Igor Ivanovic

Application of biofilm membrane bioreactor (BF-MBR) for municipal wastewater treatment

Thesis for the degree of Philosophiae Doctor

Trondheim, May 2011

Norwegian University of Science and Technology
Faculty of Engineering Science and Technology
Department of Hydraulic and Environmental Engineering

NTNU – Trondheim
Norwegian University of
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Abstract

The biofilm membrane reactor (BF-MBR) is a wastewater treatment system that combines biological wastewater treatment with a biofilm process and with membrane separation for clarification and purification of biologically treated wastewater. Work in this thesis is experimentally based on laboratory pilot scale systems specially built, developed and modified for the research purposes in this thesis.

The work in this thesis is presented through six main themes (research chapters (RC)):

RC1: Overview of previous research on knowledge on particle separation in a moving-bed-biofilm reactor (MBBR) process
RC2: Overview of previous research on membrane bioreactors (MBR) based on attached (biofilm) growth.
RC3: Results from empirical studies on the influence of aeration on membrane performances in the BF-MBR
RC4: Results from mathematical and empirical studies on influence of membrane reactor design on membrane performances in the BF-MBR
RC5: Results from empirical studies on influence of nitrogen removal on membrane performances in the BF-MBR
RC6: Results from empirical studies on influence of coagulation and flocculation on overall performances in the BF-MBR

The main contributions (C) are:

C1: An overview of the research literature on empirical studies of knowledge on MBR with biofilm implementations and separation techniques in the moving-bed-biofilm reactor
C2: Proposing a method for defining optimum aeration rates for the membrane unit based on minimizing the amount of submicron particles.
C3: Improvement in membrane performances by changing the membrane reactor geometry.
C4: Demonstration of feasibility of sustainable operation of proposed system with biological nitrogen removal treatment configurations.
C5: Improvement of membrane and overall process performance with addition of different additives.

Results from the research work in this thesis are based on five main empirical studies using a pilot scale biofilm MBR (BF-MBR) setup where overall system performance and membrane performance has been studied. The thesis is structured as a paper collection based on seven papers, where the first two are review papers and other five are results of the original research. In addition, there are three publications based on original research from the study that were published during the early stages of the project studies, presented only with the references and abstracts as secondary papers.

Abstract in Norwegian

En biofilm membran reaktor (BF-MBR) er et rensesystem for avløpsvann som kombinerer biologisk rensing ved bruk av en biofilm prosess, og med membranseparasjon for filtrering og rensing av det biologisk renset avløpsvannet. Denne avhandlingen er basert på eksperimentelle arbeider med pilot enheter og laboratoriesystemer som er spesielt bygd, utviklet og modifisert for forskningsformålene i dette studiet.

Arbeidet i denne avhandlingen er presentert gjennom seks hovedtema (forskning kapitler (FK)):

FK1: Oversikt over tidligere forskning og kunnskap om partikkelseparasjon i en moving-bed-biofilm reaktor (MBBR) prosess
FK 2: Oversikt over tidligere forskning på membran bioreaktorer (MBR) basert på vekst av fastsittende biomasse (biofilm).
FK 3: Resultater fra empiriske studier om hvordan lufting påvirker membranens prosessegenskaper i en BF-MBR
FK 4: Resultater fra matematiske og empiriske studier om hvordan membranreaktor design påvirker membranens separasjonsegenskaper i en BF-MBR
FK 5: Resultater fra empiriske studier om hvordan membranprosessen påvirkes av nitrogenfjerning i en BF-MBR
FK 6: Resultater fra empiriske studier om hvordan koagulering og flokkulering påvirker den generelle prosessen i en BF-MBR

De viktigste bidragene (B) er:

B1: En oversikt over forskningslitteratur om empiriske studier og kunnskap om MBR med anvendelse av biofilm prosesser og separasjon teknikker brukt i en moving-bed-biofilm reaktor
B2: Foreslag på en metode for å definere optimal lufting for membranenheten basert på å minimere mengden av sub-mikron partikler.
B3: Forbedring i membranens ytelsesevne ved å endre på membranreaktorens geometri.
B4: Demonstrasjon av mulighetene for en bærekraftig drift av foreslåtte system med konfigurasjoner av biologisk nitrogenfjerning.
B5: Forbedring av både membran og generelle prosessytelser ved bruk av forskjellige tilsetningsstoffer.

Resultater fra forskningsarbeidet i denne avhandlingen er basert på fem empiriske studier utført med en pilot biofilm MBR (BF-MBR) apparatur, der generelle systemytelse og membranytelse er studert. Avhandlingen er strukturert som en samling av syv publikasjoner, der de to første er ”review” artikler og følgende fem artikler er resultatene fra original forskningen. I tillegg er tre publikasjoner basert på original forskning fra studiet som ble publisert i en tidlig fase av arbeidet presentert som sekundære artikler, kun angitt med referanser og sammendrag.

Preface

This thesis is submitted to the Norwegian University of Science and Technology (NTNU) for partial fulfillment of the requirements for the degree of philosophiae doctor.

This PhD work has been performed at the Department of Hydraulic and Environmental Engineering, Faculty of Engineering Science and Technology, Norwegian University of Science and Technology, with professor TorOve Leiknes as supervisor.

Thesis has been financed by Norwegian Research Council (NRF) / NTNU faculty fellowship. Experimental part of the thesis was supported by Krüger Kaldnes, Norway, with a biofilm reactor, Zenon GE, Canada with membrane modules and Kemira, Finland with coagulants.

The thesis is based on experimental research of an advanced system for wastewater treatment that is comprised of biofilm technology for biological wastewater treatment and membrane technology for advanced particle separation and purification.

The experimental part of the thesis was carried out in the wastewater treatment laboratory of the Department of Hydraulic and Environmental Engineering, Faculty of Engineering Science and Technology, Norwegian University of Science and Technology.

Acknowledgements

First of all I would like to thank to prof. Vojislav Novakovic, NTNU for his support and encouragement to come to Norway and do PhD studies.

Very special thanks to my advisor Tor Ove Leiknes, NTNU for permanent support from the very first day of this project, for his patience, encouragement and dedication to this project.

I would like to thank to my colleagues Jirachote Phataranawik and Cheng Sun, for their enormous help in wastewater treatment laboratory and for great time we spent in the lab in last 4 years.

Further more, many thanks to, Thomas Meyn, Astrid Vik Bjørkøy, Kamal Azrague, Gema Sakti Raspati, Ciprian Scurtu and Sumihar Silalahi for great, unforgettable time that we spent together.

Gøril Thorvaldsen, Trine Margrete Hårberg Ness, Torgeir Jensen and Arne Grostad, deserve warmest thanks for helping me in the analytical lab and the workshop.

I would like to thank to my parents and my sister for constant support during very difficult moments for me, and without their help and support I would have never finished this work. Thank you.

Contents

Abbreviations

A_{436}	color, absorbance at 436 nm wavelength
aBF-MBR	assisted biofilm membrane reactor
BF-MBR	combination of moving-bed-biofilm-reactor and membrane reactor
BNR	biological nitrogen removal
BOD	biochemical oxygen demand
CAU	coefficient of additive utilization
COD	chemical oxygen demand
DOC	dissolved organic carbon
FCOD	filtered chemical oxygen demand
FI	flocculation index
HRT	hydraulic retention time
Ks	separation factor (coefficient)
LMH	membrane flux, $Lm^{-2}h^{-1}$
MBBR	moving-bed-biofilm reactor
MBR	membrane bioreactor
MF	microfiltration
MLSS	mixed liquor suspended solids
MR	membrane reactor
MSH-MR	modified sludge hopper membrane reactor
MW	molecular weight
NR	nitrification
NTNU	Norwegian University of Science and Technology
NH_4-N	ammonia concentration
NO_3-N	nitrate concentration
OLR	organic loading rate
pBF-MBR	pure biofilm membrane reactor
PDN	post-denitrification
PSD	particle size distribution
R	recovery
RO	reverse osmosis
SAD_m	specific aeration demand per membrane area
SBR	sequencing batch reactor
SH-MR	sludge hopper membrane reactor
sMBR	submerged membrane reactor
SMP	soluble microbial products
SMPc	concentration of carbohydrates in SMP
SMPp	concentration of proteins in SMP
SP	sampling point
SRT	solids retention time
SS (MLSS)	suspended solids
SUVA	specific UV absorbency
TMP	transmembrane pressure
TN	total nitrogen
TOC	total organic carbon

Introduction

By the year 2025 it is expected that 60 % of the world population will live with water scarcity if current water consumptions remain at the same current level [1]. It is expected that with further development of human society the need for fresh water will keep increasing. More than 90% of available fresh water resources will be consumed in the next fifteen years [2]. Therefore, water reuse and reclamation is inevitable in the years to come. Current wastewater technologies will have to be upgraded and/or replaced with new and more advanced water treatment technologies that can provide high quality of treated water, following sustainable practices, being less expensive to operate while meeting more strict legal regulations.

The membrane bioreactor (MBR) technology that comprises the activated sludge process and membrane technology has been recognized as a technology for advanced wastewater treatment. Several advantages of MBRs over conventional technologies are recognized, such as the high quality and hygienic effluent achieved, reduced footprint, lower sludge production, controlled biomass separation and more advanced nutrient removal ability.

Biofilm process treatment offers several advantages over other wastewater treatment processes, such as operational simplicity compared to activated sludge, higher biomass activity due to accumulation of highly specialized microorganisms and higher resistance of biomass to toxic substances. Implementation of a biofilm process instead of an activated sludge process leads to creation of a novel treatment configuration, a biofilm membrane bioreactor (BF-MBR), which has the possibility to combine the best characteristics of MBR technology and biofilm wastewater treatment processes.

Problem Outline

Implementation of a biofilm reactor instead of an activated sludge reactor in MBR technology has not been studied in detail, and has been poorly documented in available literature as of date. Understanding the interaction of the effluent from a biofilm reactor and the membrane is crucial for sustainable development of this concept. Characteristics of the effluent from a biofilm reactor differ significantly from that of an activated sludge process, which ultimately can create different membrane filtration performances. Therefore, investigation of this interaction through different process configurations and different operational conditions is important for understanding the potential capabilities of a BF-MBR process.

[1] S. Hoffmann, Planet water: Investing in the world's most valuable resources, 1st Ed. John Wiley and Sons (2009)
[2] S. J. Khan, S. Hyas, S. Javid, C. Visvanathan, V. Jegatheesan, Performance of suspended and attached growth MBR systems in treating high strength synthetic wastewater, Bioresource Technology in press (2010)

Research Context

This project has been funded by the Norwegian Research Council (NRF) and partially funded by AnoxKaldnes. The thesis is a result of experimental work on development of a new system for advanced wastewater treatment based on coupling a biofilm reactor and membrane separation technology.

The aim of this study is: 1) to investigate the potential of coupling a biofilm reactor (the moving-bed-biofilm reactor in particular) with membrane filtration, 2) to give directions for development of optimal operational conditions that lead to sustainable operations, 3) to provide a better understanding of membrane fouling phenomena in the proposed system configuration and 4) to suggest possible methods for membrane fouling minimization and control.

In order to relate findings from the pilot plants used in this study with real life situations, it was decided that the complete research work should be done under as real conditions as possible, *i.e.* with real municipal wastewater and with commercially available products such as carriers for biofilm reactor, membrane materials and commercially available additives.

Research topics (RT)

Work in this study can be divided into four research topics (RT)

RT1: Optimization of aeration intensity for membrane air scouring purposes as a tool for minimizing fraction of submicron particles and membrane fouling
RT2: Design of the membrane reactor as a tool for improved membrane filtration characteristics in BF-MBR
RT3: Investigation of nitrogen removal by biological treatment configuration and its effect on the overall performance of the BF-MBR
RT4: Addition of additives at optimal dosage for improved membrane and overall performance in BF-MBR

The research topics are covered and presented in five research papers, where each paper covers one topic except topic 4 which is covered by the two last papers. The results from the early studies are mentioned and referenced as the secondary papers in the Appendix B

Overview of papers

P1 Particle separation in moving bed biofilm reactor applications and opportunities,

I. Ivanovic and T.O. Leiknes,
Submitted to journal Separation Science and Technology

Relevance to this thesis: This paper presents a review of findings of studies on particle separations after a moving-bed-biofilm reactor, which has been used in experimental research in this study. It describes available conventional and new techniques for solids/liquid separations after biofilm reactors in wastewater treatment applications. The paper gives an outlook of application of advanced techniques such as membrane separations for solid/liquid separations after a moving-bed-biofilm process

Contribution to the content of the paper: from the review of available literature, adapted figures (illustrations) based on diagrams in original literature sources were created, and implementations of new configurations for application of submerged membrane reactors proposed.

P2 The biofilm membrane bioreactors (BF-MBR) - a review-

I. Ivanovic and T.O. Leiknes,
Submitted to journal of Desalination and Water Treatment

Relevance to this thesis: This paper presents a review of findings of experimental studies on MBRs with biofilm implementations. It describes available approaches for biofilm implementations and presents an attempt to systemize research work in this field as of date. Systematization has the purpose of positioning the research work in this thesis in perspective to current state-of-the-art in this field.

Contribution to the content of the paper: from available literature sources, figures based on the results found in literature were created and a systematization of BF-MBR in two groups proposed, based on their characteristics and defined as:

1) assisted biofilm membrane reactor (aBF-MBR) and
2) pure biofilm membrane reactor (pBF-MBR),

This distinction more precisely describes the nature of the processes and role of the biofilm in the BF-MBR process.

P3 Impact of aeration on particle colloidal fraction in the biofilm membrane reactor (BF-MBR)

I. Ivanovic and T.O. Leiknes
Published in Desalination 231 (2008) 182-190

Relevance to this thesis: This paper presents findings of experimental studies on BF-MBRs within research topic one (RT01). Submicron particles are identified in early studies as a main contribution to membrane fouling. This paper investigates the impact of aeration intensity on the characteristics of the submicron particles resulting in the membrane reactor and suggests a simple method for estimating sufficient membrane aeration for sustainable operation with minimizing enlargement of submicron particles.

Contribution to the content of the paper: I designed and performed experimental part, performed data analysis and wrote the most of the paper.

P4 Membrane reactor as a tool for better membrane performance in a biofilm - MBR (BF-MBR)

I. Ivanovic and T.O. Leiknes
Published in Desalination and Water Treatment 25 (2011) 259–267.

Relevance to this thesis: This paper presents findings of experimental studies on BF-MBRs within research topic two (RT02). Reduction of membrane fouling by alternative reactor designs was studied where mathematical calculations were experimentally verified. This paper suggests alternative reactor designs to a completely mixed flow reactor typically used in MBRs as a tool for enhanced membrane filtration performance.

Contribution to the content of the paper: I designed and performed experimental part, performed data analysis and wrote the most of the paper.

P5 Impact of denitrification on the performance of a biofilm-MBR (BF-MBR).

I. Ivanovic and T.O. Leiknes

Presented at MDIW 2010, Trondheim, Norway
Accepted to journal of Desalination (doi:10.1016/j.desal.2011.04.026)

Relevance to this thesis: This paper presents findings of experimental studies on BF-MBRs within research topic three (RT03). Impact of post-denitrification on membrane performance in a BF-MBR was not previously been studied. This paper presents results of overall process performance, comparing a process configuration for denitrification to a system for nitrification only.

Contribution to the content of the paper: I designed and performed experimental part, performed data analysis and wrote most of the paper.

P6 Effect of addition of different additives on overall performance of bioflim - MBR (BF-MBR)

I. Ivanovic and T.O. Leiknes

Presented at AMS6 and IMSTEC10, 2010, Sydney, Australia
Accepted in journal of Desalination and Water Treatment

Relevance to this thesis: This paper presents findings of experimental studies on BF-MBRs within research topic four (RT04). The effectiveness of different additives commonly used in MBR technology as membrane performance enhancers was studied in a BF-MBR configuration for the first time. Five different additives at applicable dosages were applied and their effectiveness on membrane performance compared. The effect on reduction of colloidal organic matter by applied additives was specially studied.

Contribution to the content of the paper: I designed and performed experimental part, performed data analysis and wrote the most of the paper.

P7 Improved performance through particle surface modifications by coagulation with inorganic coagulants in a BF-MBR

I. Ivanovic and T.O. Leiknes

Submitted to journal of Separation and Purification Technology

Relevance to this thesis: This paper presents additional findings of experimental studies on BF-MBRs within research topic four (RT04). The effectiveness of two chosen inorganic coagulant on reduction of number of submicron particles and their surface area was studied in the BF-MBR process. The effect on reduction of number and surface area of particles in the submicron range by applied additives was specially studied.

Contribution to the content of the paper: I designed and performed experimental part, performed data analysis and wrote most of the paper.

Thesis Structure

The thesis is structured in six chapters and two appendixes as follow:

Chapter 1: Introduction

Chapter 2: State of the Art

Chapter 3: Material and methods

Chapter 4: Results

Chapter 5: Evaluation of Results

Chapter 6: Conclusion and future work

Appendix A: Selected papers

Appendix B: Secondary papers

State of the art

Summary of the review papers

Paper P1:

Particle separation in moving bed biofilm reactor– applications and opportunities

The main goal of this thesis is to investigate membrane filtration as a new technique for solids/liquid separation after a moving-bed-biofilm reactor. Current status in the field of solid/liquid separation techniques after a moving-bed-biofilm reactor is given in this paper. Conventional techniques such as sedimentation, flotation and depth filtration are reviewed based on available literature and compared in terms of treatment efficiency, *i.e.* efficiency of solids removal, and compared based on energy consumption. It is concluded that traditional technologies are not capable of providing complete removal of suspended matter and their efficiency is evaluated based on important operational parameters for each separation technique. New techniques such as disc filtration and membrane separation are also reviewed. Application of submerged membrane reactor (sMBR) systems was emphasized and two possible configurations for implementation of this technology are suggested.

Paper P2:

The biofilm membrane reactors (BF-MBR) - a review-

With development of MBR technology, asides from main stream efforts, several research groups have been working to supplement or replace biological suspended growth in MBR with attached growth, searching for new possibilities to overcome limitations of conventional MBR processes based on activated sludge. The total number of internationally available publications in the field of biofilm applications in MBR shows that this topic has not yet received significant attention in the research community in comparison with the amount of research work related to conventional MBR. The second paper (P2), gives a review and systematization of research work done on MBR with biofilm implementations. A classification of MBR systems with biofilm implementations in two generic groups is proposed:

1. assisted biofilm MBR (aBF-MBR) - where addition of attached (biofilm) growth to conventional activated sludge MBR (AS-MBR) assist biodegradation, and
2. pure biofilm MBR (pBF-MBR) – where a biofilm reactor exclusively performs the biodegradation without presence of activated sludge

Implementations of biofilms in MBR have been shown as mostly successful in terms of enhanced nutrient removal and mainly improved membrane performances. However some studies showed a negative impact of applied biofilm, which was elaborated and commented in the paper.

Materials and methods

The basis of the experimental research in this study was a pilot scale set up (Figure 1) that consisted of a moving bed biofilm reactor and a submerged low pressure polymeric membrane reactor. The pilot plant was operated with real municipal wastewater taken from a public sewer line in Trondheim municipality, Norway. Detailed descriptions of each configuration modification and operational condition tested are given in each research paper presented. All measurements are done according national and international standards with detailed descriptions also given in each paper.

A schematic of the pilot plant with basic technical specifications and characteristics are given below. Figure 1 and Table 1.

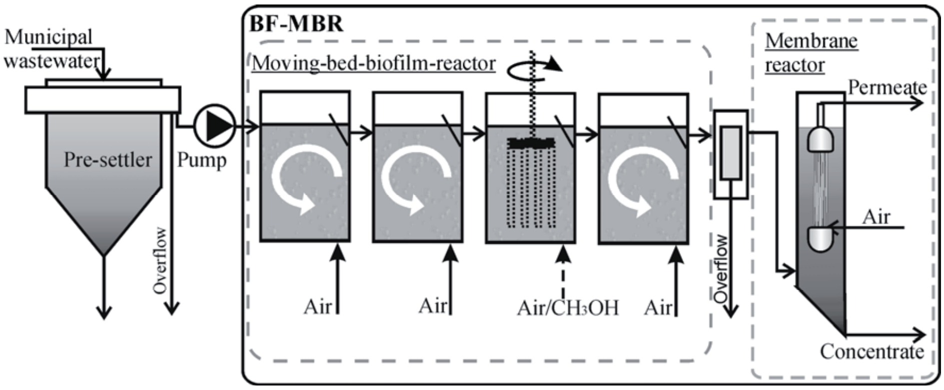

Figure 1. Basic configuration of BF-MBR used in the thesis for experimental research

Table 1. Basic characteristics pilot plant that was subject in this study

Moving-bed-biofilm reactor
Number of reactors (in series): 4
Volume of each reactor: 65 L
Type of carrier: K1: Filling fraction – 67 % of reactor volume
Surface area for biofilm growth - 335 m^2/m^3,Total growth area - 84.5 m^2
Membrane reactor
Zenon ZW10 Membrane Pilot Module
Type of membrane – Hollow fiber with configuration – outside/in
Nominal pore size – 0.04 micron
Volume of membrane reactors – 9-41 L
Operating characteristics
Flux – 22 - 52, Backwash flux – 38-58 LHM
Operating cycle: 4.75 min. production and 0.25 min. backwashing
Recovery ~ 90-96 %

Results

Summary of the research papers

Research paper P3:

Impact of aeration rates on particle colloidal fraction in the biofilm membrane bioreactor (BF-MBR)

In this study a method for estimation of aeration rate in BF-MBR is proposed, taking in account permeability drop for a certain aeration rate and distribution of particles affected by the intensity of aeration. Different aeration rates were tested (from highest to lowest) with a daily shift towards lower aeration rates. The measured change in TMP (*i.e.* permeability decline) was related to the amount of the highest fraction of submicron particles measured in the reactor. Published literature results suggest that there is a threshold rate of air scouring intensity above which no significant effect or improvement of membrane performance is observed. However, operating the membrane with air scour rates close to the threshold value could have a negative effect on particle size distribution, inducing higher amounts of submicron particles, which have been seen as potential foulants responsible for irreversible membrane fouling. Therefore, a simple and quick to perform method was proposed where peak values from particle size distribution (PSD) measurements and measured fouling rates were plotted against specific aeration demand to identify which could be the most suitable operating condition. This approach represents an alternative strategy to defining optimal operational conditions for the system with respect to aeration rates. In this study, for the system configuration that was subject of the study, optimal aeration rates were found to be not lower then 1.68 and not higher then 3.37 $Nm^3m^{-2}h^{-1}$.
Figure 2 illustrates the procedure for defining an optimal specific aeration demand based on measurements and specifications for the pilot plant used in this study.

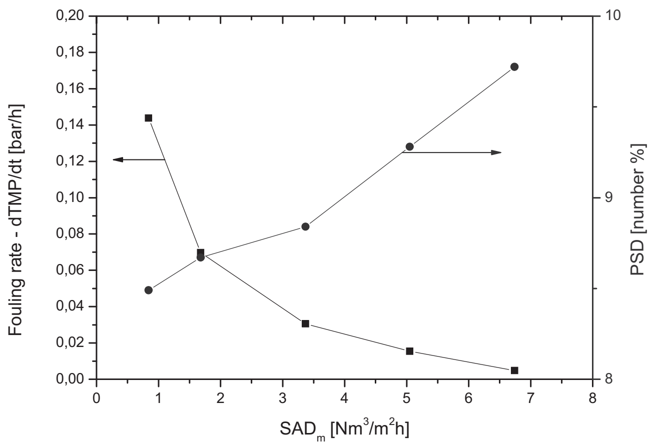

Figure 2. Fouling rate and PSD values of the most present submicron fraction for different aeration rates (SAD_m)

Research paper P4:

Membrane reactor design as a tool for better membrane performance in biofilm MBR (BF-MBR)

In this study the advantage of having an integrated flocculation zone and sludge hopper under the membrane unit is demonstrated. Redesigning a completely mixed flow membrane reactor (CM-MR) into a reactor design with a sludge hopper (SH-MR) results in a lower amount of suspended solids and total organic matter around the membrane area. Lower solids and organic loads on the membrane surface lead to lower fouling rates and longer operational cycles. By introducing the effluent from the biofilm reactor into a flocculation zone beneath the membrane unit and aeration device, a reduced amount of submicron particles around the membrane filtration area was also achieved. Additionally, a typical problem of bulking sludge from aerobic biological treatment was solved by proposing a modified sludge hopper membrane reactor design (MSH-MR). By rearranging the membrane reactor configuration design a reduction of suspended matter around the membrane area in the SH-MR was approximately 4.5 times lower than for the CM-MR design, while approximately 6 times lower in the MSH-MR design. Lower fouling rates in both the SH-MR and MSH-MR designs were attributed both to lower suspended matter and reduced amount of submicron particles.

Results for TMP development over time, measured before and after backwashing, are presented in Figure 3 for all three reactor design options investigated. The enhanced performances of the SH-MR and the MSH-MR designs under the conditions tested are quite apparent.

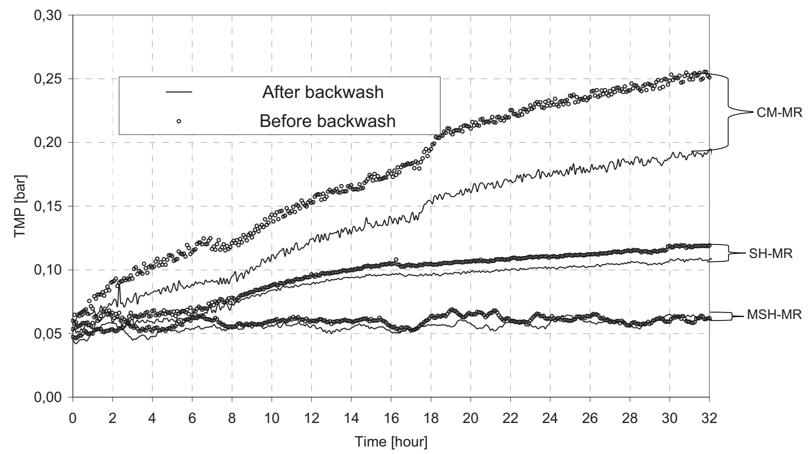

Figure 3. TMP before and after backwash for CM-MR, SH-MR, MSH-MR. Production flux 35 LMH, and backwash flux 38 LMH

Research paper P5:

Impact of denitrification on the performance of a biofilm-MBR (BF-MBR).

Post-denitrification (PDN) as an option for a compact biological nitrogen removal scheme in a biofilm-MBR (BF-MBR) has been studied. The PDN configuration was compared with a nitrification (NR) configuration with respect to membrane fouling. Full nitrification was obtained in both process configurations, while stable nitrogen removal rates about 80% were obtained by applying a constant C/N ratio of 4.5 in the PDN scheme. Both process configurations were able to manage a sustainable membrane operation over a period of one month with an applied production flux of 30 LMH. Effluent properties from the PDN process showed better settling characteristics of the solids in the concentrate in the membrane filtration unit, resulting in lower solids loading and better solids separation. However, higher membrane fouling was observed in the PDN configuration. An increased content of colloidal organic matter and mostly low molecular weight (MW) residual organics were indentified for the PDN vs. the NR processes. The higher fouling rates for the PDN configuration were attributed to higher adsorption and pore blocking mechanisms caused by these fractions. Periodical backwashing with moderate air scouring rates was found to be a suitable strategy for fouling control in a BF-MBR with a post-denitrification configuration.

Figure 4 illustrates the overall membrane performances for the two configurations that were subject of this study.

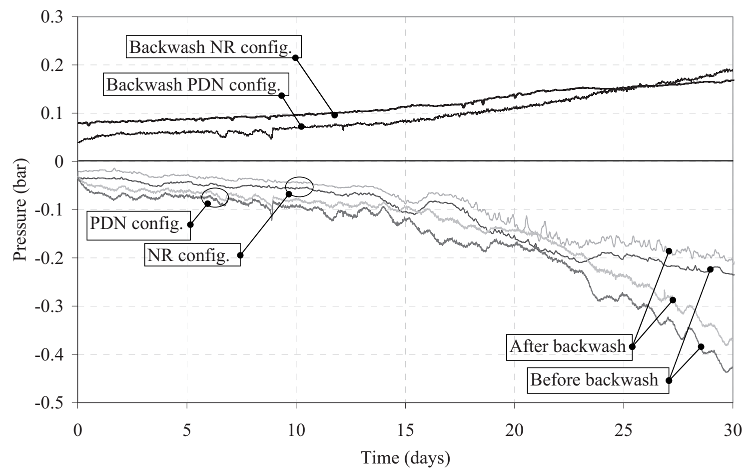

Figure 4. Overall membrane performance for PDN and NR configurations

Research paper P6:

Effect of addition of different additives on overall performance of bioflim - MBR (BF-MBR)

In MBR technology, addition of inorganic coagulants and cationic polymers has been reported in literature to lead to an improved membrane performance. In this study, five additives, two iron based and two alum based coagulants and one modified cationic polymer, were investigated in relation to reduction of soluble organic matter and membrane fouling in a BF-MBR process. Optimal dosage was determined based on maximum values of a defined coefficient additive utilization (CAU). The amount of reduction of colloidal organic mater (expressed as soluble microbial products (SMP), DOC and FCOD) and retained by the membrane was related to the measured membrane fouling rates. Iron chloride at the higher dosage chosen showed the best performance with respect to reduced fouling rates, where up to a seven times reduction was measured. For polymerized alum a reduction of about 3 times was measured. Higher basicity of the polymerized inorganic coagulant did not result in improved membrane performance. The modified cationic polymer showed a good potential for instantaneous fouling reduction, however, a continuous dosing strategy was found not to be applicable without monitoring the polymer concentration inside the membrane reactor. Reduction in fouling rates was found to correlate better to reduction of SMPc, compared to SMPp and DOC. However, FCOD was also seen as having good potential as a fouling predictor parameter. Synergetic effects of high total phosphorus removal rates and reduced fouling rates give an advantage to the iron chloride coagulant over the others tested additives in this study

Figure 5 shows measured relations between the amount of certain parameters retained by the membrane and membrane fouling rates, and the applicability of FCOD measurements as a potential membrane fouling indicator.

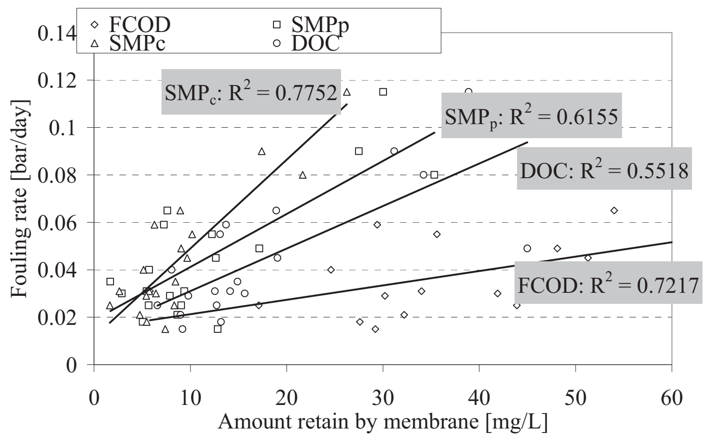

Figure 5. Correlation between given parameters and membrane fouling

Research paper P7:

Improved performance through particle surface modifications by coagulation with inorganic coagulants in a BF-MBR

In addition to the previous study, enhanced performance of the membrane reactor in a BF-MBR was studied with respect to particle surface modifications by coagulation with inorganic coagulants. In addition, the study included investigating the reduction of number of submicron particles, enlargement of suspended flocs and reduction of the organic colloidal content. Coagulants added in the membrane filtration stage of the BF-MBR had a positive effect on improvement of mixed liquor filterability and dewaterability, resulting in lower membrane fouling rates. Modified jar tests were done in aerated beakers (*i.e.* simulating conditions in the membrane reactor) to determine applicable dosages by investigating effects on flocculation index, number, size and charge of colloidal particles, surface area of submicron particles, solids and organic colloidal content. From this assessment, two dosages of 9 ppm and 22,5 ppm were chosen and tested in two sets of experiments in two membrane rectors operated in parallel. Membrane fouling was reduced in a similar manner for both lower dosages of alum and iron applied, however, iron performed better at the higher dose. Alum was able to provide a higher positive charge per unit of applied coagulant, and better addressed removal of colloidal organic content measured as FCOD. Iron was more effective in enlarging surfaces of flocs in the bulk phase, leading to more adsorption of submicron particles on floc surfaces and probably promotion of a protective cake layer on the membrane surface. Improvement in membrane performances was found to correlate to reduction of measured surface area of particles in the size range 0.04 – 0.3μm, enlargement of floc surface area for particles > 8 μm, as well as reduction of concentration of the colloidal organic content. Total phosphorus removal was measured to be in the same range for both coagulants added.

Figure 6 is an illustration of changes of particle surfaces for applied coagulant. TMP development for the chosen set of experiments is illustrated in Figure 7.

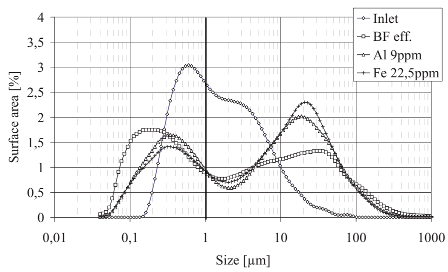

Figure 6. Surface area distribution for four sampling points

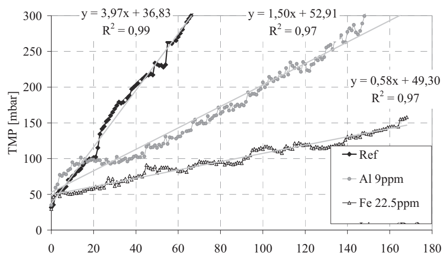

Figure 7. The TMP development

Evaluation of results

Findings from the research topic 1 (RT01) suggest a new method for estimation of optimal aeration rates with respect to minimizing the amount of submicron particles in the membrane reactor, which are considered detrimental for membrane filtration in a BF-MBR process. Method is simple to perform and gives a relatively straight forward result that can be used in optimizing the intensity of air scouring as an important fouling control method. However, the method requires further refinements where on-line monitoring of particle size changes as a function of changes in aeration rates could give more accurate predictions and thus optimal operation. The investigation did not cover the relation between aeration intensity and biochemical parameters responsible for membrane fouling such as soluble microbial product, total organic carbon or chemical oxygen demand, so supplemental studies are required to be done.

In the research topic 2 (RT02) a new approach in modifying the membrane reactor design that lead to improved membrane performance was proposed. By integrating a flocculation zone and sludge hopper under the membrane module it is possible to reduce the amount of suspended and soluble organic matter, and to some extent reduce the amount of submicron particles, around the membrane. This led to lower fouling rates and improved overall membrane filtration performance. Validation of the proposed concept should be done on larger or full scale.

Results from research topic 3 (RT03) showed that nitrogen removal in a BF-MBR process can be easily and efficiently done by implementation of a post-denitrification mode of operation. Stable denitrification rates with average nitrogen removal of about 80% were done by keeping the C/N ratio at 4.5. In comparison to a nitrification scheme, implementation of post-denitrification caused higher membrane fouling potential where pore narrowing and blocking was suspected to be a dominant mechanism. Results point out that residual organic matter from a post-denitrification configuration has higher concentration of low MW organic matter which was considered as a main reason for the higher fouling rates measured. Further investigations of the nature of organic residuals after biological treatment are required for a better understanding of the fouling nature and dominant fouling mechanisms in this configuration.

In the research topic 4 (RT04), an evaluation of five different commercial coagulants was performed in order to investigate the indirect effect these may have on membrane fouling. All tested additives showed an ability to coagulate/flocculate colloidal organic matter and furthermore reduce membrane fouling. Polymerized alum had at optimum dosages the highest ability to destabilize colloids and reduce colloidal organic matter. Iron chloride showed a better ability to flocculate suspended matter around the membrane area. The enlargement of floc sizes and floc surfaces in the upper-micron range caused a reduction of submicron particles by adsorption on/in the flocs formed. Filterability and dewaterability of the suspension with added iron chloride was better than with the other four additives tested. Iron chloride showed the best performances and therefore is suggest as additive of choice. This study showed a good potential of modified cationic polymers as fouling reducer in BF-MBR.

Conclusion

This study demonstrated the feasibility of implementing two independently developed technologies, MBBR and MBR, into a novel configuration called a biofilm-MBR (BF-MBR). Sustainable membrane operations over a period of one month with two different biological configurations at relatively high production fluxes (*i.e.* 30 LMH) and high recovery rates (*i.e.* >95%) without frequent chemical cleaning were demonstrated in paper 5 (P5). Effluent from a post-denitrification scheme showed a higher fouling potential due to a higher low MW organic content. A cyclic mode of operation with a short periodical backwash, with backwash flux around ~130% or higher of the production flux, and moderate air scouring rates was observed to give good membrane fouling control and sustainably long operation cycles of the PDN configuration.

Control of membrane fouling by reduction of particulate matter, both suspended and colloidal with new membrane reactor designs was demonstrated in paper 4 (P4). The proposed designs investigated in this study included introducing an integrated flocculation zone in the membrane reactor coupled with a sedimentation zone beneath the membrane module. The modified membrane rector design provided a significantly lower concentration of MLSS and COD around the membranes, and subsequently a more sustainable membrane performance due to much lower overall fouling rates.

Optimization of aeration for membrane scouring with respect to particles size analysis is demonstrated in paper 3 (P3). The results show the importance of finding the relationship between sufficient aeration to minimize membrane fouling, while preventing formation of colloidal particles due to excessive shear forces caused by the aeration. An approach to defining optimal operating conditions with respect to aeration rates is proposed. By evaluating overall fouling rates and PSD analysis based on an assessment of the differential number percent of submicron particles, recommendations for a design aeration rate may be defined. Optimal aeration rates found for the pilot plant configuration used in this study was found to be in the range of 1.68 to 3.37 $Nm^3 m^{-2} h^{-1}$.

Addition of different additives and their effect on reduction of colloidal and soluble matter was investigated in paper 6 (P6). Their effect on particle agglomeration and reduction of submicron particles was further investigated in paper 7 (P7). The best improvement in membrane performance was observed for the higher chosen dosage of iron chloride, while polymerized alum gave lower improvement at the same applied dosage. This was related to the highest reduction in surface area of submicron particles in the range 0.04 – 0.3μm and lower organic colloidal content. A larger surface area and larger flocs, probably formed a protective cake layer on the membrane surface which has significantly improved membrane performances at higher iron dosage. Based on the research conducted in this study the iron chloride coagulant in particular appears to be the additive of choice for a BF-MBR process.

Future Work

Results from this study demonstrate that it is possible to maintain a sustainable operation of a biofilm–MBR concept as a result of coupling of two independently developed technologies, MBBR and MBR, at various operational conditions.

Future work should include experimental evaluation of two main topics:

1) measurements for improved membrane operations, and
2) measurements for improved biological treatment

As this study has been conducted on a laboratory scale pilot plant system, all findings and results should be verified on full scale pilot plant systems.

1) Measurements for improved membrane operations

There is a need to experimentally investigate the effect of the production cycle length on minimizing of reversible and consequently irreversible fouling, since all experiments in this study were done with a production cycle of 4.75 minutes and 0.25 minutes backwashing. Investigation of optimal cycle length is expected to lead to improved membrane performances and lower operational cost.

Furthermore, there is a need for optimization of the backwash intensity and length of a backwash cycle. This would lead to better control of irreversible fouling and better overall performance.

The effect of aeration intensity as a tool for fouling control requires more detailed investigations since in this study it was demonstrated that there is a potential hazard for membrane fouling when improper empirically based aeration rates are employed. Additionally, the effect of intermittent aeration should also be studied for this application since all experiments in this study are conducted with continuous aeration rates. Future work should include knowledge from conventional MBR studies where the length of intermittent aeration may be optimized.

Backpulsing, *i.e.* very short (several seconds) with high intensity backwashing, can potentially be a good fouling control strategy as it has the capability to reduce fouling caused by pore blocking and pore narrowing of submicron particles. This technique has not yet been tested in a BF-MBR configuration and is an interesting option that should be investigated.

The effect of membrane relaxation as a strategy for fouling mitigation was not included in this study. Studies with AS-MBR have shown that this is an interesting alternative, however, its applicability to the BF-MBR process needs to be assessed. Furthermore, the investigations in this research work were based on continuous extraction of excess sludge. A combination of intermittent sludge extraction with membrane relaxation is seen as having a good potential for more advance membrane operation which could lead to lower membrane fouling rates

Membrane cleaning is a topic that has received relatively little attention in MBR technology in general. Investigating the possibility for alternative membrane cleaning strategies that can avoid classical chemicals for membrane cleaning (*e.g.* sodium hypochlorite) is recommended. Evaluation of strategies for zero discharge of chemicals used for membrane cleaning purposes from BF-MBR is therefore a topic of future work.

Investigation of an alternative process configuration with cascade series of submerged membrane reactors with completed mixed flow as proposed in Paper 1 (P1) is recommended. The proposed concept has design simplicity, could be easy to operate, would provide high effluent quality at each stage and has flexibility regarding system operations with frequent peak loads.

2) Measurements for improved biological treatment

It was demonstrated that lower organic loading rates (OLR) on the biofilm reactor creates a more favorable effluent characteristic which has good filterability and induces lower fouling rates. A more accurate assessment of the relationship between OLR and membrane fouling should be studied.

Optimization of consumption of aeration for biological treatment is an issue for future work. Combination of fine bubble aeration with mechanical agitation of suspended media could lead to lower energy consumption for the oxidation process in the biofilm reactor.

Removal of nutrients (*i.e.* nitrogen and phosphorus) has been studied in a post denitrification configuration and by applying chemical precipitation. The evaluation of the effect of pre-denitrification and optionally a combination of pre- and post-denitrification should be studied in the future.

Biological phosphorus removal in continuous flow schemes with implementation of both biofilm and activated sludge is also a topic that warrants future investigations in a BF-MBR system.

Application of different biofilm support media that can offer the possibility of simultaneous nitrification and denitrification should be tested. Furthermore, there is a media that offers good phosphorus removal without need for recirculation loops which could be an interesting alternative to media used in this study.

Application of a membrane aerated biofilm reactor for biological treatment offers a good possibility for reduction of air consumption needed for mixing suspended carriers, and also offers the possibility for simultaneous nitrification/denitrification in deeper layers of biofilm formed on membrane support layer. Applying alternative biofilm reactors such as this is an interesting development of the BF-MBR concept that warrants future investigations.

A possible BF-MBR process configuration with pre coagulation and rapid biological secondary treatment suitable for diluted wastewater (*e.g.* Norwegian conditions) is another interesting alternative that should be evaluated.

Appendix A: Selected papers

Paper P1: Particle separation in moving bed biofilm reactor – applications and opportunities

I. Ivanovic and T.O. Leiknes

Submitted to journal Separation Science and Technology

Is not included due to copyright

Paper P2: The biofilm membrane bioreactor (BF-MBR) - a review-

I. Ivanovic and T.O. Leiknes

Submitted to journal of Desalination and Water Treatment

THE BIOFILM MEMBRANE BIOREACTOR (BF-MBR)
- a review-

I. Ivanovic[1] and T.O. Leiknes[1]

[1]*NTNU – Norwegian University of Science and Technology, Department of Hydraulic and Environmental Engineering, S.P. Andersensvei 5, N-7491 Trondheim, Norway*
E-mail: igor.ivanovic@ntnu.no and torove.leiknes@ntnu.no

Abstract

Membrane bioreactors (MBR) based on the activated sludge process is a relatively new technology, with implementation worldwide increasing over the last 20 years. In parallel to commercial development, a lot of research work has been done in fundamental studies, development and optimization of this technology. Although the main focus has been on activated sludge processes, several research groups have been investigating biofilm based MBR systems. The biofilm processes have several advantages over activated sludge process and can be used as complementary, assisted to activated sludge MBR (aBF-MBR) or self standing, pure biofilm based MBRs (pBF-MBR). This article reviews the status of MBR technology with biofilm implementations. Reports published within the last 10 years are reviewed with respect to aBF-MBR and pBF-MBR studies, highlighting advantages proposed of this approach over activated sludge MBRs, identifying performance and operational characteristics given, and taking an outlook of perspectives in further development of this concept.

Keywords: biofilm membrane bioreactor, biofilm carriers, membrane fouling

1. Introduction

The MBR technology, for both municipal and industrial wastewater treatment, has seen significant growth in the last 10 years, boosted by a need for more advanced wastewater treatment, more strict legalization and increasing scarcity of fresh water resources [1]. This technology is primarily based on the conventional activated sludge concept where secondary clarifiers are normally replaced by submerged low pressure polymeric membranes for solid/liquid separation. The MBR technology has gained popularity due to several outstanding advantages over conventional process *i.e.* high quality effluent (very often hygienically highly purified), lower footprint, lower net sludge production, and improved nutrient removal [2]. Major disadvantages of the process are membrane fouling - which limits sustainability and wider applications, higher energy demand - mostly caused by air scouring demand, and higher capital costs due to the price of membranes. Better understanding of membrane fouling mechanisms, optimization of energy consumption and cheaper membrane materials have overcome some of these disadvantages, making this technology even a more realistic and viable choice by the end of this decade [3][4][5][6].

In addition to the main focus of development of conventional MBRs, several research groups have been working on biofilm assisted and biofilm based MBRs (BF-MBR), trying to combine advantages of biofilm and MBR process in order to overcome some of the limitations of conventional MBR based on the activated sludge process (AS-MBR). Implementation of biofilm processes in MBR could be done by addition of media (*e.g.* biofilm carriers) in moving or fixed bed configurations, or aerated membranes in the bioreactor as a support (*i.e.* substratum) for biofilm growth. In principle numerous materials could be used for biofilm support, however, only a few are commercially applied in full scale systems, such as cord media, RBC media, sponge and plastic media and granular activated carbon (GAC).

A combination of activated sludge (AS) and biofilm processes is also possible. Addition of biofilm carriers in activated sludge processes can assist in the biodegradation, improving nutrient removal and reducing solids retention time, which leads to increased capacity and overall improved performance of existing AS reactors [7][8]. The activated sludge can also be completely replaced by a biofilm system, shifting the process of biodegradation from suspended to attach growth only [9].

This paper presents a short review of publications in the field of assisted biofilm processes (aBF-MBR) and pure biofilm based MBRs (pBF-MBR), based on 50 scientific articles published in last 10 years. Advantages and anomalies of this approach of using biofilm processes is discussed with respect to overall system performances and membrane fouling.

2. Biological treatment by biofilm processes

Major advantages of biofilm process over activated sludge process are typically defined in that they are simpler to operate compared to activated sludge process, have higher biomass activity due to accumulation, and where the biomass has a higher resistance to toxic substances [9][10][11]. Additionally, biofilm processes favor selective development of slow growing microorganisms such as autotrophs (*i.e.* nitrogen oxidizing bacteria, NOB) and phosphorus accumulating microorganisms (*i.e.* PAO) and reduces their washout from the system [12]. Biofilm processes are comprised of moving or fixed bed schemes, with media that serves as housing for biofilm growth. Implementation of freely moving medias has an advantage over fixed media beds due to their ability to utilize the whole volume of the bioreactor, minimize or eliminate the need for biomass recirculation, and are less prone to clogging, which is typical a problem in fixed bed biofilm process with high particulate loading. In order to meet requirements for nitrification (< 2 mg NH_4-N/L in effluent) of typical municipal wastewater (*e.g.* COD=250 mgO_2/L and NH_4-N = 25 mg/L) studies have been done to determine the conditions required to achieve nitrification. A unified model that computes required surface area of biofilm as a function of mean cell residence time MCRT (or solids retention time SRT) needed for full nitrification was evaluated [7]. The results are shown in Figure 1. Design of an activated sludge process for full nitrification according to this model should be with a minimum SRT of 6 days. However, by adding biofilm media in bioreactors the activated sludge system can be converted to an integrated (bio)film activated sludge (IFAS) reactor, resulting in the opportunity to reduce SRTs for full nitrification. Shorter SRT require higher biofilm surface, where typical SRT values for an IFAS process are between 2 and 6 days. Subsequently, addition of carriers with biofilm growth reduces the SRT of an activated sludge bioreactor, which leads to increased capacity of the bioreactor or opens the possibility to operate the bioreactor on lower mixed liquor suspended solids (MLSS). This approach is beneficial for upgrading activated sludge processes. From Figure 1, it can further be seen that addition of a high biofilm surface area will lead to sufficient biodegradation where activated sludge is no longer needed. This is typically the case in a moving bed biofilm reactor (MBBR) where degradation becomes a function of the biomass available on the carriers and HRT, virtually without suspended matter (*i.e.* no activated sludge). The high biofilm surface area in MBBR is typically achieved by adding biofilm carries with a high surface area at high volumetric filling fractions, typically up to 2/3 of the reactor volume [13].

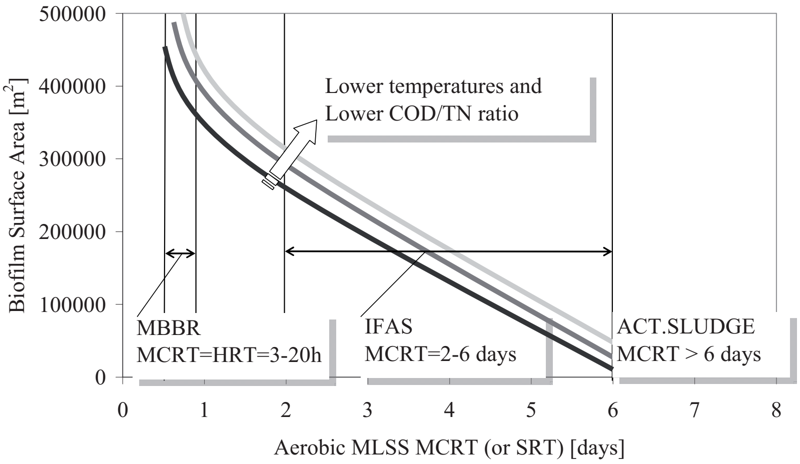

Figure 1. Threshold of required biofilm surface area for reaching full nitrification for typical municipal wastewater as a function of MCRT (SRT) – adapted from [7]

By applying the model illustrated in Figure 1 it can be concluded that by adding biofilm surface area it is possible to decrease the SRT of an activated sludge reactor, consequently increasing the capacity of existing reactors. It is also possible to design the treatment plant based on a pure biofilm process without applying activated sludge (*i.e.* decoupling SRT and HRT) if a large enough surface for biofilm growth is provided. There are different kinds of media (*i.e.* biofilm carriers) that can be used for the applications described above. Examples of types of commercially available media typically used in IFAS and MBBR applications, with basics characteristics, are given in Table 1.

Table 1. Several commercial types of media mostly used in full scale plants worldwide, adapted from [8]

Name of media	Type of bed	Specific surface area of bare media (m^2/m^3)	Typical filling fraction [%]
Ringlace	Fixed	50-100	25-50
BioWeb	Fixed	150-200	25-50
RBC	Moving	10-50	n/a
Linpor Sponge	Moving	10	15
Captor Sponge	Moving	50	5-15
Kaldnes K1	Moving	500-600	15-70

3. Assisted biofilm MBR (aBF-MBR)

By adding biofilm carriers in a conventional MBR process the result is what has been called an assisted biofilm MBR (aBF-MBR). In aBF-MBR biodegradation is carried out by suspended growth (*i.e.* activated sludge) and assisted by attached growth on carriers (*i.e.* biofilm). This approach has been studied by several research groups and often is at times defined as a hybrid MBR by some authors [2]. The motives for using carriers are different and include; reduction of the negative effect of suspended solids [9][12], improved filterability and lower membrane fouling [14][15][16][17], improved nutrient removal [15][19][20], and reduction of membrane cake layer formation by scouring effects of the suspended carriers [12][21][22].

3.1. Organic (COD) removals and nitrification

The ratio between the level of biodegradation carried out by the suspended and attached biomass depends on the amount of biomass present in either form. The filling fraction of carriers, surface area for biofilm growth, MLSS concentration in suspended growth and biomass activity are the main parameters that affect the degradation ratio. A comparison in a MBR with suspended and attached growth demonstrated that the biomass in the attached form has a higher activity, where about 1/3 lower concentration of biomass in the attached form was able to achieve the same removal rates as biomass in the suspended form [9]. Other studies also reported higher specific oxygen uptake rates (SOUR) in aBF-MBR compared to conventional AS-MBR, confirming a higher activity of the biomass in aBF-MBR type of configuration [16][20]. Generally there is no difference in the degree of organic removal between an AS-MBR and aBF-MBR. Both systems can sustainably achieve high COD removals typically around 95 - 99%, when operated at similar HRT and SRTs.

Furthermore, no significant differences in nitrification rates between aBF-MBR and AS-MBR configurations has been reported operated under similar conditions, *i.e.* the same COD/TN ratios, and HRTs and SRT, aeration rates etc. Normally a satisfactory nitrification degree >96% was achieved. However, it should be noted that some authors observed 2-4% lower nitrification rates in AS-MBR compared to aBF-MBR [19][20] (Figure 2)

3.2. Nutrients removal

Total nitrogen removal is reported to be higher in aBF-MBR systems compared to conventional AS-MBR by several authors [15][16][20]. Higher total nitrogen (TN) removal rates have been mostly attributed to simultaneous nitrification/denitrification (SND) that takes place in deeper layers of the biofilm component where anoxic/anaerobic conditions occurred. The sponge carriers seem to provide good SND conditions since they provide anoxic conditions inside the carrier element. Findings by Liang *et al.* ([18], Figure 2) differ from most reports, which is attributed to the lack of significant biofilm formation on chosen carriers in their study, as pointed out by other authors.

Phosphorus can be removed from the feedwater by assimilation for biomass growth and by phosphorous accumulating organisms (PAOs). Enhanced phosphorus removal with the addition of biofilm carries in a process has been reported by several authors [16][23][24][25]. The higher phosphorus removal was attributed to PAO organisms in the anoxic/aerobic zones found in the deeper biofilm layers. A 1.7 % to 20.1 % higher total phosphorus removal in aBF-MBR with sponge carriers compared to AS-MBR was reported, where COD/TP ratio and amount of excess sludge removed played an important role in the phosphorus removal efficiency.

Generally, organic (COD) removal and nitrification were in the same range when aBF-MBR was compared to AS-MBR under similar operating conditions. However, TN and TP removal could be significantly improved in aBF-MBR, due to the smaller floc sizes, higher microbial activity and more diverse microbial community present in the biofilm component.

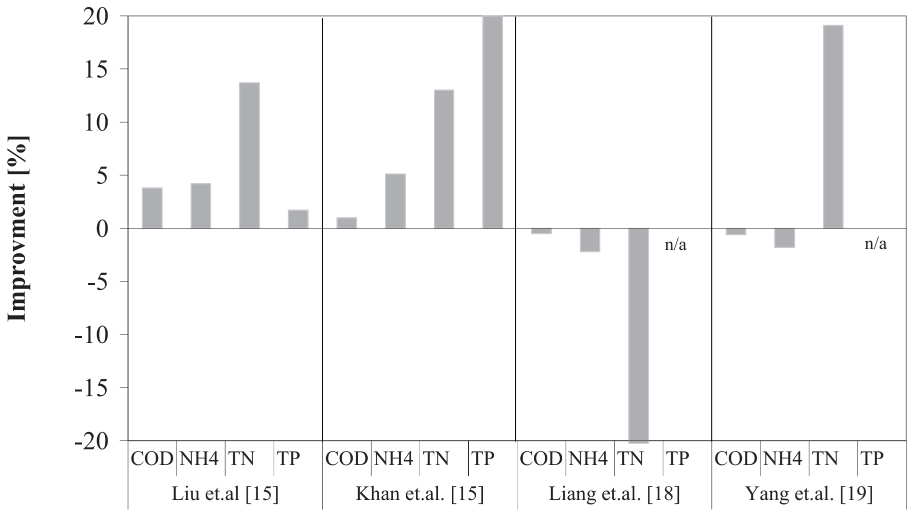

Figure 2. Efficiency of organic and nutrient removal in aBF-MBR compared to AS-MBR

3.3. Membrane performances

Membrane fouling is a common phenomenon in all membrane applications, including MBR systems [24][25][26]. Since liquid suspension (*i.e.* activated sludge and/or surplus of biofilm) is rather complex it is still unclear which fraction or compounds are mostly responsible for membrane fouling in MBR [3][5]. Colloidal and soluble organic content (*i.e.* biopolymers, SMP/EPS), suspended solids (MLSS), physical properties (*i.e.* particle size and viscosity) are mainly reported to contribute to membrane fouling.

3.3.1. Filtration characteristics

Improved filterability and lower fouling rates by implementation of attached biomass are commonly reported in aBF-MBR configurations. Reduced total resistance by 48% was reported by Wang *et al.* [14] that results it three times longer operational cycles. Better filterability was related to lower bound EPS values measured. Another study by Liu *et al*. [15] reported significant prolongation of operational cycles (from 57-65 to 92 days) as a result of adding biofilm carriers. Several other studies have reported better filterability, lower fouling rates and longer operational cycles, when MBR were operated with assistance of a biofilm [16][17][18]. Contrary to those findings, Yang *et. al.* [20] showed worse membrane performance after addition of carriers. This was explained by overgrowth of filamentous bacteria that resulted in higher EPS values [21].

3.3.2. Colloidal and soluble organic content

Soluble microbial products (SMP) and extracellular polymeric substances (EPS) are considered the main contributor to membrane fouling in MBR technology [5][26] . However, their significance and role are still debated [27]. In aBF-MBR configurations, production of SMP/EPS by attached biomass does not seem to be higher than in AS-MBR systems [9][17]. In these studies, similar values of SMP/EPS in aBF-MBR and AS-MBR are reported, suggesting no fundamental changes in biological activities. Other studies reported a reduction of SMP in aBF-MBR due to the ability of the biofilm to adsorb and bind soluble microbial products [14][15][28]. Contrary to these findings, other studies have reported measuring higher content of proteins and polysaccharides in aBF-MBR due to an overgrowth of filamentous bacteria, which could be due to a new type of non-woven carrier that was used in the study [21].

3.3.2. Effects of MLSS on fouling

In the resistance in series model, cake resistance has been indentified as a main contributor to the total resistance of flow through a membrane [9]. Reduction of the negative effect of cake formation on the membrane has been extensively studied and several methods have been proposed to reduce this impact, including air scouring, backwashing, operation below critical flux, addition of additives, novel configurations etc. [3][5][27]. The main source that creates cake formation is suspended matter (*i.e.* activated sludge), however, reduction of MLSS does not ultimately lead to better membrane performance [29]. Yang *et. al* [20] and Lee *et. al.* [9] tried to reduce the concentration of MLSS by implementing biofilm carriers in the reactor. Contrary to expectations, higher fouling rates were observed for the membranes in the aBF-MBR at lower MLSS concentrations. The membrane operated at very low MLSS concentration were exposed to formation of a dense and less porous cake layer that led to higher resistance and thus higher fouling rates. Higher MLSS concentrations led to formation of a dynamic cake layer on the membrane surface, which was confirmed by SEM and AFM images [9]. The unexpected higher fouling observed with a very low MLSS environment was additionally reviewed by Lee *et al.* [30] where the review commented a connection between lower SRT and higher SMP when the low MLSS was applied. However, in AS-MBR systems it is commonly understood that membrane operation at lower concentrations of MLSS is beneficial due to lower viscosities, lower DO diffusion resistance and lower sludging /clogging problems [5].

3.3.3. Effects of particle size distribution on fouling

It is commonly accepted that smaller particle sizes lead to a higher fouling potential [5][27][31][32]. The presence of carriers in aBF-MBR system could lead to floc brakeage and thus an increase in smaller flocs [17][18]. However, this does not seem to be the problem in aBF-MBR since studies similarly reported better filterability and lower membrane fouling rates than in comparable AS-MBR configurations. The size of the biofilm carries and filling fraction do seem to have an important role and effect on particle size distributions. Studies have reported that larger carries and lower filling fractions are able to flocculate suspended biomass, thus promoting formation of lager flocs and consequently lower fouling rates [14][16][19][21][23].

4. Pure biofilm MBR (pBF-MBR)

In pure biofilm based MBR (pBF-MBR) biodegradation is exclusively carried out by attached biomass (*i.e.* biofilm), where activity of suspended matter is neglected due to very low concentrations and low biologically active MLSS in the bioreactor [9]. An high surface area is required for growth of attached biomass, which can be achieved by addition of media in a fixed bed or moving bed configuration. Moving bed configuration has an advantage over a fixed bed since the whole volume of the bioreactor is utilized and, if it is designed properly, does not suffer from clogging problems during high particulate loadings [6][13][33]. It is desirable that the media has a high surface area that provides protection for biofilm growth and from intensive detachment mechanisms. Another important parameter is the filling fraction (*i.e.* volume of bioreactor occupied by media), where higher filling fractions are certainly desirable leading to more compact bioreactor though a free movement of the media may then be more difficult. Types of media commonly used in commercial, full scale applications are given in Table 1. In the design of such a process it has been proposed that the membrane unit in pBF-MBR should built as an external submerged unit in order to avoid accumulation of suspended matter in the biofilm reactor and to keep the attached biomass (*i.e.* biofilm) process separated from influence of suspended growth [34]. Furthermore, designing the membrane reactor as an external submerged membrane unit opens the possibility of decoupling the biological and particle separation processes, thereby creating one more level of freedom in designing pBF-MBR systems.

Initial studies of a pBF-MBR process demonstrated that it could be operated in extremely compact configurations at high organic loading rates (OLR) with the same efficiency it terms organic removals and permeate quality as for AS-MBR systems [34][35][36]. Flexibility in choice of membranes was indicated for the pBF-MBR configuration, since a microfiltration membrane of 0.1 µm pore size reduced COD in the same range (86-87%) as a 30 kDa membrane pore size at low OLR with HRT 3-4 h [36]. The study also demonstrated an ability of a pBF-MBR system to operate at higher sustainable fluxes than commonly reported in AS-MBR systems. The importance of submicron particles and their contribution to membrane fouling in pBF-MBR systems has further been demonstrated [37][38]. It was found that the relative number % of submicron particles vary as a function of OLR. A discussion of the fate of submicron particles in this process is summarized in Figure 3. Lower OLR in the biofilm reactors led to reduced residual organic loads (*i.e.* soluble COD) on the membrane surface, and consequently lower fouling rates. Higher OLR, resulted in a suspension with fragile flocs that easily brake under aeration supplied for air scouring, which led to higher production of submicron particles and higher fouling rates. This effect has also been confirmed by Ivanovic *et al.* [39] where lower filterability and dewaterability of retentate in pBF-MBR at high OLR configuration was related to a high amount of submicron particles in the range of 0,04 to 0,1µm, higher soluble organic content (FCOD) and a higher presence of filamentous bacteria. Reduction in the amount of submicron particles in a pBF-MBR system was proposed by integrating a flocculation zone beneath the membrane aeration port [40][41]. In the flocculation zone submicron particles are caught by larger particles that were retained by the membrane separation and further settled in a sedimentation zone at the bottom of the membrane reactor.

Application of pBF-MBR for shipboard wastewater treatment (including oily bilge wastewater) was demonstrated in a study by Sun *et al.* [42][43][44][45]. Good and stable biodegradability of oil and other organic compounds was ensured by application of this process using both very compact dead-end and side stream schemes. A great recovery capacity of the pBF-MBR process from oil and salt shock loads in the feed water was demonstrated. The prefered process configuration was found to be a side-stream design that employs membranes with tighter pores and combined sedimentation beneath the membrane unit.

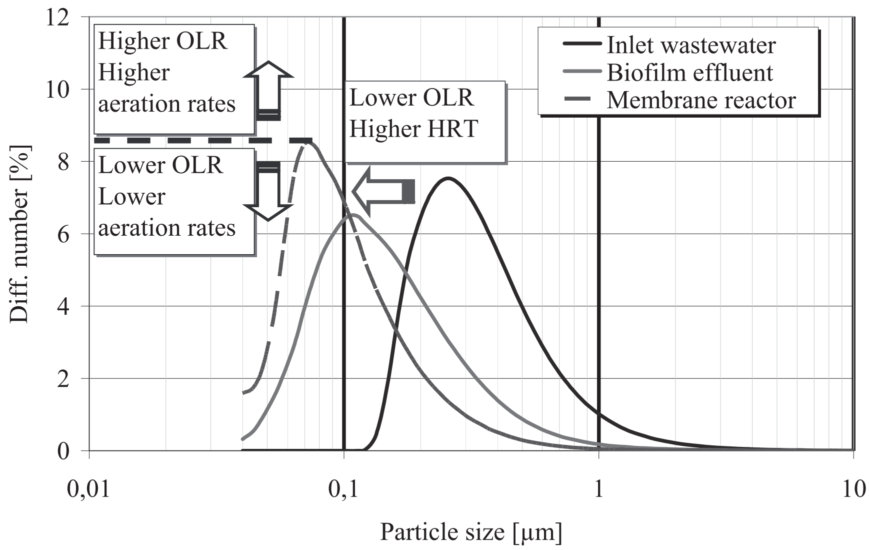

Figure 3. Fate of submicron particles in pBF-MBR in different stages of process – adapted from [38]

The flexibility in alternative designs of the biofilm reactor was demonstrated by Phattaranawik *et al.* [46] where a double-deck aerobic pure biofilm reactor was employed. The new double-deck concept was able to enhance the effect of aeration for the biological process and to minimize the load of detached suspended matter to the membrane unit. A higher packing density of a new modified flat sheet membrane module design was achieved due to an extremely low concentration of MLSS in the membrane reactor (< 50 mg/L). The modified membrane module applied for the purpose of this study displayed lower fouling rates and longer operating cycles compared to a module designed for an AS-MBR system. In another study the potential of a combination of a biofilm process with a cold digester, also in double-deck configuration, was demonstrated, that enabled reduced sludge production, lower fouling rates and higher HRT [47]. The possibility of designing pBF-MBR systems with an energy recovery unit has also been experimentally demonstrated [48]. By choosing an alternative hydrodynamic arrangement the pBF-MBR system was able to produce low MLSS (~ 100 mg/L) in the reactor, opening the possibility to use UV inactivation as a fouling control method. Addition of a UV unit resulted in 24% lower fouling rates.

An alternative approach has been proposed where a compact tertiary membrane treatment as a polishing step after a moving bed biofilm reactor in combination with disc filtration and flotation (DAF) [49]. This approach relies on a sequential removal of detached biomass from biofilm reactor, first disk filtration and/or flotation unit, resulting in low solids loads on the membrane unit. Given approach resulted in membrane fluxes in the range of 40-80 LMH being achieved. However, this approach adds another unit of operation in the treatment train and higher configuration complexity. In addition, the reported higher cleaning frequency and use of coagulant and cationic polymer for the membrane filtration are obvious drawbacks of this approach.

A recent comparative study with a fixed bed pBF-MBR and AS-MBR was conducted by Ng *et al.*, where a 71 % lower production of total SMP (60% less carbohydrate and 77,6 % less total protein) in the pBF-MBR compared to the AS-MBR was demonstrated, which resulted in 25-30% higher fluxes for the biofilm process [50]. This study further demonstrates the potential of a biofilm process compared to an activated sludge process applied to membrane bioreactor technology.

Conclusions

Implementation of a biofilm process for wastewater treatment is beneficial due to the potential of simplicity for operation compared to activated sludge, higher biomass activity, higher resistance of the biomass to toxic substances / shock loads, and development of a higher biodiversity of the microorganisms responsible for the biological treatment. Although not commonly commercially available to date, biofilm processes in membrane bioreactor technology have been shown to be potentially beneficial. Application of biofilm processes in AS-MBRs is beneficial due to the ability of the biofilm process to reduce the high SRT and MLSS values typically required for complete biodegradation of constituents in the wastewater. Inclusion of a biofilm process is practically achieved by addition of a support media that provides a high surface area for biofilm growth. Higher specific surfaces area and higher filling fractions are desirable since this can lead to more compact bioreactors and increase capacities of existing activated sludge systems. The aBF-MBR can achieve the same organic removal and nitrification rates as comparable AS-MBR designs. Higher total nitrogen and total phosphorus removals can also be achieved within a single through-put process. Other benefits include the potential for simultaneous nitrification and denitrification (*i.e.,* through existence of anoxic/anaerobic zones in the deeper layers of the biofilm component), smaller floc sizes, higher microbial activity and more diverse microbial community present in biofilm which mainly contribute to improved nutrient removals. Filterability is generally reported as improved, where lower fouling rates and higher fluxes were observed in most studies. Less bound EPS and less SMP are generated or adsorbed by the biofilm, which is considered to lead to improved membrane performances. Reduction of MLSS concentration is certainly a desirable option in MBR technology and is easily feasible by addition of biofilm carriers in existing AS-MBR systems. Operating MBRs at lower suspended solids concentration is beneficial due to lower viscosities, lower DO diffusion resistance, and lower sludging /clogging problems.

pBF-MBR systems are operated without activated sludge and where the biodegradation is exclusively carried out by a biofilm process. This system shows a great flexibility in process design and configurations, decouples the biological and particle separation processes, has the potential of membrane operation at higher fluxes / less fouling, and offers stabile operation under high organic loading. The pBF-MBR may also be used as complementary to other technologies such as activated sludge and anaerobic digestion, resulting in novel systems designs and treatment concepts, giving a flexibility and reliability for sustainable operation.

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Paper P3: Impact of aeration on particle colloidal fraction in the biofilm membrane reactor (BF-MBR)

I. Ivanovic and T.O. Leiknes

Desalination **231** (2008) 182-190

Impact of aeration rates on particle colloidal fraction in the biofilm membrane bioreactor (BF-MBR)

Igor Ivanovic[1] and TorOve Leiknes[1*]

[1]NTNU - Norwegian University of Science and Technology,
Department of Hydraulic and Environmental Engineering,
S.P. Andersensvei 5, N-7491 Trondheim, Norway
(e-mail: igor.ivanovic@ntnu.no and torove.leiknes@ntnu.no)

Abstract

In BF-MBR as in all sMBR systems sufficient aeration is necessary to maintain local cross-flow conditions for sustainable membrane operation. Aeration rate is normally based on previous experiences and manufactures recommendations. BF-MBR with an external submerged membrane unit, designed only for particle separation, could be relatively easily optimized in terms of membrane aeration. Combining observed fouling rates with measured particle size distribution and minimizing both parameters indicates a desirable sustainable aeration rate for given wastewater characteristic, operating parameters (*i.e.* production/backwash flux, recovery) and membrane module geometry.

Keywords
Biofilm-MBR, colloidal fraction, fouling rate, particle size distribution (PSD), specific aeration demand – membrane area (SAD_m)

INTRODUCTION

Biofilm membrane bioreactor (BF-MBR) is a hybrid system for municipal wastewater treatment, where the moving-bed-biofilm reactor (MBBR) for biodegradation of soluble organic matter is coupled with a submerged membrane reactor (sMBR). BF-MBR has the potential to utilize the best characteristics of a biofilm process and membrane separation resulting in compact, efficient particle removal schemes. As in all membrane processes, fouling is a main challenge that needs to be addressed. The efficiency of the process is constrained by the accumulation of materials on the surface of, or within the membrane resulting in a reduction in membrane permeability/production and the overall performance of the treatment process.

Aeration in MBR systems (both for biological and membrane operational purposes) is the most cost demanding factor in terms of energy consumption. Air scouring is necessary in submerged membrane systems in order to generate localized cross-flow conditions along the membrane surface thereby reducing cake deposit on the membrane. However, the relationship between aeration and flux or transmembrane pressure decline is still not fully understood. Aeration rates in MBR systems are based on previous experience and normally recommended by membrane suppliers (Judd, 2006).

Studies investigating membrane fouling in MBR processes have reported the significance of colloidal particles as an important factor contributing to fouling development. Wisinewski and Grasmick (1996) evaluated major components contributing to membrane fouling and estimated that colloids are responsible for 25 % of total measured fouling. Defrance *et. al* (2000) estimated this value at 30 %, while Bouhabila *et. al* (2001) reported 50 % in their results. Other studies have also shown the importance of colloidal particles in membrane fouling (*i.e.* pore blocking) for ultrafiltration membranes (Åhl *et. al.,* 2005, Juang *et. al.,* 2007). However, references in the literature that report correlations between aeration rates and effects on changes in colloidal particle characteristics as a consequence of membrane aeration can not be found. Reports have been published that show how more intensive aeration can damage floc structures and release foulants (*i.e.* EPS) bound in the floc structure (Ji and Zhou, 2006; Judd, 2006, Park *et al.* 2005).

Biofilm reactors produce an effluent with different particle characteristics compared to activated sludge, *i.e.* floc structure, particle size distribution etc. Previous studies have shown that aeration plays an important roll in particle brakeage and promotes the formation of more colloidal particles, an important component in membrane fouling (Leiknes and Ødegaard, 2007, Melin *et al.,* 2005; Åhl *et al.,* 2005). Also, it was observed in previous work that aeration rates could be a function of the configuration and geometry of the membrane reactor (Bick *et al.*, 2007, Leiknes *et al.*, 2006), though a clear connection between these two factors has not yet been fully investigated.

The aim of this study is to investigate the effect of aeration rates in the biofilm-MBR process and how this affects the colloidal fraction in the membrane filtration unit and implications this may have on membrane fouling potential.

METHODS

Experimental setup

A schematic of the process configuration and pilot plant study is shown in Figure 1. The BF-MBR process investigated in this study consists of the moving-bed-biofilm reactor (MBBR) followed by an external submerged membrane filtration reactor. This setup is based on the concept in which the biofilm reactor is responsible for the biological treatment of municipal wastewater, while the membrane reactor is used as an enhanced particle separation unit with relatively short HRT. This configuration was chosen as it has the potential for better fouling control due to the fact that almost no biological activity takes place in the membrane reactor unit and that the membrane reactor design can be focused on efficient particle removal.

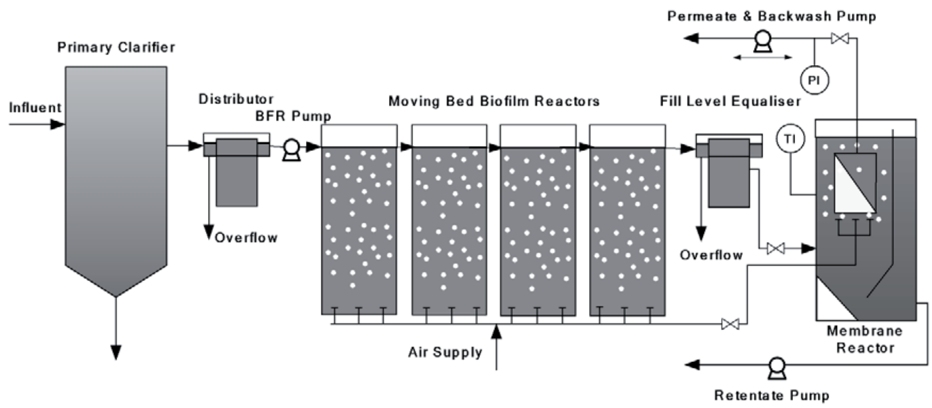

Figure 1. Flow diagram of the biofilm-MBR process configuration

Four moving-bed-biofilm (MBBR) reactors were installed in series as shown in Figure 1. The volume of each reactor was 65 L and each reactor was filled with biofilm carriers type K1, supplied by AnoxKaldnes. Filling fraction was 67 % of reactor volume giving a specific growth area for the biofilm of 335 $m^2 \cdot m^{-3}{}_{reactor\ volume}$. The hydraulic retention time (HRT) in the complete biofilm reactor was 4 hours. During the whole experimental period full nitrification was achieved. Real municipal wastewater was used during the experiments with pre-treatment consisting of a primary clarifier. The characteristics of the raw wastewater and effluent quality parameters (average, maximum and minimum) as well as the organic loading rates of the MBBR and are summarized in Table 1.

Table 1. Loading rates and characteristic of raw wastewater and effluent from MBBR

	parameter	unit	Avg.	Max	Min
Raw wastewater	SS	mg/L	114.45	286.11	45.79
	COD	mg O_2/L	274.5	382	184
	FCOD – 1.2 µm		142.2	180	80.2
	NH_4-N	mg/L	22.4	48.1	9.13
	Loading rate COD	g $O_2/m^2 \cdot d$	4.53	6.32	3.04
	Loading rate FCOD	g $O_2/m^2 \cdot d$	2.35	2.98	1.32
Effluent MBBR	SS	mg/L	156.75	438.33	62.22
	COD	mg O_2/L	204	224	140.1
	FCOD – 1.2 µm		30.5	45.2	27.2
	NH_4-N	mg/L		< 1	

The membrane reactor was designed as an external submerged membrane reactor with a volume of 33 L. A ZW-10 pilot plant membrane module supplied by Zenon Environmental Inc. was applied in this study. The membrane unit was operated under a constant flux of 52 $L \cdot m^{-2} \cdot h^{-1}$ and it was periodically backwashed with 5.8 % of the permeate production. A constant backwash flux of 58 $L \cdot m^{-2} \cdot h^{-1}$ was applied. During the operating period wastewater temperature was between 8.2 and 15.7 ^{o}C. HRT of the membrane reactor was less than 45 min. From the bottom of the membrane reactor retentate was extracted continuously with a flow rate ~2L/h. Recovery in the membrane reactor was set to 95.6 %. The filtration was operated in a cyclic mode (5 minutes) consisting of 4.75 minutes production followed by 0.25 minutes backwash. Continuous coarse bubble aeration was applied to promote local cross-flow conditions along the membrane surface. The aeration system is integrated into the membrane module at the bottom of the membrane bundle, designed as a four small holes with diameter of 2 mm. Membrane module specifications and operating modes are summarized in Table 2.

Table 2. Membrane module specifications and operating mode

Membrane reactor	Operational characteristics
Zenon ZW10 Membrane Pilot Module	Flux – 52 $L \cdot m^{-2} \cdot h^{-1}$,
- Type of membrane – hollow fiber	Backwash flux – 58 $L \cdot m^{-2} \cdot h^{-1}$
- Nominal pore size – 0.04 μm	Total permeate production – 43.2 L/h
- Configuration – outside/in	Concentrate flow - 2 L/h
Volume of membrane reactor – 33 L	Operating cycle: 4.75 min. production 0.25 min. backwashing
	Recovery = 95.6 %
	Aeration rates: 0.84, 1.68, 3.37, 5.05, 6.74 $Nm^3m^{-2}h^{-1}$

Analysis

Particle size distribution (PSD) analysis of the wastewater was done by using laser diffraction spectroscopy (Beckman Coulter LS230). The development of transmembrane pressure (TMP) was measured continuously using an online pressure transducer connected to a data acquisition system from National Instruments, FieldPoint (FP1000 and FP-AI-110), with the LabVIEW data acquisition and analysis software. TMP and temperature was logged for every two second. Data series where then extracted from the raw data depending on analysis, *e.g.* for overall performance the values from the beginning of a production cycle were extracted and plotted over time. Suspended solids (SS) were analyzed by filtering through a Whatman GF/C 1.2 μm according to the NS 4733. Chemical Oxygen Demand (COD) and ammonia (NH_4-N) were measured with the Dr Lange LCK 114, 314, 302 and 303 cuvette tests. For the Filtered Chemical Oxygen Demand (FCOD) samples were first filtered with Whatman GF/C glass microfiber filters (1.2 μm). Sludge volume index (SVI), capillary suction time (CST) and time-to-filtrate (TTF) were preformed according to the Standard methods in order to evaluate settling, dewatering and filtering characteristics of the retentate. The performance of the membrane filtration unit in the BF-MBR has been investigated with five different aeration rates; 0.84, 1.68, 3.37, 5.05 and 6.74 Nm^3/m^2h expressed as specific aeration demand – membrane area (SAD_m) according to Judd (2006). The aeration is expressed as SAD_m, however, as the pilot plant is based on a small-scale unit the values of the SAD_m measured do not represent values one can expect from full-scale systems and the trends and relationships of the values are therefore of more essence in this study.

RESULTS AND DISCUSION

Different aeration rates did not affected the characteristics of permeate due to the fact that stable permeate quality was observed during the whole experimental period. Furthermore it was not possible to observe any effect of different aeration rates on other parameters of treated wastewater e.g., SS/VSS, CST, TTF, SVI etc. The measured quality parameters for the retentate and permeate from the membrane reactor are summarized in Table 3.

Table 3. Characteristic of retentate and permeate from membrane reactor

	parameter	unit	Avg.	Max	Min
Retentate	SS	mg/L	1211,39	2583,74	568,79
	VSS		289,07	636,40	127,67
	COD	mgO_2/L	1209,11	2057,1	511,2
	CST	(second)	43,18	166,70	15,60
	TTF		56	180	44
	SVI	(ml/g)	124,53	215,03	47,00
Permeate	SS	mg/L		0	
	FCOD	mgO_2/L	22,6	30,5	17,2
	NH_4-N	mg O_2/L		< 1	
	Turbidity	NTU		< 0,1	

Membrane performance, however, expressed as TMP development over time or permeability decline (*i.e.* membrane fouling) showed a clear response when different aeration rates were applied. Figure 2 illustrates typical measurements observed for the TMP development within an operating cycle of 5 minutes. A higher fouling rate for the lower aeration rates can be observed, however, the variation for the three higher aeration rates applied are not that significantly different. Similar findings were reported by Le-Clech *et al*. (2003). Permeability decline within one cycle on average was 116.2 LMH@20^{o}C/bar for SAD_m= 0.84 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ and 75.0 LMH@20^{o}C/bar for SAD_m= 6.74 $Nm^3 \cdot m^{-2} \cdot h^{-1}$, showing that higher aeration rates give less membrane fouling. For the membrane configuration used in this study and for the given flux of operation, the effect of aeration rates higher than 3.37 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ do not show significant reduction of membrane fouling (*i.e.* permeability decline). (Figure 2)

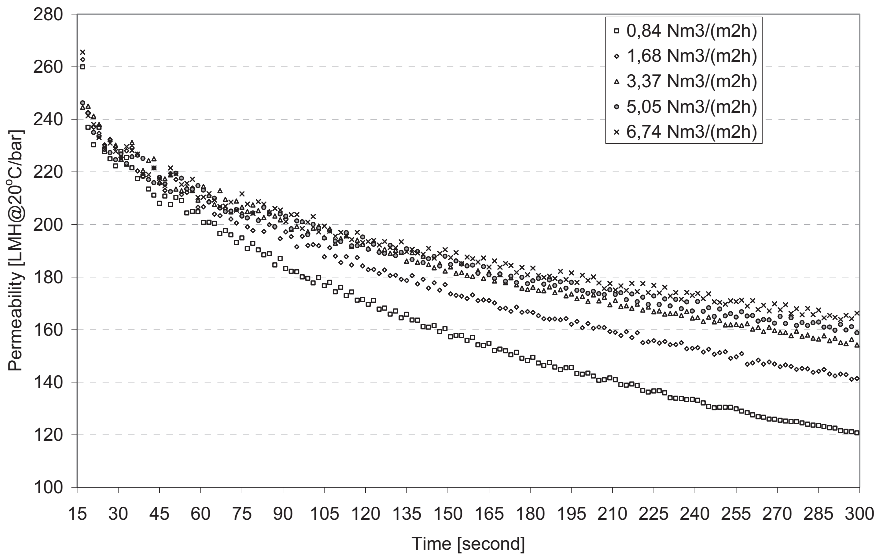

Figure 2. Permeability decline measured within a production cycle

The overall effect of varying aeration rates in the membrane rector was investigated by operating the pilot plant for five days where one aeration rate was applied per day; starting with the highest rate. Typical results measured are illustrated in Figure 3. The overall permeability decline (*i.e.* fouling rate) for the first three aeration rates is essentially the same. Based on this analysis, the performance of the membrane filtration is not significantly affected for SAD_m values higher than 3.37 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ (days 1-3). When the aeration rate is less than SAD_m values of 3.37 $Nm^3 \cdot m^{-2} \cdot h^{-1}$ the fouling rate increases significantly (days 4-5 on Figure 3). The results confirm the importance of aeration as a means to mitigate fouling in immersed membrane systems, however, there appears to be a practical limit above which the effect of increasing aeration has a minor added benefit. These results are in agreement with findings from other studies; Le-Clech *et al*. (2003), Liu *et al.,* (2003), Psoch and Schiewer (2005), and Ueda *et al.* (1997). The limit found in this study is an artifact of the membrane reactor and module design and thus system dependent. However, aeration rates may also affect the characteristics of the water in the reactor.

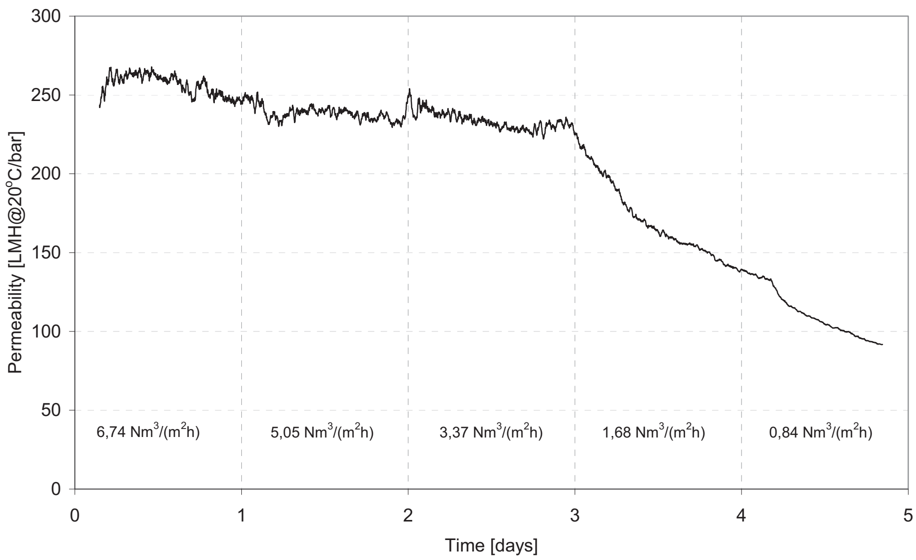

Figure 3. Overall permeability decline for different aeration rates

As fouling mitigation from aeration is commonly attributed to the shear forces generated by the air bubbles and the effect this has on the membranes, this will ultimately also affect the properties of the particulate matter in the membrane reactor. Too high shear forces will have a detrimental affect on the suspended solids by breaking them up into smaller fragments. Given that membrane fouling correlates to the submicron particles in the reactor, excessive aeration may also have a negative effect on fouling mitigation. To investigate such an affect on the system, analysis of the suspended solids around the membrane was therefore undertaken. Particle size distribution (PSD) analyses for the different operating conditions are illustrated in Figure 4. PSD results show a clear increase of the differential number percentage of particles in the colloidal fraction and in particular the fraction of particles below 0.1 µm with increasing aeration rates. The same trend is observed for all samples analyzed where representative results are shown in Figure 4. In the example shown, the differential number percentage shows that the largest fraction of particles present around the membranes has a diameter of approximately 0.07 µm. This value increased from 8.49 % to 9.72 % with increasing aeration rates showing a clear trend towards a larger number of smaller particles. The size of the particles also decreased with the largest number of particles having a diameter around 0.064 µm (Figure 4). This trend towards smaller colloids and slightly smaller diameters is a function of the increased shear forces induced during the increase in aeration intensity. Consequently, aeration in the membrane reactor has two contradicting effects with regards to membrane fouling mitigation. Higher aeration rates more efficiently remove the fouling deposition on the membrane surface and simultaneously increases the fraction of components that have been identified as major contributors to fouling. Optimal conditions for sustainable membrane operation may therefore be when these two effects are balanced, thereby giving an upper limit for aeration rates for a specific mode of operation.

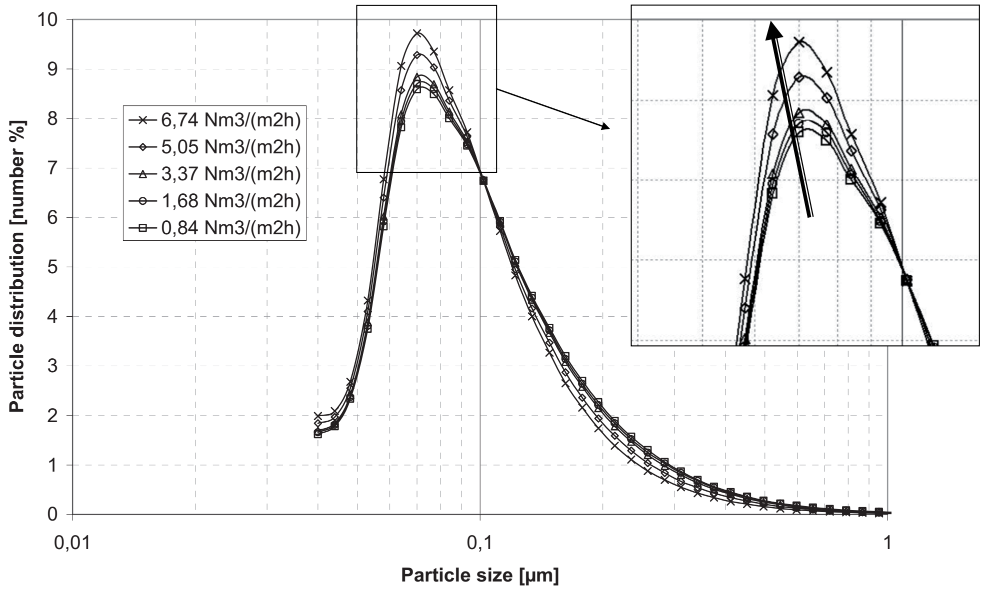

Figure 4. PSD as a function of aeration rates – zoom at submicron area below 0.1 µm

One approach to defining the optimal mode of operation with respect to aeration rates is to evaluate the relationship between membrane fouling rates and the effects of aeration on suspended solids characteristics. Results from PSD analyses combined with measured fouling rates for different SAD_m values could potentially indicate the desirable range for recommended aeration rates for the most efficient mode of operation for a system. The recommended SAD_m would ultimately define the condition that provides the least energy consumption for aeration while maintaining minimum fouling rates. Based on the results from this study, overall fouling rates measured as a function of SAD_m have been plotted together with PSD analysis results for the same conditions (Figure 5). From the presented results in Figure 5 a desirable range for SAD_m should be estimated for values higher than 1.68 but lower than 3.37 $Nm^3 \cdot m^{-2} \cdot h^{-1}$, for the given membrane reactor design and chosen operating conditions.

It is important to note that these values are specific for the system and operating conditions applied in this study. However, the approach represents and alternative strategy to define optimal operating conditions for a system with respect to aeration rates. In combination with the flux-stepping approach to define the optimal flux range of operation, analysis of PSD effects and fouling rates as a function of SAD_m may be applied to optimize membrane aeration rates. Studies are currently being conducted to generalize this approach and validate this analysis as a means to define recommended operating conditions with respect to aeration rates for a given system.

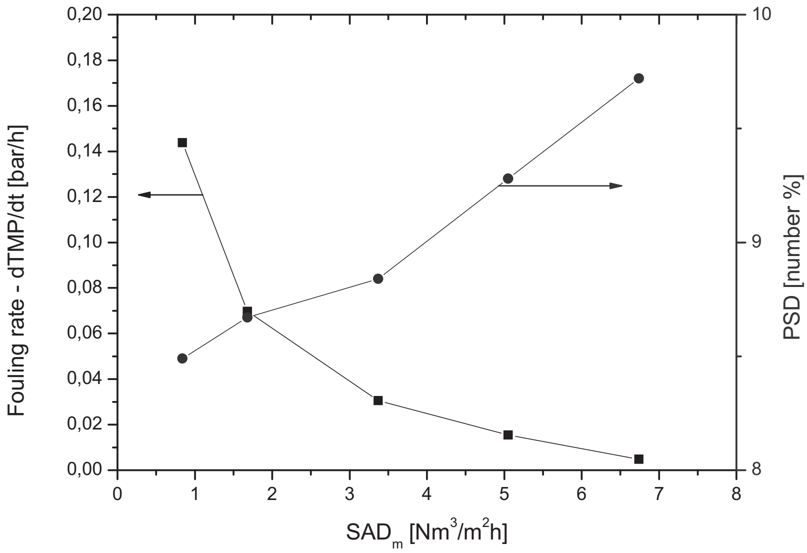

Figure 5. Fouling rate and PSD values of the most present submicron fraction for different aeration rates (SAD_m)

CONCLUSIONS

The aim of this study was to define a procedure that can determine sustainable aeration rates for a BF-MBR process with a given membrane reactor design, sustainable flux and wastewater characteristics. The results show the importance of finding the relationship between sufficient aeration to minimize membrane fouling, while preventing formation of colloidal particles due to excessive share forces caused by the aeration. An approach to define optimal operating conditions with respect to aeration rates is proposed. By evaluating overall fouling rates and PSD analysis based on an assessment of the differential number percent of submicron particle, recommendations for a design aeration rate may be defined. Further studies are currently being conducted to develop this approach and validate a general form and applicability as a design tool for optimal and sustainable operation of the membrane filtration unit for a BF-MBR process.

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Paper P4: Membrane reactor as a tool for better membrane performance in a biofilm - MBR (BF-MBR)

I. Ivanovic and T.O. Leiknes

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Membrane reactor design as a tool for better membrane performance in a biofilm MBR (BF-MBR)

I. Ivanovic[1] and T. Leiknes[1]

[1]*NTNU – Norwegian University of Science and Technology, Department of Hydraulic and Environmental Engineering, S.P. Andersensvei 5, N-7491 Trondheim, Norway*
E-mail: igor.ivanovic@ntnu.no and torove.leiknes@ntnu.no

Abstract

Coupling biofilm reactors with membrane filtration as biofilm membrane bioreactors (BF-MBR) is an interesting alternative technology to activated sludge membrane bioreactors (AS-MBR). Biofilm technology for wastewater treatment can provide a substantially lower suspended solids environment for membrane filtration compared to activated sludge processes. Potential benefits are; less membrane clogging / sludging problems, lower fouling potentials, ease of membrane cleaning, reduced energy consumption for air-scouring, and new membrane module/reactor designs. This study was aimed to investigate alternative membrane reactor designs as a tool to improve membrane performance in a BF-MBR process. Three different designs were investigated. A simplified model was developed to predict and analyze the performance of the membrane reactor designs chosen. Results showed that solids control can be achieved, in particular the MLSS concentration, as well as a reduction of the colloidal submicron particle fraction, thereby reducing membrane fouling. Modification of the membrane reactor in a BF-MBR process is beneficial. The alternative designs investigated in this study included introducing an integrated flocculation zone in the membrane reactor coupled with a sedimentation zone beneath the membrane module. The modified membrane rector design provided a significantly lower concentration of MLSS and COD around the membranes, and subsequently a more sustainable membrane performance due to much lower overall fouling rates.

Keywords: biofilm membrane bioreactor, membrane reactor, suspended solids control, membrane fouling

Introduction

A Biofilm MBR (BF-MBR), based on coupling a biofilm reactor (BF) and a submerged membrane reactor (sMR) is an alternative concept to conventional MBR systems based on an activated sludge process for advanced wastewater treatment. The concept of the BF-MBR process has previously been investigated by combining a moving bed biofilm reactor (MBBR) followed by a submerge membrane reactor (MBR) [1][2]. An operational challenge of submerged AS-MBR systems is that the process deals with liquors having high concentrations of total solids as well as dissolved compounds such as extracellular polymeric substances (EPS) leading to membrane fouling. Air scouring is commonly applied to prevent clogging and fouling of the membrane modules, an energy intensive component of AS-MBR systems. Possible advantages of the BF-MBR concept lie in the fact that biomass is attached to suspended carriers and there is no need for sludge (*i.e.* biomass) recirculation in the system. The amount of surplus biomass that become detached from the biofilm carriers also generate much lower suspended solids concentrations to be separated in membrane reactor[1][2][3][4]. Subsequently, the lower amount of suspended matter that needs to be separated gives lower viscosity, less fouling potential, *i.e.* cake deposition and clogging and less biofouling, thus reduction of the energy consumption required from the air-scouring system needed for fouling control and mitigation [3][4]. This characteristic opens the opportunity for designing a submerged membrane reactor that can operate with relatively low concentrations of suspended matter (MLSS) thereby overcoming some of the key bottlenecks in AS-MBR processes. Several references in the literature can be found where the effect of MLSS concentration on membrane performance (*i.e.* membrane fouling or permeability decline) has been evaluated [4][5][6]. It was shown that in AS-MBR higher MLSS concentrations induce higher viscosity and higher fouling potential [8], increased cake layer fouling and resistance [9], and decreased normalized permeability [6]. In general these studies report better membrane performances (i.e. less fouling) when the concentration of MLSS is lower [10][11]. However, not only the concentration of the suspended material is of significance but also the composition and characteristics of the material, in particular the colloidal fraction [11][12][13][14] .The effect of bio-solids concentration (*i.e.* MLSS) in the BF-MBR has not been fully investigated, though previous studies have shown a correlation between lower fouling rates when lower MLSS concentrations are observed around the membranes, where the significance of the colloidal fraction was demonstrated [15][16][17][18]. The potential benefit of the BF-MBR process combined with the low solids load to the membrane reactor is the opportunity to design and operate the membrane unit for enhanced particle removal and thus improved membrane fouling mitigation and control.

The membrane reactor design combined with mode of operation is an important aspect with respect to the characteristics and MLSS concentrations that can be achieved around the membranes. In this study the impact of alternative membrane reactor designs and operating modes has been investigated as tool for improving the membrane filtration performance in a BF-MBR. The approach has been to design the membrane reactor for improved solids control to reduce fouling and to investigate how this may affect the characteristics of the solids around the membrane, in particular the colloidal fraction. Three different membrane reactor designs have been investigated and operated at pilot scale under varying operating conditions.

Theory

In submerged systems the membrane modules are designed either as externally submerged units or directly immersed in the bioreactor [5]. In both cases the principle reactor design and configuration will be the same, as illustrated in Figure 1. A conventional approach of defining flow and mass balances can be applied to describe and analyze the reactor. In submerged membrane reactors operation commonly includes air-scouring for fouling control and recycling of the biomass between the biodegradation stage and the membrane filtration stage, both resulting in the reactor configuration functioning as a completely mixed membrane reactor, CM-MR.

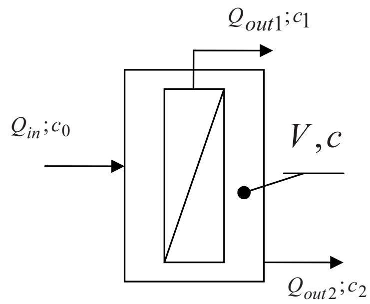


Figure 1. Principle of a typical submerged MBR reactor configuration

Following a conventional mass balance on MLSS (named as c in Figure 1) for a submerged membrane reactor designed as a CM-MR, the change in MLSS over time can be expressed as:

$$V \cdot \frac{dc}{dt} = Q_{in} \cdot c_0 - (Q_{out1} \cdot c_1 + Q_{out2} \cdot c_2) \qquad (1)$$

Where

V - volume (m^3),
c - concentration of MLSS ($kg \cdot m^{-3}$),
Q_{in} - flow rate in ($m^3 \cdot d^{-1}$),
Q_{out} - flow rate out ($m^3 \cdot d^{-1}$),
0 - subscript: inlet
1 - subscript: permeate,
2 - subscript: concentrate,

Assuming no suspended matter in the permeate, *i.e.* $c_1=0$ and completely mixed conditions, *i.e.* $c_2=c$, the concentration of MLSS inside the membrane reactor can be expressed as:

$$c(t) = c_o \cdot \frac{Q_{in}}{Q_{out2}} + c_o \cdot \left(1 - \frac{Q_{in}}{Q_{out2}}\right) \cdot e^{-\frac{Q_{out2}}{V} \cdot t} \qquad (2)$$

or,

$$c(t) = \frac{c_o}{1-R} \cdot \left[1 - R \cdot e^{-\frac{1}{SRT} \cdot t}\right] \qquad (3)$$

where SRT is solids retention time,

$$SRT=\frac{V}{Q_{out2}} \quad (4)$$

and R is recovery,

$$R=1-\frac{Q_{out2}}{Q_{in}} \quad (5)$$

By analyzing the reactor design and operating conditions using equation (3) it is possible to predict the expected performance as a function of flow rates (*i.e.* recovery) and suspended solids load. For each operating condition one can then determine the steady state concentration of MLSS in the membrane reactor based on MLSS concentration and recovery. The results of the analysis for a given condition are shown in Figure 2.

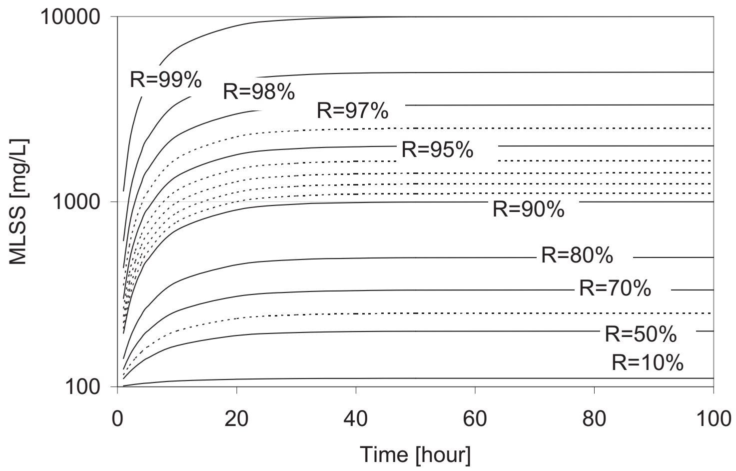


Figure 2. Steady state concentrations of MLSS as a function of recovery R, for c_o=100mg/L and SRT=9h.

Following the results in Figure 2, it is only possible to obtain lower concentrations of MLSS in the CM-MR if the membrane reactor is operated with lower recovery. From a practical point of view, operating at very low recoveries is not efficient or sustainable given that the primary objective of the process is an efficient and complete removal of MLSS in the permeate stream. Alternative strategies to reduce the MLSS concentration around the membrane therefore need to be introduced.

An alternative to such a conventional reactor design is a membrane reactor with an integrated flocculation zone and enhanced sedimentation beneath the membrane (*i.e.* sludge hopper). This modification makes it possible to reduce the concentration of MLSS around the membrane by sedimentation and to reduce the amount of submicron particles by natural flocculation [16]. A mass balance analysis of the modified membrane reactor with an integrated sludge hopper (SH-MR) can be done as above by adding a factor (K_s) that takes into account the reduction of MLSS due to the modified reactor geometry.

An expected concentration of MLSS around the membrane can be calculated based on equation (1) including K_s and expressed as:

$$c(t)=c_o \cdot \frac{Q_{in}}{K_S Q_{out2}} + c_o \cdot (1-\frac{Q_{in}}{K_S Q_{out2}}) \cdot e^{-\frac{K_S Q_{out2}}{V} \cdot t} \quad (6)$$

Where K_S is a separation coefficient equal to c_2/c ($c_2 > c$)

The value of K_s will depend on the geometry of the reactor and on the characteristics of the suspended matter coming from the MBBR biofilm reactor, which depends on the hydraulic retention time (HRT) in the MBBR and loading rates of the biofilm reactor. For a completely mixed reactor, as the case for the CM-MR option, the K_S value is equal to 1, while for a modified SH-MR design the K_S factor will have values greater than 1. The effect of steady-state MLSS concentrations around the membrane by varying K_s values in equation (6) for a fixed recovery (R) and for a given operating condition of the biofilm reactor is illustrated in Figure 3.

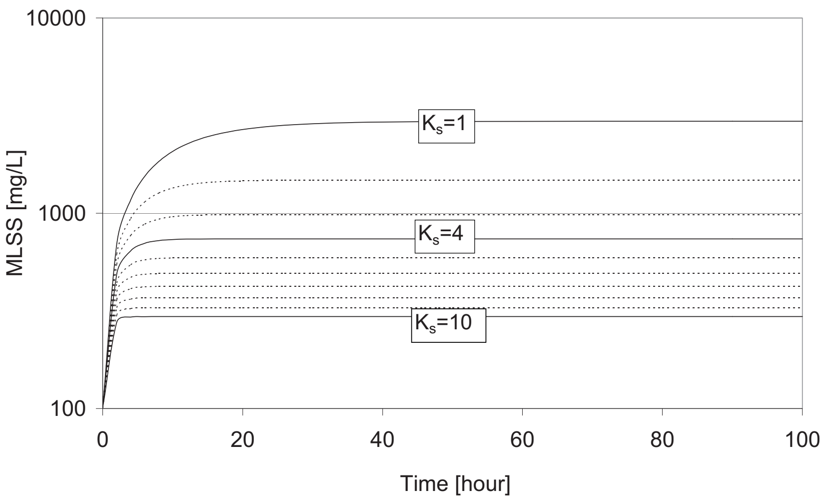


Figure 3. Steady state concentrations of MLSS as a function of separation coefficient K_s for recovery of 96%, for c_o=100mg/L and SRT=9h

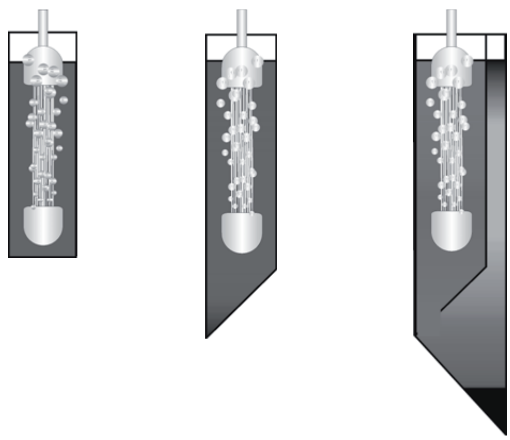
Different membrane reactor options and varying operating conditions can therefore be analyzed using the simplified model defined in equation (6) to understand the impact of alternative membrane reactor designs on the resulting MLSS concentrations around the membrane module, and subsequently the membrane filtration performance with respect to overall fouling rates.

Methods

Three membrane reactor designs were chosen and compared in this study: 1) a conventional completely mixed reactor (CM-MR), 2) a membrane reactor with integrated sludge hopper (SH-MR) and 3) a membrane reactor with a modified sludge hopper design (MSH-MR). Illustrations of the three reactor properties and differences are shown in Figure 4. The study was conducted with small-scale pilot plant setups using Zenon ZW 10 pilot plant membrane modules, and with the three reactor volumes of 9, 27 and 41 L respectively. Each membrane unit was feed with effluent from a pilot plant MBBR consisting of four moving-bed-biofilm (MBBR) reactors installed in series. The volume of each reactor was 65 L and each reactor was filled with biofilm carriers type K1, with specific surface area 335 m^2/m^3, supplied by Krüger Kaldnes. Filling fraction was 67 % of reactor volume with total surface area for biofilm growth of 84.5 m^2. Details and specifics of the pilot plant configuration and setup have been reported in previous studies [16][17].

The BF-MBR pilot plant configuration was operated first with a low strength municipal wastewater, and later with a high strength wastewater which was mixture of municipal wastewater and synthetic wastewater (Table 1).

During operation with the low strength wastewater, the membranes were operated in a cyclic mode consisting of a 4.75 minutes production time and a 0.25 minutes backwash cycle. Production flux was set at 35 $L \cdot m^{-2} \cdot h^{-1}$ and backwash flux at 38 $L \cdot m^{-2} \cdot h^{-1}$, recovery 96% and specific aeration demand of $SAD_m \sim 3.37\ Nm^3m^{-2}h^{-1}$. The MBBR reactor was operated with a 4 hour HRT, giving on average the quality parameter values and treatment efficiencies as shown in Table 1.



a) CM-MR b) SH-MR c) MSH-MR

Figure 4. Illustration of the three alternative membrane reactor designs investigated in this study with Zenon ZW-10 membrane module

During operation with the high strength wastewater, the membranes were operated with a constant flux of 22 $L \cdot m^{-2} \cdot h^{-1}$ (no backwash or relaxation), recovery 96%, and the specific membrane aeration demand of $SAD_m \sim 1.8\ Nm^3m^{-2}h^{-1}$. The HRT of the MBBR was 6 hours. Water quality parameters and treatment efficiencies are given in Table 1.

The pilot plants were equipped with National Instruments / LabVEIW data acquisition units and online measurements using various sensors, *i.e.* temperature, pressure, flow etc. All analyses were performed according to national or international standards. The development of transmembrane pressure (TMP) was measured continuously using an online pressure transducer connected to a National Instruments, FieldPoint (FP1000 and FP-AI-110) unit, with the LabVIEW 6.1 and 8.2 data acquisition and analysis software. TMP and temperature were logged for every two seconds. Data series where then extracted from the raw data with a routine written in C++ software. Suspended solids (SS) were analyzed by filtering through a Whatman GF/C 1.2 µm glass microfiber filter according to the Norwegian Standard NS 4733. Chemical Oxygen Demand (COD), ammonia (NH_4-N) and nitrogen (NO_3-N) were measured with the Dr Lange LCK 114, 314, 303, 304 and 340 cuvette tests. For the Filtered Chemical Oxygen Demand (FCOD) samples were first filtered with a Whatman GF/C 1.2 µm filter. Particle size distribution (PSD) analysis of the wastewater was done by using laser diffraction spectroscopy (Beckman Coulter LS230).

Table 1. Quality parameters and performances of MBBR for low and high strength wastewater

	Low strength wastewater			**High strength wastewater**		
	Inlet wastewater	**Effluent MBBR**	**Rem. rate [%]**	**Inlet wastewater**	**Effluent MBBR**	**Rem. rate [%]**
COD [mg/L]	217.2 ±17.8	147.9 ±32.7	31.9	522.5 ±204.1	252 ±117.2	51.7
FCOD [mg/L]	119.8 ±22.4	42.4 ±18.4	64.6	400 ±81.7	80.2 ±32.4	80
MLSS [mg/L]	68 ±8.9	103 ±54.4	-	80 ±45.5	116 ±45.9	-
NH4-N [mg/L]	29.8 ±6.4	0.16 ±0.04	99.4	38.9 ±8.7	0.21 ±0.13	99.5
NO3-N [mg/L]	<1	27 ±3.6	-	<1	34.7 ±7.8	-
HRT [h]		4h			6h	

Results

The impact of fouling rates observed within a membrane operating cycle as a function of MLSS concentration in the membrane tank was measured. For the CM-MR configuration, MLSS concentrations just under 400 mg/L up to around 3000 mg/L were investigated. Results are presented in Figure 5, showing that an increase of MLSS results in higher fouling rates, which is as expected based on reports from previous studies of MBR processes. The correlation between MLSS concentration and fouling rates is clear, indicating that a reduction in MLSS in the membrane reactor should result in a more sustainable operation of the membrane filtration unit with considerably lower fouling rates.

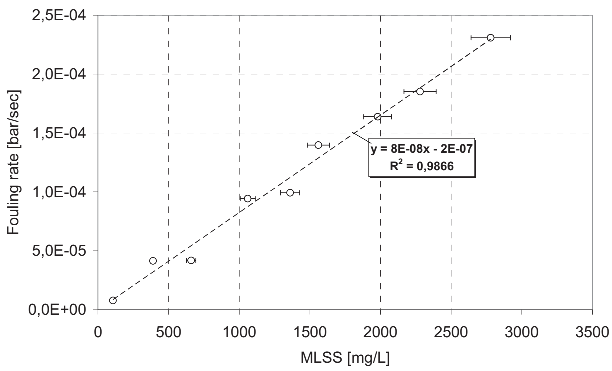


Figure 5. Example of fouling rate within an operating cycles as a function of MLSS for CM-MR

Equation (6) was used to model and predict the MLSS concentrations for the alternative reactor designs described above. For the CM-MR (Figure 4, a)) the K_S value was set to one, while the steady state concentration for the SH-MR (Figure 4 b)) was fitted for K_S=3. During experiments with the SH-MR configuration it was observed that over time the settled sludge in the sludge hopper had a tendency to float up and increase the MLSS concentration around the membrane area. A modified reactor to handle the floating sludge was therefore designed in order to separate floating sludge from reaching the membrane area, MSH-MR in Figure 4 c). Measured steady state values of MLSS in this configuration indicate that K_S has a value of ~7,5. Experimental verification of the models has been done and results are presented in Figure 6. A good fit is apparent, confirming that it is possible to estimate the concentration of MLSS inside the membrane reactor based on a given operating condition, quality of the MBBR effluent into the membrane reactor and the membrane reactor geometry/design (*i.e.* K_S factor).

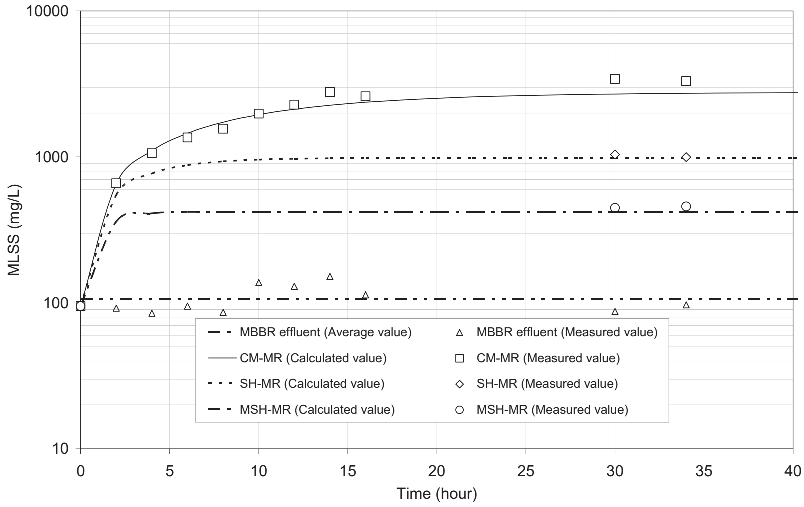


Figure 6. Calculated and measured values for MLSS for CM-MR, SH-MR and MSH-MR

The objective of this study was to determine the effect of the membrane reactor design on controlling the MLSS concentration around the membrane area and thus reducing membrane fouling rates. A comparison of the overall performance of the three reactor designs is shown in Figure 7. The fouling rate, expressed as TMP development over time, is shown both before and after backwashing during the start up period for all three reactor designs. The differences between the two measurements (ΔTMP) represent the reversible fouling formed during a filtration cycle, while the observed TMP development over time measured right after backwashing represents the irreversible fouling. The reversible fouling rates within the production cycle after 32 hours of operation were 6.18, 0.97, and 0.10 kPa/cycle for the CM-MR, SH-MR and MSH-MR, respectively. The results clearly indicate a better performance and more sustainable operation of the MSH-MR configuration due to enhanced solids control in the membrane reactor.

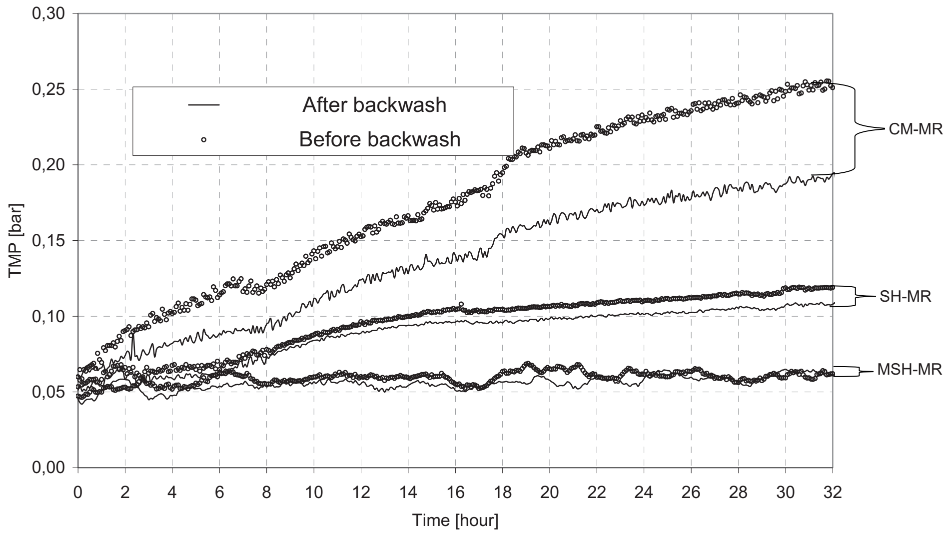


Figure 7. TMP before and after backwash for CM-MR, SH-MR, MSH-MR. Production flux 35LMH, and Backwash flux 38 LMH

The pilot plants were operated continuously over a period, setting a maximum TMP level of 30 kPa (0.3 bar) as the condition to initiate chemically enhanced backwashing (CEB) to remove the irreversible fouling. A comparison of the performance of the three reactor configurations investigated is given in Figure 8, showing results for filtration of high strength wastewater. The pilot plants were operated for a period of 7 days. The CM-MR configuration was terminated after 3 days operation as it reached the set TMP cut-off point within that period, while the other two configurations were still below this point. The observed average fouling rates were 10 kPa/day, 3.57 kPa/day and 1.42 kPa/day for the CM-MR, SH-MR and MSH-MR respectively for the operating period shown.

Table 2. Average values at steady state conditions measured around the membrane area for three membrane reactors.

	Low strength wastewater			High strength wastewater		
	CM-MR	SH-MR	MSH-MR	CM-MR	SH-MR	MSH-MR
COD [mg/L]	3400 ±306	1106 ±325	632 ±105	3630 ±448	1024 ±198	607 ±105
FCOD [mg/L]	131 ±29	91 ±15	78 ±16	225 ±46	119 ±44	108 ±26
MLSS [mg/L]	3110 ±316	757 ±234	460 ±66	2740 ±541	590 ±99	475 ±136
HRT [h]	0.3	0.7	1.3	0.3	0.7	1.3
SRT [h]	7.5	20.8	33	7.5	20.8	33
Recovery [%]	95.5-96%					

Results shown in Figure 8 confirm that the membrane reactor design plays an important role in membrane fouling control. Even when most of the operating parameters (*e.g.* HRT in MBBR, loading rates, intensity of membrane aeration, backwash/relaxation, net flux, etc.) were varied during the experiments with low strength wastewater, the overall membrane performance observed had the same trend regarding fouling dynamics. A reduction of particulate matter around the membrane module *i.e.* reduced MLSS by sedimentation in membrane reactor, can significantly improve membrane performance *i.e.* reduced fouling.

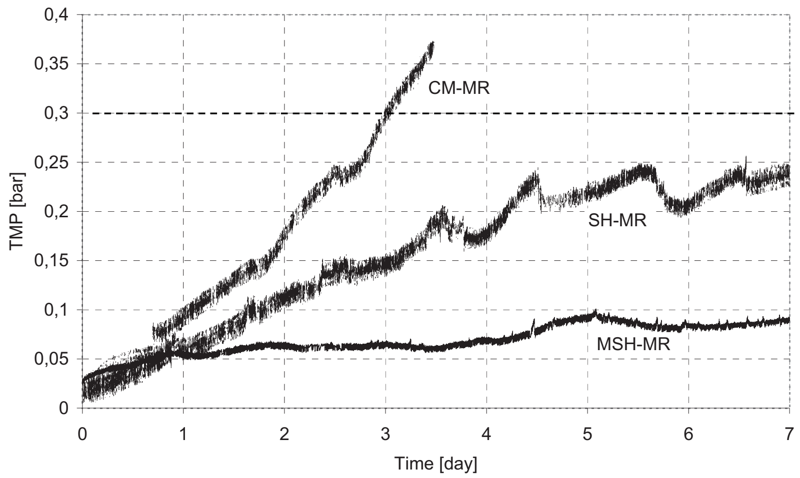


Figure 8. Overall TMP for CM-MR, SH-MR and MSH-MR at the constant flux of 22LMH

Lower concentrations of MLSS and COD (Table. 2) around the membranes as a function of the modified rector designs results in a better membrane performance. However, reduction in MLSS is not directly proportional to a reduction of fouling rates (dTMP/dt). The characteristics of suspended matter around the membrane plays an important role in membrane fouling, however, other foulants such as submicron particles and SMPs have also been identified as having an impact on fouling [10][11][13][15]. A reduction in these foulants by an enhanced membrane reactor design is also shown to have a significant contribution to controlling and minimizing fouling of the membrane. Soluble matter (FCOD) was reduced significantly in the MSH-MR compared to the CM-MR (Table 2.).

However, the submicron particles and colloidal organic matter remain significant foulants as reported in previous studies [1][2][17][18]. Previous studies have reported the impact of reducing the colloidal submicron particles around the membrane and the effect on membrane fouling. One strategy to achieve this is the integration of a flocculation zone where submicron particles from the effluent of a MBBR are captured by larger flocs that tend to settle [16]. Air-scouring for fouling control and mitigation in submerged membrane reactors is also a challenge in that too high aeration intensities may generate more colloidal material. A previous study demonstrated that there is a tradeoff between increasing aeration intensities to prevent fouling and the formation of submicron colloidal material caused by the high shear forces and particle breakage [17]. In this study the impact of introducing the MBBR effluent beneath the aeration device in the membrane reactor to avoid floc breakage and induction of smaller, submicron particles was re-investigated. Results show that it was possible to reduce the amount of submicron particles around the membrane in the MSH-MR by designing the inlet point beneath the membrane in a flocculation zone, and below the membrane aerator (Figure 9).

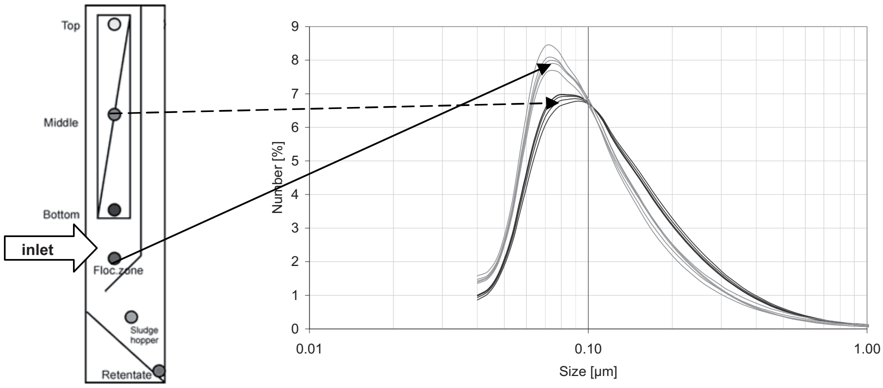


Figure 9. PSD in number percentage for submicron particles for given sampling points in the MSH-MR for four parallel measurements where each line represents average of three measurements of one sample.

The analysis of particle sizes and particle size distributions (PSD) in the respective zones of the MSH-MR design is illustrated in Figure 9. Results are shown for the sludge hopper, flocculation zone where the inlet is, and around the membrane. Based on a differential number percentage, most of the particles are in the 0.07 to 0.08 µm size range, with similar conditions prevailing in the flocculation zone. Around the membrane the PSD analysis shows a reduction in this range of submicron particle and a slight increase (0.08 to 0.09 µm) in particles in general. This is in agreement with previous findings where a reduction in this particle size range correlates with reduced membrane fouling rates. In general, this study has demonstrated that lower concentrations of MLSS, COD, FCOD and submicron particles around the membranes as a function of the modified membrane rector designs results in a better membrane performance (*i.e.* less fouling). As the amount and characteristics of suspended matter around the membrane plays an important role in membrane fouling, a reduction in these foulants by an enhanced membrane reactor design is a significant contribution to controlling and minimizing fouling of the membrane.

Conclusions

Modification of the membrane reactor design in a BF-MBR process is a potential tool to improve the overall performance of the treatment process. The alternative designs investigated in this study included introducing an integrated flocculation zone in the membrane reactor coupled with a sedimentation zone beneath the membrane module. The modified membrane rector design provided a significantly lower concentration of MLSS and COD around the membranes, and subsequently a more sustainable membrane performance due to much lower overall fouling rates. Reduction in MLSS is not directly proportional to a reduction of fouling rates (*i.e.* dTMP/dt). The characteristics of suspended matter around the membrane and other foulants also play an important role in membrane fouling, in particular the submicron colloidal fraction. This study has demonstrated that a reduction in these foulants by an enhanced membrane reactor design is a significant contribution to controlling and minimizing membrane fouling. A simple model has been proposed for calculating and predicting steady-state values of MLSS inside the membrane reactor as a function of a given membrane reactor design. This has been done by introducing a separation coefficient (K_s) which is a function of reactor design, *i.e.* hydrodynamic conditions, integration of a flocculation zone, sludge hopper etc. Further development and refinement of the model by determining adequate expressions for K_S will be investigated with the aim of developing a design tool for improved membrane reactor designs for the BF-MBR process.

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Paper P5: Impact of denitrification on the performance of a biofilm-MBR (BF-MBR).

I. Ivanovic and T.O. Leiknes

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Paper P6: Effect of addition of different additives on overall performance of bioflim - MBR (BF-MBR)

I. Ivanovic and T.O. Leiknes

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Accepted to journal Desalination and Water Treatment

Effect of addition of different additives on overall performance of bioflim - MBR (BF-MBR)

I. Ivanovic[a] and T.O. Leiknes[a]

[a] *NTNU - Norwegian University of Science and Technology, Trondheim, Norway*
igor.ivanovic@ntnu.no and torove.leiknes@ntnu.no

Abstract

The effectiveness of five additives, two iron based and two alum based coagulants and one modified cationic polymer was investigated in relation to reduction of fouling rates in biofilm-MBR. Additionally, the amount of colloidal organic matter in terms of soluble microbial products (SMP), dissolved organic carbon (DOC) and filtered chemical oxygen demand (FCOD) removal, retained by membrane was related to measured membrane fouling rates. Optimal dosage was defined based on maximum values of coefficient additive utilization (CAU). Iron chloride at higher chosen dosage showed that fouling could be reduced up to seven times, where for polymerized alum chloride it was measured reduction of about three times. The iron chloride coagulant performed better than iron chloride sulfate, in terms of fouling reduction. Higher basicity of polymerized inorganic coagulant did not result in improved membrane performance. Modified cationic polymer showed good potential in instantaneous fouling reduction, however continuous dosing strategy was found difficult to use without thorough monitoring of the system performance. Reduction in fouling rates relates better to reduction of $SMP_{carbohydrates}$, than to $SMP_{protein}$ and DOC. Also FCOD was seen as good potential fouling predictor parameter. Synergetic effect of high total phosphorus removal rates and reduced fouling rates give advantage of iron chloride coagulant over the others tested additives in this study.

Key words. Biofilm MBR, fouling control, colloidal organic matter, coagulation

Introduction

The objective of this study is to investigate the effectiveness of different additives commonly reported as filterability enhancers and fouling reducers in the MBR technology on the overall performance in a biofilm-MBR (BF-MBR).

The BF-MBR is an alternative concept to conventional MBR, where a biofilm reactor is employed instead of an activated sludge reactor. Several advantages of this approach were previously reported; *e.g.* no need for biomass/sludge recirculation, significantly lower concentration of MLSS and low viscosity of biofilm effluent giving lower energy consumption for membrane aeration and less or no membrane module sludging/clogging problems [1-3]. However, membrane fouling caused by suspended and colloidal matter remains a major challenge in development of this concept, which is also common for conventional MBR and other membrane systems [4-5].

Commonly understood techniques for fouling reduction and control include optimization of air scouring and hydrodynamics [6], backwashing and relaxation [7], membrane reactor design [8], alternative filtration modes [9], etc. Recently a strategy of adding different additives has been explored in order to adsorb and/or coagulate (flocculate) and in that way reduce certain mixed liquid compounds which are suspected to cause membrane fouling. Different research groups investigated additions of inorganic coagulants [10-11], granular or powdered activated carbon [12-13], natural and synthetic polymers [10][14-16], or combinations [17]. Even though approaches in dosing strategies, MBR configurations and membrane materials applied differ significantly in mentioned studies, a similar response was observed in that addition of certain additives at optimum dosages results in reduction of SMP (EPS), enlarges floc sizes and reduces cake porosity. Therefore, improved membrane performance was observed giving lower fouling rates, longer operational cycles or higher (*e.g.* enhanced) fluxes. Polymerized metal coagulants were found to be more effective in terms of lager floc formations and better organic removal (*i.e.* COD and DOC removal) [11][18] than non-polymerized, which could result in improved filterability. Higher basicity of polymerized alum has been reported to result in better DOC removal in drinking water applications [18], however, observations of this effect in MBR applications have not been found reported in literature. In the last few years modified cationic polymers, specially designed for MBR applications, have gained popularity as studies by several authors have shown that at optimum dosages an increase in critical flux and concentration of MLSS, reduction of SMP's, increased cake porosity and overall improved performance of MBR systems can be achieved [14-15] .

Application of inorganic coagulants and/or cationic polymers should therefore also be beneficial in improving the performance of a BF-MBR since it can be applied on mixed liquors with lower amounts of suspended matter than activated sludge systems, and thus lower dosages could be expected. In addition, the ability of coagulants to flocculate and reduce the amount of submicron particles, which have been reported as one of the major foulants in BF-MBR [1], makes the addition of flux enhancers an interesting strategy to reduce fouling. A potential negative impact on the biological treatment stage [10] is not of concern in a BF-MBR as the membrane separation process is separated from the biological process with no feedback effect on the biological treatment. Furthermore, biological phosphorus removal in a biofilm reactor is only possible in SBR schemes [19] while for systems that are continuously operated, like in this study applying a moving-bed-biofilm reactor, only chemical precipitation by iron or alum is an available option.

Materials and methods

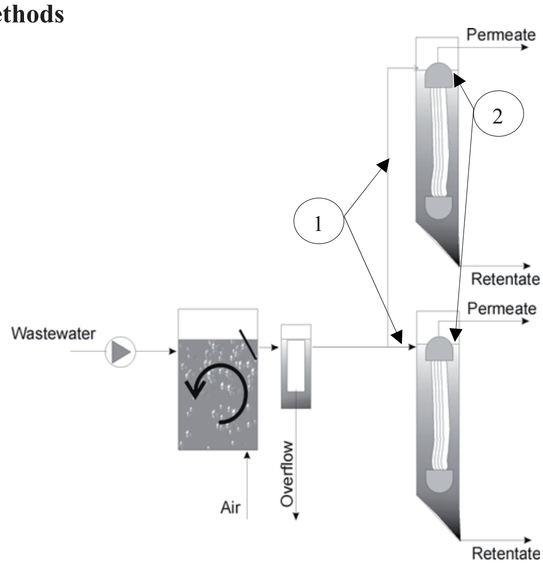


Figure 1. Experimental set up (1 and 2 refer to possible dosing points)

The pilot plant schematic used in this study is shown in Figure 1 and bioreactor configuration is described in previous studies [1][8]. Two cylindrical shaped membrane reactors with volumes of 27.5 L were connected after the biofilm reactor. The membrane reactors were designed as completely mixed reactors, however, during experimental runs sedimentation under the membrane modules was observed and measured concentrations of MLSS in the retentate was on average two times higher than the concentrate around the membrane. Membranes were operated at constant flux of 25 L m^{-2} h^{-1} (without backwash or relaxation) and with constant aeration of SAD_m ~ 1.8 Nm ^{3} m $^{-2}$ h $^{-1}$. Recovery was set at 90 % and HRT and SRT were 1 and 10.1 hour, respectively. Membranes were operated until TMP increased up to 0.3 bar or 7 operational days, after which the membranes were chemically cleaned. To alleviate uncertainties on calculated mass balances for applied additives, the experimental approach was to apply continuous dosing with dosages slightly lower than optimal dose as determined by jar tests. Additives were diluted with tap water, and then continuously dosed using a computer controlled peristaltic pump (L/S® Easy-load® II, Masterflex). Proper flocculation after applied coagulant in water and wastewater systems is normally achieved by fast and slow mixing chambers, static mixers, pipe flocculators, etc., however in this study objective was to avoid additional process units and energy requirements for that purpose. Consequently, two possible dosing point were indentified and tested, point 1 (in Figure 1) directly into the tubing connecting the distribution unit and membrane reactors (*i.e.* in-line), and point 2 (in Figure 1) at the top of the membrane reactor. Based on the calculations given in [20], the G-value for point 1 was 4 s^{-1} (Re= 290 – laminar flow) whereas for point 2 it was estimated over 5000 s^{-1}. Point 2 was therefore chosen since very good mixing and dispersion of applied additive was secured by the high G value, compared to point 1.

All analyses were performed according to national standards or Standard methods. Mixed liquor suspended solids (MLSS) were analyzed by filtering through a Whatman GF/C 1.2 µm (55 mm) glass microfiber filter according to the Norwegian Standard NS 4733. In addition, during the jar tests a Whatman 0.2 µm (55mm) was also used in order to estimate the amount of solids in the water sample between 0.2 µm and 1.2 µm. Chemical Oxygen Demand (COD), ammonia (NH_4-N), total-N (TN) and total-P (TP) were measured with the Dr Lange LCK 114, 314, 303, 304, 238, 338, cuvette tests provided by HACH LANGE GmbH, and measured with a Lasa20 spectrophotometer. For the Filtered Chemical Oxygen Demand (FCOD), Soluble Microbial Products (SMP) and dissolved organic carbon (DOC), samples were first filtered with a Whatman GF/C 1.2 µm filter. SMP_p (soluble microbial products – as protein) and SMP_c (soluble microbial products - as carbohydrate) were measured according to the Lowry [21] and Dubois [22] methods. DOC was measured by a Tekmar Apollo 9000 TOC combustion analyzer (Teledyne Tekmar, Ohio). UV absorbance (UVA_{245}) and SMP were measured with a U 3000 spectrophotometer, Hitachi. The development of transmembrane pressure (TMP) was measured continuously using an online pressure transducer connected to a National Instruments, FieldPoint (FP1000 and FP-AI-110) unit, with the LabVIEW 8.2 data acquisition and analysis software. TMP and temperature were logged every second. Four commercial metal based metal salt coagulants were chosen; two iron based using iron chloride ($FeCl_3$) and iron chloride sulfate ($FeClSO_4$), and two alum based coagulants using alum chloride and alum chloride with 50% higher basicity, both polymerized. One modified cationic polymer was used. Jar tests were conducted with iron chloride, alum chloride and cationic polymer for six different concentrations, chosen based on reported values found in the literature and preliminary jar tests with shaken flask. Prepared additives were added to mixed liquor taken from the membrane reactor in concentrations of 10, 20, 30, 40, 50 and 100 ppm metal, and polymer in 10, 30, 50, 70, 100 and 300 ppm. Jar tests were conducted in 800 ml beakers (d x h=90x120mm) aerated from the bottom with 7,5 L/min coarse bubble aeration in order to provide mixing conditions similar to that in the membrane reactor. For each sample were measured SMP_p, SMP_c, DOC, UVA_{254}, FCOD, and MLSS for two chosen filter pore sizes (*i.e.* ~ 1.2 µm and 0.2 µm). Time for reaction of the additives with the water was one hour, which is equivalent to the HRT of membrane reactors in the experimental design.

Results and discussion

Jar test results

Jar tests were designed to estimate optimum dosages that give the highest reduction of soluble organic matter (expressed as SMP_p and SMP_c, FCOD and DOC), and the highest reduction of solids fraction between 0.2 and 1.2 µm. All additives applied were observed to be able to reduce SMP's and FCOD for all dosages chosen. Thus, an optimal dose for highest organic removal was not found, suggesting that this is probably not within the range of dosages tested. The criteria for optimal dosage was therefore defined based on the highest coefficient of additive utilization CAU [mg of organic matter reduction/ mg additive], Figures 2a, 3a and 4a. Results differ for SMP's, DOC and FCOD which additionally made difficult to define what could be an optimal dosage.

From Figures 2-4 an optimum dosage not higher than 25 ppm for metal coagulants and not higher than 10 ppm for cationic polymer was defined, if all four parameters are to be taken into consideration with equal importance. Reduction of solids smaller then 1.2µm was followed by enlargement of suspended matter (>1.2µm), was observed for all applied dosages for both metal coagulants. This result indicates that colloidal matter, in general, is additionally reduced by flocculation and adsorption by suspended matter (*i.e.* fraction > 1.2µm), suggesting this is a significant mechanism for improved performance. Subsequently, the highest CAU [mg of submicron matter reduction/mg additive] value was again used as the criteria for optimal dosage. Dosages of 15 ppm for alum Figure 2b, and 10 ppm for iron Figure 3b were chosen as optimal and for the polymer was roughly estimated as 10 ppm since no significant changes in upper micron and submicron solids concentration were observed, Figure 4b.

Based on the results and analysis of the jar tests, to test possible improvements in membrane performances experimental trials with the pilot plant were performed using dosages of 9 and 22.5 ppm for both metal coagulants, iron chloride and alum chloride, and 13.5 ppm for alternative iron chloride sulfate and alum chloride with high basicity. For the cationic polymer trials were conducted with 45 ppm, and then gradually decreasing the applied dose. Based on defined optimal dosages goal of pilot plant tests was to compare which coagulant type (iron or alum based) performs better at lower and higher dosages, (by comparing iron chloride and alum chloride), if there is a significant difference in membrane performances between two iron type of inorganic coagulant (by comparing iron chloride and iron chloride sulfate) and does higher basicity of polymerized inorganic coagulant effects filterability (by comparing alum chloride and alum chloride with high basicity). Also intention was to evaluate potential of modified cationic polymer in BF-MBR and to additionally compare its efficiency to inorganic coagulants. Reduction in fouling rates that refer to overall measured fouling was related to applied dosages and reduction in amount of colloidal organic matter.

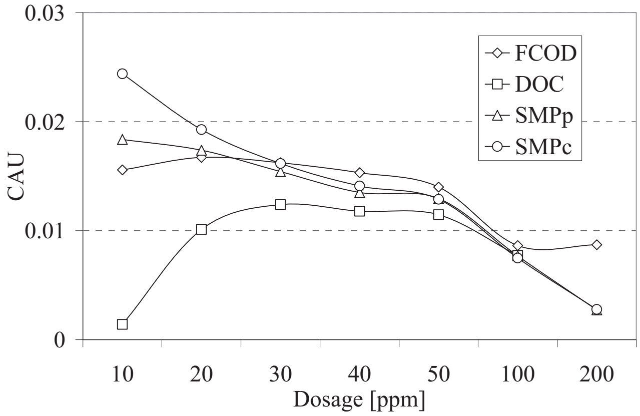
Table 1. Characteristics of inlet water, biofilm reactor effluent and removal rates

	MLSS	**COD**	**NH_4-N**	**TN**	**tot-P**
Inlet [mg/L]	176.48 (±41.22)	614.50 (±72.47)	46.29 (±8.21)	52.04 (±11.03)	22.30 (±3.21)
MBBR effluent [mg/L]	191.40 (±39.65)	326.50 (±39.60)	28.54 (±15.81)	40.11 (±7.93)	15.20 (±2.67)
R [%]	-8.45	46.87	38.34	22.92	31.82
	FCOD	**SMP_p***	**SMP_c***	**DOC**	**UV_{245}****
Inlet [mg/L]	316.80 (±54.48)	66.80 (±10.59)	10.25 (±1.66)	91.78 (±22.28)	0.69 (±0.15)
MBBR effluent [mg/L]	62.34 (±7.35)	26.52 (±4.54)	8.57 (±1.89)	23.84 (±3.30)	0.41 (±0.06)
R* [%]**	80.32	60.31	16.42	74.03	-

** - refer to total amount of proteins and carbohydrates measured by colorimetric methods [21][22]*
*** - unit **[cm^{-1}]***
****-R refer to removal rates*

Pilot plant results

The pilot plant was fed with municipal wastewater from a combined sewer system during the summer 2010. Inlet water characteristics were stable and average values were shown in Table 1. The bioreactors operated at HRT 4h had stable performance with respect to COD, FCOD, DOC, SMP's, NH-N_4, TN and tot-P removal rates (Table 1). Water temperature was on average 20±1.5 ^oC during the whole period
Alum and iron at lower dosage (*i.e.* 9 ppm), showed similar performances giving average fouling rates of 1.65 and 1.70 mbar/h, respectively, which was three times better performance then during the control runs when average fouling rate was 4.11 mbar/h.



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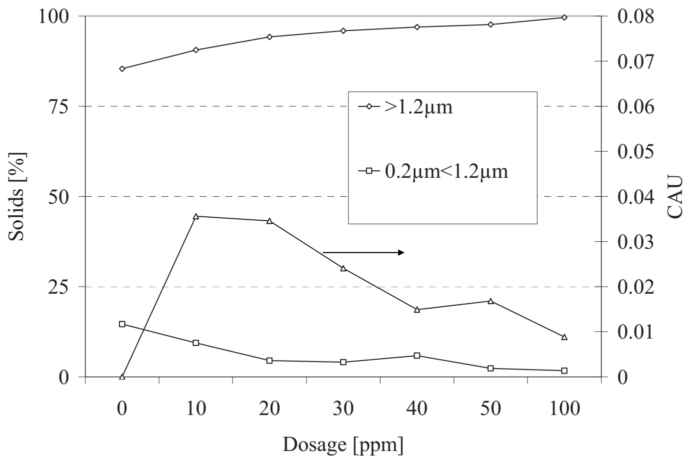

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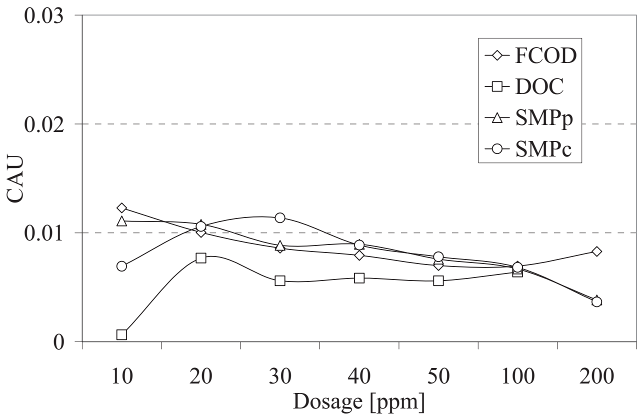
Figure 2. Removal efficiency - jar test for alum: a) soluble organic compounds, b) solids



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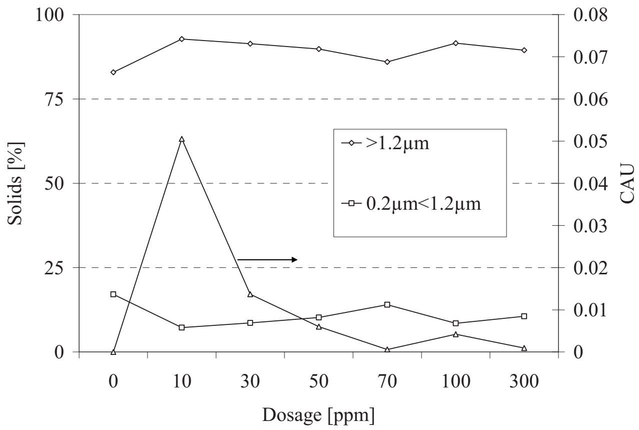
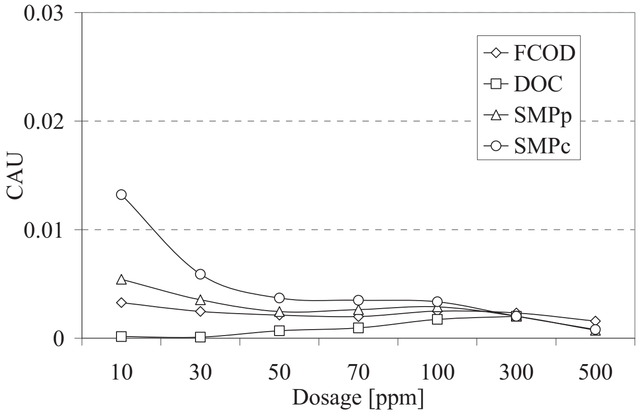


b)

Figure 3. Removal efficiency - jar test for iron: a) soluble organic compounds b) solids



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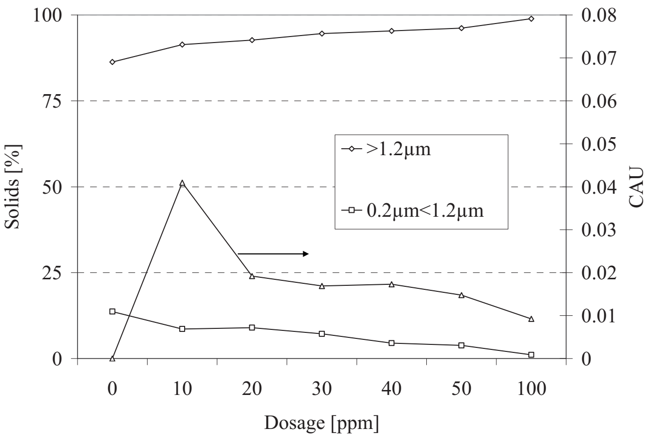

b)

Figure 4. Removal efficiency- jar test for polymer: a) soluble organic compounds b) solids

** **<u>“>1μm”</u>** refer to solids fraction higher than 1 μm and **<u>“0.2<1.2 μm”</u>** refer to solids fraction between 0.2 and 1.2 μm*

The iron based coagulant showed better performance than the alum at higher dosages (*i.e*. 22.5 ppm), giving average fouling rates of 0.58 and 1.47 mbar/h, which was 7 and 3 times lower than in control reactor. This was related to better SMP_c reduction of iron based coagulant, however not proportionally to observed reduction, again indicating complexity of membrane fouling in BF-MBR. Iron chloride sulfate at dosage of 13.5 ppm showed poorer performance in comparison to iron chloride at lower dosage (*i.e.* 9 ppm), with almost no improvement in membrane performance (*i.e*. fouling rate 3.92 mbar/h).

Alum chloride with higher basicity at dosage of 13.5 ppm resulted in almost the same fouling rates (*i.e.* 1.67 mbar/h) as regular alum chloride at lower dosage (*i.e.* 9 ppm), indicating that higher basicity not necessarily would improve filterability in BF-MBR. Alum chloride with higher basicity effected better MLSS aggregation which was measured as lower MLSS values around membrane for about 25% and in the same manner higher in retentate stream, but better DOC removal expressed thought CAU, due to this feature have not been seen as suggested in drinking water applications [18]. Results with cationic polymer indicated that only thoroughly controlled dosage of this polymer can give improvement. Continuous dosage in the beginning of the cycle gave improvement on performance since fouling rate was 1.71 mbar/h in first 24 hours, however, later sharp increments in TMP development suggested that the membrane was fouled by the polymer itself. This was observed even at much lower dosages, bellow 9 ppm. Results suggested that continuous dosing is not an option, since eventually overdose of polymer occurs which results in membrane fouling by polymer. Therefore, the effect of intermittent dosing on membrane performance was further investigated. Both membranes were operated at a higher flux of 30 $L.m^{-2} \cdot h^{-1}$, with higher recovery of 96% for 12 hours, at which a dosage of 100 and 300 mg (per L of volume reactor) was instantaneously applied (Figure 5). The sharp TMP rise was stabilized and after three hours, when TMP started to rise a continuous dose of 2.7 ppm and 0.9 ppm was applied for the next 20 hours. The strategy of 100 mg + 2.7 ppm reduced fouling rate by seven times for tested period, giving an indication that application of cationic polymer could be highly beneficial, however proper dosing strategy is crucial for successful use of this additive [14-15]. However, this finding has to be tested for longer period of time and further refinements of dosing strategies are required.

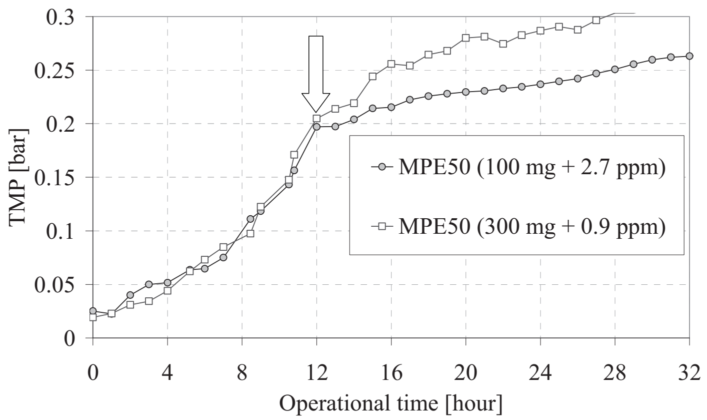

Figure 5. Suggested strategy for polymer dosage for short term experiment

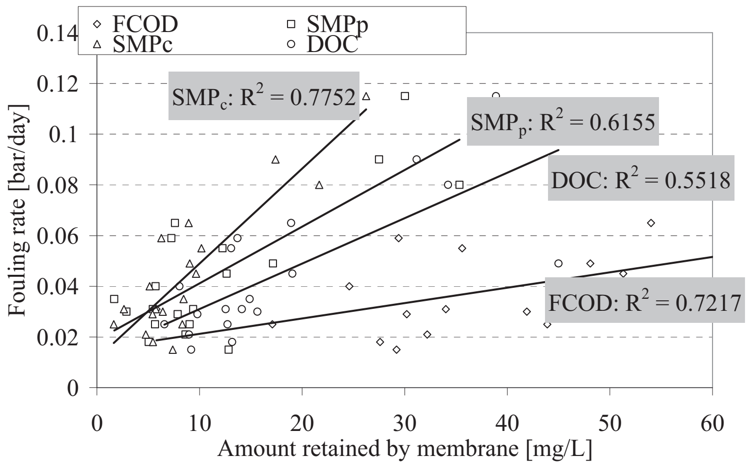

Figure 6. Correlation between given parameters and membrane fouling

This study was also used to estimate which of the parameters that represents organic colloidal matter (i.e. FCOD, SMP_p, SMP_c and DOC) could be the best with respect to predicting membrane fouling. Results from polymer were not taken in consideration, since a fouling potential by the polymer itself was observed.
Amount of compound retained by membrane was related to daily fouling rates (since chemical analysis was performed once a day from grab samples). Results suggest that SMP_c and FCOD could be used as a good fouling potential predictor, while SMP_p and DOC showed weaker correlations to fouling rates (Figure 6).

Both inorganic coagulants show a good ability to reduce total phosphorus, however, at lower dosages alum gave better removals per applied dosage compared to iron, *i.e.*1.20 to 0.98 mg P removed/mg metal, while at higher dosages this ratio was almost the same 0.58 to 0.55, respectively. The polymer tested did not affect amount of tot-P.

Conclusions

Five different additives, iron chloride and iron chloride sulfate, two polymerized alum with different basicity and a modified cationic polymer, where chosen in order to investigated a possible filterability improvement in a BF-MBR process. After extensive jar tests three dosages were chosen, 9, 13.5 and 22.5 ppm, for pilot plant trials. The best improvement in membrane performance was observed for the higher chosen dosage of iron chloride, while alum chloride gave lower improvement at the same applied dosage. Higher basicity of the polymerized alum did not give an expected improvement in filterability due to higher CAU with respect to DOC removal. A cationic polymer was found difficult to use for continuous dosing application, though a fouling reduction potential was observed for an alternative dosing strategy tested. Amount of SMP_c retained by the membrane relative to the measured fouling rates was found to be the preferred parameter with respect to predicting membrane fouling potentials. Additionally, was confirmed that FCOD could be used as good fouling predictor in BF-

MBR. Since, chemical precipitation of phosphorus by metal coagulants is common practice when applying moving-bed-biofilm reactors for wastewater treatment, a synergetic effect of phosphorus removal and improved membrane performance is foreseen when designing a BF-MBR using a moving-bed-biofilm reactor. Based on the testes conducted in this study the iron chloride coagulant in particular appears to be the additive of choice for a BF-MBR process.

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Paper P7: Improved performance through particle surface modifications by coagulation with inorganic coagulants in a BF-MBR

Igor Ivanovic[a] and TorOve Leiknes[a]

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Improved performance through particle surface modifications by coagulation with inorganic coagulants in a BF-MBR

Igor Ivanovic[a] and TorOve Leiknes[a]

[a] *NTNU - Norwegian University of Science and Technology, Trondheim, Norway*
igor.ivanovic@ntnu.no and torove.leiknes@ntnu.no

Abstract
Improved performance of the BF-MBR process by addition of inorganic coagulants, iron and alum based was studied with respect to reduction of surface area of colloidal particles, enlargement of suspended flocs, and reducing the organic colloidal content. Addition of coagulants in the membrane filtration stage of the BF-MBR had a positive effect on improvement of mixed liquor filterability and dewaterability, resulting in lower fouling rates. Modified jar tests were done in aerated beakers to determine applicable dosages by investigating effects on flocculation index, number, size and charge of colloidal particles, surface area of submicron particles, and solids and organic colloidal content. Two dosages of 9 ppm and 22.5 ppm were chosen and tested in two sets of experiments on two membrane rectors operated in parallel. Membrane fouling was reduced in a similar manner for both lower dosages of alum and iron applied, however, iron performed better at the higher dose. Alum was able to provide a higher positive charge per unit of applied coagulant, and better addressed removal of colloidal organic content measured as FCOD. Iron was more effective in enlarging surfaces of flocs in the bulk phase, leading to more adsorption of submicron particles on floc surfaces and probably promotion of a protective cake layer on the membrane surface. Improvement in membrane performances was found to correlate to reduction of measured surface area of particles in the size range 0.04 – 0.3μm, enlargement of floc surface area for particles > 8 μm, as well as reduction of concentration of the colloidal organic content. Total phosphorus removal was measured to be in the same range for both coagulants added.

Keywords. Biofilm MBR, submicron particles, inorganic coagulant, colloidal fraction

Introduction

The BF-MBR is a concept similar to conventional MBR, where a bioreactor with attached growth is employed instead of an activated sludge reactor [1]. Several advantages of the purely biofilm based MBR approach over activated sludge MBR are; 1) zero energy demand for biomass recirculation, since biomass is attached to carriers inside the bioreactor, 2) lower solids loads to membrane filtration unit, 3) lower aeration for air scouring and easier cleaning due to lower viscosity and solids content, 4) insignificant or no sludging problems of the membrane modules, 5) one more level of freedom in designing the process due to the fact that operation of biological and separation processes are decoupled [2][3]. However, membrane fouling is still the main constraint for long term sustainable operation of a BF-MBR as is the case in activated sludge based MBRs [4].
Coagulation and flocculation are commonly understood techniques that can assist

solid/liquid separation processes. In later years, coagulation has been used in MBR technologies for improvement of filterability of the activated sludge and for fouling control [4][5][6]. It has been demonstrated that different coagulants at optimum dosages are able to reduce colloidal organic content measured as SMP (EPS), DOC or FCOD [7][8], enlarge floc sizes [9][10] and reduce (bio)cake porosity [11][12]. Improvements in membrane performances are reported as lower fouling rates, longer operational cycles, higher operating fluxes or less intense mechanical and chemical cleaning demand [9]. Polymerized inorganic coagulant has the ability to provide higher positive charges than monomeric ones, and it was demonstrated that they are highly capable of reducing membrane fouling [7][9]. Positive charge delivered by hydrolysis of the coagulant in the bulk phase is partly consumed by colloids and partly adsorbed on the surfaces of flocs. Therefore, charge neutralization, bridging and adsorption on floc surfaces are the main mechanisms that are expected to induce reduction of submicron (organic) matter [13].

Application of inorganic coagulants should be beneficial for overall performance enhancement in BF-MBR, since it is expected that addition of a coagulant can reduce the amount of submicron particles in solution, which is reported to be one of the main foulants in this process [1][3]. Other positive effects are also to be expected such as enlargement of floc sizes, improved filterability and dewaterability of the solids, reduction of organic colloidal content and overall easier solids/liquid separation [14]. Negative effects of adding a coagulant are additional costs for the coagulant, higher sludge production and higher treatment costs of chemically contaminated excess sludge [15][16]. Potential toxicity of inorganic coagulants in activated sludge processes, in particular iron based, [8][13] has no effect on the quantity and activity of biomass in a biofilm process since the membrane separation stage is decoupled from the biological treatment stage in a BF-MBR.

The aim of this study is to investigate the effect of two inorganic coagulants, iron and polymerized alum based, on particle composition with respect to membrane fouling and overall system performance. Employing additional equipment (*i.e.* pipe flocculator, mixing chambers, flash and static mixers etc.) and additional energy for that purpose was intentionally avoided by applying the coagulant directly to the membrane filtration unit (Figure 1.).

Materials and methods

Two commercial metal based salt coagulants were chosen; iron based and polymerized alum based. Jar tests were done for five different concentrations, chosen based on reported values found in the literature [7][8][9] and preliminary jar tests with a shaken flask. Prepared additives were dosed to mixed liquor taken from the membrane reactor in concentrations of 10, 20, 30, 40 and 50 ppm metal. Additionally, two higher dosages were applied in order to estimate iso-electric points, *i.e.* charge neutrality of colloids. A modified jar test procedure was conducted in a 800 ml beaker (ødxh=90x120mm) aerated from the bottom with a 7.5 L/min coarse bubble aeration in order to provide mixing conditions similar to that in the membrane reactor. Monitoring of the flocs growth and stability was done by a Photometric Dispersion Analyzer (PDA 2000, Rank

Bros.Ltd., Cambridge, UK) with transparent tube of 3mm internal diameter and a flow rate of 20 ml/min according to [17]. For each dosage, changes in floc structure were measured and flocculation index logged over 30 minutes. MLSS and particle size distribution (0.04 µm-2000 µm) was measured for each sample. The sample were furthermore centrifuged @ 20000 rpm for 3.5 min and supernatant was taken for measurement of colloidal organic content (*i.e.* FCOD), unimodal size distribution of nano-particles (in range 3 nm to 1000 nm) and zeta potential. Total time for reaction for each applied coagulant dose with water was one hour.

A schematic of the BF-MBR pilot plant used in this study is shown in Figure 1 and the bioreactor configuration is described in previous studies [1][3]. Membranes were operated continuously at constant flux of 25 L m^{-2} h^{-1} (without backwash or relaxation) and with constant aeration of SAD_m ~ 1.8 Nm^3 m^{-2} h^{-1} . Recovery was set at 90 % and HRT and SRT were 1 and 10.1 hour, respectively. Membranes were operated until TMP increased up to 0.3 bar or 7 operational days, after which the membranes were chemically cleaned. Two sets of experiments, Exp 1. and Exp 2., with lower and higher chosen dosages by alternating type of coagulant. Coagulants were diluted with tap water, and then continuously dosed using a computer controlled peristaltic pump (L/S® Easy-load® II, Masterflex). The top of the membrane reactor was chosen as a coagulant adding point since very good mixing and dispersion of applied additive was secured by strong aeration for membrane air scouring purpose.

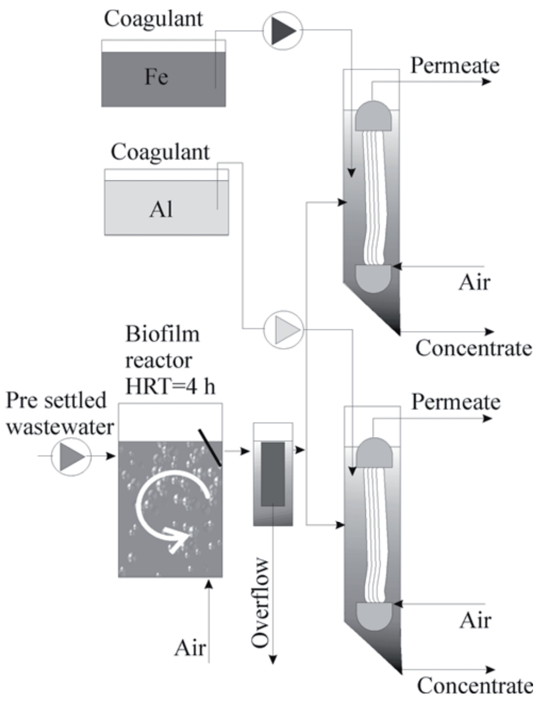


Figure 1. Experimental set up

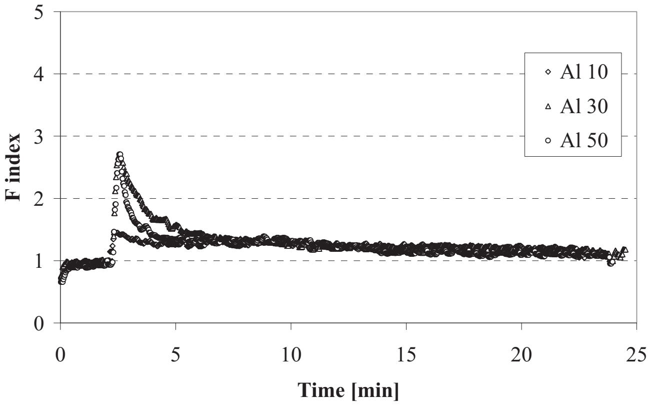
All analyses were performed according to national or Standard methods [18]. Suspended solids (SS) were analyzed by filtering through a Whatman GF/C 1.2 µm (55 mm). Samples for Filtered Chemical Oxygen Demand (FCOD) and total phosphorus were filtered with a Whatman GF/C 1.2 µm filter and measured by, the Dr Lange LCK , 014, 114, 314, 303 and 238 cuvette tests by HACH LANGE GmbH and measured with a Lasa20 spectrophotometer. Surface area % of particles in the range 0.04 – 1000 µm were done by a laser diffraction method with a LS 230 [19]. Unimodal size and total number of particles (*i.e.* counts) in the range 3 - 1000 nm were done by photon correlation spectroscopy using a N4 plus [20] and zeta potential was measured with a DelsaNano [21], all products of Beckman Coulter. Capillary suction time was measured by CST Type 304M Mains, Triton Electronics. The development of transmembrane pressure (TMP) was measured continuously using an online pressure transducer connected to a National Instruments, FieldPoint (FP1000 and FP-AI-110) unit, with the LabVIEW 8.2 data acquisition and analysis software. TMP and temperature were logged every second. Logging files were later filtered and values were hourly averaged and plotted.

Results and discussion

Jar test results

Jar tests were designed to estimate applicable dosages based on four criteria; the highest flocculation index, the highest reduction of surface area percentage of (submicron) particles measured in the range of 0.04 - 2000µm, the highest reduction in number of particles measured in the range 3 – 1000 nm, and zeta potential. The FCOD and suspended solids were also measured in order to evaluate the effect of added coagulant on colloidal matter and mixed liquor solids content. (Figures 2-6).

Polymerized alum had lower flocculation indexes (FI) for higher applied dosages than iron chloride. However, the effect of aeration applied for mixing purposes resulted in deflocculation in about 5-8 minutes. Under these conditions the flocculation index dropped to 1,1 regardless of applied dosages or type of coagulant used. The FI was set to value 1 before coagulant was added (Figure 2a) and 2b)). Based on this measurement the lower applicable dosage of 10 ppm appears to be the practical recommendation.



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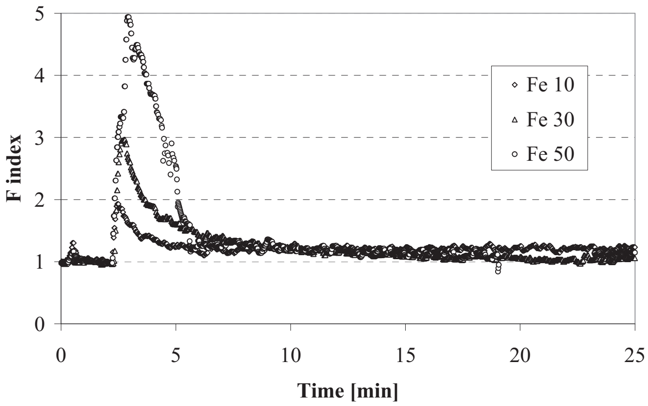

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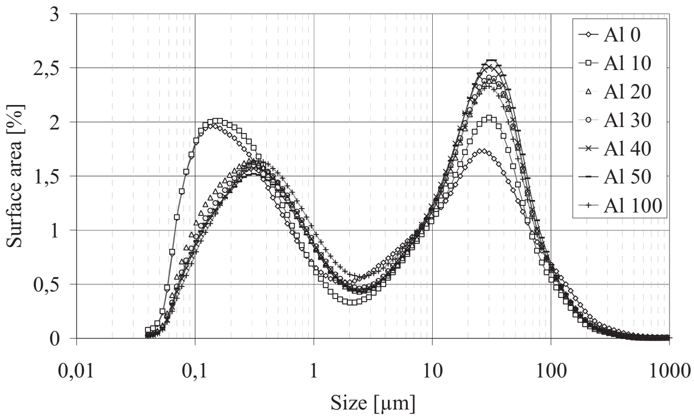
Figure 2. Jar test results – Flocculation index for: a) alum , b) iron

Taking into account the reduction of surface area of submicron particles (<1μm), the optimal dose appears to be not lower than 20 ppm for alum and between 10-20 ppm for iron, since no significant reduction of particle surface area was measured for higher applied dosages. For the larger particles greater than one micron a similar conclusion can be drawn (*i.e.* enlargement of flocs surface) (Figure 3a) and 3b).



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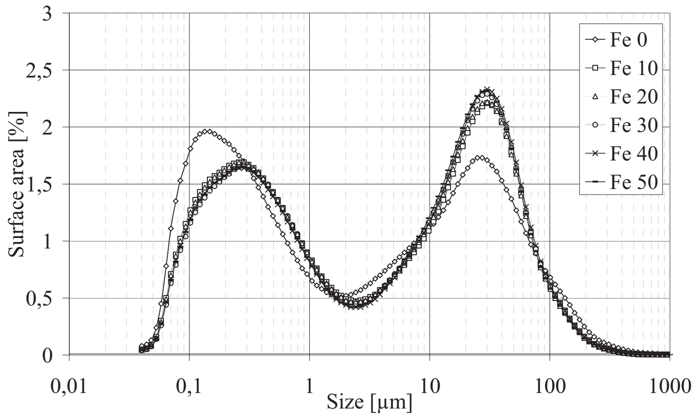

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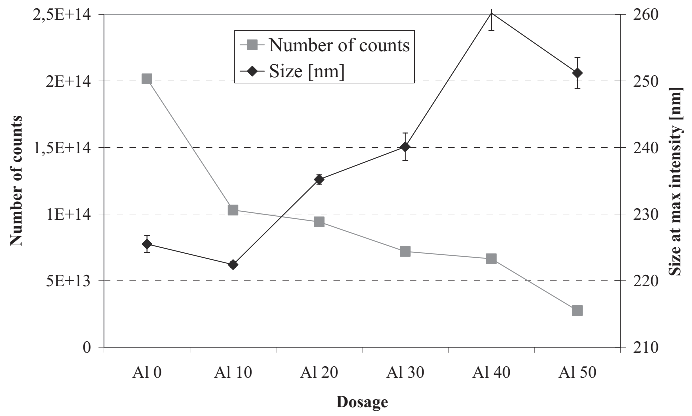
Figure 3. Jar test results – Changes in distribution of particles surface area % : a) alum , b) iron

Changes and growth in size of the nano particles in the supernatant, and consequently their reduction in number, as a function of applied dosage is presented in Figure 4a) and 4b). A unimodal size at highest laser diffraction intensity had a peak at 260 (±3.95) nm for 40 ppm of alum (Figure 4a)). For iron, the largest sizes were measured at 236.3 (±5.51) nm for a dosage of 30 ppm (Figure 4 b)). The highest reduction in number of counts (*i.e.* particles) per applied dosage was for a dosage of 10 ppm for both coagulants. Alum performed slightly better at higher applied dosages compared to iron (*i.e.* had higher reduction in number of counts).



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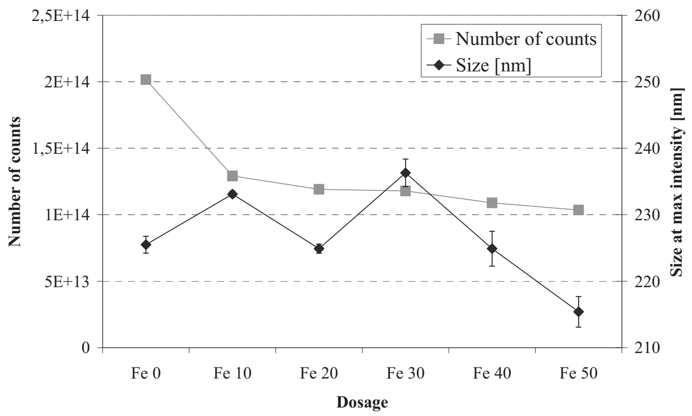

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Figure 4. Jar test results – Number of particles and size at max intensity : a) alum , b) iron

The stability of the colloids was determined by measuring zeta potentials as a function of applied dosages. No significant destabilization of the colloids was observed for most applied dosages. When additional coagulant was applied iso-electrical charge inversion took place and iso-electric points where estimated by linear interpolation for dosages of 92 ppm alum and 155 ppm iron (Figure 5). These results suggest that charge neutralization is probably not a key destabilizing mechanism and therefore not be appropriate criteria for estimating applicable doses. Adsorption and/or bridging thus appears to be the dominant mechanisms for reduction of the colloidal fraction in the complex suspension of flocs, large unassociated particles, colloids and solutes as found in the bulk phase in BF-MBR processes [9].

Sludge enlargement measured as MLSS and reduction of organic colloidal content (*i.e.* FCOD) for applied dosages are shown in Figure 6. Alum was found to be more effective in reducing the amount of organic colloidal matter with increasing dose compared to iron, while both coagulants increased the sludge content to some extend proportionally to applied dosages.

Pilot plant results

Studies on a BF-MBR pilot plant were further conducted with two sets of experiment, identified as Exp 1. and 2. Two different doses of 9 ppm and 22.5 ppm, for both coagulants tested, where chosen based on jar test results in order evaluate the effectiveness with respect to changes in particle composition and overall membrane filtration performance. In Exp 1. one reactor was operated with 9 ppm alum and the other with 22.5 ppm of iron. After that membranes were chemically cleaned and Exp 2. was then conducted with 9 ppm of iron and 22.5 ppm of alum.

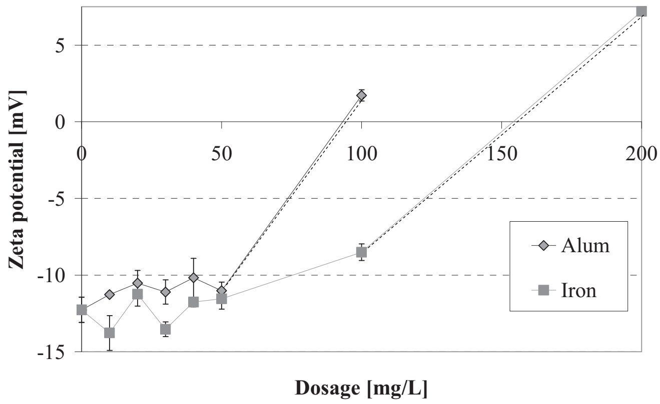


Figure 5. Zeta potential

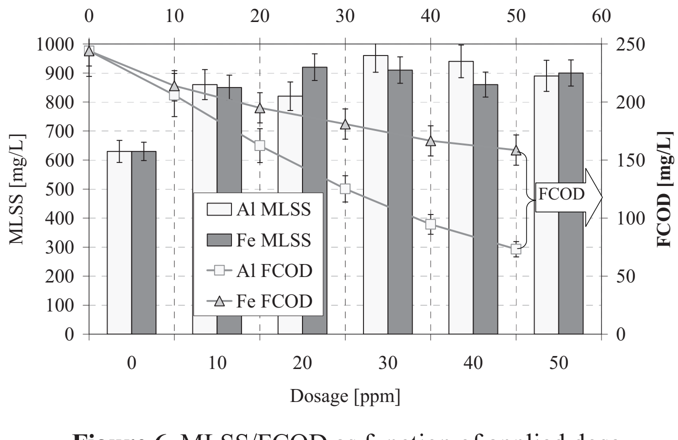


Figure 6. MLSS/FCOD as function of applied dose

Prior to these tests the pilot plant was operated for a two week period without coagulant, both to synchronize the performances of the membrane reactors and to minimize uncertainties in operation, and to obtain reference TMP values for operation without additives.

The pilot plant was fed with municipal wastewater from a combined sewer system in the municipality of Trondheim, Norway. Inlet water characteristics are shown in Table 1. The bioreactors operated at HRT of 4 hours with a stable performance with respect to COD, FCOD, and tot-P removal rates (Table 1). Water temperature was close to room temperature, on average 18.5 – 21.5 ^oC, during the whole period.

Results from the study with the additives are summarized in Table 2. Alum was found to have a better ability to bind colloidal organic matter compared to iron for both chosen dosages (Table 2). This is in agreement with similar results are reported in other studies [9]. Removal of colloidal organic matter by membrane filtration with addition of coagulant (highest to lowest) was in the order Al 22.5 ppm; Al 9 ppm; Fe 22.5 ppm; Fe 9 ppm.

Table 1. Average values for Exp 1. and Exp 2. of inlet water and biofilm reactor effluent

	MLSS [mg]	**COD [mg/L]**	**FCOD [mg]**	**TP [mg]**
Inlet [mg/L]	176.48 (±41.22)	614.50 (±72.47)	316.80 (±54.48)	22.30 (±3.21)
BF eff. [mg/L]	191.40 (±39.65)	308.50 (±39.60)	62.34 (±7.35)	15.20 (±2.67)
Rem. rates [%]	-8.45	46.87	80.32	31.82

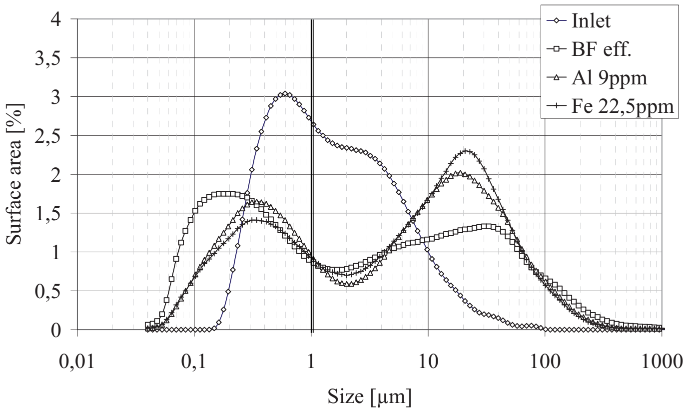
The amount of solids loaded on membrane surface as a function of applied coagulant and dose was in the order Fe 22.5 ppm; Al 22.5 ppm / Al 9 ppm; Fe 9 ppm. It should be noted thought that during Exp 2, on average about a 7% higher solids loads was coming with the effluent from the biofilm reactor, resulting in higher MLSS values than expected based on mass balance and measured values from Exp 1 (Table 2). During both experiments, MLSS concentrations in the concentrate were higher than in the bulk phase, indicating that sedimentation took place in the reactor beneath the membrane. Sedimentation factor (Ks) [3] for the reactors was in the range 1.5-2.7. For the Fe 22.5 ppm dose, MLSS was on average significantly higher compared to the other test conditions and Ks value was the lowest, suggesting that flocs were lighter and did not settle in same manner as flocs formed when alum was added. Dewaterability expressed as normalized capillarity suction time (CSTn - normalized with respect to MLSS concentration) was in good agreement with observed fouling rates for lower applied dosages, and having almost the same value for both coagulants at higher dosages (*i.e.* ~40 seconds g^{-1} L^{-1} - Table 2). Lower values represent better dewatering properties.
Both alum and iron gave similar performances in removal of total phosphorus for respective dosages applied (Table 2).

Results of measured surface areas of particulate content in the range from 0.04 to 2000 µm for four sampling points are shown in Figures 7 a) and 7b). The biological treatment significantly changes the characteristics of the particulate matter in the wastewater, particularly with a large reduction of particles in the size range 0.3 to 7-8 µm. Additionally, a clear increase in percent surface area of particles in the range 0.04 to 0.3 and flocs > 8 µm was measured.

Table 2. Average values for Exp 1 and Exp 2 of bulk phase, permeate and concentrate

		Bulk phase (around the membrane)			
		MLSS [mg/L]	**COD [mg/L]**	**FCOD [mg/L]**	**CSTn [s/g/L]**
Exp 1	**BF eff.**	184.00 ±28.86	307.67 ±12.42	59.30 ±12.21	-
	Fe 9	1040.00 ±60.83	1206.67 ±88.12	71.20 ±18.14	68.13
	Al 22.5	1271.67 ±391.16	1205.67 ±362.4	43.00 ±14.39	40.56
Exp 2	**BF eff.**	198.00 ±54.11	315.33 ±50.81	65.30 ±7.45	-
	Al 9	1315.56 ±140.33	1503.33 ±82.03	63.43 ±6,26	54.82
	Fe 22.5	2192.22 ±236.46	2409.00 ±326.51	58.67 ±3.85	41.26
		Permeate		**Concentrate**	
		FCOD [mg/L]	**TP [mg/L]**	**MLSS [mg/L]**	**COD [mg/L]**
Exp 1	**BF eff.**	-	-	-	-
	Fe 9	26.20 ±9.75	4.88 ±0.15	2186.1 ±889.5	2674.0 ±520.1
	Al 22.5	27.17 ±4.95	1.46 ±0.64	3308.9 ±280.4	3068.0 ±429.0
Exp 2	**BF eff.**	-	-	-	-
	Al 9	32.23 ±6.17	4.04 ±0.41	4046.0 ±1396.5	4313.3 ±1926.4
	Fe 22.5	29.00 ±4.51	1.66 ±0.30	3469.7 ±753.7	3165.00 ±570.3

The highest surface area % was found for particles in the size range 0.1 to 0.3 µm for Exp 1., and 0.08 to 0.3 for Exp 2, with respective peak values of 1.75 and 2.6 µm. These results show that the biofilm effluent during Exp 1. had more favorable characteristics with respect to lower submicron colloidal content than in Exp 2. However, added coagulants where not able to significantly change the fingerprint of the BF effluent. Both coagulants applied were capable of reducing the surface area of the submicron content by increasing the number of larger particles and thus increasing the floc surface area of particles in the upper micron range. Effectiveness of the added coagulant with respect to shifting towards a more favorable distribution in the submicron range was in the order Fe 22.5 ppm; Al 22.5 ppm ~ Al 9 ppm; Fe 9 ppm. Enlargement in flocs surfaces due to presences of a coagulant followed the same order.



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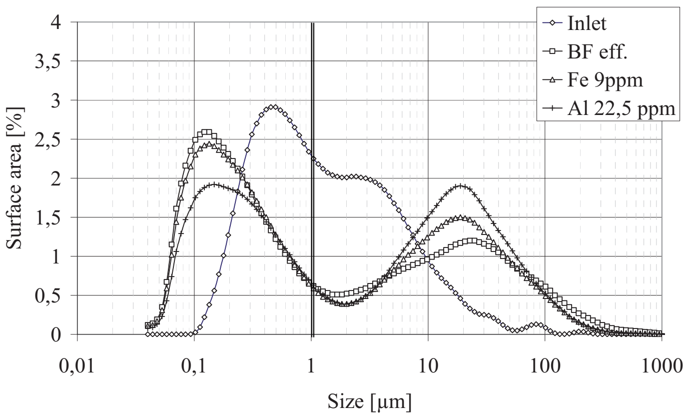

b)

Figure 7. Surface area distribution for four sampling points for a) Exp 1. and b) Exp 2.

Both alum and iron at the lower dosage (*i.e.* 9 ppm), performed similarly giving average fouling rates of ~1.5 and ~1.7 mbar/h, respectively. This was lower than average fouling rates observed during the control runs, measured at ~ 4 mbar/h. The iron based coagulant showed better performance than the alum at the higher dosages (*i.e.* 22.5

ppm), giving average fouling rates of 0.6 and 1.5 mbar/h (Figure 8a) and 8b)). The ability of the coagulants to reduce fouling based on type and dose was found to be in the order Fe 22.5 ppm; Al 22.5 ppm; Al 9ppm ~ Fe 9ppm. These results suggest that even though higher solids concentration was measured (Table 2), these probably consisted of lighter weight flocs with high surfaces area (in range 3 – 80 µm (Figure 7a)), leading to a higher adsorption and reduction of the submicron particles.

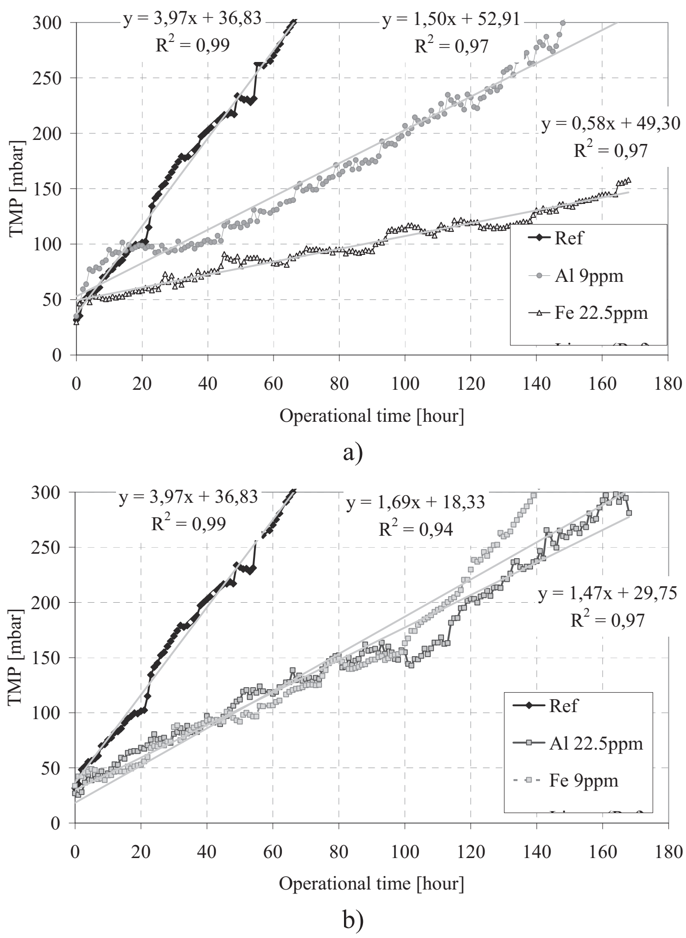

Figure 8. The TMP development for a) Exp 1. and b) Exp 2

Additionally, it is highly probable that the suspension with large surface area and light flocs has formed a protective cake layer on the membrane surface, reducing the fouling potential of the colloidal (organic) content in bulk phase and forming a dynamic cake layer with less hydraulic resistance. It is also important to note that the BF effluent during the Exp 1. had more favorable characteristics (*i.e.* less submicron particles) than during Exp. 2, and firm conclusions can therefore not be made based on this set of experiments. Further studies on the nature and characteristics of the cake layer formed on the membrane are required.

Conclusions

Two inorganic coagulants were applied in order to investigate the potential of reducing membrane fouling in a BF-MBR process by coagulation. Extensive trials using a modified jar test suggested that applicable dosages should be in the range of 10 to 25 ppm for pilot plant trials, based on FI, reduction in particle number, charge, size and reduction in surface area of submicron particles. Pilot studies with addition of both coagulants at the lower dosage range resulted in similar membrane performances improvement (*i.e.* less fouling). The best improvement in membrane performance was observed for the higher dosage range using iron chloride, while polymerized alum gave a lower improvement in performance at the same applied dosage. Results suggest that the response observed was related to a higher reduction in particle surface area of the submicron particles in the size range 0.04 – 0.3µm, and the lower organic colloidal content.

The improved membrane performances observed at the higher iron dosage as assumed to be related to the formation of lighter flocs with large surface areas which can form a protective cake layer on the membrane surface thereby reducing fouling by the submicron fractions. Both coagulants were able to satisfactory remove phosphorus. Based on the testes conducted in this study the iron chloride coagulant at doses around 20 ppm appears to be the coagulant of choice for a BF-MBR process reactor. Further studies are required to optimize the use of coagulant and to get a better understanding of the particle properties and characteristics best suited to give an overall improvement on membrane filtration performance in a BF-MBR process.

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Appendix B: Secondary papers and proceedings, poster presentations

Secondary paper P8: Investigating the effect of colloids on the performance of a biofilm membrane reactor (BF-MBR) for treatment of municipal wastewater.

Leiknes, TorOve; Ivanovic, Igor; Ødegaard, Hallvard.

Water S.A. 2006; Volume 32.(5) s. 708-714

Abstract

Performance of membrane reactor in combination with moving-bed-biofilm reactor (BF-MBR) for treatment of municipal wastewater was investigating in relation to different organic loading rates; high and low rate. Membrane was operated with constant flux 50 LMH and 96% recovery. The fouling rate was evaluated as development of the transmembrane pressure (TMP) during the operational time. Suspended solids concentrations (SS), organic matter (COD/FCOD), particle size distributions (PSD), capillary suction time (CST) and time to filter (TTF) were daily measured and further related to TMP in order to determinate major fouling factor. A higher fouling potential was observed for high rate operating conditions. Fraction of organic matter below 1.2 μm was mostly related to the changes in TMP. Furthermore higher amount of particulate fraction below 0.1 μm in membrane reactor during the high rate presented a dominant contribution to membrane fouling - colloidal fouling.

Secondary paper P9: Fouling control by reduction of submicron particles in a BF-MBR with an integrated flocculation zone in the membrane reactor..

Igor Ivanovic, TorOve Leiknes, Hallvard Ødegaard

Separation Science and Technology, 43: 1871–1883, 2008

Abstract

Submicron particles represent one of the major foulants in the biofilm membrane reactor BF-MBR. Reduction of the amount of submicron particles (colloids) adjacent to the membrane is one measure in order to provide better fouling control in BF-MBR systems. A submerged hollow fiber (Zenon Zeeweed) membrane reactor was redesigned by introducing a flocculation zone below the aeration device of the membrane module. This resulted in reduction of submicron particles around the membrane from 8.2% to 6.9 %, expressed in differential number percentage. The size of the most abundant particle fraction consequently increased from 0.70 to 0.84 μm. Furthermore, the modified membrane reactor design provided longer operational cycles, >40% reduction of suspended solids around the membrane, and improved retentate/concentrate characteristics, i.e. dewaterability (CST), settleability (SVI/SSV) and filterability (TTF).

Secondary paper P10: Influence of loading rates on production and characteristics of retentate from a biofilm membrane bioreactor (BF-MBR).

Ivanovic, Igor; Leiknes, TorOve; Ødegaard, Hallvard.

Desalination 2006; Volume 199. s. 490-492

Introduction

A hybrid biofilm membrane reactor based on the moving-bed-biofilm reactor (MBBR) combined with solids separation in an immersed membrane reactor (IMR) has been investigated for the treatment of municipal wastewater. This process enables the design of compact treatment plants with high biodegradation efficiencies that produce high quality effluents. Retentate characteristics play an important role in the efficiency of the solids separation and subsequently the overall performance of the membrane unit. The aim of this study was to investigate and compare characteristics of the retentate in the membrane reactor as a function of organic loading rates in the bioreactor of this BF-MBR process.

Summary of publications (in chronological order)

Influence of loading rates on production and characteristics of retentate from a biofilm membrane bioreactor (BF-MBR).

I. Ivanovic,; T.O. Leiknes; H. Ødegaard.

Desalination, **199** (2006) 490-492

Investigating the effect of colloids on the performance of a biofilm membrane reactor (BF-MBR) for treatment of municipal wastewater.

T.O.Leiknes,; I. Ivanovic; H. Ødegaard.

Water S.A. **32(5)** (2006) 708-714

Fouling control by reduction of submicron particles in a BF-MBR with an integrated flocculation zone in the membrane reactor..

I. Ivanovic,; T.O. Leiknes; H. Ødegaard.

Separation Science and Technology, **43** (2008) 1871-1883

Impact of aeration on particle colloidal fraction in the biofilm membrane reactor (BF-MBR)

I. Ivanovic and T.O. Leiknes

Desalination, **231** (2008) 182-190

Membrane reactor as a tool for better membrane performance in a biofilm-MBR (BF-MBR)

I. Ivanovic and T.O. Leiknes

Desalination and Water Treatment, **25** (2011) 259-267.

Impact of denitrification on the performance of a biofilm-MBR (BF-MBR).

I. Ivanovic and T.O. Leiknes

Accepted in *Journal of Desalination*

Effect of addition of different additives on overall performance of bioflim - MBR (BF-MBR)

I. Ivanovic and T.O. Leiknes

Accepted in journal *Desalination and Water Treatment*

Improved performance through particle surface modifications by coagulation with inorganic coagulants in a BF-MBR

I. Ivanovic and T.O. Leiknes

Submitted to journal of Separation and Purification Technology

The biofilm membrane bioreactor (BF-MBR) - a review-

I. Ivanovic and T.O. Leiknes

Submitted to journal of *Desalination and Water Treatment*

Particle separation in moving bed biofilm reactor – applications and opportunities

I. Ivanovic and T.O. Leiknes

Submitted to journal of *Separation Science and Technology*

Conferences and workshop presentations

Ivanovic, Igor; Leiknes, TorOve, Ødegaard, Hallavrd.

Influence of loading rates on production and characteristics of retentate from a biofilm membrane bioreactor (BF-MBR). EUROmembrane 2006, Italy.

Leiknes, TorOve,. Ivanovic, Igor;

Impact of aeration rates on particle colloidal fraction in a biofilm membrane bioreactor (BF-MBR).IWA 4th International membrane conference, Membranes for Water and Wastewater Treatment. Harrogate, UK, 2007.

Ivanovic, Igor; Leiknes, TorOve.

Effect of in-line coagulation on the membrane performance in biofilm -MBR. IMSTEC 07, 6th International Membrane Science and Technology Conference; 2007

Ivanovic, Igor; Leiknes, TorOve.

Importance of membrane reactor design on membrane performance in biofilm-MBR. ICOM 2008, Honolulu Hawaii, USA

Ivanovic, Igor; Leiknes, TorOve.

Importance of submicron particles on a membrane performance in BF-MBR. Engineering with Membranes 2008, EUROMBRA workshop

Leiknes, TorOve; Ivanovic, Igor.

New Membrane Reactor Design for Better Membrane Performance in a Biofilm MBR (BF-MBR).
Proceedings of Engineering with Membranes 2008 Membrane Processes: Development, monitoring and modelling - from the micro to the macro scale. Spain: Servicio de Publicaciones, Universidad de Oviedo 2008 ISBN 978-84-691-3670-6. s. 131-134

Leiknes, TorOve; Phattaranawik, Jirachote; Ivanovic, Igor.

Challenges and potentials of biofilm-MBR for municipal wastewater treatment. Final MBR-Network Workshop; Berlin Germany, 2009

Leiknes, TorOve; Phattaranawik, Jirachote; Ivanovic, Igor.

Prospects and Potentials of Biofilm-MBRs for Municipal Wastewater Treatment. WEFTEC 2010, Orlando Florida, USA

Ivanovic, Igor; Leiknes, TorOve

Impact of denitrification on the performance of a biofilm ‐ MBR (BF-MBR), MDIW2010, Trondheim, Norway

Leiknes, TorOve, Ivanovic, Igor;

Effect of addition of different additives on overall performance of bioflim - MBR (BF-MBR), ASM6/IMSTEC 2010, Sydney, Australia

Poster presentations

Leiknes, TorOve; Ivanovic, Igor; Ødegaard, Hallvard.

Assessment of fouling in a biofilm-MBR for treatment of municipal wastewater. 4th IWA Leading-Edge Conference on Water and Wastewater Technologies; 2007

Ivanovic, Igor; Fitz, Markus; Leiknes, TorOve.

Influence of nano particles on the membrane fouling in the biofilm membrane bioreactor (BF-MBR)., IWA- 2th National Young Water Professionals Conference, Germany; 2007