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Fouling of a hollow fibre submerged membrane during long-term filtration of activated sludge

Marin Matošić[a*], Marija Vuković[b], Mirjana Čurlin[a], Ivan Mijatović[a]
[a]*Faculty of Food Technology and Biotechnology, University of Zagreb, Pierottijeva 6, HR-10000 Zagreb, Croatia*
Tel. +385 (1) 460-5062; Fax: +385 (1) 460-5072; email: mmatosic@pbf.hr
[b]*Faculty of Chemical Engineering and Technology, University of Zagreb,*
Marulićev trg 19, HR-10000 Zagreb, Croatia

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Abstract

Membrane fouling in membrane bioreactors (MBR) is caused by cake formation on the surface, mainly attributed to suspended solids, and also by surface adsorption connected with pore blocking, attributed to soluble components of activated sludge. In this study, long-term experiments were carried out in sub-critical conditions with frequent backwashing, and the nature of fouling was investigated. The irreversible blocking by adsorption has been identified as the major cause of fouling, as it had gradually decreased membrane permeability from 417 to 55 L m^{-2} h^{-1} bar^{-1} in 123 days of continuous experiment without chemical cleaning. Cake resistance was small due to a low flux and high aeration intensity and it remained constant for most of the experiment. A sudden acceleration of fouling has been observed after 119 days of operation and attributed to irreversible fouling that caused local fluxes to exceed critical flux and lead the filtration into critical conditions, where suspended solids start to deposit. Faster fouling, in comparison to the filtration of normal sludge, has been observed when starving biomass had been filtered. That was attributed to floc breakage.

Keywords: Submerged membrane; Membrane bioreactor; Sub-critical fouling; Filtration resistance; Permeability; Municipal wastewater; Activated sludge

1. Introduction

Membrane bioreactors (MBR) which combine activated sludge process and membrane separation, thus replacing secondary clarifier, are now widely used for wastewater treatment and reclamation. Advantages of MBR over conventional activated sludge technology are better effluent quality, smaller footprint, higher concentration of MLSS (mixed liquor suspended solids), less excess sludge production and generally more

*Corresponding author.

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stable process. Membrane separation is carried out either with cross-flow filtration in side-stream MBRs or with submerged membranes, which operate in dead-end mode. Although the latter have smaller fluxes, they are generally more favoured because of their lower energy consumption required for filtration, as well as the possibility of using the aeration in the bioreactor to prevent fouling by creating turbulent cross flow over the membrane surface.

However, principal limitation of the MBR process lies in membrane fouling as a consequence of the interactions between the membrane and the mixed liquor, which affects overall process performance. Moreover, the present cost of treatment by the MBR is higher than that of the conventional process, mostly due to the initial cost of membranes and the cost of cleaning and/or replacing them. To make the MBR process economically feasible, membrane fouling has to be kept at minimum in order to make cleaning less frequent and to prolong the lifetime of the membrane.

Many studies have been carried out in order investigate mechanisms and factors that influence fouling [1]. Aeration intensity over the submerged membrane surface is recognized as a key operational parameter in preventing cake formation on the membrane surface [2–5]. Permeate flux also has an important role in the control of fouling, where the concept of critical flux [6] has been widely used to predict and prevent fouling [3,4,7]. As for membrane material, it has been reported that hydrophobic membranes were more prone to fouling than hydrophilic ones, due to the fact that most interactions between the membrane and the foulants are of a hydrophobic nature [8,9]. Also, the properties of mixed liquor have been intensively studied by many authors. Attempts have been made to measure the contribution of various constituents of mixed liquor to membrane fouling [10,11]. There is growing evidence that soluble and colloid matter is far more responsible for fouling than suspended microbial flocs where the former cause irreversible fouling by deposition in the membrane pores and onto the surface, which has to be removed by chemical cleaning [12–14]. Among the soluble species of mixed liquor, extra cellular polymeric substances (EPS) of microbial origin have been given most attention as potential foulants. Despite intensive research, membrane fouling in MBRs requires further attention in order to understand complex interactions among biologically active and constantly changing filtration media, hydrodynamic conditions of the filtration process and the membrane itself. The objective of this paper is to further the understanding of the fouling phenomenon during long term filtration conducted in nominally sub-critical conditions with normal sludge and with sludge under starvation conditions.

2. Materials and methods

Experiments were conducted on a pilot plant MBR with hollow fibre membrane (Zenon ZeeWee™-10) vertically submerged directly in the 40 L (useful volume) rectangular based (24×24×93 cm) bioreactor. Membrane properties are given in Table 1.

Flow rates of both permeate and feed water were maintained by laboratory pump and measured with flow meter while the corresponding transmembrane pressure (TMP) was measured by pressure gauge. Measured permeate flux (*J*) was normalised to 20°C using Eq. (1) where μ_T stands for permeate viscosity at a given temperature.

Table 1
Membrane properties

Dimensions	Fibre length, 0.52 m
Filtration area, m^2	0.93
Nominal pore size, μm	0.4
Cross section area, cm^2	94
Resistance (R_m), m^{-1}	6.5×10^{11}

Compressed air was supplied through a diffuser at the base of the membrane to create shear stress, thus mitigating formation of cake layer on the membrane surface, and to obtain aerobic conditions for biological treatment.

$$J\ (20^{\circ}\mathrm{C}) = \frac{J\ \mu_T}{\mu_{20}} \quad (1)$$

The pilot plant was located near the municipal wastewater collector of the city of Zagreb (south). The municipal wastewater (almost completely domestic) was used as the inlet raw water, with average COD (chemical oxygen demand) of 350 mg O_2/L. Testing was carried out in two runs lasting 50 and 123 days, and for both runs the bioreactor was inoculated with activated sludge from the municipal wastewater treatment plant with initial MLSS (mixed liquor suspended solids) concentrations of 5 and 8 g/L, respectively. No sludge has been wasted giving SRT (sludge retention time) of approximately 250 days due to sampling. Aeration was set to 3.4 m^3/h (362 $m^3/h/m^2$ of membrane cross section area), which gave high oxygen concentration, always above 4 mg/L. The membrane was backwashed with the effluent for 10 s every 9.75 min with a backwash flow rate 1.5 times higher than the permeate flow rate. The membrane was not chemically cleaned during the runs, but only in between them, and the cleaning was conducted with sodium hypochlorite solution (750 mg/L). The cleansing with hypochlorite was sufficient to restore the original permeability of the membrane.

During the runs, permeability of the system (membrane filtrating activated sludge) was estimated from the linear part of *J* vs. TMP curve by the best fit method. At least four pairs of measured flux and stabilised TMP values were used to draw the curve for each permeability calculation. TMP reported in this study is a value that was recorded at the end of the cycle, before the backwash. Critical flux was measured at the beginning of the process. Determination of critical flux was carried out by increasing the flux in the step-wise fashion (step height of approximately 5–7 L m^{-2} h^{-1} and step duration of 15 min) and observing the TMP development. The critical flux was considered to be reached when permeability at the end of the step decreased to a value less than 90% of the permeability value at the beginning of the step.

Resistance-in-series model [Eq. (2)] was used to calculate different resistances to filtration where R_t is total membrane resistance, R_m is intrinsic resistance of the new membrane, R_c is cake resistance, R_f is fouling resistance due to irreversible adsorption and pore plugging and μ is the viscosity of the permeate (in Pa s). Flux and TMP data of the new membrane for the filtration of clean water were used for the calculation according to Darcy's law by Eq. (3) to measure R_m. R_t was calculated from the filtration data during the experiment, with flux and TMP measured at the end of a 10-min cycle, before the backwash. For the calculation of the $R_m + R_f$ value (clean water resistance of fouled membrane), activated sludge was removed from the bioreactor, the membrane was washed with light spray and the bioreactor again filled with tap water to measure flux and TMP. Filtration data were used for calculation [Eq. (3)] and activated sludge was returned to the bioreactor to continue the experiment. R_c was calculated through Eq. (2) by subtraction. During the clean water filtration, sludge was kept in an aerated container with feed inflow to minimize physiological stress and special care was given to minimization of sludge loss during its transfer.

$$R_t = R_m + R_c + R_f \quad (2)$$

$$R = \frac{TMP}{\mu\, J} \quad (3)$$

For the measurement of cake resistance for different MLSS concentrations, the liquid phase of the activated sludge was continuously pumped out from the suspension through 0.4 µm submerged membrane and replaced with tap water. It was considered that complete removal of liquid phase had been achieved when the volume of pumped liquid was two times bigger than the volume of sludge. In order to obtain different dilutions of the sludge, water was pumped out from the sludge in 5 steps. After each step, concentrated sludge was transferred into the pilot plant bioreactor where the measurements for R_c calculations were done.

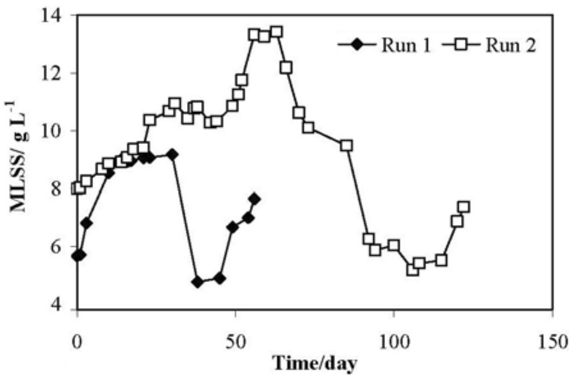

Fig. 1. MLSS development in two runs.

3. Results and discussion

To observe the membrane fouling over a long period, the MBR pilot plant was set to treat municipal wastewater in two consecutive runs. The MLSS development for both runs is presented in Fig. 1. In the first run, MLSS increased to around 9 g/L in 20 days and remained constant until day 35, when some activated sludge was lost due to a feed pump failure, and the experiment was interrupted for 3 days. After 3 days, when the sludge was aerated without the feed, MLSS concentration fell to only 4.8 g/L but when the normal feed flow was established, the MLSS increased again. In the second run, while the organic loading rate was increasing constantly (Fig. 3) until day 60 because of the feed flow increase, MLSS also increased with time. Then, it sharply decreased again when feed flow was reduced. Interruption of feed flow between days 86 and 91 caused even faster decrease of MLSS but, as in the first run, MLSS increased again along with the increase of feed flow. It can be established that MLSS concentration in operation with long SRT strongly depends on volumetric loading rate when the feed concentration is constant. The low sludge production rate, or even complete stagnation of MLSS for MBRs, has been reported earlier [15] and explained by low food to micro-organism ratio (i.e. little substrate per unit biomass), which lead to competition among the micro-organisms and resulted in a reduction of sludge production. Since very little sludge was taken from the bioreactor, accumulation of inorganic and non biodegradable particulate matter present in the wastewater seemed inevitable. However, only small increase (from 20% to 25%) in the inorganic part of activated sludge was observed. The bigger accumulation might have occurred in the dead zones on the bottom of the bioreactor.

In the first run (Fig. 2), feed water flow has been kept at a constant level of 7 L/h, but due to a decreasing temperature in the bioreactor, normalized flux has increased from 7 to 10 L m^{-2} h^{-1}. The applied flux was set well under the measured critical flux of the system (50 L m^{-2} h^{-1} for 8 g/L MLSS and 3.4 m^3/h aeration air flow) to minimize fouling and to allow long-term fouling to be observed. All the other operation parameters such as aeration intensity, back-flushing procedure and MLSS concentration were also set to ensure minimal fouling while keeping the treatment efficient.

As Fig. 2 shows, filtration was very stable with low measured TMP, which very slowly increased in time from 1.8 to 3.7 kPa, giving the average d*TMP*/d*t* of only 25 Pa/d. This proves

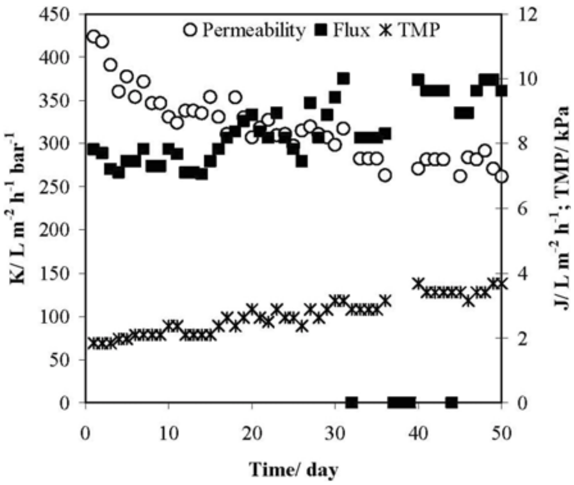

Fig. 2. Permeate flux and membrane permeability during municipal wastewater treatment in the first run.

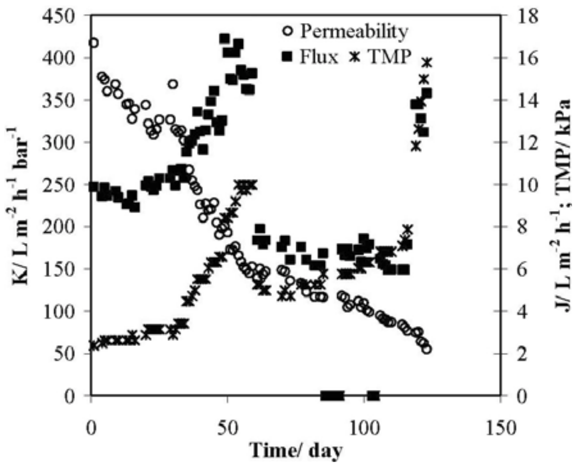

Fig. 3. Permeate flux and membrane permeability during municipal wastewater treatment in the second run.

that although some fouling occurred, prolonged MBR operation could be achieved in sub-critical conditions without chemical cleaning. Rosenberger et al. [15] also reported a very low fouling rate with the membrane and filtration media similar to the one used in this study for the first 110 days of operation at constant 18 L m^{-2} h^{-1} flux. Cho and Fane [16] experimented with a flat sheet MBR in sub-critical conditions and measured much higher fouling rate (1200 Pa/d), but they worked with different media, without backwashing and at constant flux which was only 40% lower than the critical, while in this study the flux was set much lower than the critical flux (7 and 50 L m^{-2} h^{-1}, respectively). Ognier et al. [17] reported a sub-critical fouling rate of 605 Pa/d for anaerobic tubular side-stream MBR.

In the second run (Fig. 3), a more rapid increase of TMP by day 60 was caused by intentional graduate flux increase. Nevertheless, overall fouling rate was very similar to the first run. The observation was based on the permeability decrease rates, which were 2.4 and 2.7 L m^{-2} h^{-1} bar^{-1} day^{-1} for the first and second run, respectively. However, influence of the flux on the fouling rate can be observed in Fig. 3 as permeability decline was faster during the first 60 days (average 4.7 L m^{-2} h^{-1} bar^{-1} day^{-1}), when higher fluxes were applied.

The long period of stable filtration and the slow development of fouling were interrupted when flux was again raised to 13 L m^{-2} h^{-1} on day 119. More pronounced fouling occurred and TMP increased daily to the final 15.8 kPa in 4 days of very unstable operation. The d*TMP*/d*t* value was increasing between two backwashes until it was impossible to maintain TMP below the sustainable level by the chosen backwash frequency. The filtration was stopped and the membrane cleaned chemically. It should be noted that TMP development between two backwashes was negligible before the day 119. Clearly, the system changed from sub-critical to critical behaviour. Under the same flux (13 L m^{-2} h^{-1}), imposed on the membrane in the middle of the run, filtration was easily maintained and conditions were clearly of sub-critical nature. Since the permeability was always measured in sub-critical conditions where the *J* vs. TMP plot was linear, it did not correlate well with TMP during the critical phase of the experiment because TMP was given as a value at the end of the cycle before backwash. Permeability decline rate in the critical phase remained much as it was in the sub-critical, which suggests that adsorption of trace foulants presumably responsible for the irreversible slow fouling was quite independent of cake deposition of suspended particles.

The sharp increase of TMP after long period of slow TMP development observed in the second run has been reported by several authors [18]. This behaviour can be explained by a two-step model [16]. In the first step, slow accumulation of trace foulants on the membrane surface and pores gradually increases the filtration resistance. Since flux is maintained below the critical flux, deposition of the main foulant, i.e. biomass, is prevented and the fouling rate is slow. According to the model, the fouling in the first step has to be heterogeneous in order to create the areas of membrane surface which will be exposed to different local fluxes. Such heterogeneous fouling can be expected to occur in the case of a submerged hollow fibre membrane, which is subjected to different TMP along the fibres. The shift to the second phase would occur when the local fluxes exceeded the critical flux and the biomass flocs started to deposit on the membrane causing abrupt increase of TMP. Recently, a single parameter mathematical model has been proposed to predict sudden increase of fouling in sub-critical conditions based on the concept of local flux [17].

Resistances during the second filtration run were estimated in order to assess the nature of fouling (Table 2). At the beginning of the experiment, membrane resistance (R_m) gave 75% of total resistance (R_t). As the experiment continued, R_t increased due to an increase of the fouling resistance (R_f) which was responsible for 78% of the R_t at day 110. Cake resistance (R_c) remained low and nearly constant under subcritical conditions, suggesting that cake formation has been prevented significantly with the chosen operational set-up, i.e., the transport of particles from the bulk solution towards the membrane, caused by the permeate flux, was successfully countered by back transport due to turbulent air flow. As the conditions changed to critical, R_c increased 4 times, indicating substantial cake formation. However, the main cause of fouling during most of the run was irreversible adsorption on the membrane and into its pores, represented by R_f, which eventually led to cake fouling in the end. These results confirm the two-step fouling model explained above. In the literature, R_c is usually reported as the main constituent of the total filtration resistance, but these are mostly short time experiments [19] or the ones carried out in dead end mode [8, 12]. Jiang et al. [20], however, measured R_c which was 17% and 45% of R_t depending on the backwashing frequency while Chang and Kim [14] reported R_c that comprised less than 1% of R_t for the continuous long-term operation of submerged MBR. Obviously, the absolute value and relative distribution of resistances strongly depend on system configuration and operation set-up.

Despite some doubts about whether R_c and R_f should be considered completely independent [1], we feel that the method of their measurement is simple enough and that valuable information may be gathered by decoupling them in this way.

Table 2
Resistances to filtration during municipal wastewater treatment (run 2)

Time [day]	Resistance [$m^{-1}\times10^{11}$] (fraction of R_t [%])				
	R_t	R_m	R_c	R_f	$R_c + R_f$
1	8.7	6.5 (75)			2.2 (25)
52	20.9	6.5 (31)	2.4 (12)	12.0 (57)	14.4 (69)
99	34.2	6.5 (19)	2.1 (6)	25.6 (75)	27.2 (81)
110	41.3	6.5 (16)	2.5 (6)	32.3 (78)	34.8 (84)
120	57.1	6.5 (11)	10.6 (19)	40.0 (70)	50.6 (89)

Also, as the method for measuring these resistances is simple to carry out and as the flux and TMP data collected during backwash with clean water can be used to estimate R_c and R_f, they may be used for fouling characterisation even in full scale MBRs.

Fig. 4 shows the results of an experiment carried out to estimate the influence of MLSS concentration on R_c in conditions identical to the above experiments (flux 7.5 L m^{-2} h^{-1}, aeration 3.4 m^3/h). Since the liquid phase of mixed liquor had been replaced with tap water, the concentration of soluble foulants became negligible and it was assumed that only suspended matter caused fouling. There was definitely an influence of MLSS concentration on R_c, as Fig. 4 shows. However, the influence of suspended particles on the measured total resistances was quite minor for all MLSS concentrations in experimental conditions employed. It can be established that if operational parameters are properly chosen, MLSS concentration should not have a dominant influence on fouling. Similar observations have been published previously [3,5,21]. It should also be noted that suspended particles in mixed liquor could have a positive influence on the prevention of fouling. There is growing evidence [13,19] that suspended solids can act as a dynamic layer over the membrane surface, which can slow down the penetration of soluble fouling species that cause fouling. Generally taken, the assessment of major fouling potential to soluble species in mixed liquor seems entirely justified.

The advantage of MBR over conventional biological treatment consists of the secondary settler elimination, which makes the process less dependent on biomass properties that influence sludge settling. Therefore, it is possible to operate an MBR in applications where feed water interruptions and changes occur frequently because the problem with bulking and floating sludge no longer presents a threat to the treatment process. This makes application of MBR convenient for diverse purposes. Moreover, during periods of

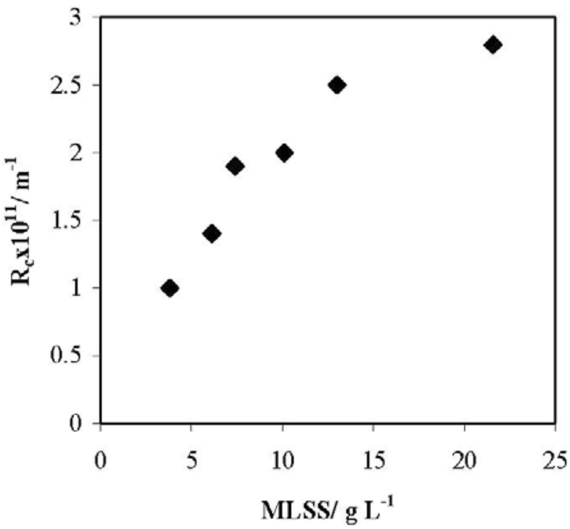

Fig. 4. Cake resistance of a membrane filtrating activated sludge of different concentrations at 3.4 m^3/h air flow and 7.5 L m^{-2} h^{-1} permeate flux.

feed interruption, biomass concentration stagnates or even decreases, thus decreasing the amount of excess sludge to be handled. On the other hand, membrane fouling seems to be greater in such biological conditions [8,9,12]. Fig. 5 shows the results of an experiment with MBR in recirculation without any feed, where biomass concentration decreased from 21.6 to 11 g/L as MLSS. The volatile fraction of MLSS decreased from 85% to 73% during 18 days, but the biomass retained some viability, since the specific oxygen uptake rate for glucose at the end was one fifth of the beginning value.

Membrane fouling in Fig. 5 is given as membrane permeability measured while filtrating activated sludge. The fouling was more pronounced during the first 7 days and afterwards it slowed down. The fouling rate for first 7 days was 18 L m^{-2} h^{-1} bar^{-1} day^{-1} and after the 7th day was 3 L m^{-2} h^{-1} bar^{-1} day^{-1}. A similar tendency can be seen in two runs during wastewater treatment (Figs. 2 and 3), but the initial fouling rates for the first days in those cases were much lower. The higher MLSS concentration might have an

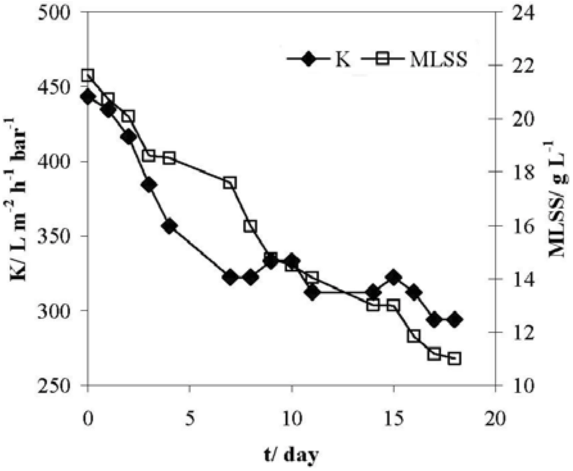

Fig. 5. Membrane permeability during filtration of activated sludge without substrate inflow at 0.1 m/s aeration intensity and 7.5 L m^{-2} h^{-1} flux in recirculation.

Table 3
Resistances to filtration during an 18-day run without substrate inflow

Time [day]	Resistance [m^{-1}×10^{11}] (fraction of R_t [%])				
	R_t	R_m	R_c	R_f	$R_c + R_f$
3	9.4	6.5 (69)	1.8 (19)	1.1 (12)	2.9 (31)
15	11.2	6.5 (58)	2.2 (20)	2.5 (22)	4.7 (42)
18	12.3	6.5 (53)	2.5 (20)	3.6 (29)	5.8 (49)

influence on the fouling rate, but, as discussed above, this does not seem to be the explanation here. The probable cause for the higher fouling rate may be the increased concentration of fouling colloid and soluble species released by floc breakage and biomass decomposition. Calculated resistances in Table 3 show an increase in both R_c and R_f. Increase of R_c does not follow the decrease of MLSS, but as MLSS decreased, the properties of mixed liquor also changed. The stable increase of R_f indicated slow irreversible fouling again. It seems that mere hydrodynamic approach is not sufficient for describing the fouling phenomenon because the properties of mixed liquor can strongly affect it.

4. Conclusions

The membrane bioreactor treating municipal wastewater was capable of long and stable performance without chemical cleaning. Extended operation was enabled by low imposed flux and sufficient aeration, all of which put the system well under critical conditions. The membrane fouling was rather slow and irreversible by nature, while fouling caused by cake formation remained constant and low. An abrupt increase in fouling rate was observed after 119 days of continuous filtration, and was attributed to irreversible fouling that caused local fluxes to exceed critical flux and lead the filtration into critical conditions, when suspended solids started to deposit. The fouling rate was influenced by a physiological state of the biomass. During the filtration of starving biomass in the condition of floc breakage, an increased fouling rate has been observed. The altered physiological state also influenced the cake resistance, which was higher than obtained in the filtration of normal sludge.

Acknowledgments

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