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New technology for wastewater treatment to decrease fouling propensity

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ABSTRACT

Membrane bioreactor (MBR) is widely regarded as an effective tool for wastewater treatment to the conventional activated sludge process thanks to its various advantages. In this paper, we study the influence of chemical oxygen demand to nitrogen ratio (ratio COD/N) on MBR fouling propensity. Therefore, two runs (I and II) were conducted with different low ratio COD/N equal to 2.3 and 0 to promote autotrophic bacteria development. The organic loading rates (OLR) were equal to 0.3 and 0kg COD.m^3d^{-1} for runs I and II, respectively and the nitrogen loading rate (NLR) was equal to 0.12 and 0.2kgNm^3d^{-1} for both runs. The results showed that the fouling rate decreased from 0.064E+12 to 0.015E+12 ($m^{-1}d^{-1}$) with the decrease of COD/N ratio. The two runs are conducted without membrane cleaning during all the experimental periods equal to 51 for the first run and 54 days for the second. The results also point out that the fouling resistance was mainly reversible for both runs such as the contribution of the resistance due to suspended solids (Rg) to the total resistance was equal to 60 and 74% for run I and II, respectively. Moreover, the contribution of adsorption resistance (Rads), decreased from 12.7 to 7.19 %. This leads to a great reduction in the cost of membrane chemical cleaning.

Keywords: MBR; Fouling; Membrane; Resistance; COD/N ratio

1. Introduction

The activated sludge process is commonly used in wastewater treatment for the removal of organic compounds. Conventional activated sludge processes usually consist of an aeration tank and a clarifier. The aeration tank is the place where the organic breakdown and the micro-organisms growth take place and the clarifier is for the separation of the activated

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sludge from the decanted clear supernatant. The effluent quality from the clarifier is, however, susceptible to large fluctuations as a result of complications arising from abnormal microbial activities, such as bulking and foaming occurring in the aeration tank [1], which makes the separation (settling) of the sludge from water difficult. This has promoted membrane filtration to be increasingly used in the activated sludge process. With a membrane filtration in the unit replacing the clarifier, the rate of sludge effluent separation is no longer limited by the settleability of the activated sludge and thus a higher and more consistent effluent quality can be obtained [2]. Due to their noticeable advantages in terms of high quality, low sludge production, and very compact plant size, membrane bioreactors (MBRs) are gaining popularity as a promising technology for municipal wastewater treatment. A recent review indicated the market value to be around $216.6 million in 2005, and rising at an average annual growth rate of 10.9% significantly faster than the larger market for advanced wastewater treatment equipment and more rapid than the markets for other types of membrane systems. Despite the numerous advantages of MBR over conventional treatment technologies, membrane fouling is still considered very problematic because of the higher operational costs associated with routine membrane cleaning [3,4]. Therefore, for a more efficient use of MBRs, problems associated with membrane fouling need to be addressed. Different fouling mechanisms, such as macro-molecule adsorption, pore plugging, and cake build-up, can take place at a membrane surface. To date, the mono-disperse particles filtration fouling mechanism is relatively clear and can be predicted from the first empirical models [5]. However, the particle matrix within the submerged MBR liquor is the biological flocs formed by large range of living micro-organisms along with soluble and colloidal compound. The bulk biomass (biological floc) physiological characteristics, such as concentrations of mixed liquor (TSS), extra polymeric substances (EPS), and possible inorganic mineral, change according to the MBR operating conditions; as a result, fouling in submerged MBR processes becomes very difficult to control and identify.

A new technology has been implemented. It consisted of the MBR-placed downstream to a physico-chemical step to extract organic carbon from wastewater. The nitrogen removal is the main biological activity in the MBR. If the nitrogen removal is limited to the nitrification, the biomass present in the reactor should be mainly composed of autotrophic species. Then, because of its specificity, the autotrophic biomass can present lower potential of membrane fouling, low sludge production; thus, the MBR can be easier to control in terms of membrane fouling and operational cost linked to air process and air membrane. The wastewater feeding the MBR is not conventional: a lower COD/N ratio can alter the biomass composition and growth favoring nitrifying microorganisms, and facilitating control of fouling owing to their specific characteristics.

In this paper, a fouling behavior of the MBR was investigated under two different low COD/N ratios equal to 2.3 and 0, which enhances the growth of nitrifying micro-organisms.

2. Materials and methods

The experimental setup consists of a 30 L working volume aerobic reactor equipped with a continuous pH controller and a 0.8 L submerged hollow fiber membrane module (0.05 μm pore size and 0.2 m^2 of surface area) (Fig. 1). Due to the high aeration rate, the reactor was considered perfectly mixed as well as the membrane module. The concentrated synthetic feed solution, the diluting water, and the permeate were injected or extracted by peristaltic pumps. The permeate pressure was recorded with sensors connected to a computer through a Labview Program which enabled to monitor the trans-membrane pressure (TMP). Aeration was continuously provided through membrane diffusers at the bottom of the reactor and just below the fibers in the membrane module.

2.1. Biological conditions

Two successive series of experiments A and B were carried out under the operational conditions listed in Table 1. At the beginning of each run, the reactor was filled with sludge inoculums from a domestic wastewater plant operated with low organic loading rate (OLR) ($<0.1\,kg\,COD\,kg^{-1}\,VSS\,d^{-1}$). The reactor was then fed with a synthetic solution containing ammonium chloride (NH_4Cl), di-ammonium hydrogen phosphate ($(NH_4)_2HPO_4$), and sodium acetate (CH_3COONa) as the sole organic compound. The other elements (Mg^{2+}, K^+, etc.) were supplied by tap water used as diluents. A sodium carbonate (Na_2CO_3) was added to the reactor to provide the alkalinity required for nitrification reaction.

While much of the existing experiments was performed under constant pressure conditions, the use of constant flux and monitoring of the resultant TMP rise have proved to be particularly useful in the context of monitoring fouling in complex fluids and are currently the mode of choice in the present study.

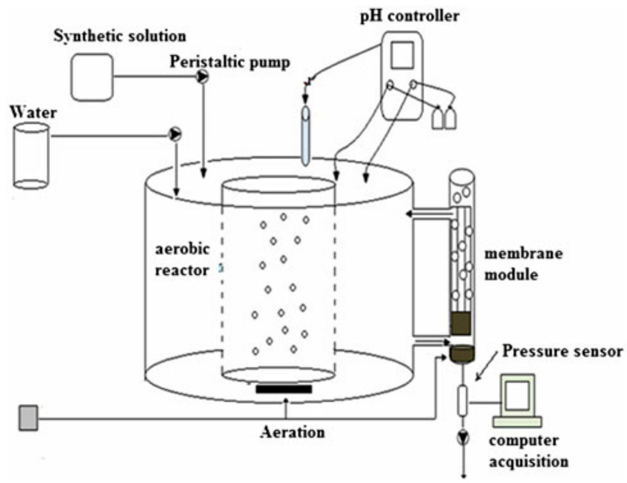

Fig. 1. Experimental unit.

2.2. Assessment of autotrophic population development

The development of autotrophic population is demonstrated by the respirometric tool. Indeed, respirometry was frequently used to quantify biological activity [6]; it was carried out to measure the mass of oxygen consumed with time by the microorganisms called oxygen uptake rate (OUR). The respirometric measurement was carried out on a 250 mL vessel; samples were aerated during 24 h without any addition of substrate enabling to reach the endogenous regime where both autotrophic and heterotrophic cells respire. Moreover, specific experiments were also carried out after adding a solution of 10 mg L^{-1} allythiourea (ATU) and sodium chlorate ($NaClO_3$) to inhibit the Nitrosomonas and Nitrobacter bacteria, respectively [6] which enabled to differentiate the OUR by autotrophic and heterotrophic populations.

Fig. 2 illustrates a test after the addition of ATU and $NaClO_3$. During the endogenous phase (A), both autotrophic and heterotrophic organisms consumed oxygen and we measured the total endogenous OUR (OUR_{endt}) while the inhibition of autotrophic led to a decrease in OUR (phase B). The difference of slopes between phases A and B enables to differentiate between autotrophic and heterotrophic OUR noted (OUR_{endaut}) and (OUR_{endhet}), respectively. The percentage of Activity autotrophic and heterotrophic is given by Eq. (1):

$$\% \text{ Activity Aut/het} = \frac{OUR_{endt} - OUR_{endhet}}{OUR_{endt}} = 1 - \frac{OUR_{enhet}}{OUR_{endt}} \quad (1)$$

Table 1
Operational conditions (pH = 7, HRT = 0.625 d)

Parameter	Run I	Run II
T(°K)	293	293–297
Duration of experiments (d)	51	54
N (mgN L^{-1})	80	125
COD (mg L^{-1})	188	0
OLR (kgCOD $m^{-3}d^{-1}$)	0.3	0
NLR (kgN $m^{-3}d^{-1}$)	0.12	0.2
COD/N ratio	2.3	0

2.3. Suspension filterability and membrane fouling

2.3.1. Molhman Index

This indicator is generally used to characterize the ability of sludge-settling if they separate more quickly from water. The Molhman Index (MI) is the sludge volume after 30 min settling (V_{30}) reported to the TSS concentration mL g^{-1}TSS given by Eq. (2).

$$MI = \frac{V_{30}}{[TSS]} \quad (2)$$

Lee et al. [7] evaluated and characterized the sludge settleability by as (i) excellent when the MI < 80 mL g^{-1} TSS; (ii) moderate if 80 < MI < 150; and (iii) bad when MI > 150 mL g^{-1} TSS.

2.3.2. Specific resistance filtration

The filterability of sludge was measured using the specific resistance filtration (SRF) αC (m^{-2}) obtained

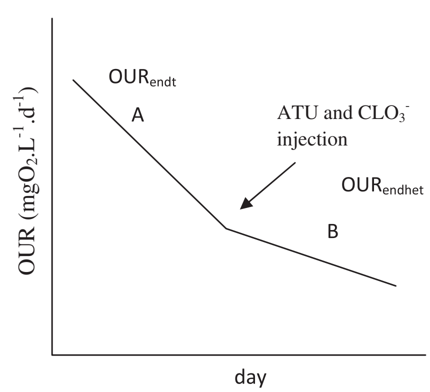

Fig. 2. Respirometric tests obtained after punctual addition of ATU and ClO_3^-inhibitor.

from experiments at constant pressure or at constant flux using the cake filtration model. For filtration at constant pressure, SRF is obtained as given in Eq. (3) [8]:

$$\frac{t}{V} = \frac{\mu\alpha C}{2\mathrm{TMPS}^2} V + \frac{\mu R_\mathrm{m}}{\mathrm{TMPS}} \quad (3)$$

The experiments were carried out in a Sartorius cell in dead-end filtration. We used a low pressure (50 kPa) in order to have filtration conditions close to those of the MBR [9]. When plotting t/V vs. V, a straight line whose slope enabled to calculate the SRF was obtained.

S is the membranes area (m^{-2}); μ is the viscosity (Pas), TMP (Pa); and C is the quantity of the accumulated matter on the membrane per volume of filtrated water ($\mathrm{kg\,m}^{-3}$).

2.3.3. Fouling rate

During the investigation fouling development on the membrane, characterized by monitoring the permeate pressure, we were allowed through the calculation of TMP (Eq. (4)), to determine the fouling rate which is expressed as $\mathrm{dTMP}/\mathrm{d}t$ or $\mathrm{d}R/\mathrm{d}t$ (Eq. (4)).

$$\mathrm{TMP} = P_\mathrm{atm} - P_\mathrm{permeat} \quad (4)$$

$$\frac{\mathrm{d}R}{\mathrm{d}t} = \frac{1}{\mu \cdot J}\frac{\mathrm{dTMP}}{\mathrm{d}t} \quad (5)$$

J is the permeate flux $\mathrm{m^3\,m^{-2}\,s^{-1}}$.

2.4. Fouling mechanism characterization

The resistance in series model was applied to investigate the fouling characteristics of the activated sludge in terms of various filtration resistances (Fig. 3). The permeate flux declined due to the following types of filtration resistances over time described by Eqs. (6) and (7):

$$J = \frac{\mathrm{TMP}}{\mu R_\mathrm{t}} \quad (6)$$

$$R_\mathrm{t} = R_\mathrm{m} + R_\mathrm{bio} + R_\mathrm{g} + R_\mathrm{ads} \quad (7)$$

where Rt is the total filtration resistance (m^{-1}), R_g is the resistance sludge accumulation on the surface of membrane, R_bio is the gel layer resistance (m^{-1}), $Rads$ is the pore-clogging resistance (m^{-1}), and Rm is the intrinsic membrane resistance (m^{-1}).

2.5. Procedure of membrane cleaning

The filtration resistance at each step was determined as follows (Fig. 4) [10]:

(1) At the end of each run, the filtration cell was isolated from the bioreactor. The total resistance was determined by measuring the pressure at different flow values imposed ($R_\mathrm{t} = R_\mathrm{m} + R_\mathrm{bio} + R_\mathrm{g} + R_\mathrm{ads}$);
(2) The membrane was washed with water to remove the accumulated sludge on the surface, and then as mentioned above, a pressure measurement at different flow imposed allowed calculating the new resistance ($R_1 = R_\mathrm{m} + R_\mathrm{bio} + R_\mathrm{ads}$);
(3) The membrane was wiped to remove a possible biofilm layer on the surface ($R_2 = R_\mathrm{m} + R_\mathrm{ads}$); and

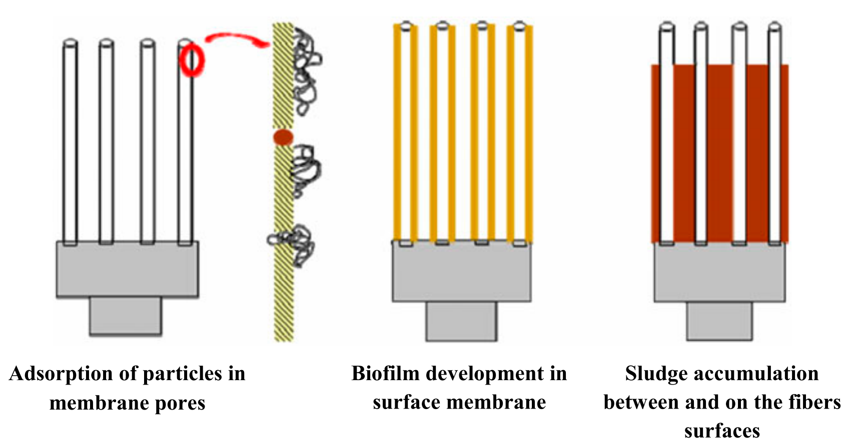

Fig. 3. Fouling mechanisms development in hollow fiber membrane.

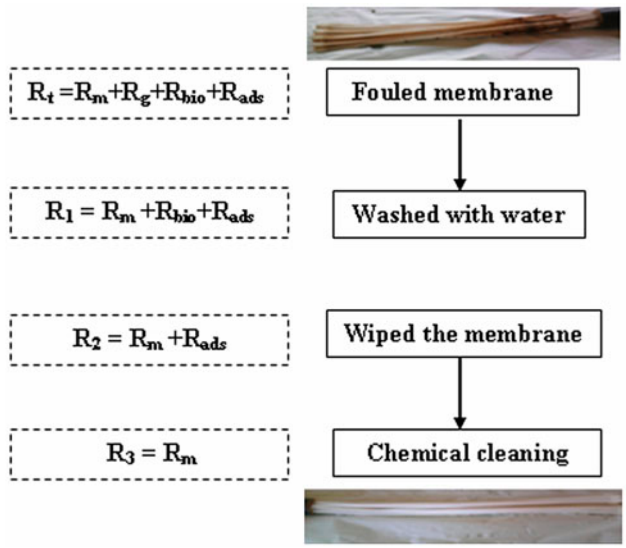

Fig. 4. Procedure of membrane cleaning.

(4) A final chemical cleaning was performed to completely regenerate the intrinsic resistance membrane ($R_3 = R_m$); $R_g = R_t - R_1$; $R_{bio} = R_1 - R_2$; and $R_{ads} = R_2 - R_m$.

3. Results and discussion

3.1. Assessment of the autotrophic population development

Fig. 5 illustrates the respirometric results obtained for runs I and II.

Fig. 5 presents the evolution of the autotrophic endogenous respirometric and the activity Aut/het percentage during runs A and B. Results show that the percentage of autotrophic bacteria activity increased from 16 to 42% for run A and reached 82% at the end of run B. This observation confirms the MBR evolution from a heterotrophic MBR toward an autotrophic MBR. These evolutions can be explained by (i) the decreasing COD/N-NH_4^+ ratio in influent passing from an urban wastewater treatment plant (COD/N–NH_4^+ = 10) at day 0 to the synthetic influent (with COD/N–NH_4^+ = 2.3 during run A and 0 during run B) and (ii) by the fact that no sludge extraction was practiced to improve autotrophic accumulation.

3.2. SRF and MI evolutions

Fig. 6 shows the evolution of SRF and MI evolution during the two experiments.

The SRF values were found in the range of 3×10^{13}–$10^{14}\,m^{-2}$. These values are still lower than other research those whose values ranged between 1.2×10^{14} and $10^{15}\,m^{-2}$ [11,12]. The MI values decreased from the first to the second run accampagned with a little decrease in SRF, and the MI values ranged between 80 and 100 for the second run which demostrates a good sludge filtrability. This confirms the interest to promote autotrophic development in order to improve the suspension filterability.

3.3. Evolution of TMP

The variations of TMP with operation time for the two runs at the flux of $10\,L\,m^{-2}\,h^{-1}$ are shown in Fig. 7.

It can be observed that the TMP was constant in the beginning of each experiment, this period was equal to 10 and 18 days to runs I and II, respectively. There was little difference in the fouling rate between the two experiments. The TMP increased and both runs showed a sharp fouling growth rate as the

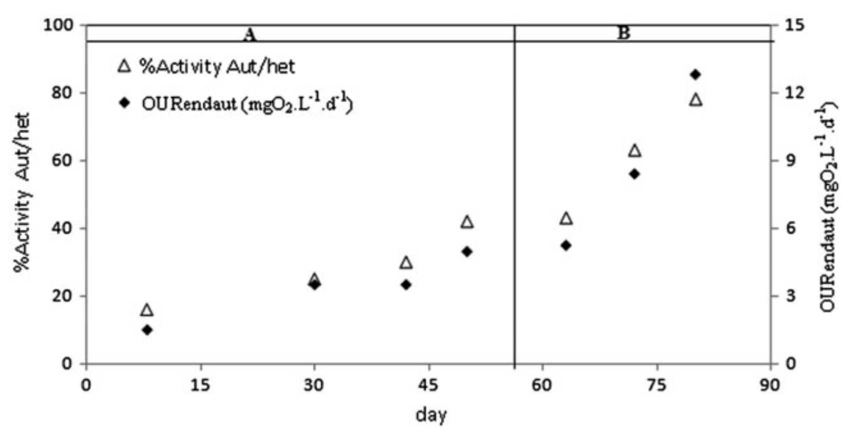

Fig. 5. Evolution of Aut/het activity and OUR_{endaut} during experiment.

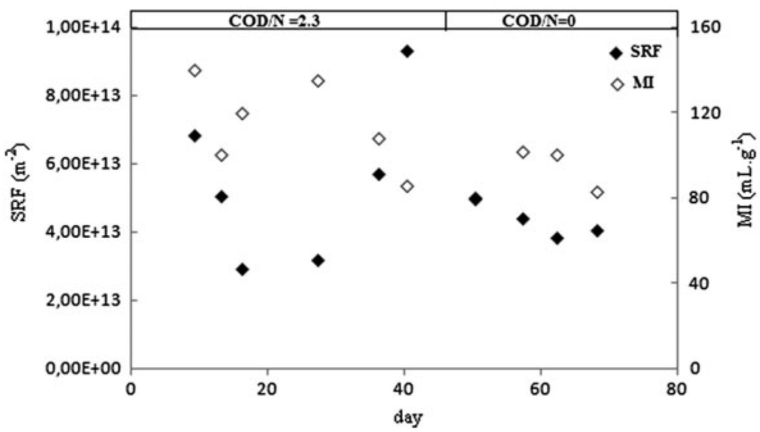

Fig. 6. Evolution of SRF and MI during the investigation.

membrane flux was kept at about $10\,\mathrm{L\,m^{-2}\,h^{-1}}$ during the experiments. The fouling rate calculated according to Eq. (5) had an average value equal to 0.064 and 0.015 E + 12 ($\mathrm{m^{-1}\,d^{-1}}$) for runs I and II, respectively. Moreover, the two runs have a low fouling rate compared to the conventional system working under high COD/N ratio. The values of the fouling rate and the cleaning frequency of previous researches taken from the literature are given in Table 2.

The fouling rate was lower than that obtained on other studies operating at high OLR [11,15]. Table 2 demonstrates that the experiments were conducted continuously without membrane cleaning (once/51 d and once/54 d) compared to other studies, this has a great influence on reducing the economic cost of membrane regeneration.

3.4. Identification of fouling mechanism

The cleaning membrane procedure developed in section (1.5) was practiced at the end of each run. The value of each resistance is given in Fig. 8.

The results point out that the fouling resistance was mainly reversible for runs I and II. Indeed, R_g was the dominant resistance with a contribution of 60 and 74% to the total resistance for runs I and II, respectively, and the irreversible fouling caused by the adsorption decreased from 7.32E + 11 to 2.16E + 11, meaning a decrease from 12.7 to 7.19% on contribution to the total resistance when the COD/N ratio decreased. Therefore, a great reduction in the cost of the chemical regeneration of the membrane can be reached.

4. Conclusion

During this study, the effect of COD/N ratio was discussed. It was shown that the reduction in the COD/N (2.3 and 0) ratio led to a reduction in the irreversible fouling compared to the conventional MBR where the ratio was more than 6. Indeed, the fouling rate was lower than depicted on other research. The R_g resistance of sludge accumulated on membrane surface was the most dominant, with

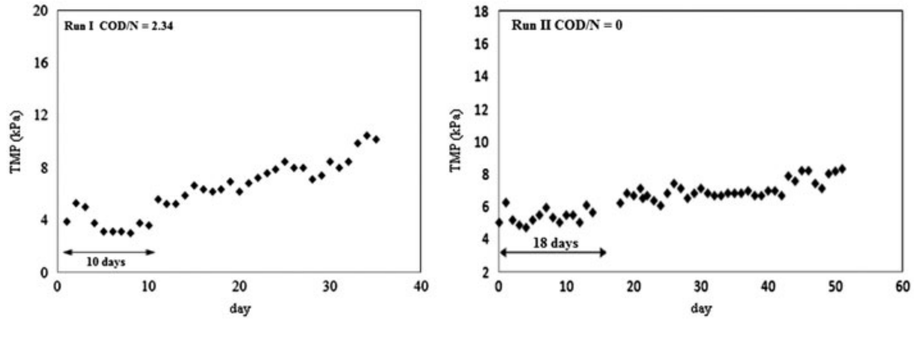

Fig. 7. TMP evolutions during the investigation for the two runs.

Table 2
Fouling rate for periods I and II and the cleaning membrane frequency in comparison with other investigation

References	Conditions	Cleaning membrane frequency	dR/dt (md)$^{-1}$E + 12
[13]	Membrane flat sheet, 0.2 μm, 0.09–0.186 kgCOD/kg^{-1}TSS d^{-1}	Not mentioned	0.161
[14]	Hollow fiber membrane (HF), 0.18 kg COD kg TSS^{-1} d^{-1}	One tine /15 d	2.56
[11]	HF, 0.5 kg COD kg TSS^{-1} d^{-1}	One time/7 d	931
[15]	HF, 1.7 kg COD kg TSS^{-1} d^{-1}	One tine /15 d One tine/30 d	0.25
This study	HF, 0.1 kg COD TSS^{-1} d^{-1}	Run I: One time /51 d Run II: One time /54 d	0.064–0.015

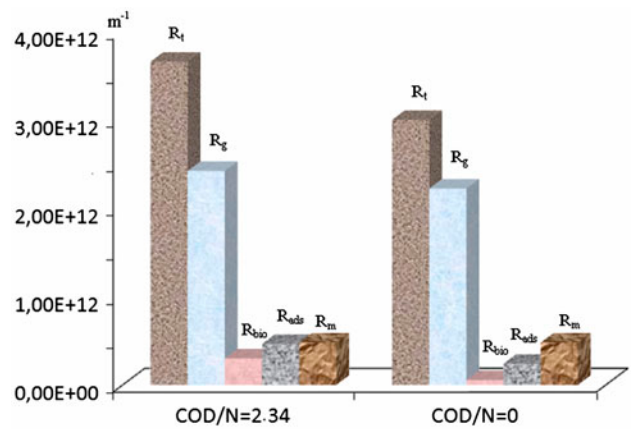

Fig. 8. Values of resistances in function of COD/N ratio.

a contribution more than 60% to the total resistance. R_{ads} was reduced from 7.32E + 11 to 2.16E + 11 and the sludge settleability was improved by the decrease in COD/N ratio.

We can conclude that this new technology of wastewater treatment, characterized by a low COD/N ratio in order to promote the autotrophic bacteria development, can lead to reduce the membrane fouling propensity and cleaning frequency.

List of symbols

α	—	specific cake resistance (m kg^{-1})
μ	—	the viscosity (Pas)
C	—	quantity of the accumulated matter on the membrane per volume of filtrated water (kg m^{-3})
COD	—	chemical oxygen demand (mg L^{-1})
J	—	permeate flux (m^3 m^{-2} s^{-1})
MI	—	Molhman Index (mL g^{-1}TSS)
NLR	—	nitrogen loading rate (kg N m^{-3} d^{-1})
R_m	—	resistance of membrane (m d)$^{-1}$
$OUR_{en}dt$	—	total endogenous oxygen uptake rate (mgO$_2$ L^{-1} d^{-1})
OUR_{endhet}	—	heterotrophic endogenous oxygen uptake rate (mgO$_2$ L^{-1} d^{-1})
OLR	—	organic loading rate (kg DCO m^{-3} d^{-1})
R_t	—	total resistance (m d)$^{-1}$
R_{bio}	—	resistance of biofilm (m d)$^{-1}$
R_g	—	resistance sludge accumulation (m d)$^{-1}$
R_{ads}	—	resistance adsorption (m d)$^{-1}$
SRF	—	specific resistance filtration (m^{-2})
S	—	membranes area (m^{-2})
TSS	—	total suspended solids (g L^{-1})
TMP	—	trans-membrane pressure (Pa)
VSS	—	volatile suspended solids (g L^{-1})
V_{30}	—	sludge volume after 30 min settling (mL)

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