JEE Journal of Ecological Engineering

Journal of Ecological Engineering 2022, 23(8), 21–29
https://doi.org/10.12911/22998993/150722
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Received: 2022.05.11
Accepted: 2022.06.17
Published: 2022.07.01

Combination of Biological Treatment and Ceramic Membrane Filtration – Performance and Maintenance of the Pilot-Scale Installation

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ABSTRACT
Membrane separation technologies equipped with ceramic membranes have shown good operational results, high robustness and reliability. In this study, a combination of Moving Bed Biofilm Reactor (MBBR) & Membrane Bioreactor (MBR) (pilot scale) was investigated for fouling control optimisation. The system was operated with flat-sheet microfiltration (MF) ceramic membranes. Because of the attached biofilm in the MBBR, it could work stably and effectively with different loading rates, which reduces the load on the membrane surface. Influences of sludge recirculation and flux on the transmembrane pressure (TMP) value and thus impact on fouling development were investigated. Intensive aeration, backwash as well as the implementation of NaOCl were used as physical and chemical cleaning, respectively. The chemical cleaning efficiency reaches up to 70% and could be increased after further research. After the determination of optimal operational conditions, the normalised permeability value (PN) was effectively maintained with a slight TMP increase.

Keywords: water reclamation, moving bed biofilm reactor, membrane bioreactor, flat-sheet ceramic MF membranes.

INTRODUCTION

Water scarcity has become more acute, especially in recent years. There are two reasons for this problem: physical (water is not always available in the required amount) and economical (water is high-priced due to expensive treatment) [Dinar et al., 2015]. This incentivises the development of new treatment techniques for water reclamation or water recycling. The ability to reuse water has a positive side, since it can improve agricultural production, reduce the consumption of energy used for water treatment and increase nutrient removal [Bastian & Murray, 2012]. Treating water for discharging purposes is no longer sufficient. Water should be cleaned to the drinking quality standards or even better. Moreover, the existing conventional treatment system cannot handle it. The only variant, in this case, is to use modern treatment technology, such as membrane separation.

Among all possible technologies, which in the future can partially or fully replace conventional treatment systems, the combination of MBBR & MBR shows good potential as a reliable way of water purification. The wide introduction of membrane processes into practice became possible because of advancements in the science of polymers and the use of synthetic polymeric membranes.

The membrane technology has become more attractive and competitive, but there are still many development constraints. Usage of such systems provides many advantages, such as increased treatment efficiency, better permeate quality, smaller footprint, less complex operation [Leyva-Díaz & Poyatos, 2015], higher reliability, and resistance to overload and toxic compounds. The MBBR serves as a reliable source of biomass for the stable work of the system and as biological pretreatment for load reduction on the membrane surface and hence decreasing fouling layer growth. Membrane fouling remains the greatest problem thatleads to a short-term life of the membrane, which can greatly increase operational, replacement, and also aeration costs for physical cleaning [Ninomiya et al., 2020; Wang C. et al., 2022].

In recent years, many different strategies for fouling control have been developed as optimisation of operational conditions, understanding of fouling mechanisms, and discovery of new membrane materials [Kimura & Uchida, 2019], as well as new ways of fouling prevention or cleaning [Wang C. et al., 2022]. One of the latest successful membrane materials is ceramics. Such membranes were made due to the unreliability of polymeric membranes. Ceramic material has unique thermal, chemical, and mechanical properties [Shi et al., 2014]; moreover. it can work with high pressure, acidity/alkalinity, and temperature. This makes it suitable for the treatment processes where polymeric membranes cannot be used. The permeability of SiC ceramic membrane can be up to 20 times higher than of conventional ultrafiltration (UF) polymeric membrane. In addition, it should be noted that ceramic membranes could be cleaned with cleaning in place (CIP) without any damage and the highly robust properties of ceramic allow for the use of back pulsing as an alternative cleaning option. One of the drawbacks of SiC membrane is the price. In summary, the properties that offer an advantage of using ceramic membrane include [Lin et al., 2018] high T, °C resistance, high stability, reliability, corrosion/abrasion resistance, can work in a wide pH range, can be backwashed, resistant to deformation; stable under different bacterial conditions, work with highly viscous fluids, greater porosity and filtration surface area of the membrane, stable pore size.

Fouling appears because of the attachment of organic matter, which is present in high concentration in the separation chamber, to the membrane surface due to its sticky properties. This is a common reason for membrane pore blockage. There are different types of fouling (Table 1) [Arribas et al., 2015; Kimura & Uchida, 2019] and each has a completely different formation time and rate.

Fouling appears on the membrane surface in the initial minutes of operation. Irreversible fouling eventually appears on the membrane surface, which cannot be removed either by chemical or physical cleaning, regardless of the efficiency of the cleaning.

There are 3 basic fouling factors, that have the most impact on the fouling rate:
- nature of the feeding wastewater;
- the hydrodynamic regime of the separation process;
- membrane characteristics [Krzeminskia et al., 2017].

Fouling develops unevenly, including [Gkotsis et al., 2014]:
- conditioning fouling (initial fouling with minor pore blockage and residues of biomaterial on the membrane surface);
- steady fouling (cake formation with sludge film and more advanced pore blockage);
- rapid fouling with TMP jump (change of flux due to unevenly distributed fouling layer, therefore flux appears to be higher than the critical value, which induces severe TMP escalation).

For fouling control, there have been developed six main strategies:
- suitable preliminary preparation of the feeding wastewater;
- permeate as the source of backwash/relaxation;
- chemical cleaning-in-place (CIP);
- chemically enhanced backwash;
- membrane cleaning by coarse aeration;
- chemical adjustment of mixed liquor [Krzeminskia et al., 2017; Azis et al., 2018].

Due to recent advances, it is possible to maintain a fouling-free system by using ceramic membranes, which have a certain number of advantages compared to polymeric membranes, including the possibility of using more harsh methods of membrane cleaning.

Table 1. Types of fouling on the membrane surface

Type of fouling	Cleaning	Structure
Reversible	Could be removed by physical cleaning	Loosely attached fouling structures, a cake layer appears on the membrane surface
Irremovable	Could be removed by chemical cleaning	Strongly attached fouling structures, pore-blocking phenomena
Irreversible	Could not be removed	Permanent fouling structures

One very important factor is the intensity of mixed liquor mixing, which plays an important role in fouling formation. Mixing keeps all fouling structures in continuous movement, which makes the attachment less possible and could remove the already attached particles on the membrane surface. Together with periodical backwash and relaxation, mixing can effectively control reversible fouling.

Chemical cleaning-in-place can be used as a method for removing irremovable fouling. CIP is highly efficient and could be performed in-situ or ex-situ. Cleaning efficiency depends on many factors, including the type of used reagent and its concentration, reagent reaction rate, characteristics of fouling on the membrane surface, etc. [Wang et al., 2014]. It should be emphasised even that the cleaning procedure should be considered carefully, for instance inadequate cleaning with NaOCl, can actually intensify the fouling rate, resulting in the rapid and repeated formation of the fouling layer [Kimura & Uchida, 2019].

MATERIAL AND METHODS

Pilot plant

The pilot-scale MBBR & MBR plant was installed in the laboratory of the Norwegian University of Life Sciences in Ås, Norway. The pilot plant was maintained for about half the year,

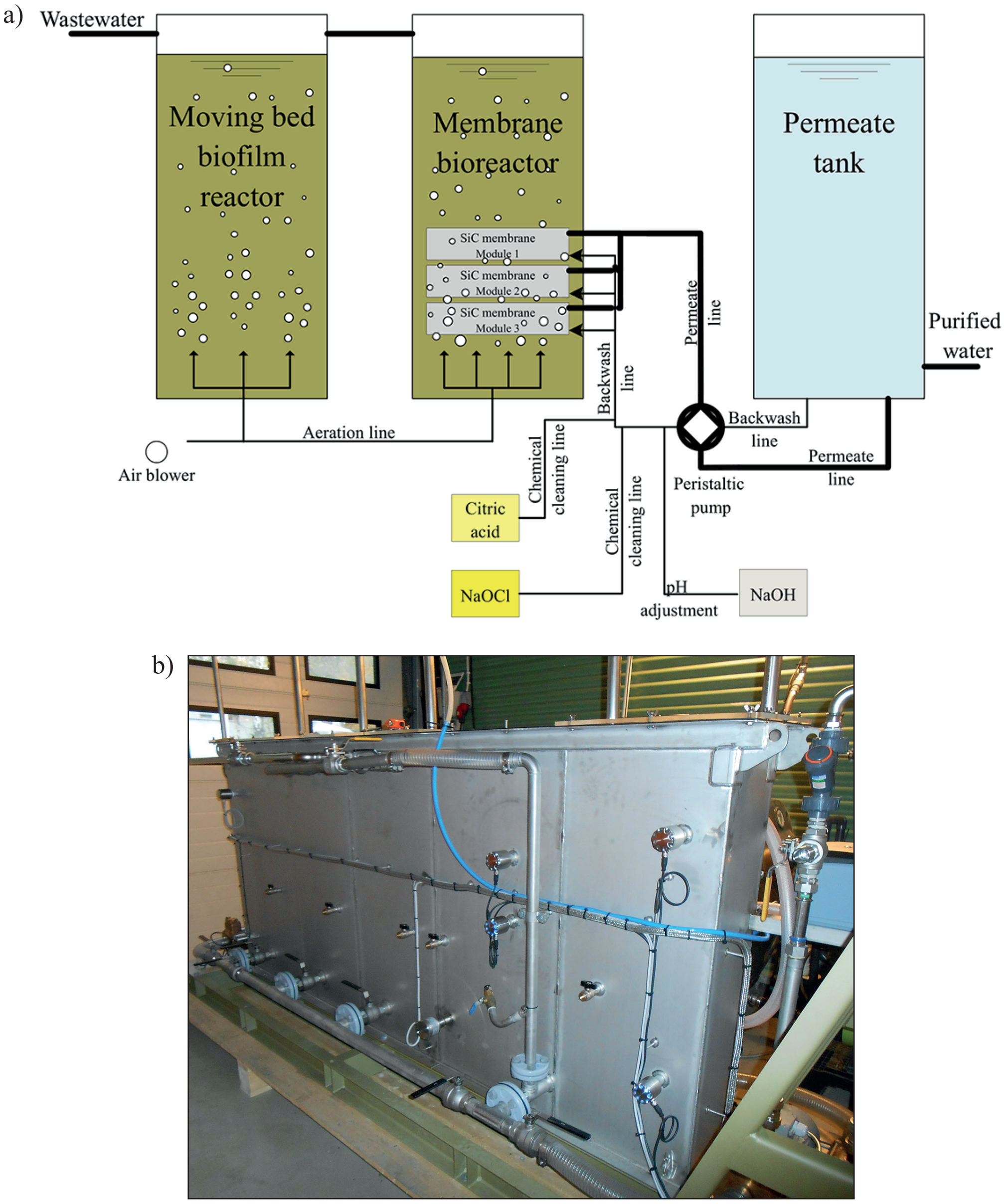

Figure 1. Schematic of ceramic MBR & MBBR pilot plant in Ås, Norway (Dzihora, 2021)

Table 2. Membrane modules and their characteristics

Number of modules	Filtration area, m^2	Theoretical flux (gross), L/m^2/h (LMH)	Theoretical capacity (gross)	
			m^3/h	m^3/d
1	0.276	150	0.04	0.96
2	0.552	300	0.08	1.92
3	0.828	450	0.12	2.88

with 130 days of continuous data. As shown in Figure 1, wastewater first enters the equalization tank (not shown on the drawing), then undergoes biological treatment in 2 subsequential MBBRs. This stage serves as pretreatment before membrane separation and also increases system stability. After biological treatment, wastewater flows to the filtration chamber, where it is sucked through ceramic membranes into the last, permeate, chamber.

Flat-sheet SiC MF membranes were used as separation membranes [Cembrane Inc., 2016]. Silicon carbide material has a relatively high active membrane surface area equal to 0.151 m^2. In one separation chamber, three membrane modules were horizontally parallel placed with the following characteristics (Table 2). The volume of each chamber is 100 liters.

During the filtration process aeration was continuously carried out via aerator diffusers with 2 mm perforation installed at the bottom of the chambers.

Before pilot plant launching, activated sludge was taken from the Bekkelaget municipal WWTP in Oslo. That biomass suspension was acclimatised to the household wastewater. The sewage from the student campus in Ås was used as the source of wastewater. Wastewater flows in the storage tanks separately as gray and black water in ratio 1:10 with average values of Mixed Liquor Suspended Solids (MLSS) 206 mg/l and Chemical Oxygen Demand (COD) 204.5 mg/l.

The operational time of pilot plant could be divided into filtration cycles, where each cycle is also subdivided into: filtration (300 sec), relaxation I (60 sec), backwash (15 sec), and relaxation II (120 sec). Permeate is used as a backwash liquid [Dzihora, 2019].

Long-term operation

System performance was assessed during 130 days of continuous operation and all data was divided into 7 main periods related to different changes in the system. For a better understanding of the situation, the data were analysed separately for each period (Table 3). The first period was the system preparation for operational mode, for sludge adaptation and biological growth until the MLSS value equals 5 mg/l. During periods 2 and 3, the first changes were conducted for system performance optimisation. Chemical cleanings were categorised as separate periods (4 and 6). The second cleaning was more optimised, resulting in a two-fold efficiency increase. Finally, the system was monitored throughout the stable operational conditions (7) in order to observe fouling mitigation and stabilisation of permeability.

Data processing

During the system operational time, all data was continuously recorded and processed. Special controllers, connected to the pilot plant,

Table 3. Operational periods of pilot plant

No.	Periods:	Day	Period description
1	Sludge adaptation	0–20	Adaptation of biomass for operational performance. It was decided that a stable mode should be reached to exclude other influences on the system
2	Increase of sludge recycle	21–34	The influence of sludge recycling on the system performance (change of pulse frequency)
3	Increase of flux	35–40	The influence of flux rate on the system performance
4	CIP-protocol development	41–51	Chemical cleaning
5	Stable operation I	52–77	Stable operation mode for fouling rate observation
6	CIP-II-protocol development	75–88	Chemical cleaning
7	Stable operation II	88–130	Stable operation mode for fouling rate observation

automatically measure main characteristics including: time [sec] and number of filtration cycle; TMP [mbar]; T [°C]; pH. For performance monitoring, representative data from each daily log file was chosen, which is represented by 8 filtration cycles, selected from every 3 hours of system operation. For each cycle, initial and final values are calculated as an average of 10 points at cycle start and cycle end, excluding the pump ramp/relaxation periods. The obtained data were used for the analysis of the system state and trend tracking.

Membrane cleaning and cleaning efficiency

Wastewater contains different pollutants of organic and inorganic nature. Hence, two types of chemicals should be used. For this purpose, a sequence of chemicals was used, namely sodium hypochlorite with 100–1000 ppm of active chlorine (to take out biological pollutants) and 0.2% citric acid to destroy inorganic substances inside the membrane). Moreover, after the addition of NaOCl, pH was adjusted by the introduction of NaOH to pH 10–11.

The backwash is necessary to maintain optimal conditions for filtration, which includes low TMP and high permeability, subsequently low fouling rate. However, after a while, TMP reaches too high values that could not be changed by simple backwash; therefore, chemical cleaning should be provided. For the performance of chemical cleaning, the filtration process should be stopped. The dosing pump directs NaOCl to the backwash line and the membrane remains in the backwash mode until the membrane will be covered with this solution.

The period of 90 days was taken and during that period 2 cleanings were performed. Membrane resistance was used for the evaluation of the cleaning efficiency (CE), which was calculated according to Equation 1 [Racar et al., 2017].

$$CE(\%) = \frac{(R_t - R_m) - R_{rev}}{(R_t - R_m)} \cdot 100\% = \frac{R_f - R_{rev}}{R_f} \cdot 100\% \quad (1)$$

where: R_t – total resistance, m^{-1};
R_m – resistance of the membrane (initial resistance), m^{-1};
R_{rev} – resistance of the reversible fouling, m^{-1}.

RESULTS AND DISCUSSION

Operational data

Influence of sludge recycle rate on system performance

After reaching stable operational performance, it was decided to provide the first changes in the system, namely, sludge recycle pulses (Table 4).

The duration of sludge recirculation (pulse interval) was gradually decreased over 15 days. At first, the change of pulse interval causes slight increases in TMP during 21–27 days (Figure 2) followed by a reduction of permeability P_N. After some time, the system stabilised and followed a TMP increase related only to continuous fouling. Therefore, it could be concluded that the system can adapt to different pulse intervals and there is no need in maintaining a high rate of sludge recirculation. Permeability after stabilisation was 160–170 LMH/bar.

Table 4. Changes in sludge recycle pulses

Day	Pulses	Day	Pulses
21	1620	32	933
23	1458	34	746
27	1166	36	550

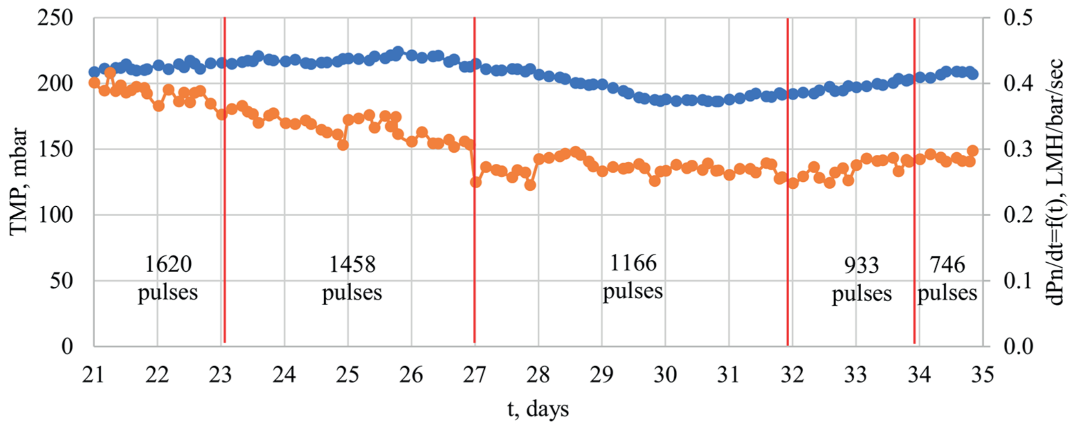

Figure 2. Dependences • TMP = f(t), and • $dP_N/dt = f(t)$ (period 2)

Table 5. Changes in flux

Day	Net flux, LMH	Gross flux, LMH	Filtration speed, Hz
34	16.6	29.71	100
35	22.99	40.94	150

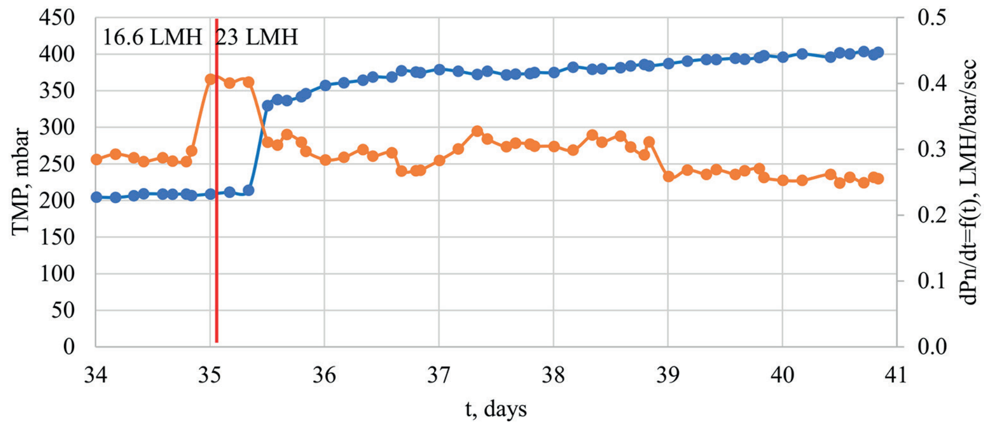

Figure 3. Dependences • TMP = f(t), and • $dP_N/dt = f(t)$ (period 3)

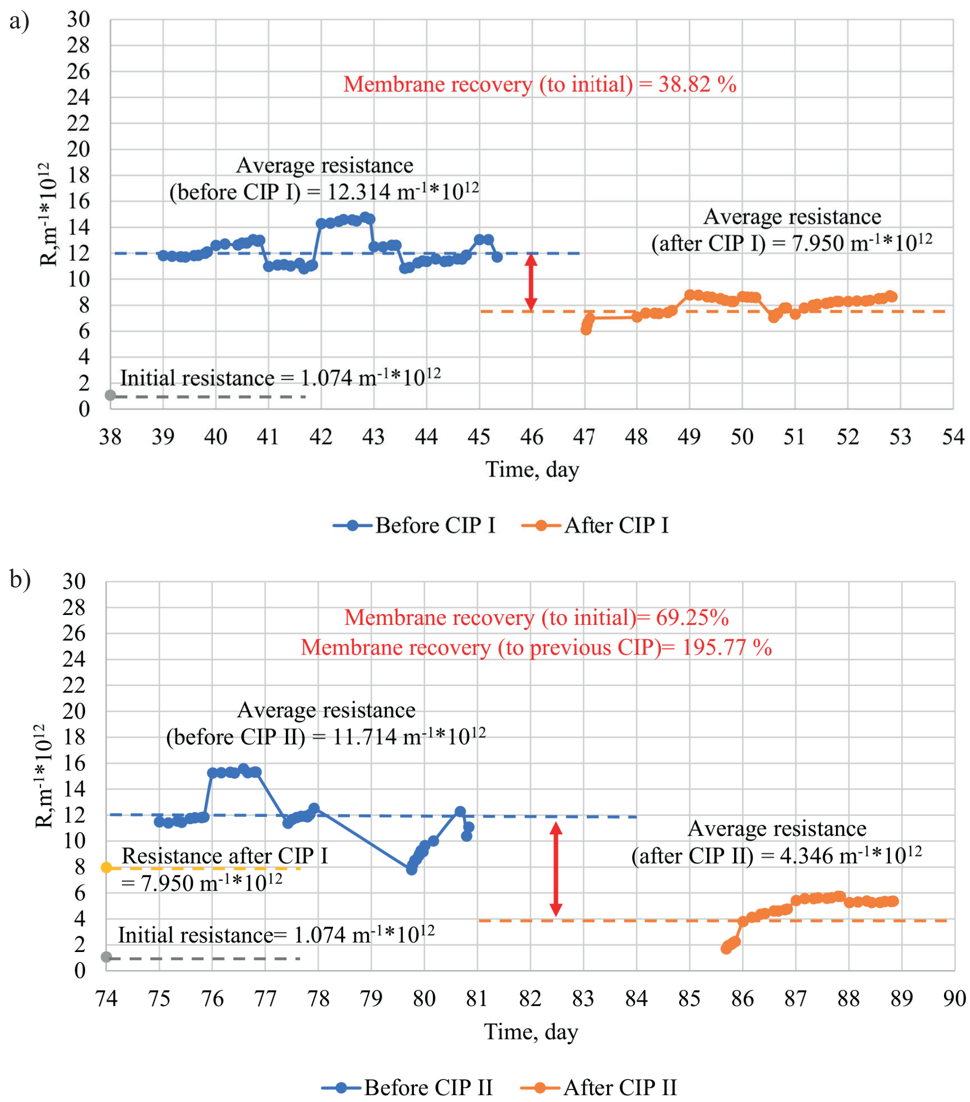

Figure 4. Cleaning efficiency for CIP I & CIP II

Influence of flux on system performance

The next period of optimal condition determination is related to flux regulation. As soon as the system stabilised and sludge recirculation decreased at the beginning of the 35th day, an increase in infiltration speed was observed (Net flux change – from 16.6 to 23 LMH) (Table 5).

This change had an interesting effect (Figure 3): in the first hours, a rapid increase in permeability P_N was observed, while the TMP value remained the same, but after some time extremely fast fouling of the membrane was discovered by the following permeability P_N drop. During the next days, TMP continued to increase, but more slightly and due to fouling only. Permeability after stabilisation was 100–140 LMH/bar.

Chemical cleaning performance

Cleaning efficiency is the percentage of fouling that was removed by the CIP or in other words membrane recovery. It is very important to provide efficient cleaning and prolong the lifetime of the membranes. Therefore, each cleaning should be analysed for further optimisation and cleaning efficiency increase. During operational time 2 cleaning was performed (Figure 4).

Treatment efficiency of the first CIP is 38.82% from initial resistance, whilst the second figure shows 69.25%, so it means that the second CIP was almost twice as effective. It happened because of a long cleaning time and an optimised cleaning protocol.

Stable operation periods

After each CIP system was operated under stable conditions for fouling growth observation (Figure 5). Period 7 has continued twice as long (period 5–25 days and period 7–52 days) and also TMP value was smaller almost by half. Normalised permeability slowly declined from around 200 to 100 LMH/bar during both periods, which proves that fouling growth was effectively mitigated.

Laboratory analyses (COD, MLSS, SVI)

During the operational time of pilot plant, there was conducted analysis of raw wastewater, mixed liquor (ML) from MBBR, activated sludge from MBR, and permeate.

COD completely correlated with the value of TMP; therefore, this laboratory analysis also could be used for system state assessment.

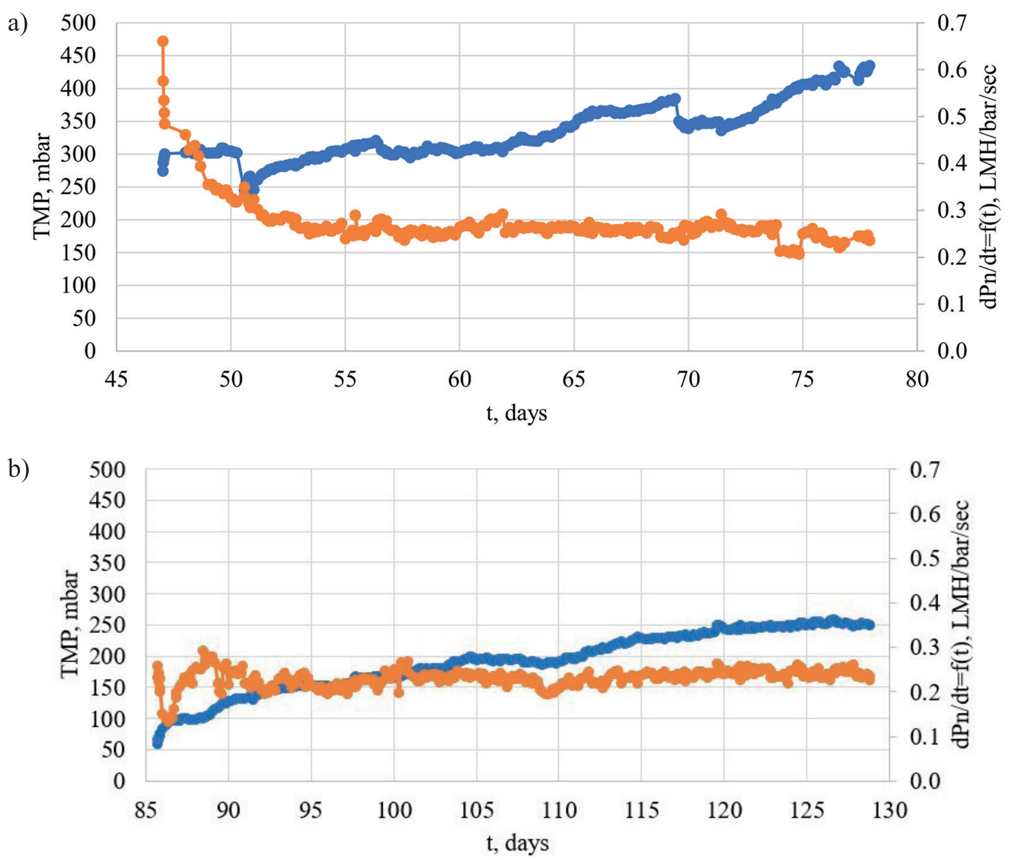

Figure 5. Dependences • TMP = f(t), and • dP_N/dt = f(t) (period 5 and 7)

Table 6. SVI value and sludge characteristics [Yousuf, 2013]

SVI	Sludge characteristics	Maintenance of the system
↑	• ↓ compact & dense; • bulking or too young sludge; • slow settlement ability.	• ↓ discharge rate; • ↑ returning sludge rate.
↓	• ↑ compact & dense; • older sludge; • fast settlement ability.	• ↑ discharge rate; • ↓ returning sludge rate.
Constant	the same characteristics	–

According to the gathered information, could be concluded that the MBBR stage plays important role in the system because different COD value in raw water does not have a significant effect on COD in the separation tank. That means that the separation tank was operated under stable performance conditions even though the loading rate was different. It is also approved by effluent quality, which stays on the same level all the time.

MLSS is one of the main characteristics of biomass (mixed liquor in MBBR and activated sludge in separation membrane tank). The MLSS value shows biological growth in the volume of treated wastewater. It is highly dependent on changes in the system. Every increase of TMP (decreasing of P_N) correlates with the MLSS value (which grows). This value helps to assess the state of the biosystem and decide which volume of activated sludge should be discharged (excess amount of sludge should be removed from the separation chamber and partially recycle to MBBRs.

Sludge volume index (SVI) analysis also was provided with a point to evaluate the settling ability of sludge. Settling ability is dependent on biomass characteristics and decreases with MLSS decreasing. In addition, this value helps to maintain the system conditions according to Table 6. For normal operational conditions, the SVI value should remain in the range of 100 to 250 mL/g. Such conditions maintain good settlement ability of the sludge as well as low permeate turbidity. The SVI values from all periods were within the range, which indicates proper operational conditions.

CONCLUSIONS

In this study, a combination of MBR & MBBR was operated with flat-sheet SiC membranes. The system operated under continuous aeration, which hinders fouling growth. However, it is not enough and other methods should be implemented. Therefore, hydrodynamical variations were performed, including changes in pulse interval and flux. A decrease in pulse interval (sludge recycling) allows fouling rate and TMP value reduction. Flux increase temporarily escalates permeability, the value of which drops after a short time due to extremely fast fouling growth on the membrane surface. Of course, it is easy to control fouling under lower fluxes, but by doing so productivity could be significantly reduced.

Three MF ceramic membranes were used for 130 days and two CIPs were conducted. Each cleaning was performed according to cleaning protocol and was quite highly effective, but still, part of irreversible fouling was left on the membrane surface. The second CIP was the most effective and showed almost 70% of membrane recovery. It is obvious that with the following studies even higher values could be achieved.

System optimisation induced to longer lifetime of membrane and more seldom chemical cleaning and therefore it decreased operational cost. The periods with physical cleaning only were prolonged almost twice with more stable values of P_N and TMP.

The MBR & MBBR combination is the most perspective wastewater treatment system, which should be developed as an option for the decentralised and centralised systems as well.

Acknowledgments

The investigation was conducted in the laboratory, which belongs to the Norwegian University of Life Sciences in Ås, Norway. The object of investigation is MBR & MBBR pilot plant. The studies were conducted with co-financing by the European Union's ERASMUS$^+$ program.

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