Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Ivanovic, Igor , Leiknes, TorOve and Ødegaard, Hallvard(2008) 'Fouling Control by Reduction of Submicron Particles in a BF-MBR with an Integrated Flocculation Zone in the Membrane Reactor', Separation Science and Technology, 43: 7, 1871 — 1883
To link to this Article: DOI: 10.1080/01496390801974704
URL: http://dx.doi.org/10.1080/01496390801974704

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

ISSN 0149-6395 print/1520-5754 online
DOI: 10.1080/01496390801974704

Fouling Control by Reduction of Submicron Particles in a BF-MBR with an Integrated Flocculation Zone in the Membrane Reactor

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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).

Keywords: Submicron particles, colloids, biofilm membrane reactor (BF-MBR), particles size distribution (PSD)

INTRODUCTION

Membrane bioreactors (MBR) combine biological treatment processes with membrane filtration to provide an advanced level of organic and suspended solids removal. The process is a refinement of the conventional activated

Received 2 September 2007, Accepted 8 February 2008
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sludge (AS) process where membranes primarily serve to replace the clarifier. The current and most commonly used MBR reactor design applied for municipal wastewater treatment is the submerged process configuration where the membrane modules are immersed in an aerated biological reactor (AS-MBR). An alternative treatment scheme to the AS-MBR is combining a biofilm reactor with membrane filtration for enhanced biomass separation (BF-MBR) (1). The moving bed biofilm reactor (MBBR) is an alternative biofilm reactor developed at NTNU Norway (2). The MBBR process consists of small plastic biofilm carriers used to create a large surface area for the biofilm to grow on and are suspended in the reactor by aeration. The moving-bed-biofilm reactor (MBBR) for biodegradation of soluble organic matter coupled with a submerged membrane reactor for enhanced particle separation and clarification of the effluent after the biological reactor represents a new concept for an alternative wastewater treatment process. The biofilm membrane reactor (BF-MBR) has the potential of utilizing the best characteristics of the MBBR processes and membrane separation. The concept for this treatment process is illustrated in Fig. 1.

Compared to the current state-of-the-art MBR processes, i.e., AS-MBR systems, the BF-MBR has the potential of being even more compact, have a higher rate of operation (i.e., higher fluxes than common in AS-MBR), being more energy efficient and having better fouling control with optional strategies for fouling minimization (1, 3, 4). A major drawback of BF-MBR is membrane fouling, which is common for all membrane systems (5). Deposition of solids as a cake layer, pore plugging/clogging by colloidal particles, adsorption of soluble compounds and biofouling are some of the main forms of fouling that have been identified. The significance of colloids and submicron particles on membrane fouling has been reported in literature where different estimations of the total measured fouling caused by these particles vary between 25% and 50% of the total (6–9). From the reports found in literature it is apparent that reduction of submicron particles around the membranes would be desirable in operation of MBR systems. Recent studies on the BF-MBR concept show that the colloidal fraction in

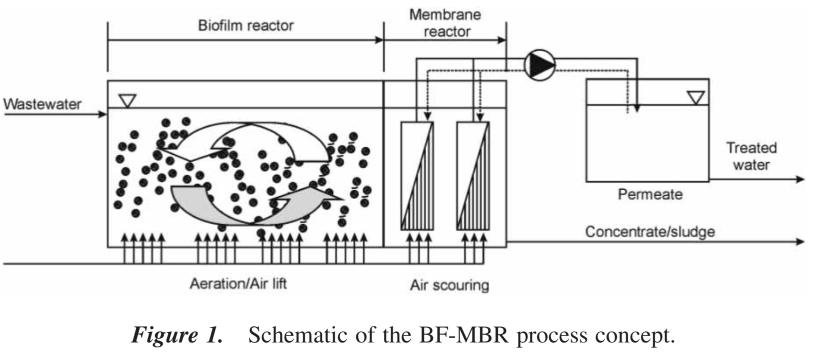

Figure 1. Schematic of the BF-MBR process concept.

wastewater, i.e., submicron particles, is an important foulant in this membrane process for wastewater treatment (1, 3).

The aim of this study is to examine the possibility of reduction of submicron particles in the membrane reactor in the BF-MBR process (i.e., around the membrane), by introducing the feed water through a flocculation zone (F-zone) placed beneath the membrane module and aeration unit and to determine the effect this may have on the overall performance and fouling of the membrane units.

METHODS

Municipal wastewater, pretreated in a primary sedimentation tank, is fed to the pilot plant where biodegradation takes place in four biofilm reactors (MBBR) placed in series, each with a volume of 65 L. The effluent from the MBBR is further treated in the membrane filtration reactor designed for enhanced particle removal. The pilot plant was operated at an organic loading rate where full nitrification was achieved, equivalent to a hydraulic retention time (HRT) in the MBBR of ~4 hours. The membrane unit was operated with a constant flux of $50\,L \cdot m^{-2} \cdot h^{-1}$ with 96% recovery in a 120 seconds cyclic mode consisting of 114 seconds production with a 6 seconds backwash sequence. The membrane performance was measured by monitoring the transmembrane pressure (TMP) development over time with on-line recording using pressure transmitters.

Various water quality parameters were measured regularly from grab samples. All analyses were performed according to the Norwegian national (NS) or international standards. Suspended solids (SS) were analyzed by filtering through a 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) samples were first filtered with Whatman GF/C glass microfiber filters (1.2 μm and 0.45 μm) before using the Dr. Lange test. Capillary suction time-CST (according to the Standard methods (10)) and time-to-filtrate-TTF (according to a modified Standard method) were preformed in order to evaluate dewatering and filtering characteristics of the concentrate. Particle size distribution (PSD) analysis of the water in the various stages of the process was also done using laser diffraction spectroscopy (Beckman Coulter LS230). TMP was logged for every two seconds. The data files were filtered by a C++ script in MS Visual Studio 6.0 where values from the beginning of a production cycle were extracted and plotted. Different sampling points were chosen; around the membrane (middle of reactor zone), in the flocculation zone and concentrate.

Two different membrane reactors were used, one with and one without a flocculation zone respectively. An illustration of the two reactor configurations with an immersed membrane module is shown in Fig. 2. The membrane reactor

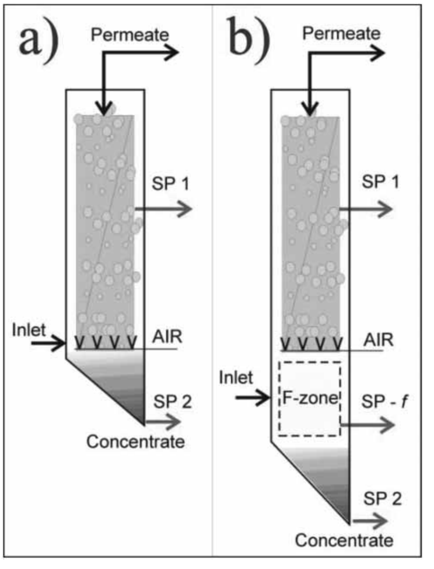

Figure 2. Two membrane reactor configurations tested – a) without and b) with flocculation zone (F-zone).

without the F-zone has a volume of 25 L and two sampling points: SP1–membrane reactor and SP2–concentrate stream (see Fig. 2). The second reactor was modified to include the F-zone by extending the bottom and placing the inlet to the membrane reactor under the membrane aeration system. The reactor with the F-zone has a volume of 33 L where the additional volume represents the F-zone. Three sampling points were included in this reactor: SP1–membrane reactor, SP–*f*-flocculation zone and SP2–Concentrate (see Fig. 2). Both membrane reactors were fitted with a ZW10 membrane module unit supplied by Zenon; UF membranes with nominal pore size −0.04 μm, outside/in operation and with a surface area of 0.93 m^2.

RESULTS AND DISCUSSION

A summary of the average feed water characteristics and average overall removal efficiencies for SS, COD/FCOD and NH_4-N for the pilot plant operation is given in Table 1. Experiments were conducted consecutively with the membrane reactor without the F-zone operated during March 2006 while the reactor with F-zone was operated during May 2006. In general, the wastewater during May was more concentrated due to prevailing dry weather conditions and subsequently higher concentrations of SS and COD/FCOD in the inlet water were measured (Fig. 3). This also resulted in slightly higher concentrations being fed from the biofilm reactor to the

Table 1. Summary of raw water quality and average removal efficiencies for the both experimental runs

		Without F-zone		With F-zone	
Parameter	Unit	Average	Average removal	Average	Average removal
SS[a]	mg/L	70	100%	106	100%
COD	mgO_2/L	242.6	91%	312.4	90%
FCOD-1.2[b]		126.5	70%	146.2	72%
NH_4-N	mg/L	22.3	>99%	30.1	>99%

[a]SS is not measured in permeate, because nominal pore size of membrane is 0.04 μm compare to nominal pore size of GF/C filter 1.2 μm.
[b]Removals in MBBR.

membrane reactor. However, the overall process removal efficiencies were in the same range during both experimental periods as can be seen in Table 1.

The results of SS analyses around the membranes (SP1) showed that the membrane reactor with the F-zone in general had lower concentrations of SS around the membrane. On average the SS concentration was 48% lower in the reactor with the F-zone. The suspended solids measured in the concentrate stream (SP2) of the reactor with the F-zone had on average 28% higher concentrations of SS compared to the reactor without the F-zone (Fig. 3). The results indicate that improved flocculation occurred in the modified reactor with the F-zone and subsequently a significantly better separation of

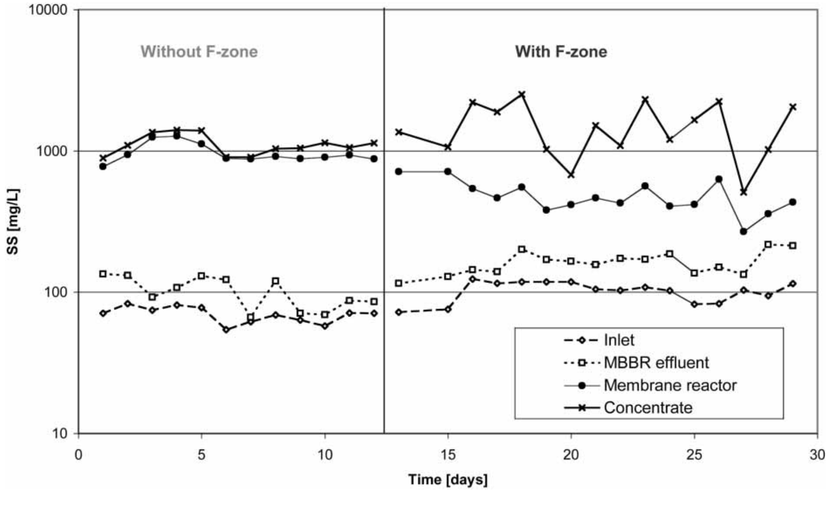

Figure 3. Suspended solids in inlet wastewater, effluent from MBBR, around the membrane (i.e., in membrane reactor) and concentrate for the two experimental periods.

suspended matter was achieved resulting in lower SS concentrations around the membrane.

Characteristics of the suspended solids were also conducted by analyzing filterability, estimated by measuring time-to-filter (TTF) of 12.5; 25 and 50% respectively of 200 ml samples. Samples were extracted from both the membrane reactor (SP1) and the concentrate stream (SP2). Analysis of samples from the membrane reactor (SP1) demonstrate that the F-zone reactor gave suspensions around the membrane that had better filterability characteristics, i.e., shorter filtering times. Results were significantly better for TTF-25 where measured values were 3 times shorter for the reactor with the F-zone compared to the one without. The concentrate streams in both reactor configurations had almost the same filterability characteristics with average TTF-12.5 and TTF-25 values slightly lower for the reactor with the F-zone while TTF-50 was a little lower for the reactor without the F-zone (Fig. 4). The higher TTF-50 values of the reactor with the F-zone could be due to higher concentration of SS measured in the concentrate.

Dewaterability was measured as capillarity suction time (CST) in both the membrane reactor (SP1) and the concentrate stream (SP2). In order to minimize the effect of different concentrations of SS on CST, the measured CST values were normalized by dividing with measured concentration of SS for each sample. A lower CST value correlates to better dewaterability characteristics. Average values as shown in Fig. 5 indicate almost the same dewatering characteristics from both membrane reactor configurations i.e., around the membrane. Even though the concentrate stream from the reactor with the F-zone had significantly higher SS values compared to the reactor without the F-zone, CST/SS values measured are lower indicating better dewatering characteristics in general for the system with the F-zone. This

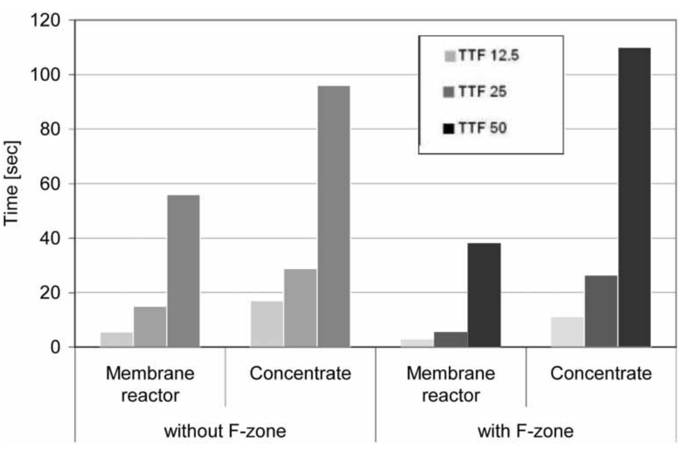

Figure 4. Time-to-filter (TTF) of 12,5; 25; 50% of sampled volumes around membrane (membrane reactor) and concentrate for both configurations.

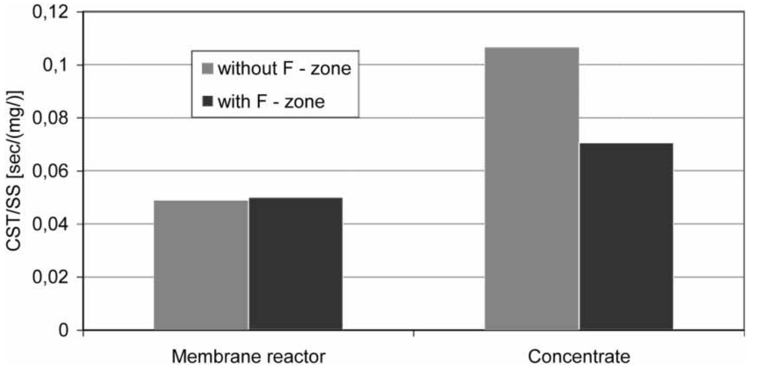

Figure 5. Normalized CST values around membrane (membrane reactor) and concentrate for both configurations.

observation confirms that there is a difference in the floc/particle structure of the suspended solids formed in the reactor with the F-zone, and that the characteristics of the concentrate are probably more favorable to treatment of the excess sludge produced.

The SVI values of the concentrate (i.e., sludge) were higher compared to what has been reported from activated sludge MBR (AS-MBR) systems (11, 12). Only the suspension around the membrane in the membrane reactor with F-zone had on average SVI-values lower then 100 (mL/g) which is often reported as a good value in AS-MBR systems (Fig. 6). In order to compare settleability, sludge settle volume (SSV) measured in Imhoff cone was recorded after 10, 20, 30, and 60 min and plotted as shown on Fig. 7. The same settling pattern was observed for both reactors, where on average particles in suspension around the membrane reactor with F-zone settled

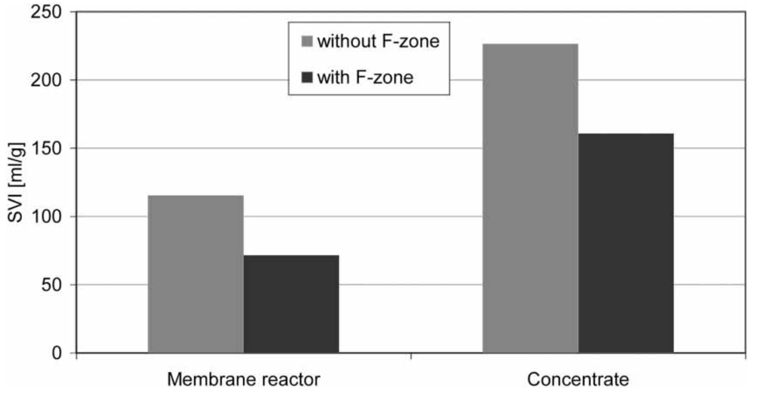

Figure 6. Sludge volume index (SVI) around membrane (membrane reactor) and concentrate for both configurations.

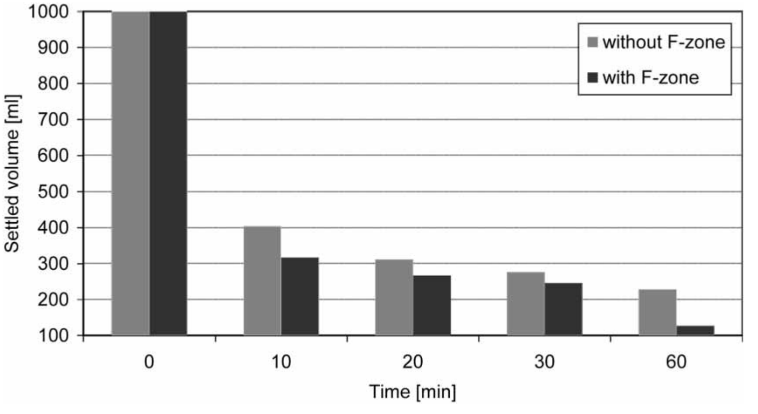

Figure 7. Sludge settled volume (SSV) after 10, 20, 30, and 60 min around membrane (membrane reactor) for both configurations.

slightly better in first 30 minutes and even faster in next 30 minutes. The reason for this is unclear but the answer could probably be found in the microscopic analysis of floc structure that has not been performed in this study.

Analysis of the particle size distributions (PSD) in the reactors (Figs. 8 and 9) reveals that in the reactor without the F-zone (sampling SP1 and SP2), a very similar PSDs are observed for the particles around the membrane as in the concentrate. A slightly lower concentration of particles between 30 and 40 μm can be observed though the difference is insignificant (Fig. 8). This observation indicates and suggests complete mixing conditions in the membrane reactor without the F-zone. Analysis of this reactor

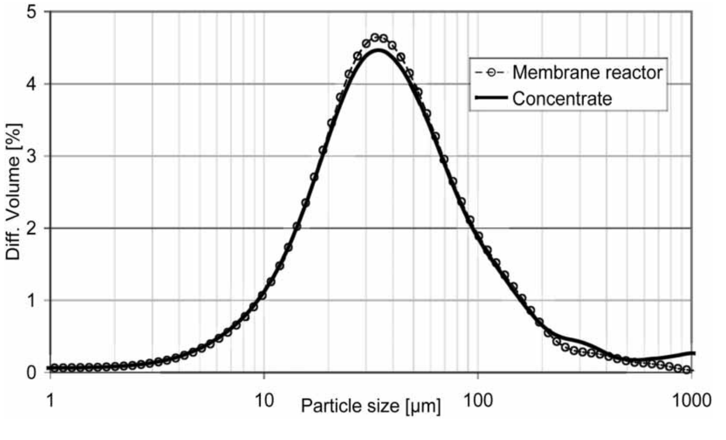

Figure 8. Particle size distribution in volume percentage around the membrane (membrane reactor) and concentrate–reactor without F-zone.

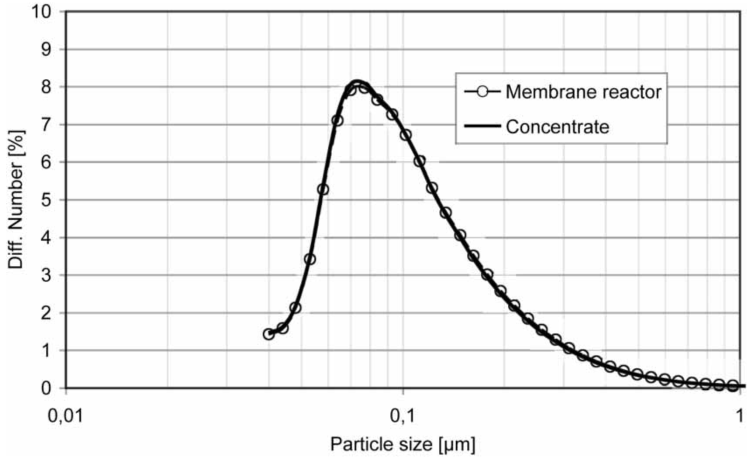

Figure 9. Number of submicron particles around the membrane (membrane reactor) and concentrate-reactor without F-zone.

configuration also show similar SS concentration values in the two sampling points (see Fig. 3). Based on the differential volume% presentation, the PSD analysis shows that the largest volume of suspended solids has a particle diameter between 5–200 μm. However, a number differential % analysis shows that the most abundant particles are the submicron particles with a measured particle diameter around 0.7 μm, and constituting about 8.2% of the total (Fig. 9).

Compared to the membrane reactor with the F-zone, the PSD analyses show a clear difference between the two sampling points; membrane reactor (SP1) and concentrate stream (SP2) (Figs. 10 and 11). The PSD analysis from sampling point SP-*f* from the F-zone is included in Figs. 10 and 11. In contrast to results observed in the reactor without the F-zone, it is apparent that a formation of larger flocs in the flocculation zone and subsequently in the concentrate stream is induced in comparison to the particles around the membrane. For all the measurements taken, the same trend can be observed in that flocculation occurred in the F-zone with a clear shift toward bigger particles in the concentrate.

The differential number % analyses (Fig. 11) show that a lower concentration of submicron particles are found around the membrane compared to the F-zone and concentrate stream. The most abundant fraction of particles was measured to have a diameter of around 0.84 μm, differential number percentage of 6.9%. The PSD analysis of the F-zone and concentrate stream show almost the same values where the most abundant fraction was measured at a diameter around 0.77 μm and 7.55% in differential number percentage. This indicates that flocculation does occur in the F-zone where submicron particles are captured and form larger flocs that settle under the membrane

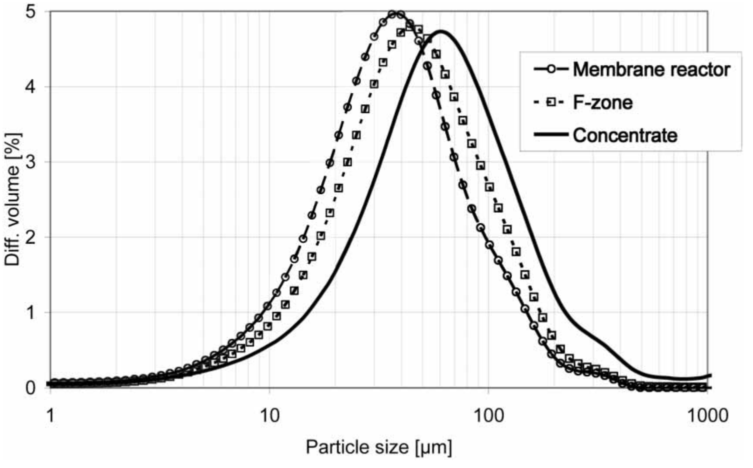

Figure 10. Particle size distribution in volume percentage around the membrane (membrane reactor), F-zone and concentrate–reactor with F-zone.

into the concentrate zone. Results from the standard sludge characteristics analysis confirm this observation in which improved dewatering and filtering characteristics of the concentrate were measured (Figs. 4–7). A direct comparison of the two experimental runs is not possible due to the fact that membrane reactors were not tested in parallel. It is evident, however, that a reduction in the numbers of submicron particles around the membrane was achieved in the membrane reactor with the F-zone. Based

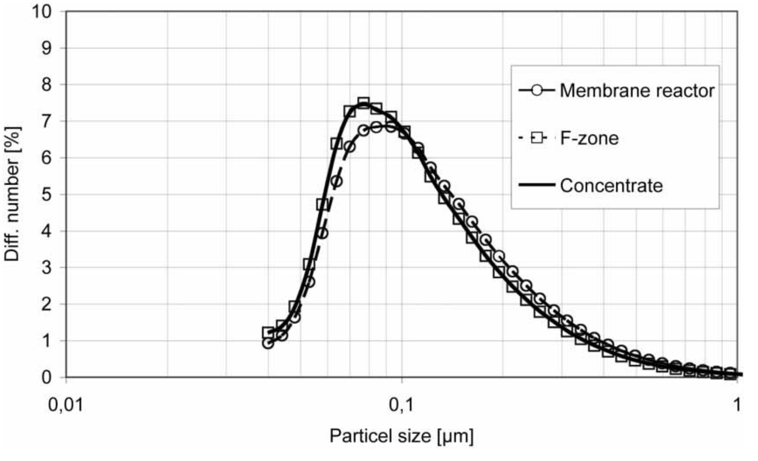

Figure 11. Number of submicron particles around the membrane (membrane reactor), in F-zone and concentrate-reactor with F-zone.

on the hypothesis that membrane fouling by submicron particles is a significant contribution to membrane fouling, the overall performance of the BF-MBR concept should be better with the membrane reactor configuration that includes the F-zone.

A sustainable operation of MBR processes correlates to low fouling rates thereby minimizing the need for implementing fouling reducing techniques that can be energy and cost demanding or frequent cleaning intervals of the membrane modules. Membrane fouling manifests itself in the increase of TMP over time and as such small increases of TMP due to operating conditions is desirable. Figure 12 shows the overall change of TMP measured for the two experimental periods (membrane reactors without and with the F-zone respectively). In this study only a combination of continuous air scouring cyclic mode of operation with backwashing was implemented as strategies for fouling control. The membrane was operated until the TMP reached a value (approximately 0.45 bar) at which time operating recommendations require chemical cleaning. The results show that the membrane reactor configuration with the F-zone was operated for about 16 days compared to the configuration without the F-zone which was operated for 12 days. The membrane reactor with the F-zone had a very low fouling rate for the first 8 days of operation. At this point, however, the fouling rate increased and stabilized to a similar rate as that measured for the configuration without the F-zone. Based on the experimental results obtained so far, it is not possible to indicate the reasons for this observation. The performance observed, however, clearly shows that a lower concentration of SS around the membrane and reduction of

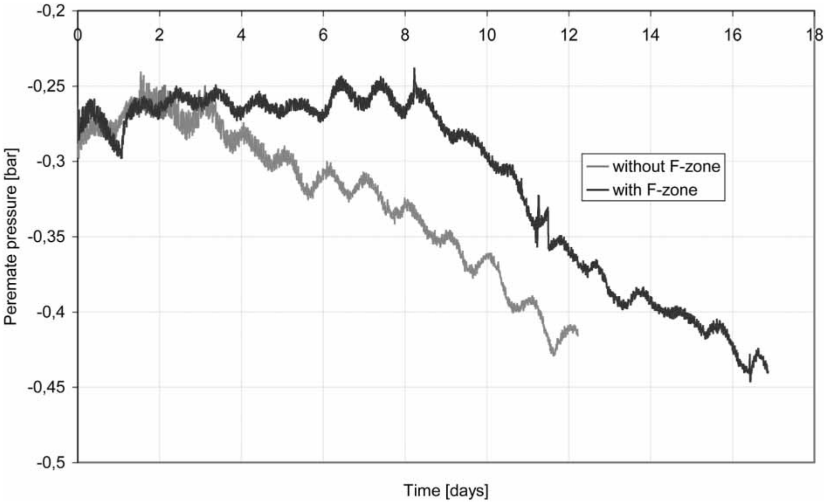

Figure 12. Overall performance of the membrane reactor expressed as TMP development (i.e., fouling rates) for both reactor configurations.

submicron particles as a result of introducing the F-zone in the membrane reactor design, did improve the overall performance of the treatment process. The results from this study reinforce the significance of submicron particles as an important foulant and their contribution to membrane fouling.

CONCLUSION

The BF-MBR concept is based on combining an optimized biofilm reactor for the removal of biodegradable constituents and a membrane reactor for enhanced particle separation. Introducing the feed water inlet port of the membrane reactor through a flocculation zone integrated in the membrane reactor resulted in a reduction of the number of submicron particles and reduction of SS concentrations around the membrane area. The effect was improved membrane performance (i.e., lower fouling rates) and a means for better fouling control in the BF-MBR process. The results confirm that submicron particles in the feed water to the membrane reactor are a significant foulant and that a reduction of this component in the water had a positive effect on membrane performance. A side effect of the alternative membrane reactor design was improved concentrate characteristics with respect to sludge treatment in that better dewatering and filterability characteristics were measured. Overall, the alternative reactor design and enhanced flocculation resulted in less fouling and is therefore a potential strategy for fouling control and minimization by reduction of submicron particles in the wastewater effluent.

More detailed studies are currently being undertaken in order to make a more detailed assessment of the particles measured in the BF-MBR and to gain a better understanding of the nature and fouling behavior of this fraction and of particulate matter.

ACKNOWLEDGMENT

The authors would like to acknowledge AnoxKaldnes, Norway, for support with the biofilm reactor and ZENON Environmental Inc., Canada, for supplying the membrane modules.

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