Fouling membrane in an anaerobic membrane bioreactor treating municipal wastewater treatment

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Abstract
An anaerobic membrane reactor treating municipal wastewater was evaluated. The experiments were performed using a pilot-scale UASB reactor with a submerged tubular ultrafiltration membrane operated with a hydraulic retention time (HRT) of 8 hours. The system was operated under intermittent filtration mode (4 min on/1 min off) with and without nitrogen gas bubbling during the relaxation time (IF4NP and IF4P, respectively). Removal ratios in AnMBR of 68.6% and 87.9% for IF4P and IF4NP 79.5% were founded. Nitrogen bubbling improved the filtration performance. The elapsed time to reach 40 kPa for conditions IF4NP and IF4P were 443 and 108 hours, respectively. According with results, the intermittent filtration combined with nitrogen bubbling during the period of relaxation was an effective operation strategy in order to minimize membrane fouling.

Keywords
UASB; gas bubbling; ultrafiltration; wastewater treatment.

INTRODUCTION

In warm climates countries, up-flow anaerobic sludge blanket (UASB) reactors have been used for treating municipal wastewaters at environmental temperature. However, the BOD removal efficiencies reaching up to 70% (Chernicharo, Van Lier, et al., 2015) and biomass can be loss in the effluent. To improve the removal efficiencies, anaerobic membrane bioreactors (AnMBR) have been used recently for wastewater treatment and provide an effluent with lower COD, pathogens and suspended solids concentration compared with conventional UASB treatment meeting the discharge standards (Stuckey, 2012).

Although the use of AnMBR treating municipal wastewater at real scale has been limited, in the last decade research has increased especially with submerged membranes. Different membranes configuration has been used in submerged AnMBR: flat sheet (Hu and Stuckey, 2006), hollow fiber (Giménez, Robles, et al., 2011; Wen, Huang, et al., 1999), and tubular modules (Jeison and van Lier, 2006; van Voorthuizen, Zwijnenburg, et al., 2008). In addition, these membranes can be used directly immersed into the bioreactor or immersed in a separate membrane tank (Liao, Kraemer, et al., 2006).

Liao et al. (2006) reported that AnMBR used completely stirred tank reactor (67%), anaerobic filters (15%), UASB reactors (10%), fluidized bed (7%) and septic tanks (2%). Wen et al. (1999) investigated an UASB reactor with a submerged membrane treating domestic wastewater. The results shown COD removal above 88% with an effluent concentration below 20 mg·L^{-1} and without suspended solids. Lin et al. (2009) evaluated two systems with a submerged membrane into a UASB reactor to determinate the properties of biomass and its effects on membrane fouling.

Despite the use of submerged AnMBR treating municipal wastewater show many advantages, there are drawbacks such as low flux, membrane fouling, high capital and operational costs that limit the extensive use of AnMBRs (Ozgun, Dereli, et al., 2013). According with Stuckey (2012), membrane fouling is caused by a combination of components in the reactor (soluble organics, colloidal particles from the feed and cell lysis, and inorganic precipitates), and these in turn are influenced by parameters such as the composition of the biological system, membrane type, hydrodynamic conditions, reactor operating conditions and the chemical system. Soluble organics like soluble microbial products (SMP) and extracellular polymeric substances (EPS) may reduce the membrane porosity. Significant fouling by EPS and an unequal distribution of these were found in fouled membranes suggesting that the initial and gradual increase of TMP in filtration trials is due to the deposition of the SPE in the early hours (Cho and Fane, 2002). SMP size and concentration is an adverse factor to the permeate flux because it significantly contributes to the membrane fouling (Liang, Liu, et al., 2007). On the other hand, the operation mode of AnMBR is another important factor affecting fouling membrane. Intermittent filtration with backwashing or bubbling during relaxation time are used as strategies to decrease fouling. During the relaxation time, efficient cake layer removal from the membrane surface can be achieved (Cerón-Vivas, Morgan-Sagastume, et al., 2012).

The aim of this study was to understand membrane fouling in an UASB with a submerged ultrafiltration membrane treating municipal wastewater at environmental temperature.

MATERIAL AND METHODS

Experimental set-up

The experiments were performed in an up-flow anaerobic sludge blanket reactor (UASB) fed with municipal wastewater. The pilot UASB reactor located in the wastewater treatment plant of the National University of Mexico (UNAM) Campus was a cylindrical PVC column with 0.50 m internal diameter and 3.6 m working liquid depth (V= 0.7 m^3) operated at a hydraulic retention time (HRT) of 8 h. The reactor was inoculated with granular sludge coming from an UASB treating municipal wastewater. An ultrafiltration tubular membrane module (0.2375 m^2, diameter of 9 mm, polyvinylidene fluoride, PVDF, 100 kDa as molecular cut-off manufactured by MEMOS GmbH, Germany) was immersed on the upper section of UASB reactor. The UASB reactor was maintained at environmental temperature.

Real domestic wastewater was pumped into the UASB reactor using a peristaltic pump (Masterflex 77410-10, 0.35 HP, Cole-Parmer, USA). The suction was performed by a peristaltic pump (Masterflex 7553-80, USA), which controlled the permeate flux through speed adjustment. The permeate flux was collected and storage for further analysis. The transmembrane pressure (TMP) was recorded every 30 seconds by a pressure transducer (PT) (OMEGA PX319) located in the permeate line. Analog signals were processed by a data acquisition card (DAC) connected to a computer (PC) using a Lab-View application. The reactor set-up is shown in Figure 1. The system was operated under intermittent filtration mode (4 min on/1 min off) with and without nitrogen gas bubbling (0.75 L· min-1) during the relaxation time (IF4NP and IF4P, respectively). Tests were conducted until TMP reached 40 kPa.

Analytical methods

Samples of raw wastewater, UASB effluent and permeate were taken daily. Chemical oxygen demand

(COD), total and volatile suspended solids (TSS, VSS) and pH analysis were made using Standard Methods (APHA, AWWA, et al., 2012). Soluble microbial products (SMP) were obtained by filtration through a 0.45 µm filter (Nitrocellulose, Millipore, USA). Extracellular polymeric substances (EPS) were extracted using the heating method, following a procedure similar to the EPS extraction method used of Zhang et al. (1999) and Ng et al.(2010) The same filter used for SMP determination was introduced in an Erlenmeyer flask with an equivalent sample volume of MilliQ water and heated at 80°C for 10 min and filtered again through another 0.45 µm filter. The filtrate collected was the EPS sample used in the study. Soluble microbial products (SMP) and extracellular polymeric substances (EPS) were determined as total organic carbon (TOC). TOC was measured using a TOC analyser (Analytic Jena Multi N/C 2100). Particle size distribution was measured with Mastersizer 2000. Samples were analyzed in duplicate. SEM imaging was performed on fouled membranes in a JEOL JSM-7600F SEM. The details of sample preparation and operation procedures can be found in our previous study (Cerón-Vivas, 2014)

Figure 1. Submerged anaerobic membrane bioreactor

Fouling rate

Membrane resistance was calculated by Darcy`s law (Eq. 1).

$$Rt = Rm + Rf = \frac{\Delta P}{\mu J} \qquad (1)$$

Where R_t is the total hydraulic resistance (m^{-1}), ΔP is the TMP (Pa), μ is the viscosity of permeate (Pa·s) and J is the permeate flux ($m^3 \cdot m^{-2} \cdot s^{-1}$). R_m and R_f are membrane and fouling resistances (m^{-1}) respectively. The fouling rate was calculated as the slope of total hydraulic resistance over time. A new membrane module was used for each experimental condition. Prior to filtration runs, R_m was determined by measuring the water flux with desionized water so R_t was evaluated when the test finished.

The specific cake resistance, α ($m \cdot kg^{-1}$) to constant flux were calculated from equations 2 and 3 derived from Darcy's law.

$$\Delta P_L = \Delta P_C + \Delta P_M \tag{2}$$

$$\Delta P_L = \alpha\mu C_s \frac{Q^2}{A_M^2} t + \mu R_m \frac{Q}{A_M} \tag{3}$$

Where ΔP_L is the TMP (Pa), μ the dynamic viscosity (Pa·s), C_s the suspended solid concentration (kg·m^{-3}), Q is the flow (m^3·s^{-1}), A_M is the membrane area (m^2), t is time (s) and R_m is the membrane resistance (m^{-1}) (Kovalsky, Bushell, et al., 2009). In this equation the two terms represent the sum of the pressure drop through fouling cake (ΔP_c) and the TMP through the membrane (ΔP_M) being the last constant. A plot of ΔP_c versus t should yield a straight line with slope equal to α.

RESULTS AND DISCUSSION

AnMBR Performance

Table 1 shows the performance of the pilot-scale AnMBR at different operating conditions. The temperature varied between 18^{o}C and 21 ^{o}C and pH was always kept near neutrality. The COD removal in UASB reactor was higher in IF4NP test and the AnMBR had the same behavior (68.6% and 87.9% for IF4P and IF4NP, respectively). The results are according with other authors who have found removal efficiencies between 80 and 90% in pilot-scale AnMBR treating municipal wastewater (Giménez, Robles, et al., 2011; Salazar-Peláez, Morgan-Sagastume, et al., 2011; An, Wu, et al., 2010). Despite removal in IF4NP test was higher than IF4P, considering only the membrane unit around 40% of the COD coming from the anaerobic reactor was removed in both tests, indicating retention of soluble and particulate material on the membrane. Raw wastewater had high solids suspended concentration but an important fraction was removed the UASB reactor. Membrane retained solids from the upper section of the UASB but permeate present some TSS due to the growth of a biofilm on the pipe walls.

Table 1. AnMBR performance

Parameter	IF4P			IF4NP		
	Influent	Effluent	Permeate	Influent	Effluent	Permeate
Temperature (^{o}C)	20.0 ± 1.0	19.0 ± 1.0	18.8 ± 1.1	19.6 ± 1.0	18.6 ± 0.8	18.1 ± 2.2
pH (units)	7.9 ± 0.3	7.7 ± 0.2	8.0 ± 0.2	7.8 ± 0.3	7.4 ± 0.1	7.6 ± 0.2
COD (mg O_2·L^{-1})	525 ± 174	222 ± 61	150 ± 33	657 ± 235	130 ± 55	78 ± 35
TSS (mg·L^{-1})	515 ± 472	70 ± 12	16 ± 6	1307 ± 729	47 ± 9	15 ± 5
VSS (mg·L^{-1})	208 ± 52	69 ± 10	16 ± 6	737 ± 311	47 ± 9	14 ± 4

Membrane fouling

Permeate flux were stable along filtration runs (2.5 L·m^{-2}·h^{-1}). However, the permeate pump speed had to be frequently adjusted in order to maintain a constant flux. The R_m values for clean membranes were (E12 m^{-1}) 1.31 and 1.97 for conditions IF4P to IF4NP, respectively. Figure 2 show the Rf for the tested conditions. The TMP and the fouling resistance increased more rapidly in condition IF4P while condition IF4NP had a longer filtration time (108 vs. 443 hours) indicating that relaxation with nitrogen bubbling improved the filtration performance lowering the TMP at constant flux. Similar results were obtained in previous studies at bench-scale with synthetic wastewater (Cerón-Vivas, Morgan-Sagastume, et al., 2012). These results agree with those found by Fawehinmi et al. (2007) who reported a linear relationship between the fouling rate and gas bubbling.

The fouling rate of the membrane used in an AnMBR decreased when the bubbling gas was used. Figure 3 shows the results of specific resistance (α) and fouling rate (FR). According with Tiller *et at*. (1987), the initial structure of the cake layer under zero stress during the filtration depends mainly on size, form and aggregation state; furthermore, when the deposited particles over the membrane increase, the cake layer can be compressed increasing the value of α (Chellam y Xu, 2006).

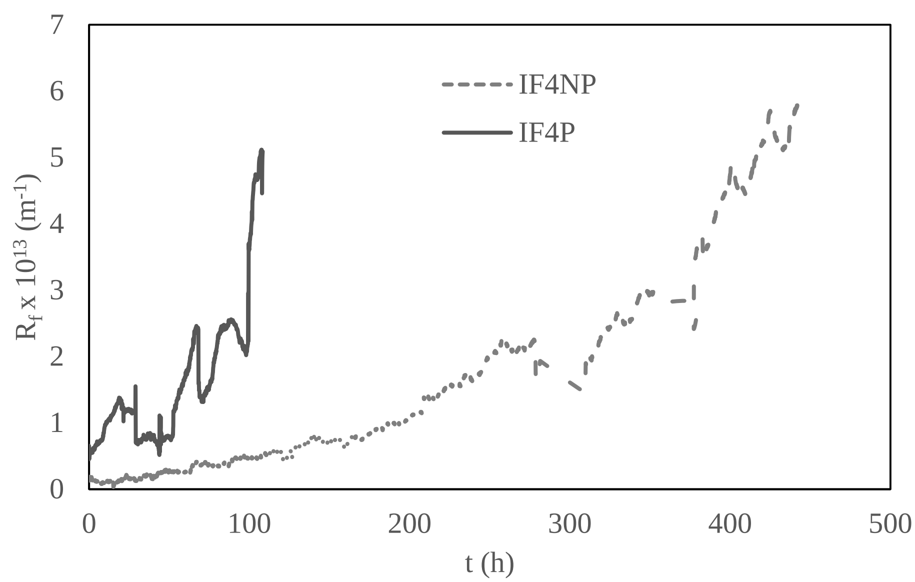

Figure 2. Fouling resistance for IF4P and IF4NP

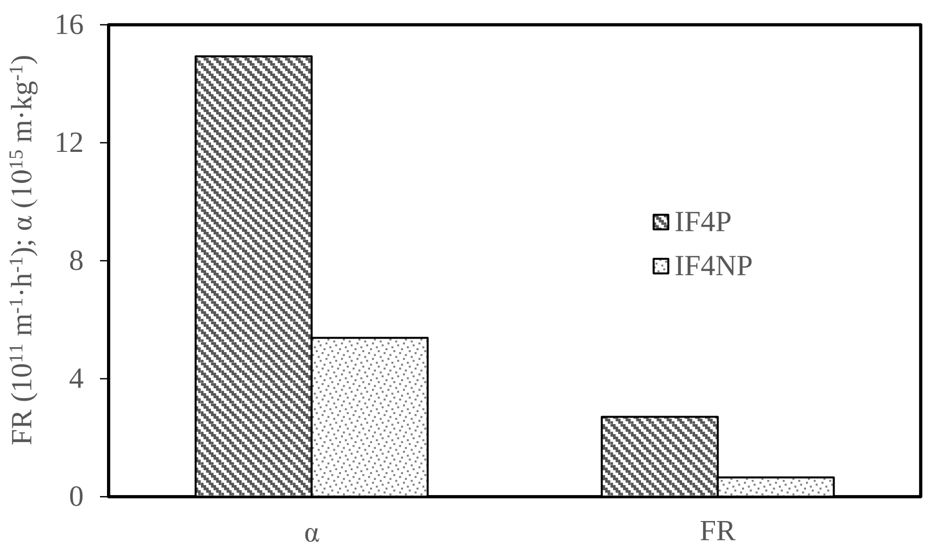

Figure 3. Specific resistance and fouling rate

Figures 4 and 5 show the morphology of the fouled membrane surface for IF4P and IF4NP. It can be seen the formation of a cake layer on the membrane surface. In the magnified images different microorganisms and inorganic compounds can be seen.

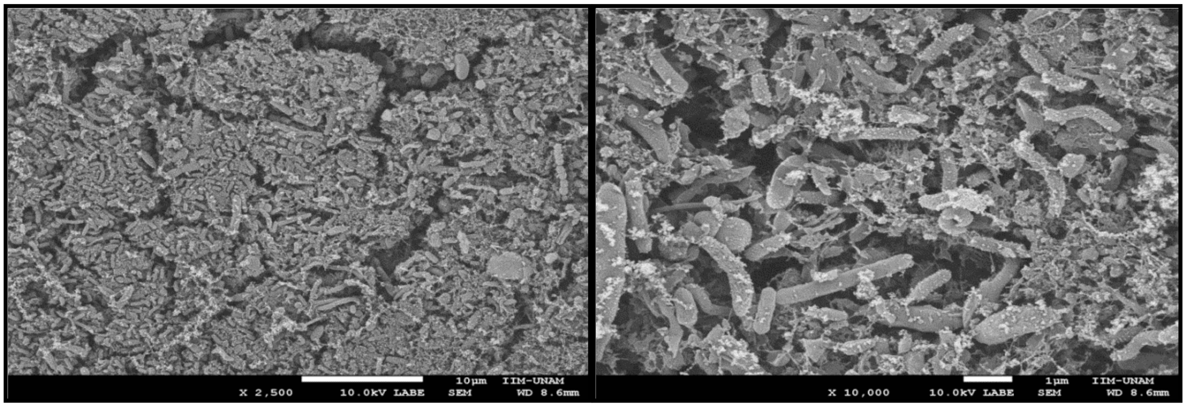

Figure 4. SEM image of the fouled membrane surface for IF4P

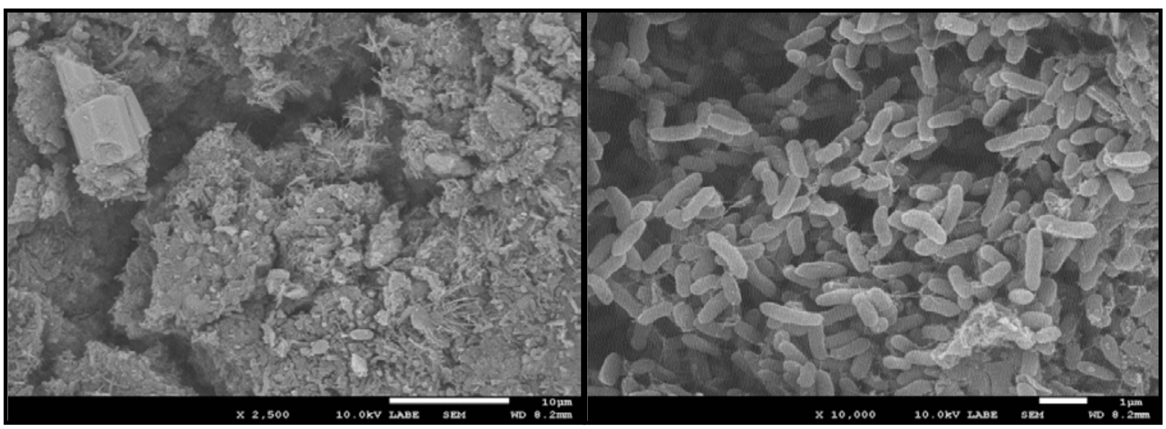

Figure 5. SEM image of the fouled membrane surface for IF4NP

Foulant substances

Figure 6 shows the SMP and EPS concentration in the UASB effluent. SMP concentrations in UASB effluent (SMPe) and permeate were higher than EPSe. However, the membrane retained more EPS than SMP (52.6% for EPS vs 17.4% for SMP) indicating that it acts as a barrier to these compounds causing accumulation on the surface of the membrane (Le-Clech, Chen, et al., 2006). According with the results, there were clear statistically significant differences between the values of SMPe for IF4P and IF4NP conditions which can be affected by the turbulence generated by bubbling nitrogen. The EPS have no statistically significant differences between the tests, supporting that the SMP are major contributors in the fouling membrane. SMP and EPS concentration obtained in this study were higher than those reported for bench-scale (Cerón-Vivas, Morgan-Sagastume, et al., 2012) due to washout of soluble material that can be produced from the sludge bed because the upflow velocity in the UASB reactor was higher (0.5 $m \cdot h^{-1}$ in the pilot UASB vs. 0.1 for bench-scale). According with Ozgun et al. (2013) the upflow velocity in membrane coupled UASB reactors seem to be the critical parameters determining the efficiency and effluent fouling propensity due to the efficiency of solids entrapment determines the amount and properties of solids leaving the UASB with the effluent.

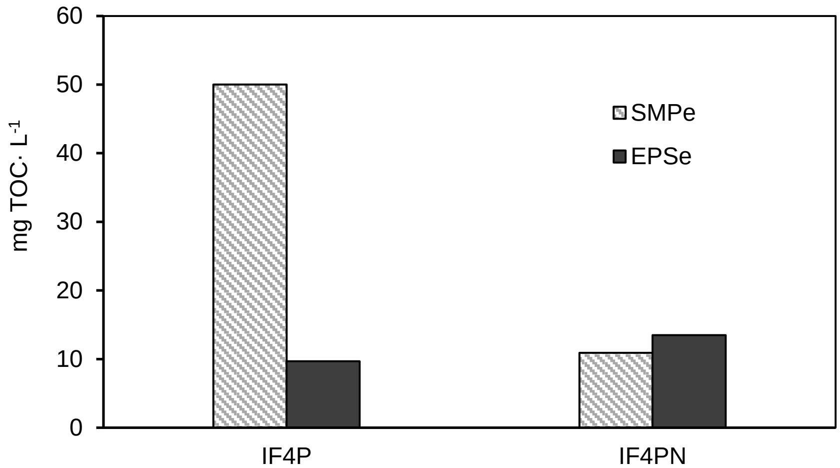

Figure 6. SMP and EPS in UASB reactor

The particle size distribution in the UASB effluent for IF4P and IF4NP were 40 µm and 50 µm respectively, indicating the presence of large aggregate, which are important in the formation and structure of the cake layer (Lin, Xie, et al., 2009). The values found in this study were higher than other pilot AnMBR (Martinez-Sosa, Helmreich, et al., 2011) but were according with results obtained at bench scale (Jeison and van Lier, 2006; Hu and Stuckey, 2006).

CONCLUSIONS

AnMBR achieved high COD and TSS removal efficiencies and may be considered as a suitable technology for municipal wastewater treatment. Gas bubbling in the relaxation time promoted mixing in the UASB reactor and it was an effective strategy to minimize the membrane fouling.

ACKNOWLEDGEMENTS

This research was funded by a grant from the Instituto de Ingeniería UNAM (Fondo de Proyectos Internos). The authors thank Roberto Briones, Lauro Santiago and Margarita E. Cisneros for their technical support. The first author appreciates the support given by the Universidad Pontificia Bolivariana (Colombia) and the scholarship provided by the Instituto de Ciencia y Tecnología del Distrito Federal, México.

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