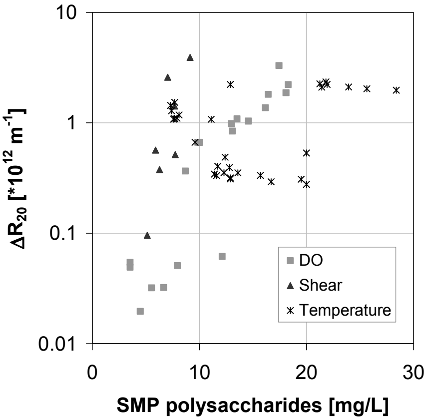


Figure 8.17: Compilation ΔR_{20} with SMP polysaccharides

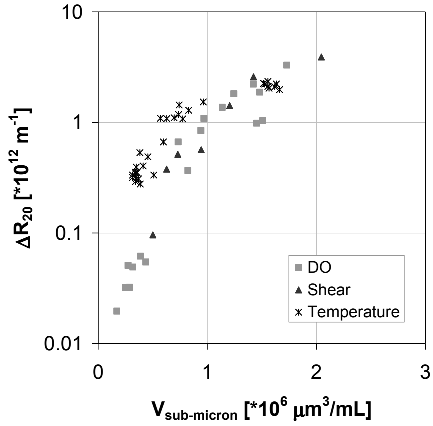


Figure 8.18: Compilation ΔR_{20} with sub-micron particle volume

8.6.3 Conclusions

The stress experiments discussed in this chapter demonstrate that activated sludge filterability is a dynamic parameter that can change rapidly due to various circumstances. The activated sludge quality analyses show that the filterability is closely related to the amount of fine substances in the free water (in the form of colloidal particles and SMP). The sub-micron particle volume seems a better indicator to characterise the filterability of activated sludge than the SMP concentrations, because many of the SMP substances are so small that they can pass the membrane and thus do not participate in the fouling process.

The strong relation between filterability and the amount of fine particles in the free water shows that the mechanisms of flocculation and deflocculation play a crucial role with respect to the filterability of activated sludge. When the filterability of the starting sample collected from MBR Heenvliet was good the filterability deterioration due to the imposed stress circumstances in the laboratory could be restored by simply aerating the sample. However, when the filterability of the collected sample was already poor the aeration had no effect on the filterability. This shows that in this case the activated sludge had different unfavourable flocculation characteristics. The reason for the poor filterability and flocculation properties of the collected samples could not yet be retrieved.

In conclusion it is mentioned that in practice stress occasions can not be avoided. The primary attention point is to recognise the occurrence of stress situations. Subsequently operational measures can be taken. Based on the experiments described in this chapter these operational measures should aim at improving the flocculation (characteristics) of the activated sludge. Operational measures that can be mentioned are an extension of the residence time in aerobic circumstances or (temporary) addition of chemical coagulants to enhance the activated sludge flocculation.

9 Discussion, conclusions and recommendations

9.1 Introduction

In this chapter the DFCm assessment (chapter 5) and the findings of the three measurement campaigns (chapters 6 to 8) are combined to discuss and answer the research questions that were posed in chapter 1.4. Subsequently the main conclusions of the research are summarised and several recommendations are proposed. Section 9.2 deals with the assessment of the DFCm. In section 9.3 the relation between the filterability as measured with the DFCm and the development of the permeability in the considered full-scale MBR-plants is discussed. The relation between filterability and activated sludge properties is discussed in section 9.4. In section 9.5 the main conclusions of the research described in this thesis are summarised. This chapter concludes with some recommendations in section 9.6, both related to future research with the DFCm as well as to optimisation of the MBR filtration process in general.

9.2 DFCm assessment

In this section the research questions related to the DFCm assessment as posed in chapter 1.4 are discussed. Aspects that were discussed in chapter 5 are recalled and considered again on the basis of the experience gained in the campaigns at MBR Varsseveld and MBR Heenvliet and in the stress experiments.

9.2.1 DFCm possibilities and limitations

In order to judge the value of the DFCm results it is important to discern the strong points and the shortcomings of the method. In this context the first research question *i* posed in the research objectives in chapter 1.4 reads:

> *What are the limitations and the possibilities of the DFCm?*

The basic feature of the DFCm is the characterisation of the *filterability* of an activated sludge sample. It is emphasised that the DFCm does not simulate or measure fouling; filterability is a *characteristic*, whereas fouling is a *process*.

When comparing the MBR process with the conventional activated sludge process the filterability can be considered a similar parameter as the settleability of the sludge: the *sludge volume index* (SVI). A low value for the SVI is a prerequisite but not a guarantee for a well-functioning sludge settling process. The same counts for the filterability and the functioning of the filtration process: good filterability can be considered a precondition for a satisfactory filtration process, but it has to be accompanied by good operational circumstances and membrane characteristics.

Paradoxically the characteristic filterability is not measured in practice in full-scale installations and no standardised measurement to do so exists. The conventional parameter to monitor and control the filtration process is the *permeability*. However, a decline in the permeability is not necessarily related to poor activated sludge filterability, but can also result from shortcomings in the operation of the filtration process or poor membrane characteristics. Two important undesired events in this respect are inefficient cleaning measures and the occurrence of clogging. Both events have in common that they reduce the membrane surface that is available for filtration, which leads to irregular distribution of the flux and a higher total fouling rate, irrespective of the activated sludge filterability. The power of the DFCm is that it offers the possibility to clarify whether fouling problems should be attributed to the activated sludge properties (i.e. filterability) or to inadequate operation of the filtration process.

As discussed in chapter 3 the total fouling process consists of a reversible, irreversible and irrecoverable component. In a standardised DFCm experiment only the short-term reversible fouling potential of an activated sludge sample is comprehended, but evidently the other two mechanisms gain significance when the time scale is extended. On the basis of the DFCm experiments a potential theoretical relationship between filterability and irreversible fouling can not be assessed. However, this issue can be investigated on an empirical basis by analysing the filterability evolution in relation to the permeability development tin time. This issue is discussed in section 9.3.

> *The DFCm is a tool to characterise the filterability of an activated sludge sample. Filterability is only partly determines the actual degree or rate of fouling so it is not a decisive parameter with respect to the filtration process. On the other hand good filterability can be considered a precondition for a satisfactory filtration process. Unlike permeability measurements the DFCm can clarify the role of the activated sludge filterability in the fouling process.*

9.2.2 Assessment operational circumstances

The operation of the filtration process is an important factor determining the extent of fouling. With respect to the formation of short-term fouling especially the balance between the forces towards (flux) and away from (crossflow) the membrane surface is crucial. It is thus important to assess the differences in these two parameters between the DFCm and the considered full-scale MBR plants. In this context the second research question *ii* stated in section 1.4 reads:

> *Is the filtration process as applied in the DFCm representative for the MBR filtration process in practice?*

The differences between the DFCm operational circumstances and the considered full-scale plants are considerable. In the first place the means of creating crossflow are different. In the DFCm the crossflow circumstances are created by means of liquid crossflow, whereas in the considered full-scale plants this is done by means of coarse bubble aeration. In addition also the fluxes applied in full-scale MBRs (generally around 20 L/m^2·h) are substantially lower than in the DFCm (80 L/m^2·h). Both considerations lead to the conclusion that the operational circumstances in the DFCm membrane tube can not be considered equal to the filtration regime in practice. However, the question is whether this is an objection. As mentioned before the purpose of the DFCm is not to simulate the filtration or fouling process, but to characterise the potential of an activated sludge sample to cause fouling. Evidently the *fouling* process is affected by the operational circumstances, but the question is whether the *filterability* of the activated sludge is affected by the operational circumstances.

Liquid crossflow versus air-liquid crossflow

In order to demonstrate differences in filterability a crucial precondition is that the operational circumstances (shear forces and the flux) in the DFCm experiments are constant for all experiments and controllable with high accuracy. Since the crossflow can be controlled more accurately with liquid crossflow this method is preferable over coarse bubble aeration. In the basis liquid crossflow and coarse bubble aeration crossflow pursue the same goal: the creation of a shear force along the membrane surface. Filterability is an activated sludge characteristic and is not related to crossflow circumstances. It can be expected that when the filterability of activated sludge is poor for liquid crossflow circumstances it will also be poor for coarse bubble aeration crossflow circumstances. This hypothesis could not be investigated on the basis of DFCm experiments, but is confirmed in literature (see chapter 5.2).

High flux versus low flux

The relatively high flux applied in the DFCm experiments is required to create cake formation, a prerequisite to compare the filterability of different activated sludge samples. A consequence of a high flux is that the forces transporting the particles to the membrane surface increase and bigger particles can accumulate in the cake layer. A theoretical analysis on the basis of backtransport mechanisms indicates that at filtration with the DFCm at the standard flux of 80 L/m^2·h particles smaller than 5 μm are prone to be involved in cake layer formation. At a flux of 20 L/m^2·h this critical particle size decreases to 2 μm. This implies that the DFCm experiments would provide a deceptive characterisation of the filterability if the activated sludge contains a high concentration of particles with a size range between 2 and 5 μm. Particle counting experiments however indicate that the volume of particles between 2 and 5 μm is negligible compared to the volume of particles smaller than 2 µm (see Figure 4.7 in chapter 4.6). This implies that a higher flux does not significantly change the size of constituents accumulating in the cake layer. It can be expected that an activated sludge that exhibits poor filterability at a high flux will also have a poor filterability at a low flux. This hypothesis is confirmed by several DFCm experiments in which the filterability was characterised at different fluxes (see Figures 6.4 and 7.4).

> *The operational circumstances in the DFCm membrane tube are not equal to the (immersed) MBR filtration process in practice. This is however not an objection since the scope of the DFCm is not membrane operation but activated sludge properties. Literature and experimental results support the proposition that the filterability as characterised with the DFCm forms a reliable indication for the filterability in practice in a full-scale MBR plant.*

9.2.3 DFCm applicability

When knowing the possibilities and limitations of the DFCm the range of application of the method can be specified. In this context research question *iii* in the problem statement in chapter 1.4 reads:

> *How should the DFCm be applied to yield optimal profit from its possibilities?*

In the research described in this thesis the DFCm has been applied in two ways:

- As a *practical* tool to monitor the activated sludge filterability during everyday operation of two full-scale MBR plants.
- As a *research* tool to identify the activated sludge characteristics that are related to its filterability.

The most practice-oriented application of the DFCm is putting it in operation in-situ at a full-scale MBR plant during everyday operation. The major advantage of this approach is that the DFCm results can be analysed in relation to the process operation, external circumstances and the full-scale permeability development. This application has been tested at the full-scale MBR plants of Varsseveld and Heenvliet. Despite the similar approach the two research campaigns some differences. In the case of Varsseveld the most interesting results were obtained for some isolated measurements with intervening periods of several months (after uncoupling of the cheese factory, see chapter 6.4). The isolated measurements provided a momentaneous indication of the activated sludge filterability, but evidently did not provide detailed information about the development and the dynamic changes of the filterability throughout the total period. For the Heenvliet measuring campaign the measurement frequency was approximately one week; in this way a more accurate and more reliable characterisation of the filterability development over a longer period was formed. From a critical point of view the measurement frequency in Heenvliet (one week) can still be considered low, because several stress experiments demonstrated that filterability is a characteristic that can change considerably on a time-scale of hours. From this point of view it is thus desirable to have a DFCm measurement frequency as high as possible. Ideally the DFCm is extended and developed into a method for on-line measurements (further discussed in section 9.6). In this way dynamic alterations in the activated sludge filterability can be identified instantly and the process operation can be adjusted to minimise or prevent fouling problems.

A drawback of the in-situ measurements at full-scale MBR plants is that the filterability properties are formed by a combination of numerous external and operational factors that can impossibly be all surveyed. This drawback can be overcome by using a different research approach. By imposing well-defined stress conditions in lab-scale experiments the filterability and properties of an activated sludge sample can be manipulated. By comparing the manipulated samples with unaltered reference samples the changes in activated sludge properties can be attributed exclusively to the imposed stress conditions. The stress experiments are favourable for identification of the activated sludge properties that play a role in the filtration process (further discussed in section 9.4). On the other hand it has to be recognised that the imposed stress circumstances can not be considered fully representative for the stress that may occur in practice in a full-scale MBR plant.

The most powerful and practice-oriented application of the DFCm is to put it in operation in-situ at a full-scale MBR plant. In this way the filterability can be characterised in relation to the permeability development in the full-scale plant and to external and operational circumstances. Poor filterability quality can be identified in an early stage and the process operation can be adjusted to improve the filterability and/or to minimise fouling problems.
In addition the DFCm can be employed as a lab-scale research tool to identify specific activated sludge characteristics that are related to its filterability.

9.2.4 DFCm output

The output of a DFCM experiment consists of a dataset that plots the development of the filtration resistance as a function of the specific permeate production. Evidently it is important to understand what information this dataset provides about fouling mechanisms. In this context research question *iv* in chapter 1.4 reads:

What is the dominant fouling mechanism in a DFCm experiment and what is the significance of the DFCm output?

In a short-term DFCm experiment the irreversible fouling rate is negligible compared to the reversible fouling rate. The fouling created in a DFCm experiment can be characterised according to the theory of cake layer filtration. In this approach the total filtration resistance is formed by the concentration of substances c_i in the activated sludge liquid accumulating on the membrane surface [g/L] and the specific resistance α caused by these substances [m/g]. The specific resistance is a function of the prevailing ΔR and the compressibility coefficient s (an activated sludge characteristic). ΔR can be isolated to rewrite the resistance increase as a function of the specific permeate production V with two coefficients (see chapter 5.7 for deduction and denotation symbols):

$$\Delta R = p \cdot V^{\frac{1}{1-s}} \qquad \text{, with: } p = (\alpha_r \cdot c_i)^{\frac{1}{1-s}} \text{ and } s = \text{compressibility coefficient}$$

Each DFCm experiment produces a dataset from which the values for coefficients p and s can be derived. Subsequently the value for $\alpha_r \cdot c_i$ can be calculated from p and s. The coefficient $\alpha_r \cdot c_i$ represents a combination of the concentration of substances accumulating in the cake layer and the specific resistance of these substances at a reference cake layer resistance.

The compressibility coefficients s measured in the three measurement campaigns vary between 0 and 0.45 (see Figure 9.1). These values are lower than compressibility coefficients mentioned in several literature references. For example, Roorda (2004), Mendret et al. (2009) and Ramalho (1983) found values between approximately 0.6 and 0.75. However, it has to be kept in mind that these values were obtained for dead-end filtration experiments. As a consequence of the crossflow circumstances in the DFCm only relatively small particles are attracted to the membrane. These smaller particles can be expected to have less inner void space than bigger particles that can accumulate in the cake layer for dead-end filtration and thus also a lower propensity to be compressed.

Since coefficient p is partly dependant on the compressibility coefficient s a relation between these two parameters can be expected, but this is not confirmed by the experimental results. Figure 9.1 shows the combinations of p and s for the three measurement campaigns discussed in this thesis. For the Varsseveld experiments the compressibility coefficient s was reasonably constant (approximately 0.2) with varying values for p. The results for the Heenvliet campaign and the stress experiments show an opposite trend; coefficient s varies and coefficient p is more or less constant (and low) for all DFCm experiments. On the other hand it has to be realised that the compressibility coefficient only delivers a relative contribution to the total resistance increase. The varying values for s in the Heenvliet campaign and the stress experiments are accompanied by relatively low values for p. This implies that the *absolute* contribution of s to the total filtration resistance is limited. This is illustrated in the Figure 9.2 and Figure 9.3, in which the relation between the coefficients s and $\alpha_r \cdot c_i$ with the ΔR_{20} values are plotted. No relation is demonstrated between *s* and the increase of the filtration resistance. The results indicate that the total resistance increase is predominantly determined by the coefficient $\alpha_r \cdot c_i$.

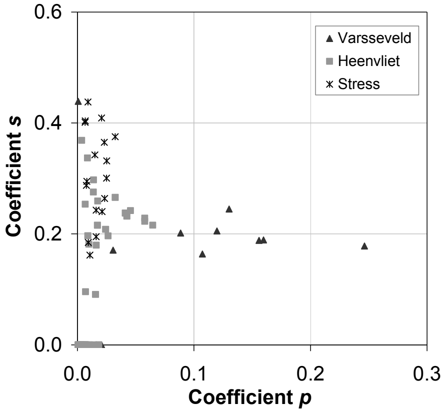


Figure 9.1: DFCm output, coefficient *s* with coefficient *p*

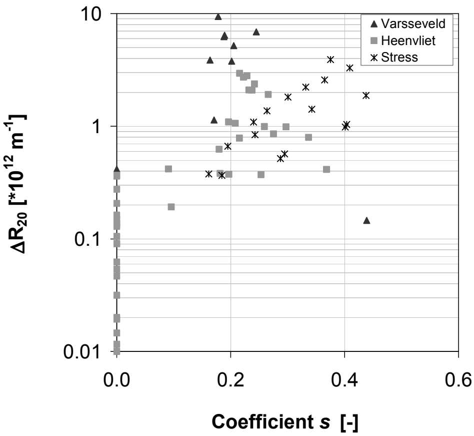


Figure 9.2: DFCm output, relation between coefficient *s* and ΔR_{20}

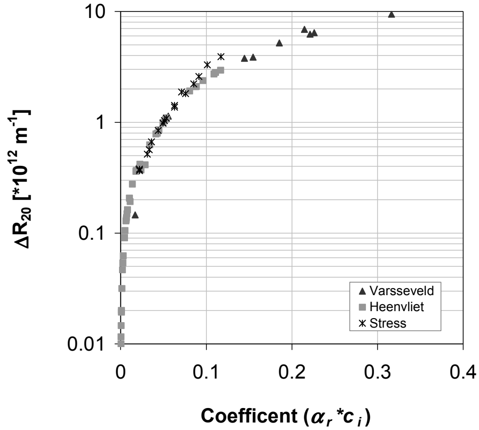


Figure 9.3: DFCm output, relation between coefficient $\alpha_r \cdot c_i$ and ΔR_{20}

In addition to the results presented in Figure 9.1 and Figure 9.2 it is mentioned that cake compression is a function of the prevailing TMP. In the DFCm experiments the TMP reaches values up to 0.6 Bar. However, in practice the TMP values occurring in a submerged full-scale MBR plant for municipal wastewater treatment are considerably lower. This implies that in practice also the influence of cake layer compression will be of minor influence compared to the concentration of substances accumulating in the cake layer and their specific reference resistance.

The dominant fouling mechanism in a DFCm experiment is cake layer formation. The total cake layer resistance is predominantly determined by the concentration of substances accumulating in the cake layer. The compressibility coefficient shows no relation with the total resistance increase in the relevant range.

9.3 Filterability and full-scale permeability

The DFCm experiments do not provide direct information about the potential of an activated sludge sample to cause (long-term) irreversible fouling. However, a possible relation between filterability and irreversible fouling potential can be investigated on an empirical basis. The permeability development in a full-scale MBR plant in between two chemical cleanings forms a reflection of the irreversible fouling rate. If filterability is related to irreversible fouling a relation between the DFCm experiments and the permeability development can be expected. In this context question *v* in chapter 1.4 reads:

> *What is the relation between filterability as characterised with the DFCm and the permeability development as occurring in full-scale plants?*

In a short-term DFCm experiment the irreversible fouling rate is negligible compared to the reversible fouling rate. The irreversible fouling rate can only be assessed on the basis of the recovery after physical cleaning measures. Since a well defined physical cleaning protocol is not implemented in the measuring protocol a DFCm experiment does not provide direct information about the potential of the activated sludge to cause irreversible fouling. Nonetheless, in practice a relation between these two “forms” of fouling in a full-scale MBR plant is not unthinkable since the irreversible fouling rate is directly determined by the efficiency of the (physical) cleaning measures to remove reversible fouling. In this research a potential relation between reversible and irreversible fouling was assessed on the basis of an empirical comparison of the DFCm results and the permeability development in the full-scale MBR plants of Varsseveld and Heenvliet. The development of the permeability in between two chemical cleanings can be considered a reflection of the irreversible fouling rate occurring in the plant. Although this analysis has been performed for only two plants the results are interesting.

In the Varsseveld measuring campaign the DFCm results could be clearly linked with the permeability development. In the first few months after start-up of the plant the extreme poor filterability as measured with the DFCm was accompanied by a distressing decline of the permeability. As soon as the cheese factory was uncoupled from the sewer both the filterability and the permeability showed strong recovery.

In the Heenvliet measuring campaign the DFCm results did not correspond with the permeability development. In general the filterability was good, whereas the permeability showed a continuous decreasing trend. A closer analysis of the situation in Heenvliet however indicated that the decreasing trend in the permeability could probably not be attributed to poor activated sludge filterability. When the membrane tank was emptied for maintenance cleaning the membranes appeared to be subject to severe clogging problems. At least 25% of the total membrane surface was covered with a thick sludge layer and suspected to be practically excluded from the filtration process. As a consequence the local fluxes in the available membrane surface increased correspondingly to cope with the total permeate flow. This situation is evidently highly unfavourable for the filtration process, since (apart from the filterability quality) the flux is a crucial parameter affecting the fouling rate. The problems encountered in Heenvliet strongly indicate that the decreasing trend in the permeability was not caused by fouling or a poor activated sludge quality, but by clogging. Although clogging is partly dependant of the activated sludge properties (viscosity through MLSS concentration) it is predominantly the result of inadequate turbulent circumstances in the membrane modules.

For the MBR Varsseveld measuring campaign the development of the permeability in time was clearly related to the filterability as characterised with the DFCm. The results support the hypothesis that filterability and irreversible fouling are related.
For MBR Heenvliet no link between filterability and permeability development was demonstrated, but the occurring clogging problems strongly indicate that the permeability decrease could not be attributed to poor activated sludge filterability.

9.4 Filterability and activated sludge properties

A major objective of the research described in this thesis is to improve the understanding about the relation between activated sludge filterability and its characteristics. In this context question *vi* in chapter 1.4 reads:

> *Which activated sludge properties are linked to filterability and how?*

This research question is discussed in several steps. After discussing some generalities the attention is focused on soluble microbial products, which are commonly linked to fouling. From the SMP the attention is shifted to the role of sub-micron particles and their relation with SMP. In conclusion the relation between filterability and the mechanisms of flocculation and deflocculation is discussed.

9.4.1 General

Fouling results from the complex interaction between the membrane, the activated sludge and the way the filtration process is operated. Despite the multitude of factors involved in the process the basic principle that stands is that MBR fouling is caused by “substances” that are present in the activated sludge suspension. As discussed in chapter 3.8 the physical nature and especially the size of the foulants is assumed to have the greatest impact on the fouling potential of activated sludge.

The size of the substances involved in the fouling process can roughly be marked out. Thanks to crossflow circumstances employed in the MBR filtration process the relatively big particles (compared to the size of the membrane pores) are not involved in the fouling process. A theoretical analysis indicates that in a DFCm experiment at standard operational circumstances (J = 80 L/m^2·h, CFV = 1.0 m/s) particles with a diameter exceeding approximately 5 µm are excluded from the fouling process (see section 5.6). Also the smallest (i.e. dissolved) substances are not involved in the fouling process, because they pass the membrane pores. General agreement exists that the attention should be focused on the free water phase of the sludge (Rosenberger et al., 2005; Evenblij, 2005). Activated sludge bulk characteristics such as MLSS concentration, floc size distribution, floc composition (bound EPS concentration), pH and dissolved oxygen concentration all play a role in the fouling process, but can all be considered indirect influence factors.

> *In theory the filtration resistance in a DFCm experiment is caused by particles that are retained by the membrane (approximately 0.03 µm) up to a size of approximately 5 µm.*

9.4.2 Filterability and SMP concentrations

Focusing on the free water phase can hardly be considered a simplification since it is still a very comprehensive fraction. It contains a wide range of substances such as proteins, polysaccharides, various acids, nutrients, RNA, DNA, carbohydrates, vitamins, exocellular enzymes, viruses, cell fragments and individual bacteria (Metcalf and Eddy, 2003).
Currently the attention in MBR fouling research is to a large extent concentrated on the role of soluble microbial products (SMP) in the free water, which are generally considered to predominantly consist of proteins and polysaccharides. In this research the protein and polysaccharide concentrations in the free water were determined according to the colorimetric methods described by Lowry et al. (1951) and Dubois et al. (1956) respectively. Figure 9.4 represent a compilation of the ΔR_{20} values plotted with the SMP concentrations for the three measurement campaigns discussed in this thesis.

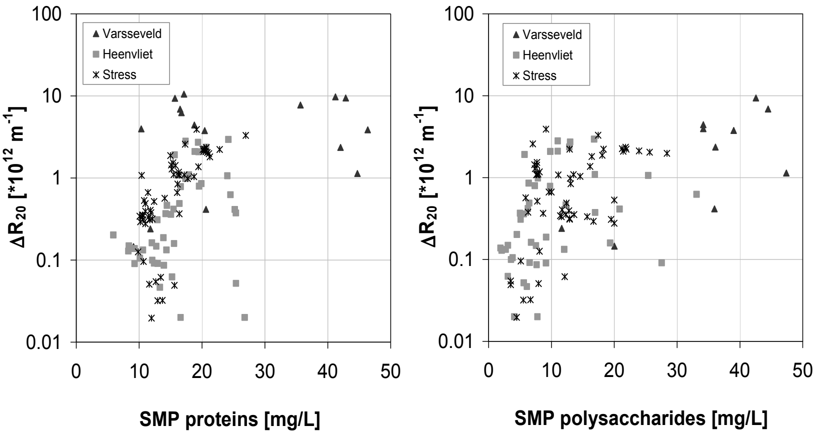


Figure 9.4: Compilation of ΔR_{20} values with SMP concentrations for all three measurement campaigns

For proteins the relation is weak. Some exceptional high concentrations encountered at MBR Varsseveld are indeed corresponding with the poor filterability, but the generally obtained concentrations between 10 and 20 mg/L are accompanied by filterability qualities ranging between excellent (<0.05) and very poor (>>1). Similar considerations apply when comparing the polysaccharide concentrations with the filterability, although the correlation is slightly higher. Again the highest concentrations (encountered at MBR Varsseveld) are accompanied by the highest ΔR_{20} values. As with the proteins also polysaccharide concentrations around 10 mg/L are accompanied by filtration qualities ranging from excellent (<0.05) to very poor (>>1). On the other hand the overall results for the polysaccharides give a more distinct indication that the filterability deteriorates with increasing concentration compared to proteins.

The relation between activated sludge filterability and the SMP concentrations in the free water is weak. Exceptional high SMP concentrations are indeed accompanied the poorest filterability, but on the other hand the prevailing concentrations are accompanied by filtration qualities ranging from excellent to very poor.

9.4.3 Filterability and total sub-micron particle volume

Information about the colloidal substances in the activated sludge free water was obtained on the basis of the particle size measurements. The particle counter used in this research is capable of measuring particles with a diameter from 0.4 to 5.0 µm. This implies that only the upper part of the total colloidal range is covered by the measurements[6]. Nonetheless, the volume of particles in the range between 0.4 and 1.0 µm at least provides some information about the colloidal particle volume. The experimental results show a distinct correlation between the sub-micron particle volume and the filterability. Figure 9.5 shows a compilation of the sub-micron particle volume with the ΔR_{20} values for the measurement campaign in Heenvliet and the stress experiments (no particle counting experiments were performed in the Varsseveld campaign).

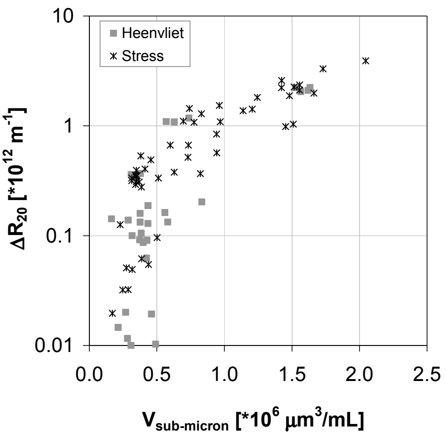


Figure 9.5: ΔR_{20} values with sub-micron particle volume

Comparing Figure 9.5 with Figure 9.4 leads to the conclusion that the sub-micron particle volume appears to be a better indicator for the filterability of activated sludge than the SMP concentration. This observation is discussed more in detail in section 9.4.4.

[6] The colloidal rang is assumed 0.01-1.0 mm, see chapter 3.8

The volume of sub-micron particles in the free water with a diameter ranging from 0.4 to 1.0 µm shows a more distinct relation with the filterability than the SMP concentrations.

9.4.4 SMP concentration and size

An important conclusion that can be drawn from the SMP analyses and the particle counting experiments is that the parameter *volume* (i.e. particles) appears to provide more information about activated sludge filterability than the parameter *concentration* (i.e. SMP). This can be explained on the basis of the pore size of the membrane, in the case of the DFCm (and the far majority of the MBR plants in practice) this is ultrafiltration. Although the pore sizes in the ultrafiltration membranes are small[7], the filtration process can still be considered a physical sieving process. Substances with a size smaller than the membrane pores can easily permeate through the membrane without participating in the fouling process, while bigger substances are retained and can contribute to fouling. This indicates that the distinction between *soluble* and *colloidal* substances in the free water with respect to filterability is crucial. Actually the term "soluble microbial products" can be considered a misnomer, since with the sludge-water separation methods used in this research both soluble and colloidal substances are present in the free water.
In addition it points out the major shortcoming of the colorimetric SMP analyses on activated sludge free water: it does not distinct soluble SMP that can pass the membrane and colloidal SMP (or SMP attached to colloids) that are retained by the membrane. This implies that additional information is required about the size of the SMP to clarify their influence on filterability.

More information about the size of the SMP in the free water can be obtained on the basis of fractionation. In the research campaign in Varsseveld extensive fractionation experiments for several different cut-off sizes were conducted, but due to the exceptional circumstances with the cheese factory (see chapter 6.4) it is questionable whether the obtained results are representative.

The most straightforward available information about the size of the SMP can be obtained by comparing the SMP concentrations in the free water and the DFCm permeate. Figure 9.6 shows the dataset for the three measurement campaigns. The Varsseveld case shows deviant results, but the data gathered from the campaign in Heenvliet and the stress experiments indicate that on average 60% of the proteins present in the free water pass the membrane. This thus implies that at least 60% of the SMP proteins are smaller than the membrane pores (0.03 µm) and do not participate in the (short-term) fouling process. Based on these findings it seems that the SMP protein concentration being a week indicator for the filterability quality of activated sludge is

[7] Ultrafiltration (UF) membranes nominal pore size is assumed to range from 0.01 and 1.0 µm

explained by the fact that they are to a large extent in solution and thereby too small to play a role in the fouling process.

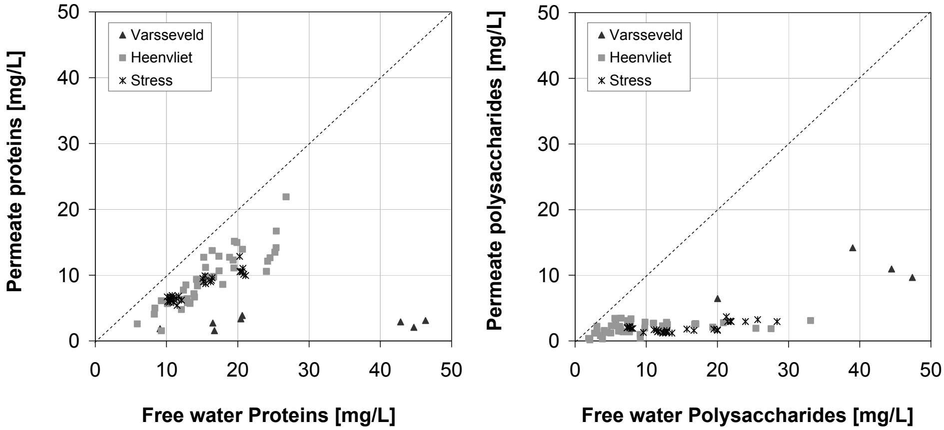
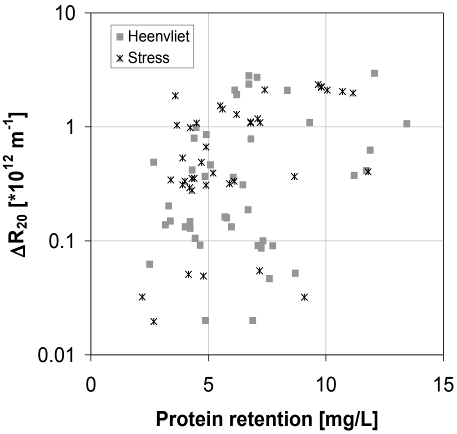


Figure 9.6: Compilation of SMP retention by the DFCm membrane

The polysaccharides show totally different retention behaviour than the proteins. The results indicate that the amount of polysaccharides breaking through the membrane is always low and does not depend upon the concentration in the free water. This observation is surprising since the size range of polysaccharides is considered to be similar to proteins (Metcalf and Eddy, 2003). An explanation could be that, contrary to proteins, the polysaccharides are attached to colloidal material in the free water. From this point of view the SMP polysaccharide retention would seem a good indicator for the filterability. However, the differences in filterability can also not be explained on the basis of SMP retention. Figure 9.7 shows the ΔR_{20} values plotted with the measured retained SMP concentrations. The protein retention shows no relation at all with the filterability. The slightly better relation between polysaccharide retention and filterability support the hypothesis that filterability is related to colloidal material, but the coherence of the data can still be considered weak.



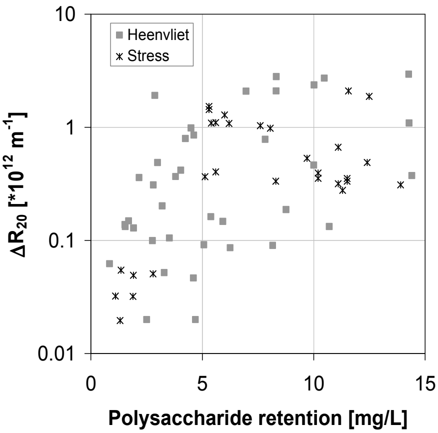


Figure 9.7: Compilation ΔR_{20} values plotted with SMP retention (by the DFCm membrane)

The results indicate that the distinction between retained and breakthrough SMP is not sufficient to explain filterability. Apparently a more detailed understanding about the colloidal particle volume and size distribution is required.

> *The explanation for the SMP concentration being a poor indicator for activated sludge filterability is that the used analyses do not provide information about the size of the SMP (i.e. the ratio between soluble and colloidal SMP). Soluble SMP can pass the membrane whereas colloidal SMP are retained. However, filterability could also not be explained on the basis of retained SMP. Additional knowledge about the colloidal size distribution is required*

9.4.5 Sub-micron particle volume and SMP concentrations

A drawback of the particle counting results is that they only provide information about a part of the colloidal fraction (0.4- 1.0 μm). Nonetheless, it can be expected that the total volume of particles with a size between 0.4 and 1.0 μm forms an indication for the volume of particles smaller than 0.4 μm. From this point of view a relation between the sub-micron particle volume and the SMP concentrations can be expected. The results indeed confirm a link, see Figure 9.8, but the coherence can not be considered evident.

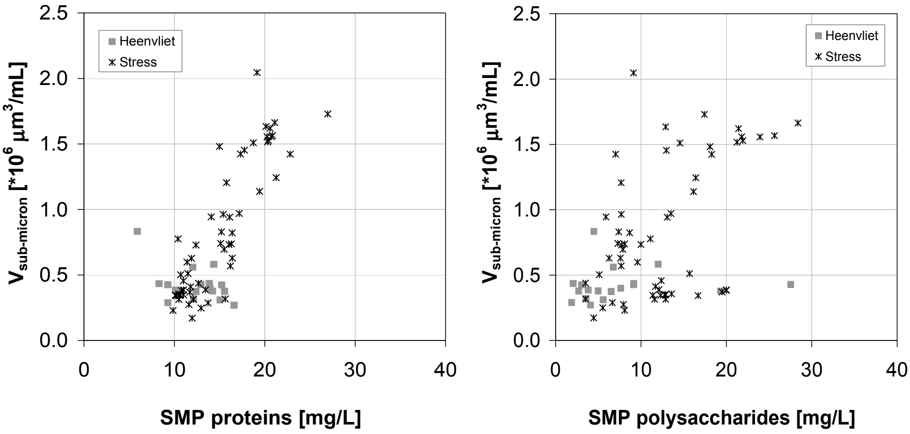


Figure 9.8: Sub-micron particle volume with SMP concentrations

Evidently the sub-micron particles represent a certain concentration. This concentration can be estimated by assuming the particles to be spherical and to have a density in the same order of magnitude as water (1000 mg/L). Figure 9.9a presents the typical output of a particle counting experiment in the covered colloidal range. For each range the volume distribution can be calculated ($V = counts \cdot 1/6 \cdot \pi \cdot D^3$), see Figure 9.9b.

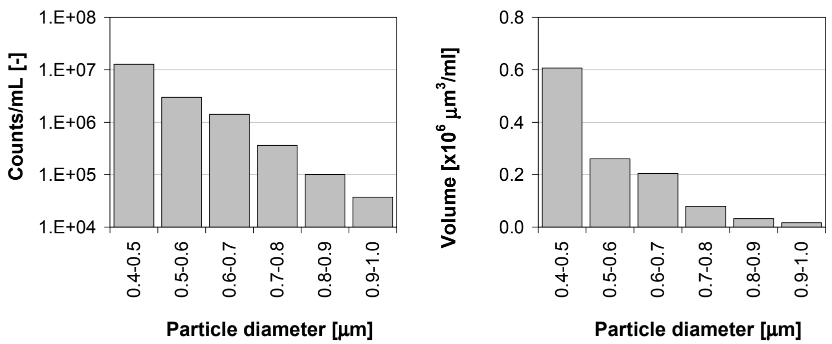


Figure 9.9: Typical sub-micron size and volume distribution in the range 0.4 to 1.0 µm

The obtained values for sub-micron particle volumes are in the order of magnitude of $1 \cdot 10^6$ µm^3/mL. This corresponds with a concentration of only 1 mg/L, significantly lower than the SMP concentrations that are measured with the colorimetric methods (see Figure 9.8). Possible explanations for this discrepancy are:

- The far majority of the SMP in the free water are smaller than the detection limit of the particle counter (0.4 µm).

- The particle counting results represent an underestimation of the actual colloidal particle volume in the free water.

Both explanations can be defended. In the first place the retention measurements indicate that on average 60% of the proteins pass the (DFCm) membrane and are thus smaller than 0.03 µm. Intuitively this indicates that the remaining 40% of proteins have a size that is in the same order of magnitude as the membrane pores, indeed considerably lower than the detection limit. In addition the particle volume distribution (Figure 9.9b) indicated that the total sub-micron volume (0.4-1.0 µm) is predominantly formed by particles with a size between 0.4 and 0.5 µm. This seems to indicate a higher particle volume in the range below 0.4 µm. This issue is discussed more detailed in the next section 9.4.6.

On the other hand the high retention of polysaccharides and the high absolute SMP concentrations in the free water would seem to indicate the presence of colloidal material in higher concentrations than the 1 mg/L as measured with the particle counter.
Besides this the particle counting experiments hold several uncertainties, see chapter 4.6. The values from the particle counting experiments have to be considered with some reserve. It is possible that the particle counting data represent an underestimation of the actual colloidal particle volume, due to the low dilution rate and the transparent nature of the particles. Additional basic research is required to optimise the methodology of the particle counting, especially with respect to respect to fluids with high particle concentrations such as activated sludge (free water).

9.4.6 Colloidal and sub-micron particle size distribution

The particle counting experiments and SMP analyses indicate that the colloidal particle size distribution in the free water plays a crucial role with respect to the filterability of activated sludge. Information about this topic in literature is however lacking.

As discussed in the previous section the results of the particle counting experiments have to be considered with reserve. Despite the uncertainties the particle counting experiments still might form a basis for further analysis of the colloidal particle size distribution. Apart from the absolute data all particle counting experiments at least have in common that they show a similar size distribution in the range between 0.4 and 1.5 µm: an exponential decrease of the number of particles with increasing size range (see Figure 9.9 and Figure 4.6 in chapter 4.6).

Based on this trend the distribution in the range below 0.4 µm can be estimated, by assuming a backward extension of the exponential trend within the range between 0.01 to 0.4 µm (see Figure 9.10a). From the data in Figure 9.10a the volume distribution can be calculated, see Figure 9.10b. The results indicate that the colloidal volume is predominantly formed by particles with a size in the range between 0.1 and 0.5 µm (>80%). This indicates that the volume and thereby the resistance of the cake layer is highly influenced by the presence of particles in the free water with a size between 0.1 and 0.5 µm (in all unities: number, concentration and volume). It is again emphasised that the volume distribution as presented in Figure 9.10 is based on an *assumed* exponential colloidal particle distribution. Additional research is required to verify this assumption.

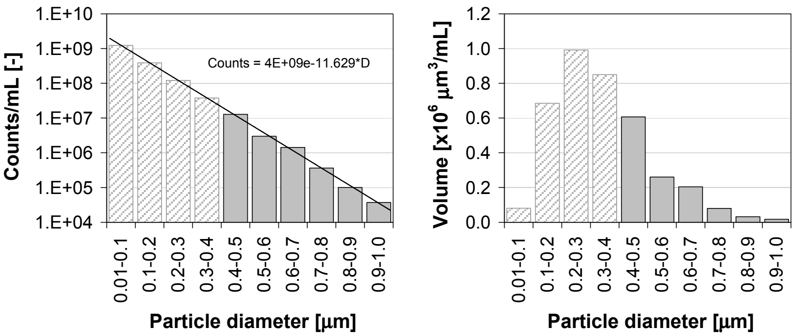


Figure 9.10: Counts and volume distribution based on an assumed exponential particle size distribution

Nonetheless, what stands is that the potential contribution to the total cake layer decreases with decreasing particle size. This especially weighs for particles with a size between 0.01 and 0.1 µm. In the presumed exponential size distribution the relative contribution of this smallest size range to the total colloidal particle volume is only 2%. This indicates that even if the size distribution in the smallest range differs strongly from the presumed exponential size distribution the relatively big particles will have a dominant contribution to the volume of particles that might accumulate in the cake layer.

Based on an exponential particle size distribution, as obtained for part of the colloidal range in the particle counting experiments, the particles that determine the volume of the cake layer (i.e. the filtration resistance) are predominantly in the size range between 0.1 and 0.5 µm. Additional knowledge is required about the particle size distribution in the range below 0.4 µm, but even if it significantly differs from an exponential one the contribution of relatively big particles will be substantial.

9.4.7 Flocculation

Understanding the fundamentals, mechanisms and activated sludge properties that play role in the fouling process is only a first step. Evidently the next step is to apply this knowledge in practice to improve the MBR filtration process. In this context research question *vii* posed in chapter 1.4 reads:

> *Can the properties and the filterability of the activated sludge collected from full-scale MBR plants be related to the process operation or to external circumstances?*

The experimental results presented in this thesis indicate that the filterability of MBR activated sludge is closely related to the amount of fine particles in the free water. Filterability improves with a decreasing volume of colloidal material in the free water. In practice the MBR process operation should thus aim at minimising the amount of fine material in the free water.

The stress experiments discussed in chapter 8 clearly demonstrate that the amount of fine material in the free water is closely related to the operational or biological circumstances that the activated sludge is experiencing. When activated sludge is experiencing stress the floc structure is damaged and *deflocculation* occurs, expressed in the release of fine material from the activated sludge matrix into the free water. The experiments however also show that this is a reversible process: by imposing favourable conditions the fine material is absorbed again by the activated sludge flocs. This process of *flocculation* is accompanied by an improvement of the filterability.

The stress circumstances imposed in the experiments discussed in chapter 8 can be considered extreme, but also in practice activated sludge is continuously subject to some degree of stress conditions. This stress is in the first place related to continuous variations in the influent flow (rate, composition, temperature). In addition also inadequate process control (both with respect to the biological and the filtration process) can stress the biomass. The starting point for a satisfactory filtration process is a well-considered and balanced operation of the biomass to create an optimal flocculation process in reference conditions. In addition supplementary operational measures can be taken to enhance the flocculation process in case of stress circumstances caused by fluctuations in the influent properties. Examples of these measures are the extension of the aerobic residence time and the addition of chemicals to enhance the flocculation process.

The fundamentals of the activated sludge flocculation process go beyond the scope of this research. The stress experiments demonstrated that in case of a good starting filterability (i.e. filterability at the time of sampling) the filterability deterioration created by stress conditions could be reversed by simply aerating the sample. However, in some cases the filterability was already poor when the samples were collected from the full-scale plant. Apparently the biomass was already experiencing stress conditions at the moment of sampling. Remarkably in these cases the filterability could not be improved by imposing continuous aerated circumstances. This indicates that the flocculation properties of these activated sludge samples were totally different from the ones with a good starting filterability. Supplementary research is required to improve the understanding of MBR activated sludge flocculation process.

MBR activated sludge filterability is closely related to the mechanisms of flocculation and deflocculation. The process control should aim at an optimal flocculation process once the activated sludge reaches the membrane area.

9.5 Main conclusions

Membrane fouling in membrane bioreactors is a complex process resulting from the interaction between three main parameters: (1) the membrane characteristics, (2) the membrane operation and (3) the activated sludge properties. The conventional parameter used to monitor and manage the filtration process, the permeability, does not provide a distinction between the influences of each of these three factors in the fouling process.

A method was developed to characterise the filterability of activated sludge: the Delft Filtration Characterisation method (DFCm). Samples collected from any MBR plant are filtrated in a filtration unit with a single sidestream membrane under constant and well-defined operational circumstances. In this way differences in resistance increase that occur during filtration can be attributed exclusively to differences in the activated sludge properties (i.e. filterability). The most powerful application of the DFCm is to employ it in-situ at a (full-scale) MBR plant to monitor the development of the activated sludge filterability in relation to the development of the permeability. In this way the influence of the activated sludge activated sludge properties on the filtration process can be verified.

The DFCm has proven to be a useful tool to comprehend the filtration process at any (full-scale) MBR plant. In the measurement campaign at MBR Varsseveld the DFCm results demonstrated that the decline of the permeability could indeed be attributed to poor activated sludge filterability. In the second campaign at MBR Heenvliet a good filterability was accompanied by a disturbing decline of the permeability, but the later observed clogging problems in the membrane modules revealed that the decreasing trend of the permeability had to be attributed to inadequate membrane operation rather than to poor activated sludge filterability.

Particle counting experiments in the size range between 0.4 and 1.0 μm provide a better indication of the activated sludge filterability than the SMP concentrations. This can be explained by the fact that the SMP analyses comprehend both soluble and colloidal material. Soluble substances can pass the membrane without participating in the fouling process, whereas colloids are retained by the membrane and thus have the potential to accumulate in the cake layer. However, filterability could also not be explained on the basis of SMP retention. This indicates that more detailed information is required about the particle size distribution in the colloidal range to explain the filtration properties of activated sludge. Particle size distribution data indicate that the particles that determine the volume of the cake layer (i.e. the filtration resistance) are predominantly in the size range between 0.1 and 0.5 μm. However, additional studies on the colloidal particle size distribution of activated sludge are required.

Activated sludge filterability is closely related to the mechanisms of flocculation and deflocculation. The mechanism of deflocculation is closely related to the conditions that the biomass is experiencing. Fluctuations in the process (shear force, biological treatment) and in the influent properties (quality, quantity, temperature) can all be considered stress circumstances and promote the process of deflocculation. In order to generate activated sludge with good filterability a balanced biological treatment process is required, preferably with a low loading rate. In case of (sudden) stress conditions supplementary measures are required to enhance the flocculation process.

9.6 Recommendations

To finalise this chapter several future directions are proposed to improve MBR technology. These recommendations are related to the Delft Filtration Characterisation method itself, to scientific fouling research and to the optimisation of the filtration process in general.

9.6.1 DFCm development

Irreversible fouling

The irreversible fouling rate in a DFCm experiment is negligible compared to the reversible fouling rate. In this research the relation between these two forms of fouling could only be investigated on an empirical basis, by comparing the DFCm filterability as measured over time with the development of the permeability in the considered full-scale plants. The potential of an activated sludge sample to create irreversible fouling could not be investigated on the basis of the DFCm experiments. In order to do so the DFCm measuring protocol can be extended with a physical cleaning step. A major attention point in this context is that as with the filtration circumstances also the physical cleaning measures are well-defined and controllable with high accuracy in order to preserve the possibility to compare different DFCm experiments with each other.

A suggestion for extension of the measuring protocol is to perform a series of filtration cycles with periods of relaxation in between. The recovery of the filtration resistance can be considered a reflection of the activated sludge potential to cause irreversible fouling. On the other hand it has to be kept in mind that the irreversible fouling rate in an MBR plant is more a reflection of the physical cleaning efficiency than an activated sludge characteristic.

Online filtration characterisation

As demonstrated in chapters 6 to 8 the filterability of activated sludge is a dynamic parameter. The highest measuring frequency that was applied with the DFCm was at MBR Heenvliet, with measurements approximately once every week. These weekly measurements provided a reasonable insight in the development of the filterability but evidently increasing the measurement frequency improves the understanding of the filterability dynamics. Ideally the DFCm is employed as an automated on line tool in an existing MBR plant to monitor the filterability continuously. In this way the filtration process can be optimised by anticipating (sudden) changes in the activated sludge filterability.

9.6.2 Fouling research

Particle size

The experimental results discussed in this dissertation demonstrate that the filterability of activated sludge is determined by the particle size distribution in the colloidal range (0.01-1.0 µm). As already mentioned in subsection 9.4.6 more knowledge is required about this size range. The SMP analyses that were performed to determine the retention by the DFCm membrane demonstrated that the soluble microbial products in the free water are smaller or in the same size range as the pores of the UF membrane (0.03 µm). More elaborate fractionation experiments might improve the understanding of the (SMP) size distribution in the free water. The particle counter used in the research has a lower limit of 0.4 µm. A particle counter in the lower range can provide better understanding of the particle size distribution in the smallest size range.

Flocculation

The amount of fine material in the activated sludge free water and thereby the filterability is closely related to the mechanisms of flocculation and deflocculation. Several experiments demonstrated that the flocculation process can be enhanced by simply aerating the activated sludge (i.e. bioflocculation). However, in several other experiments the properties of poor filterable (deflocculated) sludge could not be influenced by continuous aeration. The reason for these poor flocculation properties could not be retrieved. Additional research is required to understand the mechanisms and influence factors with respect to MBR activated sludge flocculation.

9.6.3 Process control improvement

As discussed in chapter 5 the permeability can be considered a weak parameter to operate and assess the MBR filtration process. In this final section another approach for operation of the filtration process is proposed. The starting point for this approach is a comparison with the conventional clarification step in the activated sludge process. Important design parameters for conventional (secondary) clarifiers are the required *surface area* of the tank and the *surface loading rate*. In addition the performance of a clarifier is determined by the *activated sludge properties* (suspended solids concentration and sludge volume index). The three variables mentioned here can be analysed in close comparison to the MBR filtration process.

Standardised filterability analysis

Evidently in the MBR filtration process the activated sludge properties play an important role, although in a different way compared to the sedimentation process. Contrary to the conventional clarification process in MBR filtration the sludge concentration is not a limiting factor. More important, the crucial characteristic of the sludge is not settleability but filterability. With the *sludge volume index* the conventional activated sludge process already has its standardised analysis to quantify the suitability of the activated sludge to be separated from water. MBR technology requires a similar kind of standardised analysis or parameter to assess the filterability (apart from the discussion what the methodology of that method would be). A bottleneck in this respect is that the settleability is a parameter that can be determined very easily, whereas the filterability is a more complex characteristic. A simple, cheap and at the same time reliable method to characterise filterability is not available yet, but is essential to improve the control over the MBR fouling process.

Effective membrane surface

The *surface area* (m^2) and the *surface loading rate* ($m^3/(m^2 \cdot h)$), two important design parameters for clarifiers, can directly be compared with the variables involved in the MBR filtration process. As with the clarification process also in the MBR process a certain surface area is required to separate the activated sludge and the water. The surface loading rate has the same unity as the flux and is very comparable with it. A crucial aspect of the membrane area (i.e. surface area) in the MBR process is that it is a *dynamic* parameter. This is in contrast with the clarification process, where the surface area is a constant. In the ideal situation the *effective* membrane surface equals the *total* membrane surface, but in practice the effective membrane surface is continuously under pressure as a result of clogging and fouling. Inefficient membrane cleaning measures and poor hydrodynamic circumstances both lead to a decrease of the effective membrane surface. This subsequently leads to uneven distribution of the local fluxes and an increase of the fouling rate.

It is difficult or even impossible to detect a decline of the effective membrane surface on the basis of gross permeability measurements. Evidently these problems gain significance when the scale (i.e. total membrane surface) of the MBR plant increases and the process is more difficult to control. In order to assess the efficiency of the filtration process it is necessary to improve the understanding about the distribution of the total flow over the membrane surface. The process operation (i.e. flux measurements) has to be refined to assess the distribution of the flow over the membrane surface to detect potential clogging problems or to detect insufficient cleaning.

> *Operation of the MBR filtration process should not be based on permeability but on the two following factors: (1) the filterability of the activated sludge and (2) the effective membrane surface available for filtration.*

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Samenvatting

Membraan bioreactoren

Het membraan bioreactor (MBR) proces is een innovatieve technologie voor de behandeling van huishoudelijk afvalwater. De basis voor MBR technologie is het bekende en veel toegepaste actiefslib proces, waarbij een geconcentreerde suspensie micro-organismen de verontreinigingen in het afvalwater biologisch afbreekt. Na het biologische zuiveringsproces dient de biomassa gescheiden te worden van het gezuiverde water. In het conventionele actiefslib proces gebeurt dit door bezinking, terwijl in het MBR proces de scheiding plaatsvindt met behulp van een filtratiestap (voornamelijk door middel van ultrafiltratie membranen). De voornaamste voordelen van het MBR proces zijn de goede effluentkwaliteit en de ruimtebesparing die bereikt kunnen worden ten opzichte van het conventionele actiefslib proces. Het voornaamste nadeel van MBR technologie is dat het proces relatief duur is vanwege de hoge kosten die gerelateerd zijn aan het voorkomen en verwijderen van membraanvervuiling.

Membraan fouling

Membraanvervuiling (*fouling*) kan worden beschreven als het neerslaan van bestanddelen op het oppervlak of in de poriën van het membraan gedurende filtratie of de terugloop van het functioneren van het filtratieproces dat hieruit volgt. Fouling is een complex proces dat het gevolg is van de interactie tussen drie hoofdfactoren: (1) de karakteristieken van het membraan, (2) de bedrijfsvoering van het membraan en (3) de eigenschappen van het te filtreren actiefslib mengsel.
Bovendien kan binnen het totale fouling proces onderscheid gemaakt worden tussen (1) een fysiek verwijderbare (*reversible*) component, (2) een chemisch verwijderbare (*irreversible*) component en (3) een niet verwijderbare (*irrecoverable*) component, die elk met een specifieke snelheid optreden tijdens filtratie.

MBR fouling is een onderwerp dat intensief en op sterk uiteenlopende manieren wordt onderzocht. Echter, elke MBR installatie heeft zijn eigen unieke combinatie van de drie genoemde invloedsfactoren. Dit betekent dat de invloed van een bepaalde factor op fouling altijd in relatie zal staan tot de andere twee en de beschouwde tijdschaal en dus niet zonder meer gegeneraliseerd kan worden. Dit verklaart dan ook de uiteenlopende en soms zelfs tegenstrijdige onderzoeksresultaten die in de vakliteratuur over MBR fouling terug te vinden zijn.

Methodologie

De Technische Universiteit Delft heeft een methode ontwikkeld die het mogelijk maakt om de filtreerbaarheid van actiefslib te karakteriseren: de *Delft Filtration Characterisation method* (*DFCm*). De DFCm bestaat uit een kleinschalige filtratie installatie met een enkelvoudig tubulair ultrafiltratie membraan en bijbehorend meetprotocol. Met de DFCm kunnen actiefslib monsters afkomstig uit elke willekeurige MBR installatie gefiltreerd worden onder identieke hydraulische

omstandigheden en membraan startcondities. De verschillen in de toename van de weerstand kunnen daarom exclusief toegekend worden aan verschillen in de eigenschappen van het slibmonster. De DFCm vormt een bruikbare methode om de invloed van de parameter *filtreerbaarheid* nader in kaart te brengen voor een MBR installatie op praktijkschaal.
De huidige parameter die gebruikt wordt om het filtratieproces te sturen en te analyseren, de *permeabiliteit*, verstrekt geen informatie over de exacte rol van elk van de drie invloedsfactoren op het fouling proces. Met de DFCm kan beter inzicht verkregen worden in de rol van de slibeigenschappen in het filtratieproces.
De belangrijkste beperkende factor van de DFCm is dat de methode geen directe informatie verschaft over het *irreversible* en *irrecoverable* fouling proces, welke de dominante mechanismen zijn op de langere termijn. Een eventuele relatie tussen filtreerbaarheid en lange termijn vervuiling kan op empirische basis geanalyseerd worden door de DFCm resultaten te beschouwen in relatie tot de het verloop van de permeabiliteit in de beschouwde MBR installatie.

DFCm data analyse

Het belangrijkste vervuilingmechanisme op de korte termijn in MBR filtratie is *reversible* koeklaag vorming, ofwel de ophoping van (relatief eenvoudig verwijderbare) deeltjes op het membraanoppervlak. Volgens een theoretische analyse op basis van *backtransport* mechanismen komen in een DFCm experiment alleen deeltjes met een grootte kleiner dan ongeveer 5 μm in aanmerking komen om te accumuleren op het membraanoppervlak (bij het standaard meetprotocol: stroomsnelheid = 1,0 m/s en flux = 80 $L/m^2 \cdot h$).
De totale koekweerstand ΔR [m^{-1}] kan worden uitgedrukt als een functie van de specifieke permeaatproductie V [L/m^2], met drie constanten die gerelateerd zijn aan de eigenschappen van het actiefslib monster: (1) de concentratie aan deeltjes die accumuleert in de koeklaag c_i [g/L], (2) de specifieke weerstand α_R [m/kg] die veroorzaakt wordt door deze deeltjes bij een bepaalde referentie totale weerstand en (3) de samendrukbaarheid coëfficiënt van deze deeltjes s [-].

$$\Delta R = (\alpha_R \cdot c_i \cdot V)^{\frac{1}{1-s}} = (\alpha_R \cdot c_i)^{\frac{1}{1-s}} \cdot (V)^{\frac{1}{1-s}}$$

Elk DFCm experiment levert een dataset op waaruit de waarden van $\alpha_R \cdot c_i$ en s kunnen worden afgeleid. Een nadere analyse geeft aan dat in het relevante bereik van ΔR de samendrukbaarheid coëfficiënt een ondergeschikte rol speelt en dat de totale weerstandstoename voornamelijk bepaald wordt door de coëfficiënt $\alpha_R \cdot c_i$.
Om verschillende DFCm experimenten te kunnen vergelijken op basis van een enkele waarde is de zogenaamde ΔR_{20} waarde [$\cdot 10^{12}$ m^{-1}] geïntroduceerd, welke de weerstandstoename voorstelt na filtratie van 20 L/m^2 permeaat. Op basis van ervaringen met diverse MBR pilot- en praktijkinstallaties is een praktische classificatie opgesteld om de kwaliteit van actiefslib wat betreft filtreerbaarheid te kwalificeren: voor ΔR_{20} lager dan 0,1 is de filtreerbaarheid goed, voor waarden tussen 0,1 en 1,0 redelijk en als ΔR_{20} hoger is dan 1,0 wordt de filtreerbaarheid aangeduid als slecht.

Onderzoeksresultaten

De DFCm is toegepast als instrument om over een periode van enkele maanden de filtreerbaarheid te meten in relatie tot het verloop van de permeabiliteit van de twee MBR praktijkinstallaties in Varsseveld en Heenvliet. Daarnaast zijn meerdere experimenten uitgevoerd met het actiefslib van MBR Heenvliet waarbij de filtreerbaarheid van de monsters gemanipuleerd is door het opleggen van stress omstandigheden in labschaal experimenten.

MBR Varsseveld

In de eerste maanden na het opstarten van MBR Varsseveld werd het functioneren van de installatie sterk beïnvloed door de aanvoer van afvalwater van een lokale kaasmakerij. Dit afvalwater bleek een chemisch polymeer te bevatten dat door de membranen werd tegengehouden maar niet door de biomassa afgebroken kon worden. In de full-scale installatie werden hierdoor aanzienlijk fouling problemen ondervonden. Ook met de DFCm werd aangetoond dat de filtreerbaarheid van het actiefslib extreem slecht was. Nadat besloten was om de kaasfabriek van het riool af te koppelen vertoonden zowel de permeabiliteit in de full-scale installatie als de filtreerbaarheid (zoals gemeten met de DFCm) een aanzienlijke verbetering. Aan de hand van de DFCm experimenten kon aangetoond worden dat de filtreerbaarheid van het actiefslib inderdaad de beperkende factor was in het filtratie proces (en niet de bedrijfsvoering van het filtratieproces). Gezien de uitzonderlijke omstandigheden konden er geen duidelijke conclusies getrokken worden wat betreft de exacte karakteristieken die de filtreerbaarheid van het actiefslib beïnvloed hebben.

Mbr Heenvliet

Het actiefslib van MBR Heenvliet is gedurende een jaar geanalyseerd op filtreerbaarheid, met een bemonsteringsfrequentie van circa een maal per week. Met uitzondering van de opstartperiode was de filtreerbaarheid van het slib goed tijdens de meetperiode. Toch vertoonde de permeabiliteit een sterk dalende trend. Naderhand bleek dat er problemen waren met de integriteit van de membranen (loslatende lijmlagen) en dat er op grote schaal clogging (slibophoping) was opgetreden in de membraanmodules als gevolg van gebrekkige beluchting. In dit geval heeft de DFCm aangetoond dat de filtreerbaarheid van het actiefslib niet de beperkende factor was in het filtratieproces. De filtreerbaarheid vertoonde correlatie met de temperatuur en de slib volume index van het actiefslib, maar niet met de concentratie *Soluble Microbial Products* die door veel onderzoekers in verband wordt gebracht met fouling.

Stress experimenten

Actiefslib monsters afkomstig uit MBR Heenvliet zijn onderworpen aan drie verschillende stress omstandigheden in labschaal experimenten: (1) langdurige zuurstofloze omstandigheden, (2) kortstondige hoge schuifspanning en (3) abrupte temperatuursverandering.

De resultaten tonen aan dat alle drie de opgelegde stress omstandigheden in meer of mindere mate zorgen voor deflocculatie van de slibvlokken die gepaard gaat met een verslechtering van de filtreerbaarheid. De deflocculatie uit zich in het vrijkomen van *Soluble Microbial Products* en *colloïdale* deeltjes vanuit de slibvlokken in het vrije water. Andersom toont het slib een sterke neiging tot flocculatie (en een bijgaande verbetering van de filtreerbaarheid) wanneer het na de stress omstandigheden weer wordt blootgesteld aan voortdurende beluchting. Het volume aan colloïdale deeltjes toont een betere relatie met de filtreerbaarheid dan de concentratie SMP.

Conclusies

The DFCm heeft zich bewezen als een geschikte methode om de filtreerbaarheid van slibmonsters afkomstig uit full-scale installaties te karakteriseren. Met deze kennis over de filtreerbaarheid kan het functioneren van een full-scale installatie beter in kaart gebracht worden omdat een veronderstelde terugloop van de permeabiliteit specifiek kan worden toegewezen aan slechte filtratie eigenschappen van het slib of tekortkomingen in de bedrijfsvoering van het filtratieproces.

Fysische en chemische analyses tonen aan dat de filtreerbaarheid van actiefslib (met een ultrafiltratie membraan) nauw gerelateerd is aan het volume colloïdale deeltjes (0.01-1 μm) in het vrije water. De concentratie *Soluble Microbial Products*, een parameter die door veel onderzoekers in verband wordt gebracht met fouling, blijkt een zwakke indicator voor de filtreerbaarheid omdat de algemeen gebruikte analyse methode geen onderscheid maakt tussen opgeloste SMP bestanddelen die niet en colloïdale bestanddelen die wel door het membraan worden tegengehouden.

De concentratie colloïdale bestanddelen en daarmee de filtreerbaarheid van actiefslib is sterk gerelateerd aan de mechanismen van flocculatie en deflocculatie. Om een actiefslib te genereren met goede filtratie eigenschappen is een stabiel biologisch zuiveringsproces nodig, bij voorkeur met een lage slibbelasting. In geval van (plotselinge) stress omstandigheden kunnen aanvullende operationele maatregelen toegepast worden om het flocculatie proces en de filtreerbaarheid te verbeteren (bv. het verlengen van de aërobe hydraulische verblijftijd of het doseren van een coagulant).

Aanbevelingen

De huidige parameter die toegepast wordt om het filtratieproces te analyseren en te regelen is de permeabiliteit. De permeabiliteit kan echter beschouwd worden als een zwakke parameter omdat deze alleen informatie oplevert over het gevolg van fouling en niet over de oorzaken ervan.
In dit opzicht zou de bedrijfsvoering van de filtratiestap meer overeenkomst moeten vertonen met het bezinkingsproces in conventionele actiefslib systemen. Het conventionele bezinkingsproces heeft zijn eigen gestandaardiseerde methode om de bezinkbaarheid van het actiefslib te karakteriseren (de *slib volume index*). Ook het MBR proces heeft een dergelijke gestandaardiseerde kwaliteitsparameter nodig. Een goede filtreerbaarheid kan beschouwd worden als het uitgangspunt voor een goed functionerend filtratieproces. Naast de filtreerbaarheid is een belangrijke parameter in het filtratieproces het membraanoppervlak dat beschikbaar is voor filtratie. Dit zogenaamde effectieve membraanoppervlak staat voortdurend onder druk als gevolg van clogging en fouling. Wanneer het effectieve membraanoppervlak daalt zal de lokale flux bij het membraanoppervlak dat nog wel beschikbaar is voor filtratie

toenemen. Hierdoor zal ook de snelheid van vervuiling toenemen, ongeacht de filtreerbaarheid van het slib. Om ongelijkmatige verdeling van het effectieve membraanoppervlak te detecteren zijn gedetailleerdere (lokale) fluxmetingen nodig.

Dankwoord

Den Haag, December 2009.

Na een periode van ruim vier jaar onderzoek komt er met dit proefschrift een einde aan mijn tijd als promovendus aan de TU Delft. Voordat ik iedereen wil bedanken die heeft bijgedragen aan de totstandkoming ervan is dit dankwoord natuurlijk ook een goed moment voor een korte teruglik.

Mijn onderzoeksdoel, de vervuiling van membraanfilters doorgronden, bleek een hele uitdaging. Microbiologie, chemie en vloeistofmechanica zijn afzonderlijk al complexe vakgebieden, in de wondere wereld van de afvalwaterzuivering komen ze allemaal samen. Dat maakt het er niet eenvoudig op, maar zeker wel interessant. De basis van de afvalwaterzuivering, bacteriën die de verontreinigingen in ons afvalwater biologisch afbreken, is voor mij (als civiel-technicus) op zich al een wonderlijk proces. Dit principe vervolgens gebruiken om op grote schaal afvalwater steeds beter en duurzamer te zuiveren ervaar ik als een uitdagend vakgebied om in werkzaam te zijn. Vooraf is de uitkomst van een promotie onderzoek geen zekerheid, maar ik kijk met een goed gevoel terug op de resultaten in dit proefschrift. De membraanvervuiling puzzel is nog niet opgelost, maar ik denk dat dit onderzoek een goede bijdrage levert aan het sorteren van de stukjes en er her en der ook een aantal inpast.

De persoonlijke hoogtepunten van mijn promotietijd zijn eenvoudig aan te wijzen: de congressen! (en deze in combinatie met de locaties ervan en het schappelijke aantal vakantiedagen bij de TUD) Dankzij mijn onderzoek heb ik het voorrecht gehad om te reizen naar plekken waar ik anders waarschijnlijk niet snel op of aan toe gekomen zou zijn. Reizen door Korea is een onvergetelijke ervaring. En ik had nooit gedacht om een paar jaar na mijn stage al weer terug te mogen zijn in het fantastische Australië. Hoogtepunt was ook zonder meer mijn laatste congres op Hawaii. Het was praktischer geweest als ik Maui en The Big Island nu definitief had kunnen afvinken op mijn to do lijstje, maar ik heb mezelf stellig beloofd dat ik daar nog eens terugkeer…

Al voelde het soms niet zo als ik veel te vroeg in de regen een slibmonster stond te scheppen: promoveren doe je niet alleen. Velen hebben mij de afgelopen tijd bijgestaan en die wil ik graag bedanken.

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culinaire smaak en interessante theorieën over voetbal, tennis en wielrennen. Ik koester dan ook zeer goede herinneringen aan de vele Afvalwaterberaad diners, de congressen die we bezocht hebben en de studiereizen naar Toulouse en Berlijn.

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Stefan

Curriculum Vitae

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