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Study of the influence of different variables on membrane fouling in submerged membrane bioreactors

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ABSTRACT

Membrane fouling is still the greatest handicap to overcome in membrane bioreactor (MBR) technology. In this study, a multi-factorial experimental design was carried out in order to optimize the MBR process, studying the effect of major variables involved in the fouling phenomenon as well as the interaction between them. To that end, a submerged membrane bioreactor was operated at three different levels of: sludge retention time (SRT) [25 and 60 d], mixed liquor suspended solids (MLSS) [6 and 14 g/L], and ratio between soluble and particulate chemical oxygen demand (sCOD/pCOD) [1 and 4] in the wastewater. The concentration of extracellular polymeric substances (EPS), as one of the most relevant factor responsible for membrane fouling, and the permeate flux were measured. After an exhaustive statistical analysis, it was confirmed that there is a relationship between the concentration of EPS and membrane fouling. It also appears that the ratio sCOD/pCOD in the composition of the wastewater plays a very important role in the fouling which becomes less pronounced as the soluble fraction increases. Moreover, high fouling potential was observed at high SRT values. Finally, it was proved that the membrane fouling was slightly less pronounced at higher MLSS concentration.

Keywords: Activated sludge; Extracellular polymeric substances (EPS); Fouling; Membrane; Microfiltration; Mixed liquor suspended solids (MLSS); Particulate matter; Soluble matter; Sludge retention time (SRT); Submerged membrane bioreactor (SMBR)

1. Introduction

Membrane bioreactors (MBRs) are suspended growth activated sludge treatment systems that rely upon membrane filtration prior to discharge of the treated effluent, thereby replacing the solids separation function of the secondary clarifier. The MBR technology for wastewater treatment is becoming increasingly popular. However, the widespread application is still restricted, mainly by membrane fouling, which reduces membrane permeability [1–3]. This fouling phenomenon during membrane filtration is due to the interaction between the membrane material [4] and the mixed liquor components [5]. In fact, the process is strongly influenced by three factors:

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biomass and wastewater characteristics, operational conditions, and membrane characteristics [6,7].

Understanding and control of membrane fouling is a complex process due to the amount of parameters, and interactions among them that have to be taken into account, such as membrane material, configuration, and characteristics (pore size, surface energy, and charge, hydrophilicity, etc.).

It is known that sludge retention time (SRT) is an important variable in membrane fouling. However, the results from several studies have been unclear. Although many researchers reported that membrane fouling is less pronounced as SRT increases [8–14], others reported the opposite [15,16]. Therefore, up to date, the effect of SRT in membrane fouling is controversial. Even though its direct effect in fouling is not elucidated, it has been proved that SRT may affect other factors that influence membrane fouling such as viscosity and relative hydrophobicity [7,11,16].

Another factor strongly affecting membrane fouling is the concentration of mixed liquor suspended solids (MLSS). In this case, there are also discrepancies due to the complexity and variability of the biomass nature; some studies reported that lower MLSS concentration may improve membrane filtration [17–19], whereas others [20] observed that there was no impact of MLSS on membrane fouling for values of 4–8 g/L, and even a larger permeate flux was obtained for 12 g/L.

Finally, it is clear that the composition of feed water has an influence on the activated sludge composition in the mixed liquor, which consequently influences the membrane biofouling [5,21]. In this context, it would be interesting to know the effect of the soluble and particulate COD ratio (sCOD/pCOD) on the membrane fouling, so this issue has been addressed within this work.

Many submerged membrane bioreactor (SMBR) studies have identified extracellular polymeric substances (EPS), together with soluble microbial products (SMP), as the most relevant factors that are responsible for the membrane fouling [12,22–25]. EPS are a matrix of large polymeric molecules containing variable proportions of proteins, polysaccharides, nucleic acids, humic-like substances, lipids, and heteropolymers such as glycoproproteins, which are secreted by micro-organisms and located at or outside microbial cell surfaces. The accumulation of EPS in the sludge suspension and on the membrane can raise the viscosity of the mixed liquor and block membrane pores and/or form a fouling layer, leading to the increase in filtration resistance [23,25]. The EPS generation will depend on different parameters such as SRT, feed composition, and MLSS.

Given the complexity of the fouling phenomena, the main objective of this work is to use an experimental design to determine the influence of MLSS, SRT, and the ratio sCOD/pCOD, as well as their interactions, on membrane fouling in SMBR, as well as the relationship between these factors and the concentration of EPS, which are one of the main substances causing membrane fouling.

2. Materials and methods

2.1. Experimental plant

As shown in Fig. 1, the pilot plant consisted of a cylindrical aerated reactor with a submerged hollow fiber microfiltration membrane. The capacity of the reactor was 75 L and the polyvinylidene fluoride (PVDF) hollow fiber membrane module supplied by Micronet Porous Fibers, had a surface of 1 m^2 and pore size of 0.40 μm. Synthetic wastewater was fed into the biological reactor and water level was kept constant by a level sensor. An air diffuser was installed below the membrane module so as to provide dissolved oxygen and agitation in the reactor, as well as to remove attached sludge out of the membrane by shear force. The reactor was operated with filtration–backwashing cycles, with a filtration period of 12 min followed by 0.5 min backwashing. The backwashing flux ratio was 2.5:1 with respect to the permeate flux. This ratio was given by the membrane manufacturer. Air demand was of 0.4 m^3/h.

2.2. System operation

An experimental design was carried out in order to evaluate the contribution of SRT, MLSS, and sCOD/pCOD to membrane fouling (Table 1). Two different levels were set up for each parameter, which were 25 d and no sludge withdrawal for SRT, 7 ± 1 and 14 ± 1 g/L for MLSS, and with a ratio of sCOD/pCOD about 1 and 4. For all these experiments, the concentration of EPS and the permeate flux was measured. The hydraulic retention time was set at five hours. A different membrane module was used for each trial and the same initial water permeability of the module was ensured.

The low and high levels of MLSS were adjusted by regulating the concentration of synthetic wastewater, maintaining the MLSS concentration practically constant for each experiment. When the system was operated at 25 d SRT, a portion of sludge was removed daily from the reactor, while there was no sludge wasted in case of a 60 d SRT. The composition of the synthetic wastewaters for the low and high levels of

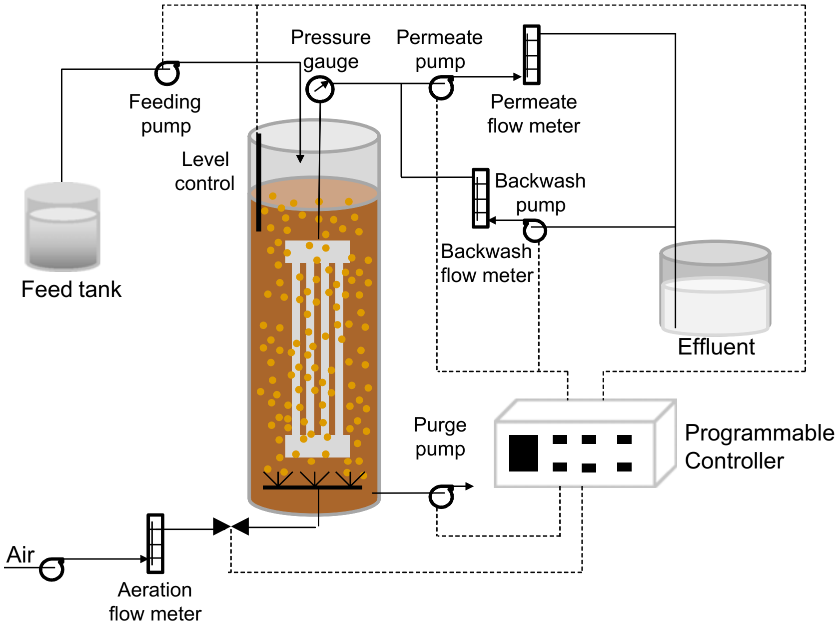

Fig. 1. Schematic diagram of the submerged hollow fiber membrane bioreactor.

Table 1
Experimental multi-factorial design

Data matrix Run	MLSS (g/L)	SRT (d)	sCOD/pCOD
− Low level	7 ± 1	25	≈1
+ High level	14 ± 1	60	≈4
1	−	+	−
2	−	−	−
3	−	−	+
4	−	+	+
5	+	+	+
6	+	−	+
7	+	−	−
8	+	+	−

the sCOD/pCOD ratio is shown in Table 2. These solutions were diluted with tap water to a desired COD concentration. It should be noted that the COD removal efficiency was above 90% in all cases.

2.3. Analytical methods

The extraction of the EPS solution was based on a thermal treatment method [14]. In this study, the EPSs were determined as the sum of carbohydrates and proteins, which were analyzed by colorimetric methods. The carbohydrate fraction was measured using the phenol/sulfuric acid method developed by DuBois et al. [26] for which glucose was used as a standard. The protein content was analyzed by the Folin method developed by Lowry et al. [27] and bovine serum albumin was used as a standard. The measurement of the EPS in the trials without sludge purge (SRT = 60 d) was carried out after the stabilization of the sludge, as once the purge is stopped, SRT will rise, modifying the F/M ratio and consequently, the amount of EPS within the system.

Standard methods [28] were used to determine the COD and MLSS, which were measured regularly. The permeate flux and suction pressure were measured daily during each run. The statistic analysis was carried out by Statgraphics XV software.

3. Results and discussion

3.1. Evolution of permeate flux and membrane pressure under different operating conditions

The evolution of the permeate flux (J_p) and membrane pressure with time was measured under different operating conditions. The results are shown in Figs. 2 and 3, where a similar trend can be observed for both, J_p and suction pressure, in all trials. The figures show an increase in suction pressure and a decrease in the permeate flux. This phenomenon occurs due to the membrane fouling, which can be attributed mainly to sludge cake deposition on the membrane and clogging of membrane pores [4]. The response of the permeate flux is not linear for all operational conditions, since two stages can be clearly

Table 2
Composition of the synthetic wastewater used for the low and high levels of sCOD/pCOD

Levels Composition	sCOD/pCOD ≈ 1 Concentration (mg/L)	sCOD/pCOD ≈ 4 Concentration (mg/L)
Glutamic acid	40	120
Celulose	44	–
Starch	45	–
Casein	35	–
Sucrose	10	
Peptone	44	
Yeast extract	45	
Oleic acid	4.5	
Acetic acid	6	
Ethanol	3	
$NaHCO_3$	450	
K_2HPO_4	100	
KH_2PO_4	40	
$MgSO_4{\cdot}7H_2O$	112	
$CaCl_2$	27	
$FeCl_3{\cdot}6H_2O$	1.5	
NH_4Cl	76	
$(NH_4)_2{\cdot}SO_4$	47	
Characterization of the synthetic wastewater		
Total COD (mg/L)	520	510
Soluble COD (mg/L)	250	400

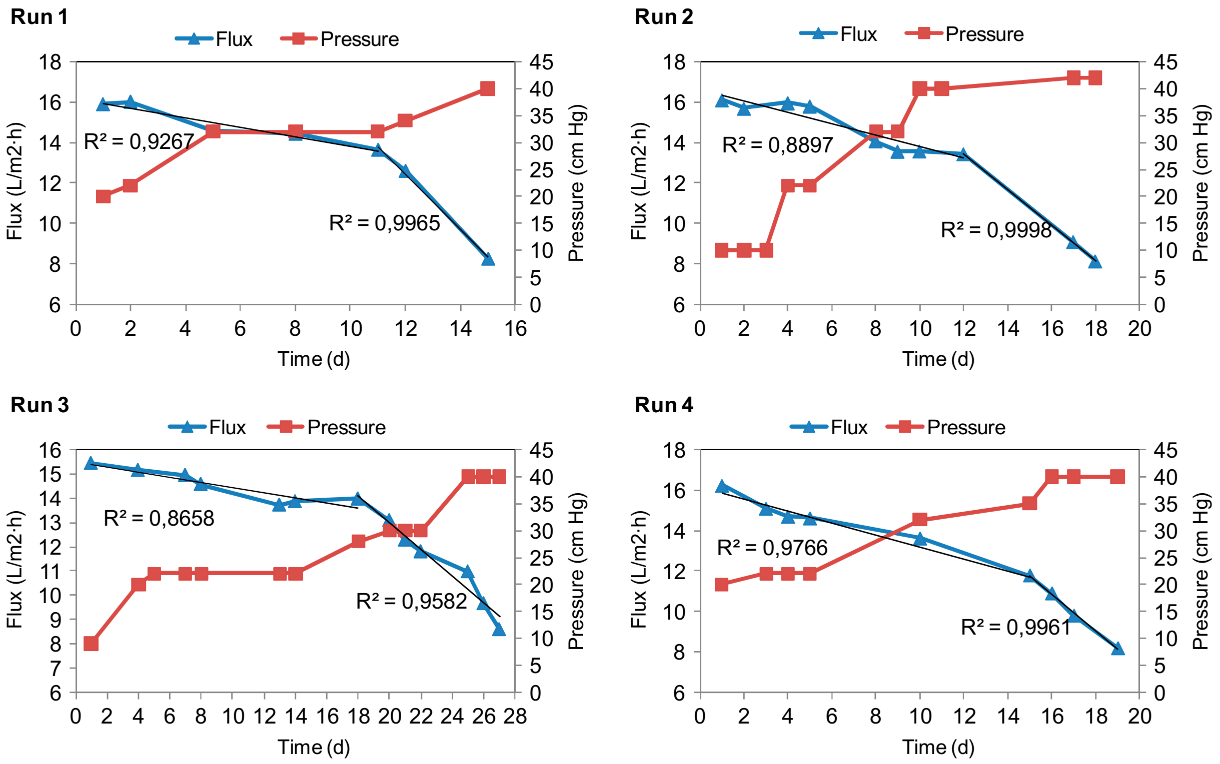

Fig. 2. Evolution of the permeate flux and suction pressure for low MLSS concentrations.

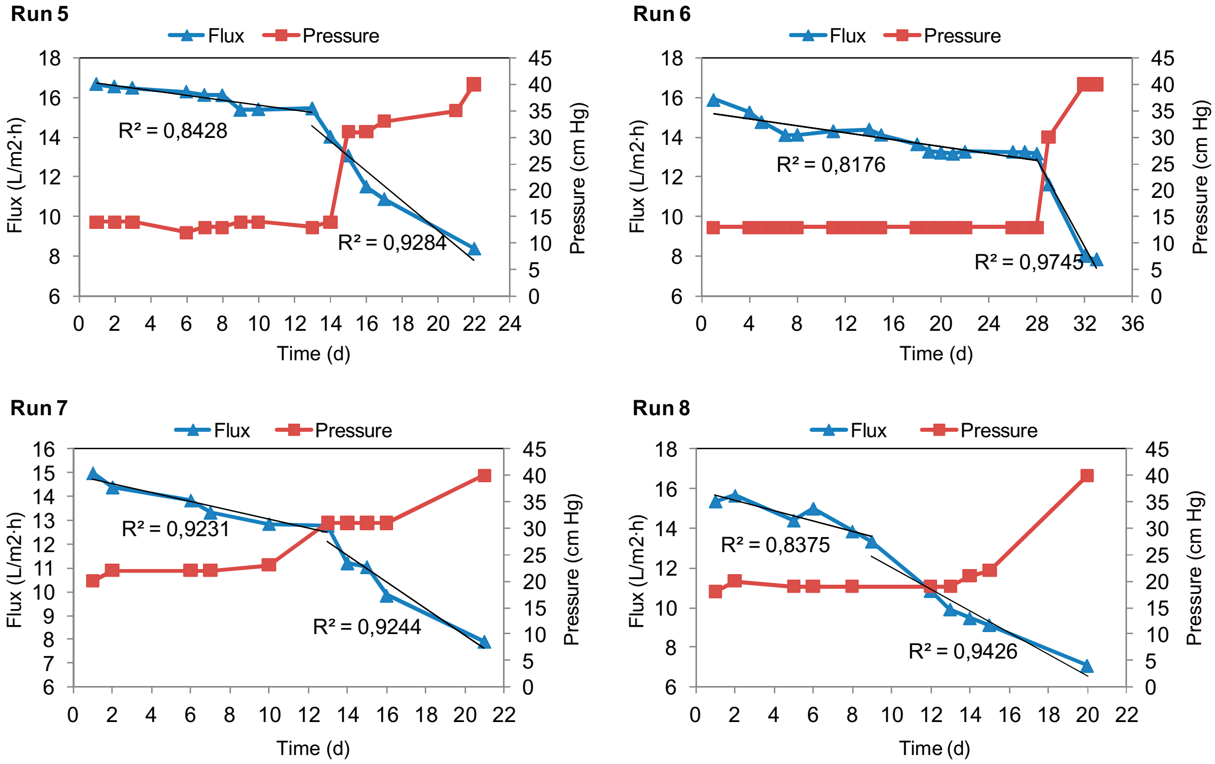

Fig. 3. Evolution of the permeate flux and suction pressure for high MLSS concentrations.

distinguished. In the first stage—slow fouling—the membrane surface and pores are progressively covered by organics like EPS causing an easier adhesion of microbial flocs to the membrane surface. These flocs can promote the cake formation with a slight permeate flux drop in this stage. In the second stage—quick fouling—the membrane flux decreases suddenly as a consequence of the formation of a more compact cake layer [29].

As shown in these figures, trials 1, 2, and 4 were the ones, where a large flux decrease was obtained in the shortest time, and trial 6 was the one with a longest time.

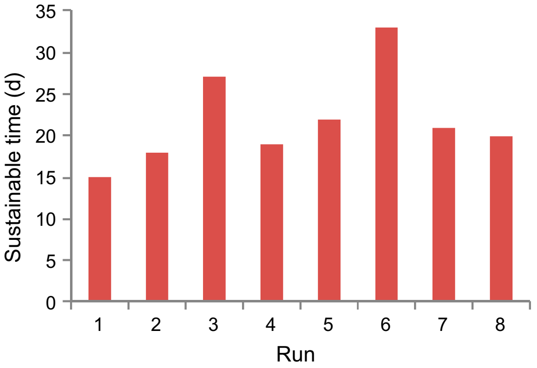

Fig. 4. Sustainable time for each run.

3.2. Sustainable time

The sustainable time was defined as the time at which the permeate flow decreased by 50% with respect to the initial permeate flux. The results were obtained from graphics of flux vs. time (Figs. 2 and 3) and are illustrated in Fig. 4. It can be observed that best results were achieved at runs 3 and 6, both at low SRT and sCOD/pCOD ≈ 4 but at different MLSS concentration, low and high, respectively. In fact, the results of the statistical analysis showed that the effect of the interaction between the SRT and sCOD/pCOD ratio was relevant (Fig. 5). However, these variables independently had even more effect in sustainable time and consequently in membrane fouling. The higher SRT was, the faster membrane fouling occurred, most likely due to large amounts of foulants and high fluid viscosity [7,11,16]. Regarding the ratio sCOD/pCOD, the higher the particulate fraction was, the faster membrane fouling was observed. This could be explained by the adhesion of particulate matter on membrane surface and pores. Moreover, it was reported that membrane fouling was a little less pro-

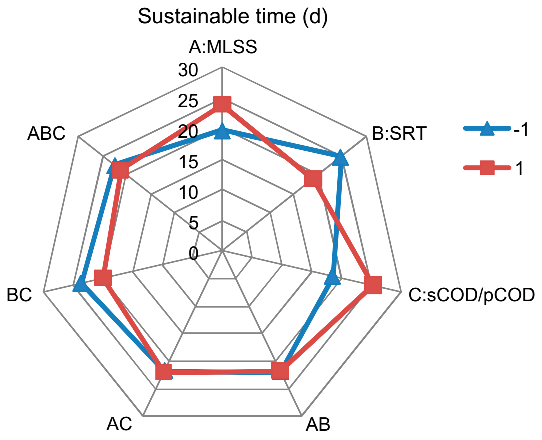

Fig. 5. Radar graph of the main effects on the sustainable time.

nounced at higher MLSS concentration. In this regard, as previously mentioned, several researchers have reported that fouling increases with MLSS concentration [17–19], although there is a controversial issue as there are other works that show just the opposite [30], and they attribute this fact to scouring effects of the large sludge particles. This scouring effect will increase with increasing MLSS, since more flocs are present. In any case, MLSS itself is currently considered only weakly correlated with membrane fouling in SMBR in its common ranges such as 6–15 g/L.

3.3. EPS concentration

The concentrations of EPS, as the sum of carbohydrates and proteins, are summarized in Fig. 6. EPS concentration has been expressed per gram of volatile suspended solids instead of MLSS, so as to be more representative. As shown in Fig. 7, the main factor to influence total EPS concentration was MLSS. It can be observed that the highest values of EPS have been obtained for the trials with lower concentration of MLSS (6 g/L), with the exception of trial 3, which has been carried out with low SRT. On the contrary, its concentration was smaller with high MLSS values. This can be attributed to the process entering endogenesis, which modifies the F/M ratio and thus the extracellular metabolites are consumed by the microorganisms, decreasing its concentration.

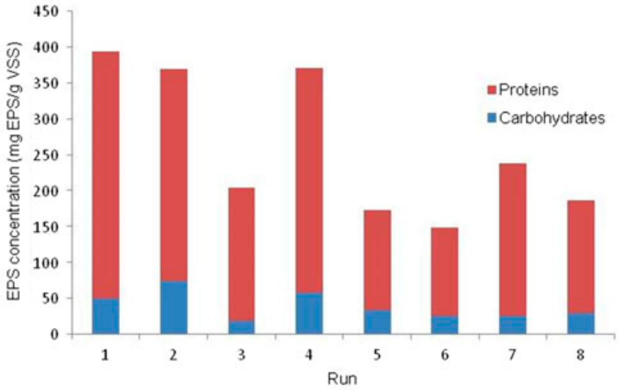

Fig. 6. Effect of EPS concentration in membrane fouling.

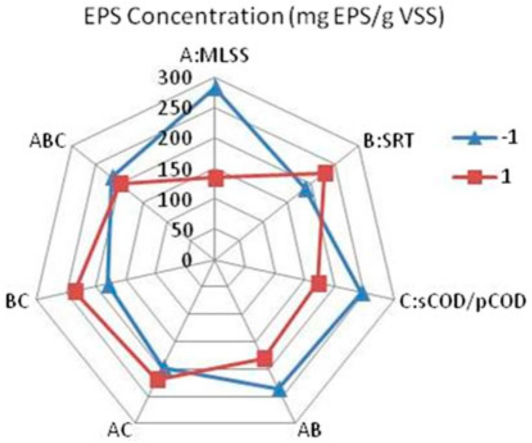

Fig. 7. Radar graph of the main effects for total EPS concentration.

The sCOD/pCOD ratio also brought about a higher soluble fraction and lower EPS concentration, though to a lesser extent.

3.4. Correlation between EPS concentration and sustainable time

As it is shown, there is a direct relationship between sustainable time—and consequently membrane fouling—and EPS concentration. The higher the EPS concentration, the lower the sustainable time was calculated. The trial with the highest EPS concentration and with the highest fouling was the one with the following operational conditions: low MLSS (7 g/L), high SRT (60 d), and low SCOD/pCOD ratio (≅1).

The statistical analysis was carried out by fitting the results to a linear model. There was a significant relationship between the total EPS concentration and sustainable time, since the p value was 0.0296 ($p < 0.05$) with a 95% of confidence level. In addition, a correlation coefficient of −0.757244 also showed a reasonably strong interaction between these two variables.

On the other hand, as shown in Fig. 6, Runs 2 and 4 had almost the same total EPS concentration;

however, in Run 2 the carbohydrates concentration was higher than in Run 4 and therefore, the membrane fouling occurred slightly faster. Furthermore, the membrane fouling was less pronounced in Run 3 in comparison with Runs 5 and 8, even though the total EPS concentration was higher. This could be due to the lower fraction of carbohydrates. Indeed, several researchers have reported that the carbohydrates fraction of both EPS and SMP contribute to fouling more than the protein fractions [18,31].

4. Conclusions

In this study, the membrane fouling was investigated under different operational conditions based on a multi-factorial experimental design, so the effects of SRT, MLSS, and sCOD/pCOD ratio, as well as the interaction between them were studied. The results lead to the following conclusions:

(1) In all cases, the decline of permeate flux due to the membrane fouling happened in two stages. In the first one—slow fouling—the permeate flux slightly decreased due to the gradual deposition of organics like EPS on the membrane surface and pores. In the second stage—rapid fouling—there was a pronounced permeate flux drop as a consequence of the compaction of the cake layer.

(2) The major variables affecting the membrane fouling were the SRT, the sCOD/pCOD ratio, and the interaction between them. Regarding the sCOD/pCOD ratio, as the soluble fraction increased the membrane fouling was less pronounced. Moreover, high fouling potential was observed at high SRT. Although the MLSS concentration had no relevant influence on membrane fouling, less membrane fouling was observed at high MLSS concentration.

(3) Higher MLSS concentrations in MBR brought about lower EPSs concentrations. In addition, the sCOD/pCOD ratio also had an influence —to a lesser extent—on EPS concentration: the higher the soluble fraction, the lower the EPS concentration.

(4) The statistical analysis shows that there was a reasonably strong relationship between total EPS concentration and membrane fouling.

(5) To sum up, in this study, the optimum operational conditions to minimize the membrane fouling were low level of SRT (25 d), high level of MLSS (14 g/L), and high level of the sCOD/pCOD ratio (4).

For a better knowledge of biological processes in MBRs, different molecular biological tools will be used in future studies so as to understand the metabolic pathways involved in several biological reactions and to know which micro-organisms and enzymes are responsible for that.

Acknowledgments

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