Tracking particle size distributions in a moving bed biofilm membrane reactor for treatment of municipal wastewater

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Abstract
A study has been conducted to investigate the effect of loading rates on membrane fouling in a moving bed biofilm membrane reactor process for municipal wastewater treatment, especially analyzing the fate of submicron colloidal particles and their influence on membrane fouling. Two operating conditions defined as low and high organic loading rates were tested where the development and fate of the particulate material was characterized analyzing the particle size distributions throughout the process. Analysis of the membrane performance showed higher fouling rates for the high-rate conditions. The fraction of colloidal submicron particles was higher in the membrane reactor indicating that fouling by these particles was a dominant contribution to membrane fouling.

Keywords
Particle size distribution, wastewater, membrane bioreactors, fouling.

INTRODUCTION
Membrane bioreactors (MBR) are commonly understood as the combination of membrane filtration and biological treatment using activated sludge (AS) where the membrane primarily serves to replace the clarifier in the wastewater treatment system (van der Roest et al., 2002). A variety of process configurations exist where the membrane is installed either in an external unit or immersed in the aeration tank and where the systems are designed to be operated under low-pressure vacuum. One of the benefits of AS-MBRs compared to conventional activated sludge systems is that they can be operated at higher suspended biomass concentrations resulting in long sludge retention times even at small reactor volumes. However, as common with all membrane processes, the efficacy of the MBR is largely constrained by fouling, i.e. the accumulation of materials on the surface of or within the membrane resulting in a reduction in the membrane permeability. Fouling is particularly a problem in MBRs since the process deals with liquors having high concentrations of total solids as well as dissolved compounds such as extracellular polymeric substances (EPS). Fouling is therefore caused by different substances and the mechanisms are rather complex and interrelated. Certain types of fouling (reversible) can be removed by backwashing, i.e. cake formation and loose depositions, while others are permanent (irreversible, fouling which is only recoverable by chemical cleaning). The latter is often attributed to EPS (Nagaoka et al., 1996, Chang et al., 2002). A second limitation, clogging – which refers to the filling of the membrane interstices with solids – must also be suppressed for successful operation. All these phenomena limit the flow of water through the membrane demanding physical and/or chemical membrane cleaning to restore permeability. Both the concentration and the characteristics of the solids are therefore important parameters for designing MBR processes and for optimizing fouling control and cleaning strategies.

An alternative strategy to reduce the effect of membrane fouling by high biomass concentrations as in AS-MBR systems is combining membranes with biofilm processes (BF-MBR). Conventional biofilters are prone to clogging at high loads of organic and particulate matter resulting in frequent backwashing. The moving bed biofilm reactor (MBBR) is an alternative biofilm reactor which tolerates high particulate and organic loading rates (Ødegaard et al., 2000). In the Kaldnes MBBR process, the biofilm carriers are small plastic cylinders which are suspended in the reactor by aeration (Ødegaard et al., 1994). The process is very compact compared to conventional AS
treatment process and may be used for low-rate as well as high-rate systems. The solids concentrations in the effluent from a MBBR reactor (and influent to the membrane reactor) would typically be around 100-200 mg/l depending on water characteristics and loading rate. This is dramatically lower that in an activated sludge/membrane reactor and if particulate fouling is an important fouling mechanism, there is a potential with this process combination for efficient biomass separation by membrane filtration while managing particulate fouling. By combining a compact biofilm process with efficient particle separation stable high quality effluent can be achieved. Membrane separation after a moving bed biofilm reactor offers therefore outstanding control of the effluent.

The objective of this study has been to investigate the effect of loading rates and membrane fouling on the performance of a moving bed biofilm membrane reactor for the treatment of municipal wastewater. In particular, potential fouling by the particulate matter has been evaluated by tracking variations in particle size distributions throughout the process. A high and low loading rate on the biofilm reactor was tested where the performance of the membrane separation unit was defined by fouling rates measured for the given operating conditions.

MATERIAL AND METHODS

Experimental setup

The schematic of the treatment process and experimental configuration is presented in Figure 1. Municipal wastewater from a combined sewer system is fed to a pilot scale moving bed biofilm membrane reactor (MBB-M-R). The wastewater is pretreated using a small gravity settler and is then pumped from an overflow into the moving bed biofilm reactors (MBBR). The outlet of the bioreactors is subsequently led into the membrane reactor tank from where permeate is extracted through the membrane under vacuum. A small volume of the concentrate, i.e. retentate, is removed as excess sludge.

Figure 1. Schematic of the MBB-M-R concept and experimental configuration

Table 1. Operating conditions

<table>
<thead>
<tr>
<th></th>
<th>High-rate operation (45 min HRT)</th>
<th>Low-rate operation (180 min HRT)</th>
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<tbody>
<tr>
<td>Average load on MBBR:</td>
<td></td>
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<tr>
<td>COD</td>
<td>24 gO₂/m².d (7.8 kgO₂/m³.d)</td>
<td>COD 7 gO₂/m².d (2.3 kgO₂/m³.d)</td>
</tr>
<tr>
<td>FCOD¹</td>
<td>12.1 gO₂/m².d (3.9 kgO₂/m³.d)</td>
<td>FCOD¹ 3.4 gO₂/m².d (1.1 kgO₂/m³.d)</td>
</tr>
<tr>
<td>Average SS in membrane reactor ~ 800 mgSS/l</td>
<td>Average SS in membrane reactor ~ 500 mgSS/l</td>
<td></td>
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</tbody>
</table>

¹ FCOD: COD in filtered sample (Whatman GF/C-filter)

The biological treatment process. Four pilot plant biofilm reactors were installed in series. Each reactor had a volume of 65 liters giving a total of 0.26 m³. The Kaldnes K1 biofilm carrier was used which has a surface area for biofilm growth of 325m²/m³ of reactor volume. The filling fraction of media was 67% as normal for this process, resulting in a biofilm growth area of 21.1 m² per reactor and a total growth area of 84.5 m². To evaluate the effect of varying loading rates on the overall performance of the process two conditions were chosen, defined as high-rate and low-rate operation.
respectively. A summary of the two operating conditions is presented in Table 1. When operating in
the high-rate mode, only one of the four MBBR’s was used while all reactors were used in the low-
rate mode. The organic loading was such under high-rate conditions that no nitrification took place
while complete nitrification was ensured under the low-rate conditions. The two operating
conditions were chosen such that a distinction between the two effluents with respect to solids
characteristics was achieved.

The membrane separation process. The membrane reactor was based on a submerged reactor design
where the treated wastewater effluent is removed as permeate. A ZW-10 pilot module (supplied by
Zenon Inc.) was applied in this study with an operating flux set to 50 l m⁻² h⁻¹ for a pressure range
varying from 0.1-0.5 bar. The performance of the membrane module was determined by measuring
the transmembrane pressure (TMP) for constant flux operation. The development of TMP for
different fluxes was measured continuously using an online pressure transducer connected to a data
acquisition system from National Instruments, FieldPoint (FP1000 with FP-AI-110 analogue input),
in combination with the LabVIEW 6.1 data acquisition and analysis program. The water
temperature was also logged continuously with a temperature transducer. Water flow rates were
measured manually with rotameters on the respective lines of flow. Fouling rates for the membrane
performance was calculated as rate of permeability decline, expressed as normalized flux divided by
the transmembrane pressure (1 m⁻² h⁻¹ bar⁻¹). The membrane reactor was operated in a cyclic mode
consisting of a production period of 9.5 minutes with a 0.5 minute backwash.

<table>
<thead>
<tr>
<th>Membrane reactor specifications</th>
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<tr>
<td>Reactor volume</td>
</tr>
<tr>
<td>Hydraulic retention time</td>
</tr>
<tr>
<td>Sludge retention time</td>
</tr>
<tr>
<td>Configuration</td>
</tr>
<tr>
<td>Nominal Membrane Surface area</td>
</tr>
<tr>
<td>Nominal pore size</td>
</tr>
</tbody>
</table>

Analysis
All analyses were performed according to national or international standards. Suspended solids
were analysed by filtering through Whatman GF/C 1.2 µm according to the NS 4733. Chemical
Oxygen Demand (COD) was measured with the Dr Lange LCK314 cuvette test. For the Filtered
Chemical Oxygen Demand (FCOD) the sample was first filtered with a Whatman GF/C glass
microfiber filter. Total phosphorus was analysed according to NS-EN ISO 4725. Ammonia was
analysed by Ammonia selective electrode (ORION, model 95-12) and by Dr Lange Dosi Cap Zip
Ammonium LCK 303. Particle size distribution analysis was also done using laser diffraction
spectroscopy (Beckman Coulter LS230).

TMP was logged for every second. The data files were filtered by an awk-script in cygwin where
values from the beginning and end of a production cycle were extracted. Out of a filtration cycle of
600s (30s backwash and 570s production) the TMP plot represents values from time step 59s and
589s in each cycle, i.e. 30 seconds after production start and 10 seconds before backwash.

RESULTS AND DISCUSSION
Process performance
The treatment and performance data for the process are shown in Table 3 and Figure 2. As expected
the low rate process has slightly better results on removing organic matter. The inert soluble COD-
concentration has been found to be around 30-40 mg COD/l in earlier investigations with this
wastewater. This means that essentially all biodegradable COD was removed in the low rate process while a small biodegradable COD residual might be left in the high rate process. No suspended COD was detected after membrane filtration which means that COD was equal to FCOD.

### Table 3. Average treatment efficiency of the overall process (min and max values given in parenthesis)

<table>
<thead>
<tr>
<th></th>
<th>Inlet biofilm reactor</th>
<th>Outlet biofilm reactor</th>
<th>Outlet membrane reactor</th>
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</thead>
<tbody>
<tr>
<td><strong>LOW RATE</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>COD</td>
<td>242 (157-312)</td>
<td>178 (111-310)</td>
<td></td>
</tr>
<tr>
<td>FCOD</td>
<td>118 (58-173)</td>
<td>53 (23-72)</td>
<td>31.9 (22.8-42.3)</td>
</tr>
<tr>
<td>SS (mg/l)</td>
<td>74 (30-136)</td>
<td>88 (24-211)</td>
<td>0</td>
</tr>
<tr>
<td><strong>HIGH RATE</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>COD</td>
<td>251 (190-296)</td>
<td>222 (119-358)</td>
<td></td>
</tr>
<tr>
<td>FCOD</td>
<td>127 (102-152)</td>
<td>65 (55-78)</td>
<td>37.8 (29-46.5)</td>
</tr>
<tr>
<td>SS (mg/l)</td>
<td>92 (35-151)</td>
<td>126 (16-377)</td>
<td>0</td>
</tr>
</tbody>
</table>

**Figure 2.** Treatment efficiencies of COD and FCOD

A typical example of membrane performance for the high-rate and low-rate process respectively is shown in Figure 3. The membrane unit has been operated in the same way with respect to aeration, permeation flux, back pulsing, operation cycles, recovery etc. Results are shown by comparing permeability decline expressed as change in TMP over time. The points plotted represent the TMP value immediately after a backwash and at the end of a cycle prior to the backwash cycle. The change in time of TMP measured immediately after a backwash represents an overall fouling rate which determines the degree of irreversible fouling as a function of the operating condition. The difference between TMP measured after and before a backwash gives the fouling rate for an operating cycle and expresses the efficiency of the backwashing to remove instantaneous fouling.

**Figure 3.** Membrane performance for low-rate and high-rate loadings

The variation in difference between the two values is due to the varying influent wastewater quality in the test period. From Figure 3 it is apparent that in general the rate of permeability decline is
greater during high-rate operating conditions compared to the low-rate condition. The TMP differential in a production cycle is also in general greater than for low-rate operation. This difference in fouling behavior must be due to the differences in wastewater quality (i.e. composition of soluble organics, EPS etc) and suspended solids characteristics for the two operating conditions tested.

**Particle analysis**

Various studies have investigated the effect of wastewater quality and membrane fouling for AS-MBR processes. These studies were aimed at identifying the dominating fouling mechanisms and operational factors that influence the overall performance of the membrane filtration. Wisniewski and Grasmick (1998) reported that the soluble fraction was the most important, which relates to the extracellular substances (EPS) content in the wastewater and the fouling effect of soluble sugars and proteins. Experiments carried out by Defrance *et al.* (2000) conclude that suspended solids contributed most to the membrane fouling and Bouhabila *et al.* (2001) reported that the colloidal fraction was the most important. Cicek *et al.* (2003) have reported that fluxes in an AS-MBR system are strongly correlated with the COD fraction below 0.1 µm. An evaluation of the submicron particle fraction is therefore important in understanding membrane performance for the MBB-M-R process under varying loadings and operating conditions. The aim of this study was to evaluate submicron colloidal fractions as a function of operating conditions and to determine how this correlates to the membrane filtration.

Recent studies have shown that there is a shift towards larger particles with increasing organic loading in AS-MBR (Wisniewski *et al.*, 2000) or HRTs for the moving bed biofilm reactor (Melin *et al.*, 2005). The effect of HRTs on particle size distributions for the bioreactor used in this study was conducted and results are shown in Figure 4. There is a clear trend that particle aggregation correlates with increasing HRT with a clear shift towards larger particles as shown in the differential volume % analysis. Aggregation of the smaller particles also results in a redistribution of the numbers of particles in the various size fractions where there is a relative increase in the number of smallest size fractions, particularly at HRTs higher than 2 hours (right graph in Figure 4). Although particle aggregation is clearly taking place in the biofilm reactor the question remains whether the relative increase in number of submicron particles in the 0.5-0.05 µm size range impacts membrane filtration at higher loading rates, *i.e.* short HRTs.

![Figure 4](image-url). Development of particle size distributions as a function HRT

The two loading rates chosen in this study represent the shorter and longer HRTs which were investigated and shown in Figure 4, and as such the wastewater effluent qualities with respect to particulate material should differ. Particle and colloidal fractions of the wastewater were tracked throughout the process by analyzing samples from the inlet to the bioreactors, the bioreactor outlet and in the concentrate within the membrane reactor. Typical results of particle size distributions as a function of loading rates on the bioreactors are shown in Figure 5 and Figure 6.
Operating the MBB-M-R process with low loading rates has a distinct effect on the particle size distributions. From Figure 5 it is apparent that aggregation of particles occurs in that the volume percent increases (shown in the differential and cumulative curves) indicating a shift to larger particles. The differential number analysis however shows that the relative number of submicron particles increases indicating that the smallest particle size fractions are not readily incorporated into the aggregates. This apparent inefficiency to include the smallest fractions in the aggregates may be due to several reasons, i.e. non-ideal flocculation conditions, and/or particle stability, biomass characteristics. The transformation of the solids for the high loading rate is to a lesser degree in that some of the larger size fractions aggregate while the relative number of submicron particles remains essential the same. The effect of the particle size characteristics in the effluent from the biofilm reactor as a function of loading should therefore ultimately have an affect on the membrane performance. If membrane fouling correlates with the particle size fraction that is below 0.1 µm, membrane fouling will depend on the development of the smallest fractions (< 1 µm) and to a lesser degree on the aggregation of the larger particle size range as these will not have the same impact on fouling as the submicron particles. Results shown in Figure 5 and Figure 6 indicate the potential for membrane fouling based on particle size distribution. Operating the process with low loading rates increases the relative amount of submicron particles producing feed water to the membrane reactor with a greater fouling potential. This appears to be contradictory to the performance results obtained for the membrane filtration (Figure 3) on the assumption that the submicron particles represent the most important affect on permeability decline (Cicek et al., 2003).

The fate of the particles in the biofilm reactor effluent after being fed to the membrane reactor was therefore investigated. Membrane filtration of the bioreactor effluent is performed with a
submerged membrane module in a cyclic mode as described above. The system is operated with approximately 95% recovery and under near steady-state operation the solids concentration in the reactor is on average around 500 mgSS/l for low-rate conditions and 800 mgSS/l for high-rate. Continuous aeration of the membrane module is also applied to reduce fouling and enhance the system flux. The increase in solids concentration combined with the aeration will ultimately induce flocculation in the membrane reactor thereby changing the characteristics of the biofilm effluent. Visual observations confirm that the solids in the membrane reactor for low-rate conditions have better settling properties, giving better accumulation in the sludge pocket of the reactor. This observation agrees with the results from the analysis of the particles size distribution as well as average suspended concentration values measured in the reactor for the two operating conditions. The effect of the membrane reactor design and operation on particle size distributions in the membrane reactor are shown in Figure 7, plot A for high-rate and plot B for low-rate. The membrane reactor was operated in the same mode for all tests and samples were taken after the system reached near steady-state with the set recovery.

**Figure 7.** Particle size distributions in the membrane reactor as a function of operating condition

For high-rate operation the differential volume% result shows a slight decrease of the larger particles suggesting that the degree of aeration in the membrane reactor breaks up the larger aggregates, but they remain relatively large (> 20 µm). At the same time, analysis of the differential number % shows there is a slight decrease in the total number of submicron particles but with the fraction of submicron particles being relatively unchanged. In comparison, for low-rate conditions results of the differential volume % indicate an additional increase of larger particle sizes, which may explain the better settling properties observed. The differential number % also shows a significant decrease in the number of submicron particles around the 0.1 µm particle size range. In the event that fouling by submicron particles is a dominant fouling mechanism it is apparent that the effect of this particle size range on membrane permeability will be less under low-rate compared to high-rate loading conditions. The results are in agreement with the TMP development data registered for the membrane performance as shown in Figure 3.

This study has focused on evaluating the effect of particle size distributions on membrane filtration as a function of two process operating modes. Solids characteristics such as particle strength, particle density, sludge volume index (SVI), floc structure, zeta potentials and so on have not been included in the analysis so far. Ultimately these parameters will give important additional information in gaining a better understanding of how sludge properties relate to solids behavior in the membrane reactor and consequently fouling.

Although membrane fouling is caused by several phenomena and mechanisms, previous studies have shown that fouling in MBR systems is primarily caused by the soluble fraction (EPS) in the
wastewater in combination with the suspended solids, in particular the submicron colloidal fractions where permeability decline has been strongly correlated to this fraction. Which form of fouling is dominant will vary as a function of wastewater quality, membrane reactor design and operating conditions. The design and operating modes of the membrane reactor induce a degree of flocculation, however, the degree of flocculation will also depend on sludge characteristics related to stability, floc structure, floc density etc. The effects of these characteristics combined with varying operation modes on the submicron particle fractions are currently being investigated.

CONCLUSION
This study has investigated the potential of membrane fouling in an MBB-M-R process where two modes of operation were investigated. Focus has been on the potential effect of the colloidal submicron particles by tracking particle size distributions in the MBB-M-R treatment concept. Results show:

- particle size distribution was found to vary as a function of the loading rate on the biofilm reactor where low-rate had a relatively higher fraction of submicron particles, i.e. increased potential for membrane fouling by colloidal fraction
- membrane fouling rates, however, were lower for low-rate operation compared to high-rate
- particle aggregation and reduction of submicron particles in the membrane reactor was more pronounced during low-rate operation indicating better flocculation, i.e. less fouling potential
- membrane performance, i.e. fouling, correlates with the relative number of submicron particles in the membrane reactor as found in other studies
- other factors affecting the submicron particle fraction in the MBB-M-R process need to be investigated, i.e. influence of soluble organic composition, floc structure, floc stability, alternative operating modes and reactor designs etc.

ACKNOWLEDGMENTS
Kaldnes Miljøteknologi AS, Norway, for support with the biofilm reactor and ZENON Environmental Inc., Canada, for supplying the membrane modules

REFERENCES