

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

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### 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 reactor 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

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### 1. 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

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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 [1–4]. 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 [5–7]. 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 [11–14]. 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 [15–18]. 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.

## 2. 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 Fig. 1. A conventional approach of defining flow and mass balances can be applied to describe and analyse 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.

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

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

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:

$$c(t) = c_0 \cdot \frac{Q_{in}}{Q_{out2}} + c_0 \cdot \left(1 - \frac{Q_{in}}{Q_{out2}}\right) \cdot e^{-\frac{Q_{out2}}{V} \cdot t} \quad (2)$$

or

$$c(t) = \frac{c_0}{1-R} \cdot \left[1 - R \cdot e^{-\frac{1}{SRT} \cdot t}\right] \quad (3)$$

where SRT is solids retention time

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

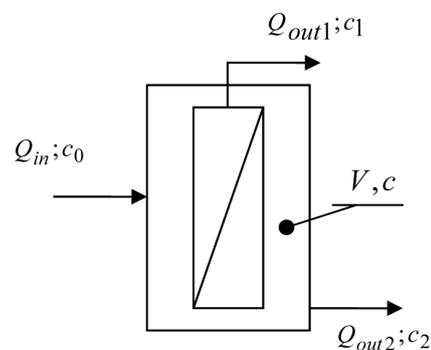


Fig. 1. Principle of a typical submerged MBR reactor configuration.

and  $R$  is recovery

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

By analyzing the reactor design and operating conditions using Eq. (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 Fig. 2.

Following the results in Fig. 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 Eq. (1) including  $K_s$  and expressed as:

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

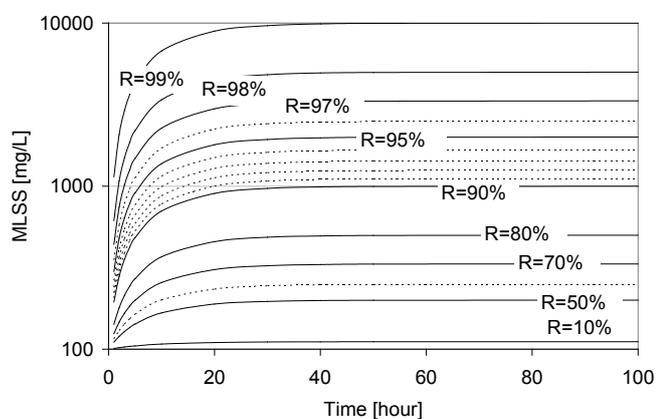


Fig. 2. Steady state concentrations of MLSS as a function of recovery  $R$ , for  $c_o = 100$  mg/L and SRT = 9 h.

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 Eq. (6) for a fixed recovery ( $R$ ) and for a given operating condition of the biofilm reactor is illustrated in Fig. 3.

Different membrane reactor options and varying operating conditions can therefore be analyzed using the simplified model defined in Eq. (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.

### 3. 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 Fig. 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  $335 \text{ m}^2/\text{m}^3$ , supplied by Krüger

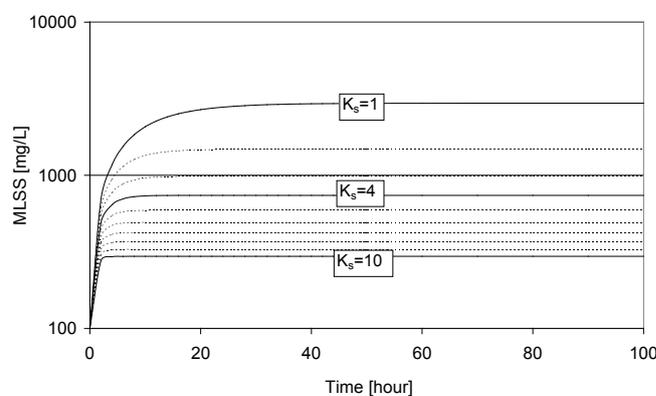


Fig. 3. Steady state concentrations of MLSS as a function of separation coefficient  $K_s$  for recovery of 96%, for  $c_o = 100$  mg/L and SRT = 9 h.

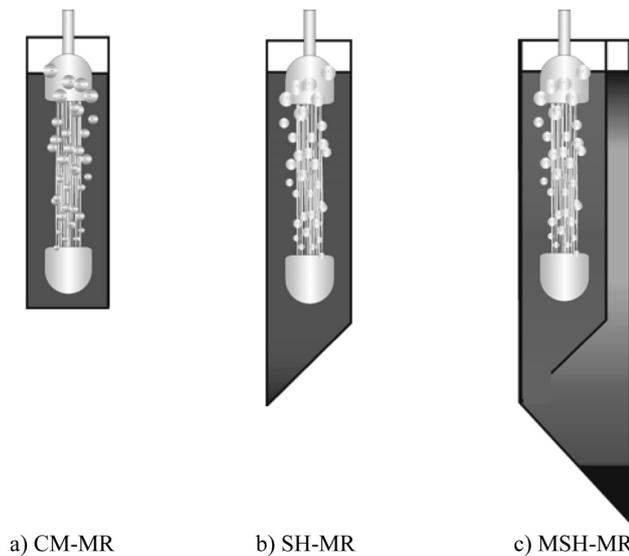


Fig. 4. Illustration of the three alternative membrane reactor designs investigated in this study with Zenon ZW-10 membrane module

Kaldnes. Filling fraction was 67% of reactor volume with total surface area for biofilm growth of 84.5 m<sup>2</sup>. 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 min production time and a 0.25 min backwash cycle. Production flux was set at 35 L.m<sup>-2</sup>.h<sup>-1</sup> and backwash flux at 38 L.m<sup>-2</sup>.h<sup>-1</sup>, recovery 96% and specific aeration demand of SADm ~ 3.6 Nm<sup>3</sup>m<sup>-2</sup>h<sup>-1</sup>. The MBBR reactor was operated with a 4 h HRT, giving on average the quality parameter values and treatment efficiencies as shown in Table 1.

During operation with the high strength wastewater, the membranes were operated with a constant flux of

22 L.m<sup>-2</sup>.h<sup>-1</sup> (no backwash or relaxation), recovery 96%, and the specific membrane aeration demand of SADm ~ 1.8 Nm<sup>3</sup>m<sup>-2</sup>h<sup>-1</sup>. The HRT of the MBBR was 6 h. Water quality parameters and treatment efficiencies are given in Table 1.

The pilot plants were equipped with National Instruments/LabVIEW 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 were 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<sub>4</sub>-N) and nitrogen (NO<sub>3</sub>-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).

#### 4. 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 Fig. 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

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	—
NH <sub>4</sub> -N, mg/L	29.8 ± 6.4	0.16 ± 0.04	99.4	38.9 ± 8.7	0.21 ± 0.13	99.5
NO <sub>3</sub> -N, mg/L	<1	27 ± 3.6	—	<1	34.7 ± 7.8	—
HRT, h	4			6		

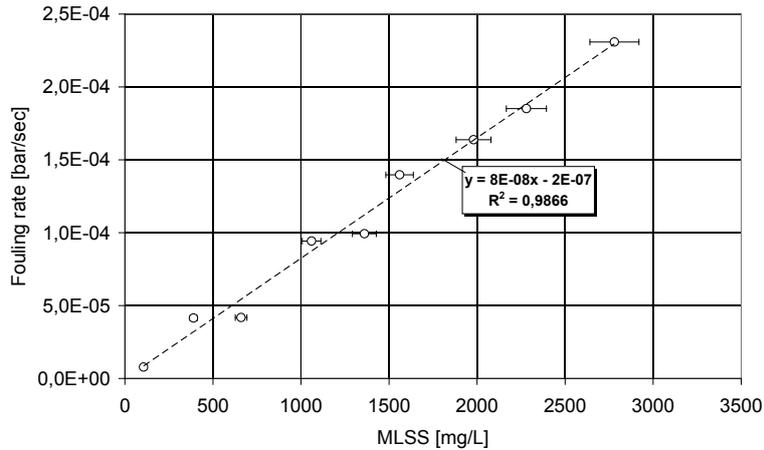


Fig. 5. Example of fouling rate within an operating cycles as a function of MLSS for CM-MR.

operation of the membrane filtration unit with considerably lower fouling rates.

Eq. (6) was used to model and predict the MLSS concentrations for the alternative reactor designs described above. For the CM-MR (Fig. 4a) the  $K_s$  value was set to one, while the steady state concentration for the SH-MR (Fig. 4b) 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 Fig. 4c. Measured steady state values of MLSS in this configuration indicate that  $K_s$  has a value of  $\sim 7.5$ . Experimental verification of the models has been done and results are presented in Fig. 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).

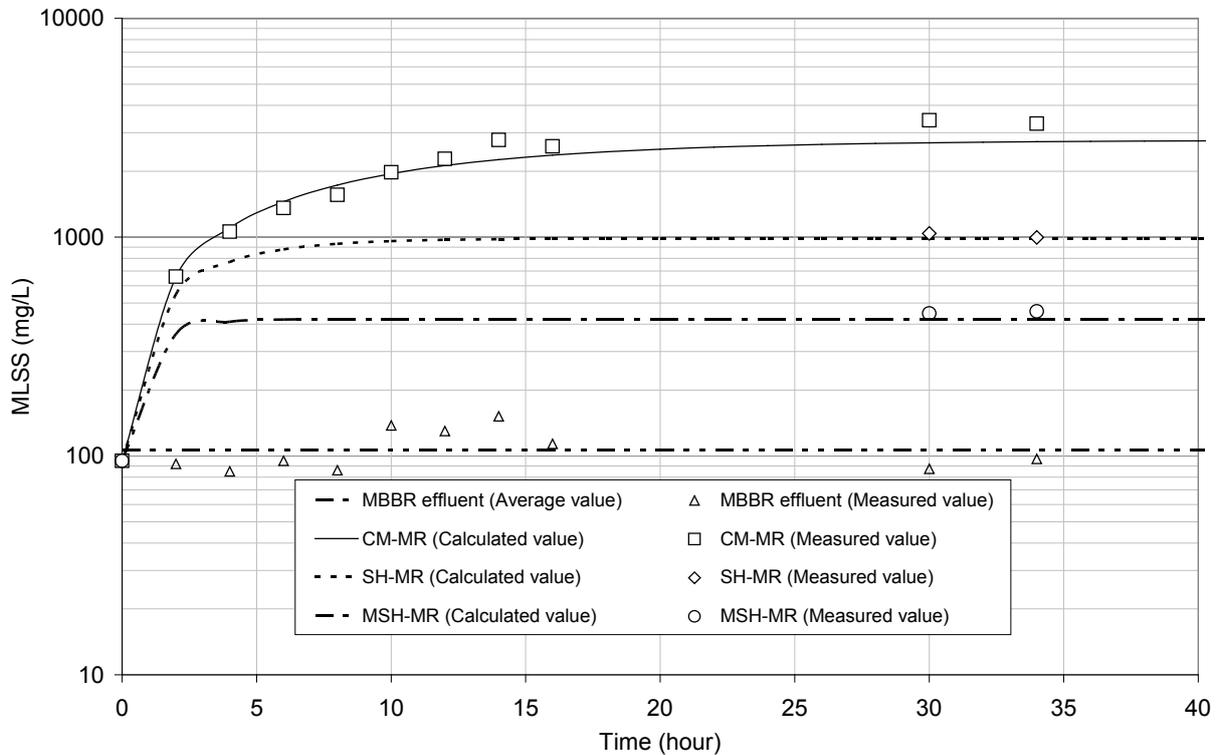


Fig. 6. Calculated and measured values for MLSS for CM-MR, SH-MR and MSH-MR.

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 Fig. 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 ( $\Delta$ TMP) 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 h 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.

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 Fig. 8, showing results for filtration of high strength wastewater. The pilot plants were operated for a period of 7 d. The CM-MR configuration was terminated after 3 d 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/d, 3.57 kPa/d and 1.42 kPa/d for the CM-MR, SH-MR and MSH-MR respectively for the operating period shown.

Results shown in Fig. 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.

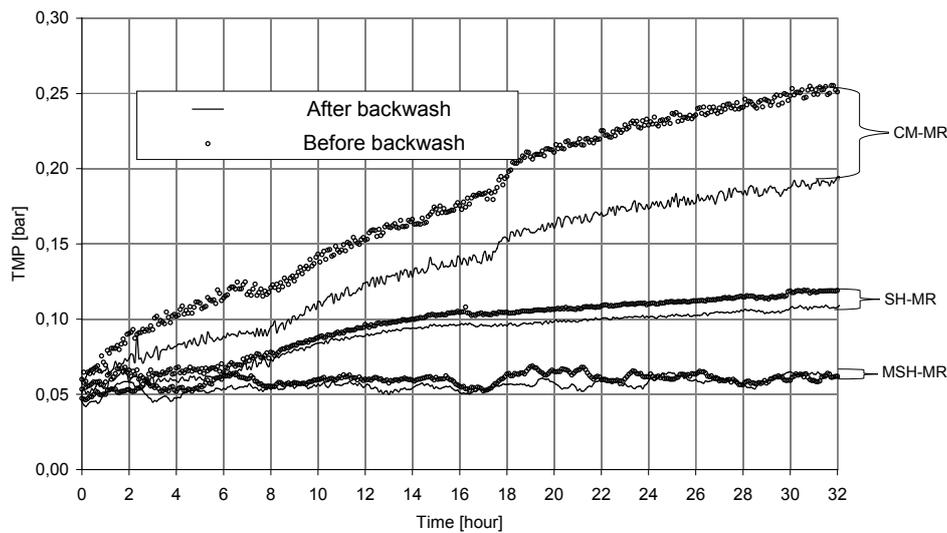


Fig. 7. TMP before and after backwash for CM-MR, SH-MR, MSH-MR. Production flux 35 LMH and backwash flux 38 LMH.

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		

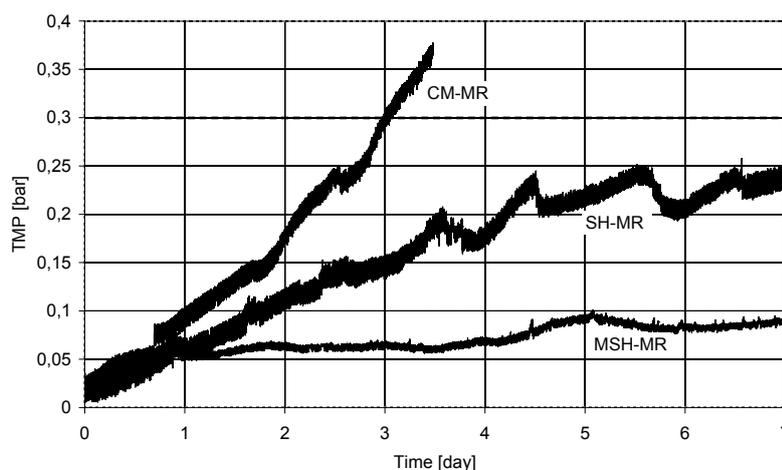


Fig. 8. Overall TMP for CM-MR, SH-MR and MSH-MR at the constant flux of 22 LMH.

reduced MLSS by sedimentation in membrane reactor, can significantly improve membrane performance i.e. reduced fouling.

Lower concentrations of MLSS and COD (Table 2) around the membranes as a function of the modified reactor 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 (Fig. 9).

The analysis of particle sizes and particle size distributions (PSD) in the respective zones of the MSH-MR design is illustrated in Fig. 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 0.07–0.08  $\mu\text{m}$  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 (0.08–0.09  $\mu\text{m}$ ) 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 reactor 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.

## 5. 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 reactor design provided a significantly lower concentration of MLSS and

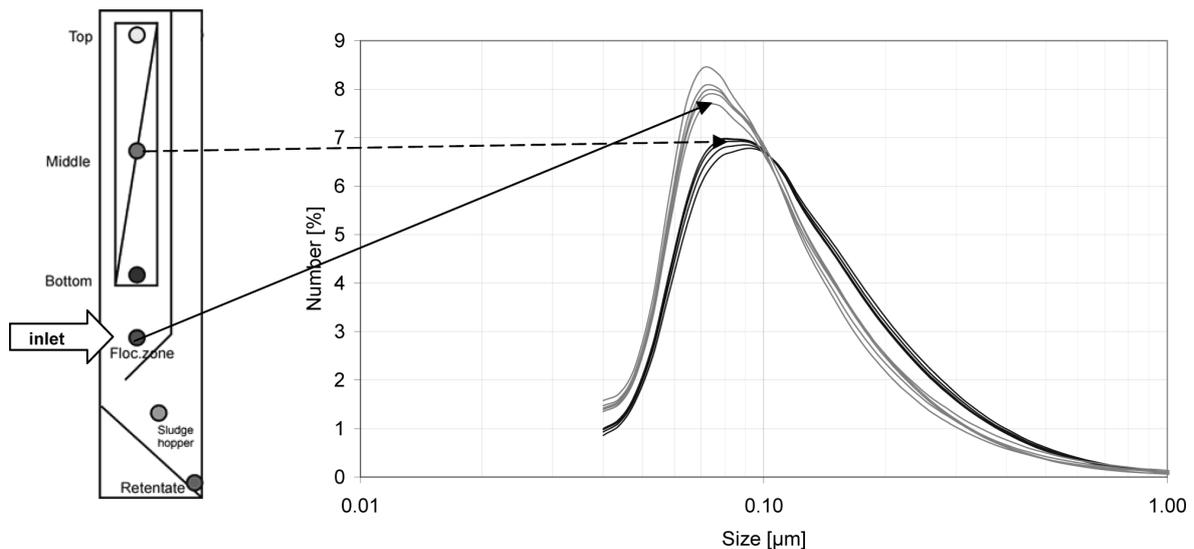


Fig. 9. PSD in number percentage for submicron particles for given sampling points in the MSH-MR for four parallel measurements where each line represents average of three measurements of one sample.

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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