Three stages MBR (methanogenic, aerobic biofilm and membrane filtration) for the treatment of low-strength wastewaters
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ABSTRACT

The use of a new three stages MBR process with a first methanogenic UASB stage, a second stage with aerobic biofilm growing on small carrier elements maintained in suspension and third stage with membrane filtration module is presented. The objective of the first methanogenic chamber is to diminish COD of the raw wastewater, producing a biogas rich in methane, and decrease the sludge production. In the second stage, the remaining soluble biodegradable COD is oxidized by heterotrophs. In the third stage, the membrane modules could be operated at higher fluxes than those reported for AnMBR systems, and similar to those obtained in aerobic MBRs. In this sense, the concept of these three stages MBR is to join the advantages of the methanogenic and aerobic membrane bioreactor processes, by reducing energy requirements for aeration, producing biogas with high methane percentage and a permeate with very low COD content. A synthetic wastewater was fed to the three stages MBR. COD in the influent was between 200 and 1,200 mg/L, ammonium ranged from 10 to 35 mg/L and phosphorous concentration was 8 mg/L. OLR in-between 1 and 3 kg COD/(m² d) and a HRT of 13–21 h were applied. Temperature was between 17.5 and 23.2 °C. During the whole operating period the COD removal efficiencies were in the range of 90 and 96% of which in between 40 and 80% was removed in the first methanogenic chamber. Biogas production with methane content between 75 and 80% was observed. With regard to membrane operation, average permeabilities around 150 L/(m² h bar) were achieved, operating with fluxes of 11–15 L/(m² h).

Key words | anaerobic, biogas production, membrane bioreactor, membrane fouling

INTRODUCTION

The application of anaerobic processes for treating diluted waste streams has received high attention in recent years. One of the reasons is that it may guarantee the process sustainability with regard to the use of the aerobic processes due to the lower energy consumption, generation of a biogas with a high methane content and diminution of biomass production. One of the most popular anaerobic systems for treating wastewaters is the Upflow Anaerobic Sludge Blanket (UASB) reactor. Due to its simplicity and compactness this technology has been proposed and applied to the treatment of various industrial wastewaters and even domestic wastewater in warm regions (Seghezzo et al. 1998). However, anaerobic effluents require additional treatment due to the presence of residual biodegradable organic matter. Moreover, they are not recommended for treating domestic wastewater in mild or cold regions due to the diminution of activity with temperature and wash out of a fraction of the anaerobic biomass with the effluent. Both effects, diminution of activity and biomass wash out, may decrease the methanogenic bioreactor capacity of treating wastewaters, especially at low temperatures. In this sense, the use of filtration membranes allows avoiding the observed loss of biomass, and could make wastewater treatment feasible even at lower temperatures (Judd 2006).

Over the last decade, the adaptation of membranes coupled with anaerobic biological processes has made membrane reactors a promising alternative to conventional wastewater treatment. Hu & Stuckey (2006) achieved 90% soluble COD (Chemical Oxygen Demand) removal efficiency at a 3 h Hydraulic Retention Time (HRT) with an
inlet concentration of 460 mg/L, using two Anaerobic Membrane Bioreactors (AnMBR) with both, flat sheet and hollow fibre modules. Ho & Sung (2010) investigated the performance of a cross-flow AnMBR treating synthetic municipal wastewater. They achieved more than 95% COD removal, with permeate concentration lower than 40 mg/L. Hu et al. (2009) proposed a bioreactor, based on the installation of hybrid aerating membrane into an anaerobic baffled reactor (HMABR). The results demonstrated that after the installation of membrane module, total Volatile Fatty Acids (VFA) and COD concentration in the HMABR effluent were decreased by 68.1 and 59.5% respectively, with increased nitrogen removal efficiency by 83.5%, at influent COD concentration of 1,600 mg/L and NH4–N concentration of 80 mg/L. This demonstrates that the AnMBR can treat low-strength wastewater with similar treatment performance as aerobic MBRs.

One of the main drawbacks of using AnMBR is related with membrane fouling and the maximum operating flux that can be achieved. Fluxes have a strong influence on both the capital and operation costs of the process. Most of the authors working with AnMBRs reported fluxes in the range of 5–15 L/(m2 h) at temperatures above 30 °C (Zhang et al. 2005; Saddoud et al. 2007; Trzcinski & Stuckey 2009). Jeison & van Lier (2006) obtained critical flux values in the range 16–23 L/(m2 h) under mesophilic (30 °C), and 5–21 L/(m2 h) under thermophilic (55 °C) conditions. In the case of domestic wastewater treated at ambient temperatures, operating fluxes are significantly lower. Lew et al. (2009) reported 11.25 L/(m2 h) at 25 °C, while Wen et al. (1999), operating a laboratory scale anaerobic bioreactor coupled with a membrane filtration worked with flux of 5 L/(m2 h). Similar results were obtained by Ho & Sung (2010), who operated with flux set on 5 L/(m2 h) and the temperature of 15 and 20 °C. Moreover, Spagni et al. (2010) demonstrated that the applicable fluxes obtained in AnMBR ranged between 2 and 5 L/(m2 h) depending strongly on operational conditions and rapid membrane fouling was usually observed. Therefore, the fluxes obtained in AnMBR are lower than those observed in aerobic MBR, that are in the range between 20 and 30 L/(m2 h) (Judd 2002; Wen et al. 2004).

In this work an alternative to overcome problems related with the operation of AnMBR (fouling) and aerobic MBR (high energy consumption and sludge production) is proposed: the use of a new three stages MBR process with a first methanogenic UASB stage, a second stage with aerobic biomass growing onto plastic support carrier and in suspension, and a third stage with the membrane modules. The objective of the first methanogenic chamber was to reduce the COD of the raw wastewater, producing a biogas rich in methane, and diminish the sludge production. In the second stage, the remaining soluble biodegradable COD was oxidised by heterotrophs. In the third stage, the membrane module was operated at higher fluxes that those reported for AnMBR systems, and similar to those referred for aerobic MBRs. In this sense, the concept of the three stages MBR was to join the advantages of the methanogenic technology and aerobic membrane bioreactor processes and make the anaerobic treatment feasible even for diluted wastewaters at low temperatures.

**MATERIALS AND METHODS**

**Bioreactor configuration and operation**

A 170 L three stage membrane bioreactor was operated, with a methanogenic UASB, an aerobic biofilm and a membrane filtration chamber connected in series (Figure 1). A 120 L volume UASB system was used for the first methanogenic stage. The effluent of the UASB reactor was led to the aerobic biofilm stage which consists of a 36 L aerobic bioreactor with 18.5 L (50 % of the effective volume) of Kaldnes K3 support. During the experiment the fraction of

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**Figure 1** | Schematic diagram of the three stage pilot-scale MBR. (1) UASB chamber, (2) Biofilm aerobic chamber, (3) Membrane chamber, (4) Feeding and recirculation, (5) Permeate (backwashing) and (6) Biogas. P1, P2, P3, P4 and P5 refer to the sampling ports.
Mixed Liquor Suspended Solids (MLSS) from aerobic chamber was recycled to the UASB stage using a peristaltic pump. Finally, the filtration stage was carried out in a 20 L aerobic chamber, where a membrane module Zenon ZW10 with a surface area of 0.9 m$^2$ was employed. This aerobic chamber, where a membrane module Zenon was aerated to minimize membrane fouling. The operation of the system was controlled by a PLC (Siemens S7-200) connected to a computer. Trans-membrane Pressure (TMP) data was measured with an analogue pressure sensor (Efector500 PN-2009) and collected in the PC via an analogue PLC module Siemens EM 235.

The UASB reactor was seeded with 50 L of anaerobic biomass (27 g VSS/L) from the Internal Circulation anaerobic reactor of a brewery industry located in Galicia (Spain), whereas 5 L of biomass from a MBR pilot plant treating urban wastewater was employed as an aerobic biomass inoculum.

The study was performed during 175 days and the operation could be divided in three different periods:

**Period I: Days 0 to 77. Start up period.** During the start-up permeate flux was fixed at 11 L/(m$^2$ h bar), and increased to 15 L/(m$^2$ h bar) after day 30. Feeding COD concentration was around 600 mg/L. The reactor was fed using synthetic wastewater composed of diluted skimmed milk, NaHCO$_3$, and trace elements. During the first operation days, some other chemicals were added to the feeding (NH$_4$Cl, Na$_2$HPO$_4$, KH$_2$PO$_4$) but were eliminated on day 40, since ammonia and phosphorous were present in the effluent of the system. From day 58 to 84, sludge purges from the sampling port P4 (Figure 1) in the UASB reactor were performed.

**Period II: Days 78 to 114.** During this period the COD concentration was increased from 600 to 900 mg/L because of the addition of methanol to the synthetic wastewater used during the Period I. The additional COD concentrations were 300 mg/L from day 78 till day 98, and 30 mg/L from day 98 onwards. Methanol was added in order to carry out micropollutants removal experiments (data not shown). During this period permeate flux varied between 12 and 15 L/(m$^2$ h bar) due to the increase of membrane fouling.

**Period III: Days 115 to 175.** During this period the COD concentration in the feeding was increased from 900 to 1,200 mg/L. The system was purged from day 141 on, because of the high MLSS concentration in the reactor. These purges took place from the sampling port P5 (Figure 1) in the UASB reactor and from membrane chamber due to the accumulation of biomass. Permeate flux varied between 12 and 15 L/(m$^2$ h bar).

**Analytical methods**

Temperature, pH, alkalinity, and concentrations of Dissolved Oxygen (DO), Volatile Suspended Solids (VSS), total and soluble Chemical Oxygen Demand (COD), Biological Oxygen Demand (BOD$_5$), ammonium, nitrite, nitrate, phosphate and total phosphorous were determined according to the Standard Methods (APHA-AWWA-WPCF 1998). The soluble COD concentration (sCOD) was measured by filtering the samples using 0.45 μm nitrocellulose membrane filters (HA, Millipore). Concentrations of total Dissolved Organic Carbon (tDOC) and total dissolved Inorganic Carbon (IC) were measured with a Shimadzu analyser (TOC-5000). Biogas composition was measured in a gas chromatograph HP 5890 Series II with the column of Porapack Q 80/100 2 m × 1/8” (SUPELCO).

With respect to the membrane operation, transmembrane pressure and permeability were measured continuously. The critical flux was determined according to the modified flux-step method proposed by van der Marel et al. (2009).

**RESULTS AND DISCUSSION**

The system was operated at ambient temperature, and wastewater temperatures changed with seasons (23–17.5 °C, which corresponds to the end of summer and winter). The pH of the effluent from UASB was around 6.7. Aerobic chamber and permeate pH varied from 6.7 to 7.7 and from 7.0 to 8.2, respectively, depending on the system performance.

**COD removal performance**

The results of 175 days of the three stages MBR operation are presented. The average Organic Loading Rate (OLR) was between 1 and 3 kg COD/(m$^3$ d), the HRT being in the range of 13 and 21 h. The total COD (tCOD) and soluble COD (sCOD) fed fluctuated from 200 to 1,000 mg/L from 150 to 870 mg/L, respectively, and the average sCOD/tCOD ratio was around 0.73. During the first 114 days of operation large variations of the tCOD fed to the reactor were observed. During this period the synthetic wastewater was...
stored at environmental temperature and the milk used was subjected to rapid degradation in the feeding tank. From day 114 on, the total COD fed did not change so much with time as the synthetic wastewater was stored in a fridge assuring that no such degradation occurred.

The difference between tCOD and sCOD concentrations of the effluent of the anaerobic chamber and recirculation from the aerobic biofilm chamber was attributed mostly to the presence of suspended solids. The sCOD measured in both, anaerobic effluent and recirculation, was similar, indicating that most of the soluble biodegradable COD was removed in the first anaerobic chamber. COD measured in the permeate was very low, around 10 mg/L, and did not vary during the whole experimental period. The differences observed between sCOD in the chambers, and that observed in the permeate indicated that the membrane retained a fraction of colloidal matter present in the mixed liquor.

Strong fluctuations of the sCOD in the chambers were caused by various factors. Between operating days 78 and 98 of Period II, 300 mg/L COD as methanol were introduced into the system with the feeding, causing partial inhibition of the methanisation stage and the accumulation of sCOD. Consequently, the COD concentration measured in UASB effluent was affected, as well as an overall system performance (Figure 2). As a result, the sCOD removal rate in anaerobic reactor decreased drastically and accumulated, reaching values below 20%, however no VFA was detected in the UASB effluent. On the other hand, the biogas composition analysis showed a diminution of methane percentage from more than 70% to less than 50%. This could probably be explained by the toxic properties of methanol versus anaerobic bacteria (Enright et al. 2005), especially taking into account that the system was operated in psychrophilic conditions. Therefore, from the day 98 onwards the concentration of methanol was decreased to the tenth part and the efficiency of the system was recovered, reaching sCOD removal efficiencies of 70% in UASB reactor and more than 95% in the entire system. Methane reached more than 70% of the biogas composition. At the beginning of the Period II, the decrease of DO concentration (from 3.5 to less than 1.0 mg O₂/L) in aerobic reactor was observed due to a COD overloading of the second aerobic stage. As a consequence, accumulation of tCOD (data not shown) and sCOD were observed, caused mainly by the elevated MLSS concentration in both, UASB effluent and recirculation. From the day 134 sCOD of both the effluent of the anaerobic chamber and recirculation of the aerobic chamber diminished to the values previously observed. The efficiency of the sCOD removal was recovered, reaching over 99% in the entire system, while around 80% was removed in the anaerobic bioreactor (Figure 2).

Biogas production in the UASB chamber was detected during the whole experimental period, but its production rate was not quantified with the required accuracy. On the other hand, the biogas produced contained a large fraction of methane that was around 80% during the Period III.

Macronutrients removal efficiency

One of the aims of this research was to produce nutrient rich wastewater, free of microbial indicators that could be reused in agriculture. Neither nitrogen nor phosphorous removal was obtained and the presence of both nutrients was observed in the effluent. The average concentrations of N-NH₄⁺ were 10–20 mg/L till day 114 and between 20 and 35 mg/L when the feeding COD was increased. All
the ammonia produced as the effect of protein hydrolysis in the anaerobic system was detected in the permeate, except from Period III, when nitrification in aerobic chamber was observed.

**Biomass characteristics**

In the UASB chamber the VSS concentration measured at the bottom part was maintained at 30–35 g/L. Granules used as seed grew in the anaerobic chamber and their retention was observed during the whole experimental period. In both aerobic and membrane chamber the average VSS concentration was around 1–2 g/L, except during certain moments of Period III, when excessive MLSS accumulation occurred (reaching around 4 and 8 gVSS/L in the aerobic and membrane chamber, respectively), mostly due to the increase of the OLR and instantaneous decrease of the UASB efficiency. Biofilm growing on the support carrier in aerobic chamber was well developed within the experiment. The average Mean Cell Retention Time (MCRT) was above 200 days for the whole system. Moreover, the overall biomass yield was calculated being 0.094 gVSS/gCOD, that is much lower than the typical values determined for aerobic MBRs (0.25–0.61 gVSS/gCOD) (Judd 2006), and close to those observed for the anaerobic treatment of wastewaters, that are in the range between 0.11 and 0.14 gVSS/gCOD (van Haandel & Lettinga 1994).

**Membrane performance**

The flux was maintained between 12 and 15 L/(m² h) during the whole operational period, being more variable on periods II and III due to the higher fouling in the unit during these two periods. The flux achieved was higher than those observed in anaerobic membrane bioreactors, with values between 5 and 10 L/(m² h) (Zhang et al. 2007; Lew et al. 2009; Spagni et al. 2010; Ho & Sung 2010), but lower than those typically reported in aerobic membrane bioreactors operating with similar membrane modules, being between 20 and 25 L/(m² h) (Judd 2002; Wen et al. 2004). On the other hand, the observed fluxes were much lower than those referred by Leiknes & Ødegaard (2007), who worked with a biofilm membrane bioreactor with a first Moving Bed Bioreactor (MBBR) followed by a filtration chamber connected in series and obtained fluxes of 50 L/(m² h).

Permeabilities between 100 and 200 L/(m² h bar) were normally observed during the three operational periods. These values were slightly better than those observed during the operation of similar membrane modules, (Bouhabila et al. 2001; Judd 2002), and also were higher than permeabilities observed in anaerobic membrane bioreactors (Zhang et al. 2007; Spagni et al. 2010). The reason of such a behaviour is still unclear, but it is considered that aerobic biomass, both in suspension and in biofilm retained or degraded some foulants generated in the methanogenic stage. This fact made feasible to achieve the observed permeabilities.

After a chemical intensive cleaning on day 57, permeability rose up to 400 L/(m² h bar) whereas the original permeability of the membrane was only 250 L/(m² h bar). Therefore, the behaviour of the membrane on period I was possibly related with the lower initial permeability of the membrane used.

Membrane critical flux was determined and the value at which irreversible fouling occurs was 19 L/(m² h). The criterion employed was that the increment of TMP with respect to time was higher than 10 Pa/min (Le-Clech et al. 2003). During the whole experimental period, the flux applied was below the critical flux, thus it was expected that reversible fouling was predominant. In fact, it was observed during all the operation time that permeability was almost fully recovered when a physical cleaning with tap water was carried out.

**CONCLUSIONS**

The system presents a high tolerance to loading changes and temperature fluctuations. Moreover, excellent COD removal performance, comparable with aerobic MBR, can be obtained using a combination of an anaerobic UASB bioreactor with membrane filtration for treating low-strength wastewater. On average, the permeate COD was less than 10 mg/L and the sCOD removal was above 95%, reaching 99% during the stable operation. Additionally, the effluent was free of suspended solids. Very low COD concentration and the level of nutrients in the effluent allow reusing purified wastewater (e.g. in agriculture).

Biogas production was detected during the whole operating period, with average methane content of 75–80%. Due to effective retention of biomass by the UASB reactor and membrane module, sludge concentration in the anaerobic bioreactor could be kept at high values, reaching more than 30 g/L. Moreover, granular sludge growth was observed.

With respect to the performance of the membrane, the highest permeabilities were achieved in the periods when the biomass was purged of the system. The predominant
fouling that took place in the membrane was reversible fouling, since permeability was recovered with physical cleaning. The membrane was operated at environmental temperature with fluxes of 15 L/(m² h), lower than those achieved in aerobic MBRs treating municipal wastewater, but higher than fluxes obtained in methanogenic AnMBRs.

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