Experimental study of the anaerobic urban wastewater treatment in a submerged hollow-fiber membrane bioreactor at semi-industrial scale

J.B. Giménez[a], A. Robles[b,*], L. Carretero[b], F. Durán[b], M.V. Ruano[a], M.N. Gatti[b], J. Ribes[a], J. Ferrer[b] and A. Seco[a]

[a] Departament d'Enginyeria Química. Escola Tècnica Superior d'Enginyeria. Universitat de València. Doctor Moliner, 50. 46100. Burjassot. València. Spain. (E-mail: *juan.b.gimenez@uv.es; m.victoria.ruano@uv.es; josep.ribes@uv.es; aurora.seco@uv.es*)

[b] Institut Universitari d'Investigació d'Enginyeria de l'Aigua i Medi Ambient, IIAMA. Universitat Politècnica de València. Camí de Vera, s/n. 46022. València. Spain. (E-mail: *ngerobma@upvnet.upv.es; laucarma@upvnet.upv.es; fredurpi@upvnet.upv.es; margat@upvnet.upv.es; jferrer@hma.upv.es*)

* Corresponding author: Tel.: +34 96 387 99 61; Fax: +34 96 387 90 09 E-mail: *ngerobma@upvnet.upv.es*

Abstract

The aim of this study was to assess the effect of several operational variables on both biological and separation process performance in a submerged anaerobic membrane bioreactor pilot plant that treats urban wastewater. The pilot plant is equipped with two industrial hollow-fiber ultrafiltration membrane modules (PURON® Koch Membrane Systems, 30 m^2 of filtration surface each). It was operated under mesophilic conditions (at 33 ºC), 70 days of SRT, and variable HRT ranging from 20 to 6 hours. The effects of the influent COD/SO$_4$-S ratio (ranging from 2 to 12) and the MLTS concentration (ranging from 6 to 22 g·L^{-1}) were also analysed. The main performance results were about 87% of COD removal, effluent VFA below 20 mg L^{-1} and biogas methane concentrations over 55% v/v. Methane yield was strongly affected by the influent COD/SO$_4$-S ratio. No irreversible fouling problems were detected, even for MLTS concentrations above 22 g·L^{-1}.

1. Introduction

Anaerobic treatments, which are commonly applied to high-loaded wastewaters
(sludge digestion and industrial wastewater treatment), have the following main benefits
compared to aerobic treatments: minimum sludge production due to the low biomass
yield of anaerobic organisms, low energy demand since no aeration is required, and
biogas production that can be used to fulfil process energy requirements (Ho and Sung,
2010). Furthermore, anaerobic processes are also seen as a sustainable approach for
low-strength wastewater treatment since they involve a lower environmental impact
than aerobic processes in many aspects such as net balance of greenhouse gas emissions
as well as the possibility of total nutrient recovery from urban wastewaters. However,
due to the low-growth rate of anaerobic bacteria, considerable biomass concentrations
and/or high temperatures are required in order to achieve suitable organic matter
removal rates, especially for low-strength wastewaters like urban ones. Membrane
technology applied to wastewater treatment by the so-called membrane bioreactors
(MBR) is a promising alternative to obtain high biomass and COD concentrations by
decoupling both hydraulic retention time (HRT) and solids retention time (SRT). The
complete retention of the microorganisms inside the MBR system allows high SRT to
be obtained with reduced working volumes. In recent years, submerged MBR
technology has been reported as a successful application for anaerobic wastewater
treatment (Huang *et al*., 2011). In addition, among the different types of membranes, the
hollow-fiber ones have been identified as the most suitable membranes to achieve high

effluent flows and high quality with low filtration energy demands (Lew *et al*., 2009).

The application of membrane separation processes in wastewater treatment has increasingly been applied during the last decade (Lesjean and Huisjes, 2007) even to large urban WWTPs. The installations of this type have grown from small-sized WWTPs to very large-sized WWTPs (maximum design flow rates up to 45000 m^3 day^{-1}) within only a few years (Huisjes *et al*., 2009). It is important to highlight that all these MBR urban WWTPs are based on aerobic processes where a high amount of aeration energy is required not only for organic matter removal but also for proper membrane performance. Even though the MBR technology has not yet been applied to anaerobic urban wastewater treatment at a full-scale plant, it is of emerging interest to the scientific community due to the above-mentioned advantages. Several studies have been published where anaerobic treatment of organic matter coupled to membrane separation processes is evaluated either with high-strength wastewater or with synthetic wastewater at laboratory scale (see e.g. Liao *et al.*, 2006; Jeison *et al*., 2010; Huang *et al*., 2008).

Several issues have been recognized elsewhere as potential drawbacks which must be solved in order to successfully apply anaerobic MBR technology to urban wastewater treatments. The first key issue that comes up with this technology is the dissolved methane in the effluent (Forster *et al*., 2007). A post-treatment process will be required to oxidize this methane before it is discharged to the atmosphere. Several studies have shown that methane is biologically oxidized by means of Methane Oxidizing Bacteria (MOB), which are capable of using methane as carbon and energy source (Hanson and Hanson, 1996). Other studies have shown that methane could be used as a carbon source for biological denitrification (Rhee and Fuhs, 1978; Meschner and Hamer, 1985; Modin *et al*., 2010). A recent study has shown that methane can be oxidized by a partnership of

methanotrophs and microalgae that grow together in bioflocs. In this study, algae provided the necessary oxygen for the methane oxidation process and almost all the carbon that originated from methane was assimilated into biomass without overall release of carbon dioxide (van der Ha *et al.*, 2011). Another key issue is the competition between Methanogenic Archaea (MA) and Sulphate Reducing Bacteria (SRB) for the available substrate (Hulshoff, 1998) when there is significant sulphate concentration in the influent. For urban wastewater, which can easily present low COD/SO_4-S ratio, this competition can critically affect the biogas production. Moreover, the presence of SRB can lead to several problems that must be solved, such as odor and corrosion problems, inhibition of MA, and decrease of the amount and quality of the biogas produced. With regard to the physical separation process, membrane fouling is the main key issue of MBR technology since it decreases membrane permeability and increases operational and maintenance costs (Chang *et al.*, 2002). The necessity of working at high SRT for anaerobic treatment of low strength wastewaters could lead to high MLTS concentrations and then, to low membrane permeability. Thus, the effect of MLTS over membrane fouling must be assessed. Membrane fouling can be reduced by different strategies (Vallero *et al.*, 2005; Liao *et al.*, 2006; Dvořák *et al.*, 2011), such as the following: optimizing the frequency and duration of the physical cleaning stages (back-flush and relaxation); optimizing different operational variables (e.g. gas sparging intensity); and operating membranes under sub-critical filtration conditions (which are bounded by the so-called critical flux). However, the effect of these operational variables on the membrane fouling is not properly evaluated at lab scale since it strongly depends on the membrane size. Specifically in hollow-fiber membranes the hollow-fiber length is the main design parameter. Among the reported anaerobic MBR studies to treat urban wastewater, most of them have been assessed at laboratory scale plants (Hu and Stuckey, 2006; Fawehinmi *et al.*, 2007; Huang *et al.*, 2011). Nevertheless, no

references have been found in the literature concerning the application of anaerobic MBR technology with industrial-scale membrane modules to treat real urban wastewater. Since the membrane performance cannot be directly scaled-up from laboratory to real plant, especially with hollow-fiber-based technology, further studies are needed on membrane technology at industrial-scale in order to facilitate the design and implementation of this technology at full-scale WWTPs.

The main objective of this paper is to study the feasibility of Submerged Anaerobic MBR technology (SAnMBR) applied to urban wastewater treatment. The novelty of this work lies in studying the feasibility of this technology under specific conditions that are similar to the ones expected at full scale plants. To this aim, a pilot plant that incorporates industrial membrane modules has been designed and operated with the effluent of the Carraixet WWTP pre-treatment (Valencia, Spain). Thus, the influent load variability, which is typical of urban WWTPs, is considered in this study.

In this work, the pilot plant operation results are presented and the main technical problems that SAnMBR technology could present are identified and evaluated, such as the dissolved methane in the effluent, the SRB and MA competition for substrate at low COD/SO_4-S ratio, and the membrane fouling.

2. Materials and methods

2.1. Pilot plant description

Figure 1a shows the SAnMBR pilot plant that was used in this study. The pilot plant was designed to treat a maximum flow-rate of 1200 L h^{-1}, assuming a net flux of

20 L $m^{-2} h^{-1}$ in both membrane tanks, which would lead to a minimum HRT of 2 hours. It consists of an anaerobic reactor of 1.3 m^3 total volume (0.4 m^3 head-space volume) connected to two membrane tanks of 0.8 m^3 total volume each (0.2 m^3 head-space volume). Each membrane tank includes one industrial hollow-fiber ultrafiltration membrane module (PURON® Koch Membrane Systems (PUR-PSH31), 0.05 μm pore size). Each module consists of 9 hollow-fiber bundles of 1.8 m length that give a total of 30 m^2 membrane surface. A rotofilter of 0.5 mm screen size has been installed as pre-treatment system. One equalization tank (0.3 m^3) and one Clean-In-Place (CIP) tank (0.2 m^3) are also included as main elements of the pilot plant. In order to control the temperature when necessary, the anaerobic reactor is jacketed and connected to a water heating/cooling system.

Figure 1b shows the flow diagram of the pilot plant. The pilot plant is fed with the effluent of the Carraixet WWTP pre-treatment (screening, degritter, and grease removal). After further pre-treatment in the rotofilter (RF) and homogenization in the equalization tank (ET), the wastewater is pumped to the anaerobic reactor (AnR). In order to improve the stirring conditions of the anaerobic reactor and to favour the stripping of the produced gases from the liquid phase, a fraction of the produced biogas is recycled to this reactor. The sludge is continuously recycled through the external membrane tanks (MT) where the effluent is obtained by vacuum filtration. In order to minimise the cake layer formation, another fraction of the produced biogas is also recycled to the membrane tanks from the bottom of each fiber bundle. With the aim of recovering the biogas bubbles extracted with the membrane effluent, a degasification vessel (DV) was installed between the MT and the vacuum pump. This DV consists of a pipe-section widening that is conic-shaped, which favours the biogas accumulation at the top of this element. The obtained permeate is stored in the CIP tank. By using two

membrane tanks in parallel, the pilot plant has been designed and automated with high operational flexibility, which allows the pilot plant to work with either one membrane tank or both tanks. Hence, different transmembrane fluxes can be tested without affecting the hydraulic retention time (HRT) of the plant. In order to control the solids retention time (SRT) in the system, a fraction of the sludge is intermittently extracted from the anaerobic reactor throughout the day.

2.2. Membrane performance

The membrane operation was programmed in order to allow the study of different relaxation and back-flush frequencies and durations. Normal membrane operational mode is carried out by a defined schedule of different individual stages that are combined from a filtration-relaxation (F-R) basic cycle (see Figure 2). Besides classical membrane operational stages (filtration, relaxation and back-flush) the two following stages were also considered in the membrane operation:

Degasification: a typical disadvantage of dead-end, hollow-fiber membranes is the accumulation of biogas at the top of the fibers which reduces the effective filtration area. The degasification stage consists of a period of high flow-rate filtration that is carried out to enhance the filtration process efficiency by removing the accumulated biogas.

Ventilation: This stage is similar to a back-flush, but the permeate is pumped to the membrane tank through the degasification vessel instead of through the membrane. The aim of the ventilation stage is to recover the biogas accumulated in the degasification vessel.

The membrane performance is then established by a proper selection of the membrane operational mode. Figure 2 shows the possible membrane operational modes to be selected. For instance, the operational mode defined by X=2; Y=10; Z=50 implies that a back-flush is carried out every two F-R cycles; a ventilation is carried out every ten F-R cycles; and a degasification followed by a ventilation is carried out every fifty F-R cycles.

Besides the membrane operational mode, the separation process is also controlled by the transmembrane flux (J), the transmembrane pressure (TMP), the sludge flow-rate recycled through the membrane tanks, and the recycled biogas flow-rate.

2.3. Pilot plant instrumentation, automation, and control

Numerous on-line sensors and automatic equipment was installed in order to automate and control the pilot plant operation and to obtain on-line information about the state of the process. The instrumentation is connected to a network system that includes several transmitters, a Programmable Logic Controller (PLC), and a PC to perform multi-parameter control and data acquisition. Both the operational data logging and the pilot plant control are carried out by a SCADA system installed in the PC, which centralises all the signals from the different sensors and actuators that are installed in the plant. The on-line sensor consists of the following: 3 pH-Temperature transmitters that are located in the anaerobic reactor and the two membrane tanks; 1 oxidation-reduction potential sensor that is located in the anaerobic reactor; 6 flow-rate transmitters (one for each pump); 5 level transmitters (one for each tank: ET, AnR, MT1, MT2, and CIP); 2 liquid pressure transmitters to control the TMP; and 3 gas

pressure transmitters (one for each of the two blowers, and the third one for the reactor). With regard to actuators, the pilot plant consists of the following: 8 frequency converters, which command the rotational speed of the 6 pumps and the 2 blowers; one regulatory valve to control the biogas discharge according to the pressure in the system; and 6 on-off control valves, which are used to establish the flow direction aimed to control the sludge wastage and the different membrane operational stages (filtration, back-flush, ventilation, standby...). Based on this instrumentation, the pilot plant includes several control loops that are hierarchically organized in lower layer and upper layer controllers. Lower layer controllers consist of several classical PID and on-off controllers that were designed to control the main operational variables: flow-rates (influent, sludge recycling, and wastage, permeate, recycled biogas through the reactor and membrane tanks); biogas pressure in the system, transmembrane pressure in both membrane tanks; reactor temperature; and level in all the tanks. Upper layer controllers consists of a supervisory fuzzy-logic-based control system that establishes the different set-points for the above-mentioned operation variables according to all the information gathered from the different sensors installed in the plant.

2.4. Pilot plant operation

The SAnMBR pilot plant was started up with a considerable biomass inoculum (40% of the total working volume), which was taken from the anaerobic digester of the full-scale WWTP. The pilot plant was operated at SRT of 70 days and controlled temperature of 33 ºC. The HRT was gradually decreased over the operational period from 21 to 6 hours. Only one membrane module was necessary to obtain the required treatment flow-rates. The treatment flow-rate was controlled by the scheduling of the different membrane operational stages as explained above, i.e., changing the frequency

and duration of the filtration, relaxation and back-flush stages. The recycled biogas flow-rate through the anaerobic reactor was set to 4 $Nm^3 h^{-1}$ to obtain proper mixing conditions. The biogas sparging intensity in the membrane tank was kept at 0.23 $Nm^3 m^{-2} h^{-1}$ (recycled biogas flow-rate of 7 $Nm^3 h^{-1}$) to provide suitable shear conditions over the membrane surface.

2.5. Sampling and laboratory measurements

In order to evaluate the biological process performance, 24-hour-composite samples were collected from influent and effluent streams and grab samples of the biogas produced and the anaerobic sludge were collected from the reactor once a day. The following parameters were analysed daily for the influent, effluent, and the anaerobic sludge: Total Solids (TS), Volatile Solids (VS), Total Suspended Solids (TSS), Volatile Suspended Solids (VSS), Volatile Fatty Acids (VFA), alkalinity (Alk), sulphate (SO_4-S), sulphide (HS^-), and nutrients (ammonium (NH_4-N) and orthophosphate (PO_4-P)). The following parameters were determined once a week: total and soluble chemical oxygen demand (COD_T and COD_S, respectively); total nitrogen (TN) and filtered total nitrogen (TN_f); and biogas composition (CH_4, CO_2, and H_2S). Furthermore, a sludge sample was fixed for microbiological analysis once a week.

2.6. Analytical and microbiological methods

Solids, COD, sulphate, sulphide, and nutrients were determined according to Standard Methods (APHA, 2005). The carbonate alkalinity and VFA concentration were determined by titration according to the method proposed by WRC (1992). The methane fraction of the biogas was measured using a gas chromatograph equipped with

a Flame Ionization Detector (GC-FID, Thermo Scientific). 1 mL of biogas was collected by a gas-tight syringe and injected into a 30 m x 0.319 mm x 25 μm HP-MOLESIEVE column (Agilent Technologies) that was maintained at 40 ºC. The carrier gas was helium at a flow-rate of 40 mL min^{-1}. CH4 pure gas (99.9995%) was used as standard.

The microbiological analysis was performed by the FISH (Fluorescent In Situ Hybridization) technique to identify the different species of SRB and MA. For Gram negative cells, 1 ml aliquots of the sample were fixed in freshly prepared 4% paraformaldehyde (PFA) for 1-3 hours at 4ºC. Gram positive cells were fixed with ethanol (1 volume of sample and 1 volume of ethanol) at 4ºC for 4-16 hours. After fixation, cells were washed with Phosphate Buffered Saline (PBS), resuspended in PBS-absolute ethanol (1:1, v/v) and stored at -20ºC (Harmsen *et al*., 1996). Hybridizations were carried out as described in Amann *et al*. (1990). Oligonucleotide sequences employed in this study are listed in Table 1. Hybridized cells were enumerated by the following procedure. For each sample, a minimum of 20 images of randomly chosen microscopic fields were captured with a Leica DFC420c digital camera connected to a Leica DM2500 epifluorescence microscope. Then, these images were analysed using an automated bacteria quantification software (Borrás, 2008) based on thresholding techniques that is programmed in Matlab®. The countable pixel area of the specific probe-fluorochrome signal was expressed as a mean percentage of the pixel area count from the EUBmix plus ARCH915 probes signal. Error of the quantification was calculated by dividing the standard deviation by the square root of "n", where "n" is the number of examined fields.

3. Results and discussion

3.1. Biological process performance

Table 2 shows the average wastewater characteristics of the influent to the anaerobic reactor during the experimental period. This table highlights the significant sulphate concentration and the relatively low COD concentration of the influent, as well as the strong variability of the influent load as can be deduced from the high values of standard deviation associated to each parameter. The uncertainty associated to each value includes both the standard deviation of the different samples analysed throughout the experimental period and the coefficient of variation associated to the analytical methods.

Figure 3a shows the treatment flow-rate, effluent VFA and the total solids concentrations throughout the experimental period. As this figure shows, the treatment flow-rate was gradually increased from 2000 to 5000 L day^{-1}. Effluent VFA was used as an indicator of the adaptation of the system to the different organic loading rates. Once the effluent VFA concentration remained practically stable below 20 mg L^{-1}, the next increase in the influent flow-rate was applied. The MLTS concentration in the system throughout the operational period increased from 8 to 22 g L^{-1}. As Figure 3a shows, the effluent VFA concentration was maintained at low values for every treatment flow-rate studied. It indicates a suitable COD removal even for working at HRT of around 8 hours. However, COD removal rate depends on other operational variables, such as the temperature or the influent COD/SO$_4$-S ratio. Therefore, further studies are needed in order to establish the most recommendable value of HRT for different operation conditions.

Figure 3b shows the amount of biogas produced per unit of treated wastewater as a

function of the influent COD/SO$_4$-S ratio. As this figure illustrates, the biogas

production rate was significantly affected by the variability of the influent COD/SO$_4$-S

ratio. This phenomenon can be attributed to the presence of SRB in the system, which

competed with MA for the available substrate. The continuous line in Figure 3b

represents the threshold value above which the sulphate reduction becomes

stoichiometrically fulfilled (2.01 mgCOD mg^{-1}SO$_4$-S). When the influent COD/SO$_4$-S

ratio is below this threshold, almost the entire amount of COD is removed by SRB, and,

consequently, the methane production decreases due to the low COD available for MA.

In contrast, when the COD/SO$_4$-S ratio is above this threshold, the remaining COD is

available for MA, and, thus, methane production increases. Despite the presence of

SRB, a significant methane-rich (over 60% v/v) biogas production was observed (an

average of 100 L d^{-1}) throughout this operational period. Moreover, this biogas

production rate showed that the sulphide concentration in the system (over 90 mgS L^{-1})

did not critically affect the methanogenesis.

Figure 4a shows the MA and SRB percentages determined by the FISH technique

throughout the experimental period. FISH analyses revealed that, during the starting-up,

the MA population was higher than SRB one due to the biomass inoculum, which was

characterised by a complex community of MA formed by Methanobacteriales (1 ± 1%),

Methanomicrobiales (1 ± 1%), and Methanosarcinales (5 ± 1%). After 6 days of

operation, only acetotrophic methanogens (order *Methanosarcinales*) and a small

amount of *Methanobacteriales* were detected. The percentage of acetotrophic

methanogens decreased throughout this period reaching values under 1% on day 110.

This decrease was related to the decrease observed in the influent COD/SO$_4$-S ratio,

which also resulted in a slight reduction of the methane production. In general, between

days 60 and 80, as well as between days 130 and 150, MA proliferations were detected,

which were correlated with an increase in both the influent COD/SO_4-S ratio and the organic loading rate.

With regard to SRB activity, during the start-up a sulphate removal lower than 50% was observed (see Figure 4b). This initial low sulphate removal was attributed to the low presence of SRB in the inoculum. FISH analyses proved that SRB were present in the inoculum, although in a low percentage. This inoculum was mainly composed of bacteria belonging to the *Desulfobacteraceae* family ($2 \pm 1\%$), within the order *Desulfobacterales*. The *Desulfovibrionales* order was also present, but in low percentages (less than 1 %). A slow development of SRB was observed within the *Desulfobacterales* order (up to $4 \pm 1\%$) and the *Desulfovibrionales* order (up to $1 \pm 1\%$). After 20 days of operation 98% of sulphate removal was obtained, which correlates with an increase in SRB observed in the system (see Figure 4).

In order to evaluate the global process performance, the mean values of the main operational variables and the average results of the treatment performance were obtained for the period comprised between days 75 and 135 (see Table 3 and Table 4), which was considered a relatively stable period. During this period, the pilot plant had enough operating days and the treatment flow-rate was maintained stable before it was sharply increased from 150 to 220 $l \cdot h^{-1}$. Table 3 shows an almost complete sulphate reduction, as well as low effluent COD and VFA concentrations. The pH values were stable over the entire period (around 6.7), which is high enough to avoid methanogenic inhibition and low enough to avoid chemical precipitation, which could lead to inorganic fouling in the membrane. The high variability of the methane % in the biogas shown in Table 3 was attributed to the strong dynamics of the influent load (see Table 2) and mainly to the influent COD/SO_4-S ratio variability as mentioned above.

Table 4 shows that the average total COD removal efficiency for this period was around 87%, which is comparable to other studies (Liao *et al*., 2006; Hu and Stuckey, 2006, Lin *et al*., 2009). However, the average methane yield observed was relatively low (0.069 LCH$_4$ g^{-1}COD compared with the expected (theoretical) value of 0.389 LCH$_4$ g^{-1}COD). This low methane yield can be explained by several factors such as the strong competition between SRB and MA for the available substrate, the loss of dissolved methane in the effluent, and the strong variability of the influent COD load. Indeed, if the COD that is removed by the SRB is not considered in this yield estimation, the resulting methane yield value is higher than the observed one (0.294 LCH$_4$ g^{-1}COD) but still lower than the above-mentioned expected value. This fact can be explained by the loss of the methane that is dissolved in the effluent. Since the produced biogas is used for membrane physical cleaning by gas sparging, it can be assumed that dissolved methane in the liquid phase will be in equilibrium with methane in the gas phase, i.e. the effluent methane concentration will be close to the methane solubility concentration. According to the methane solubility in water at different temperatures, it is expected that the dissolved methane in the effluent will be increased at lower temperatures. Methane molar fraction in pure water (no data are available for wastewater) can be calculated by means of Henry's Law:

$$X^{CH_4} = \frac{P_g^{CH_4}}{K_H^{CH_4}(T)}$$

where $P_g^{CH_4}$ is the methane partial pressure, and $K_H^{CH_4}(T)$ is the temperature-dependent Henry's law constant, which can be calculated as follows (Tchobanoglous *et al*., 2003):

$$\log K_H^{CH_4}(T) = -\frac{675.74}{T(K)} + 6.88$$

According to these theoretical considerations, Figure 5 shows the methane

solubility as function of the gas-phase methane content for temperatures of 15, 20 and 33 ºC. As can be seen in this figure, the dissolved methane concentration in equilibrium with 55 % of methane in the gas phase corresponds to 12.3 mg CH_4 L^{-1} (49.2 mg COD L^{-1}). With this percentage of methane in the gas phase, an increase of the dissolved methane concentration of 3.1 mg CH_4 L^{-1} (12.4 mg COD L^{-1}) will be obtained when the temperature is decreased from 33 to 20 ºC. At 15ºC the methane in the effluent would be increased by 18.3 mg COD L^{-1} with respect to the value at 33ºC.

Since temperature will be the main deciding factor to apply this technology in the different climate regions of the world, the pilot plant is currently being operated successfully at 20 ºC in order to assess the feasibility of SAnMBR technology to treat urban wastewater in a full scale plant (data not shown). This temperature has been selected since it is considered a common water temperature in most of the regions, where this technology could be implemented.

3.2. Separation process performance

With regard to the physical separation process, Figure 6 shows the TMP profile obtained at the end of the experimental period (on day 135) as well as the membrane operational mode used on that period. The membrane operational conditions were the following: a permeate flux of 10 LMH (normalised to 20 ºC); a constant biogas sparging intensity of 0.23 Nm^3 m^{-2} h^{-1}; and a MLTS in the anaerobic reactor of 22 g L^{-1}. The MLTS in the membrane tank was around 26 g L^{-1}, according to the ratio between the sludge flow-rate fed to the membrane tank and the net permeate flow-rate. The critical flux (normalised to 20 ºC) under these conditions was determined as 13 LMH. Hence, the membrane was operated under sub-critical filtration conditions to minimise fouling

problems. Figure 6a shows a total TMP recovery after the relaxation stage. The maximum value of TMP reached was 0.08 bar. This is really low compared to 0.6 bar, which is the maximum value advised by Koch Membrane Systems. The membrane behaviour can be better observed in Figure 6b, where an amplified period of one hour from Figure 6a is shown. This figure shows how the TMP remains practically constant during the filtration periods. It is also important to note that this low TMP value was obtained for the maximum MLTS of the experimental period, which highlights the good performance of the membrane over the entire experimental period. It indicates the possibility of operating membranes at higher MLTS concentrations than in aerobic processes, since anaerobic systems do not present the oxygen transfer limitation problem that limits the MLTS concentration in aerobic processes (see e.g. Stephenson *et al*., 2000). As can be observed in the figure, physical fouling is removed from the membrane surface due to the physical cleaning mechanism (relaxation, back-flush, shear intensity of gas sparging). This fouling removal highlights the importance of establishing a proper membrane operational mode in order to minimise filtration problems (fouling and clogging). Throughout the entire experimental period, non-irreversible fouling on the membrane surface was detected, even for high MLTS. This is mainly the result of working under sub-critical filtration conditions. MLTS concentrations above 25 $g \cdot L^{-1}$ are not recommended because the critical flux decreases to values below 10 LMH, thus, making the filtration process unnecessarily expensive. For instance, a critical flux of about 8 LMH was obtained for a MLTS concentration of 30 $g \cdot L^{-1}$. For these operational conditions, 10 MLH of J_{20} resulted in a continuous increase in the TMP.

Further research is needed in order to gather more information under different operation conditions which will be needed to carry out an exhaustive economical

analysis on the proposed technology compared to the existing ones.

4. Conclusions

The pilot plant performance demonstrates that SAnMBR can be a promising technology for urban wastewater treatment. The competition between MA and SRB must be considered, especially when the influent wastewater presents a low COD/SO_4-S ratio. Under mesophilic conditions and 70 days SRT, almost 90% of COD removal was achieved, but with a low methane yield, which is mainly due to the COD removed by SRB. No irreversible fouling problems were detected, even for high MLTS concentrations. A flux of 10 LMH and a MLTS concentration above 22 gL^{-1} (sub-critical conditions) led to a TMP that was lower than 0.1 bar.

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Figure and table captions

Figure 1. General view of **(a)** the pilot plant and **(b)** the flow diagram. (Nomenclature: **RF**: rotofilter; **ET**: equalization tank; **AnR**: anaerobic reactor; **MT**: membrane tanks; **DV**: degasification vessel; **CIP**: clean-in-place; **P**: pump; and **B**: blower).

Figure 2. Sequence of the different individual stages in a normal membrane operational mode.

Figure 3. (a) Evolution during the operational period of the treatment flow-rate, the effluent VFA concentration, and the MLTS concentration; **(b)** biogas production as function of the influent COD/SO_4-S ratio.

Figure 4. Evolution of: **(a)** MA and SRB percentage; **(b)** influent and effluent sulphate concentration, and SRB percentage.

Figure 5. Solubility of methane in water as a function of the percentage of methane in the biogas at 15, 20 and 33 ºC.

Figure 6. **(a)** TMP evolution on day 135 of operation. **(b)** Zoom to one hour of operation. The membrane operational mode was: F-R basic cycles of 300 seconds in length (250 s filtration and 50 s relaxation); 40 seconds of back-flush with $(F-R)_{10}$; 40 seconds of ventilation with $(F-R)_{10}$; and 40 seconds of degasification with $(F-R)_{50}$. (Nomenclature: **S**: StandBy; **D**: Degasification; **V**: Ventilation; **B**: Back Flush; **R**: Relaxation; **F**: Filtration).

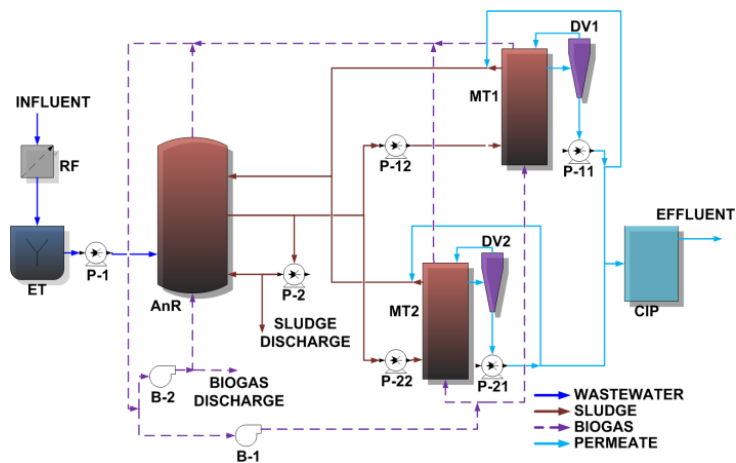**Table 1.** Oligonucleotide sequences employed in this study

Table 2. Average influent wastewater characteristics

Table 3. Mean values of the main operational variables from the period between day 75 and day 135.

Table 4. Average treatment efficiency values from the period between day 75 and day 135.
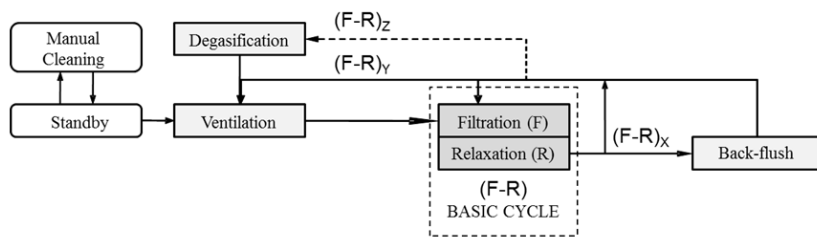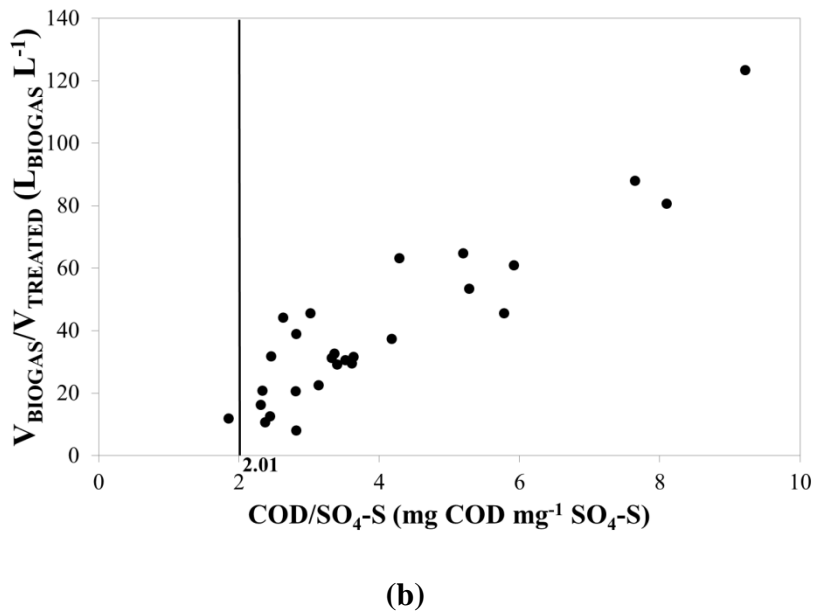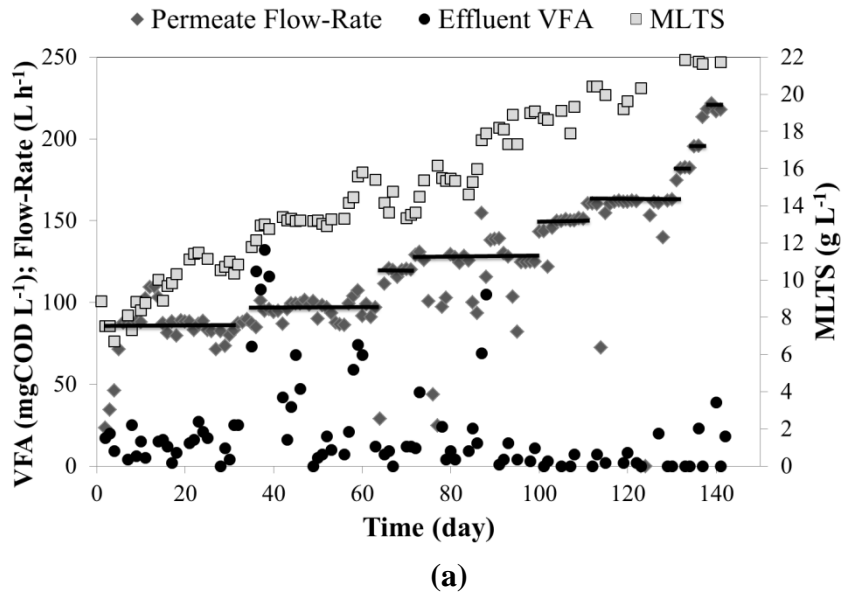
(a)

(b)

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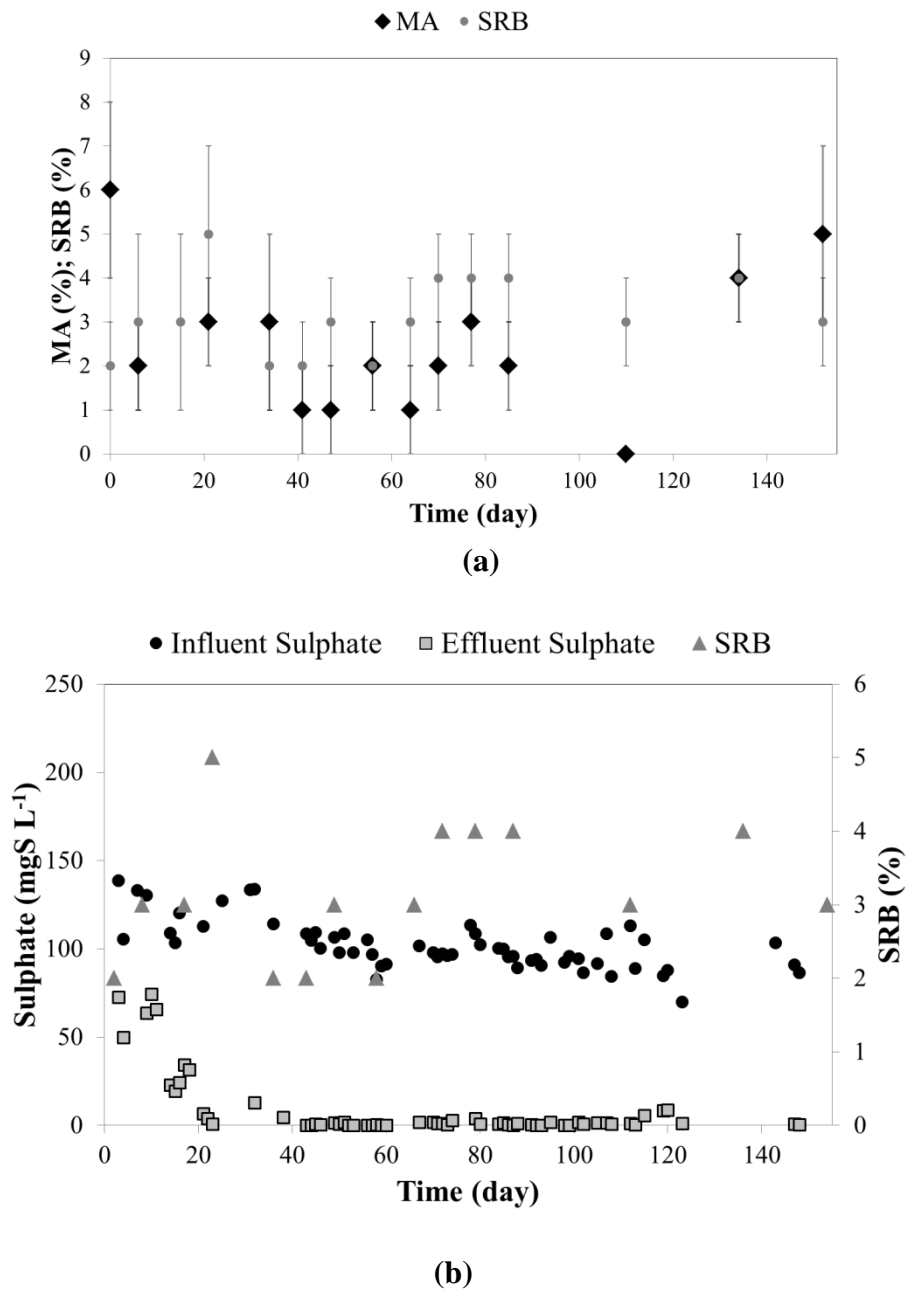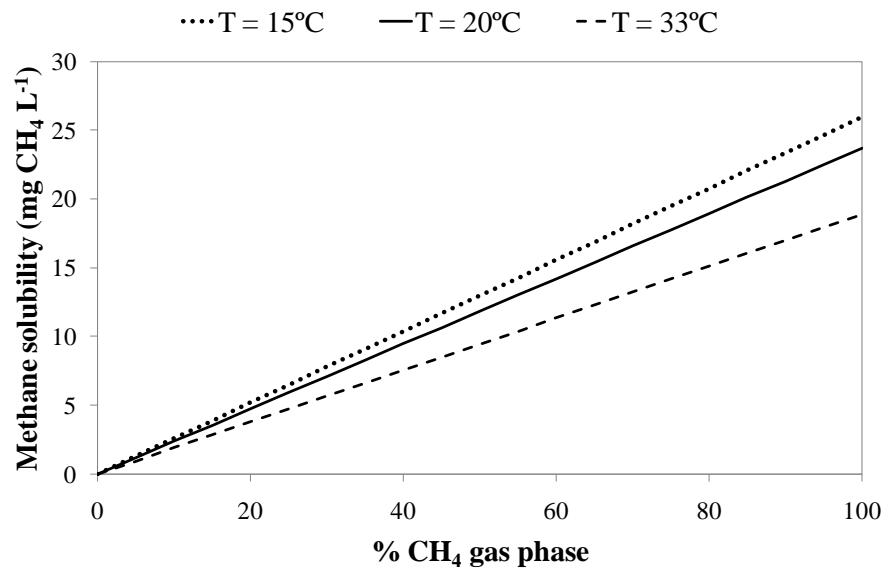
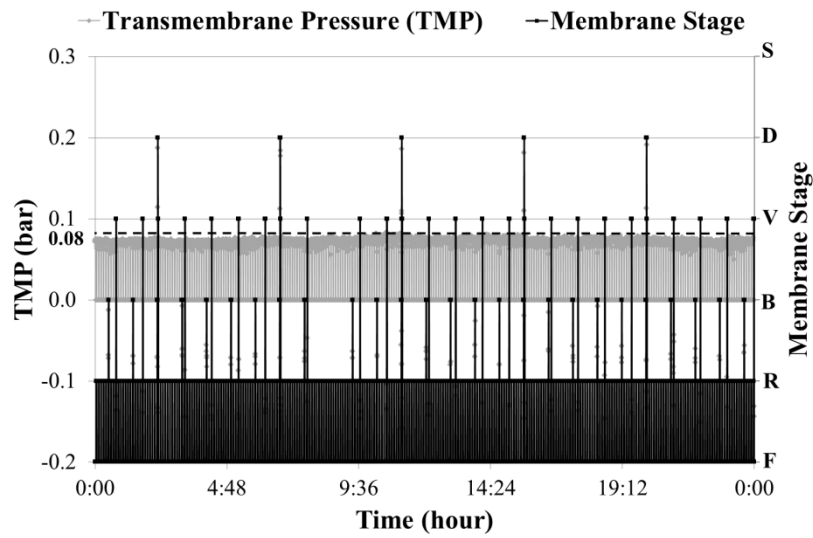**Figure 2.** Sequence of the different individual stages in a normal membrane operational mode.

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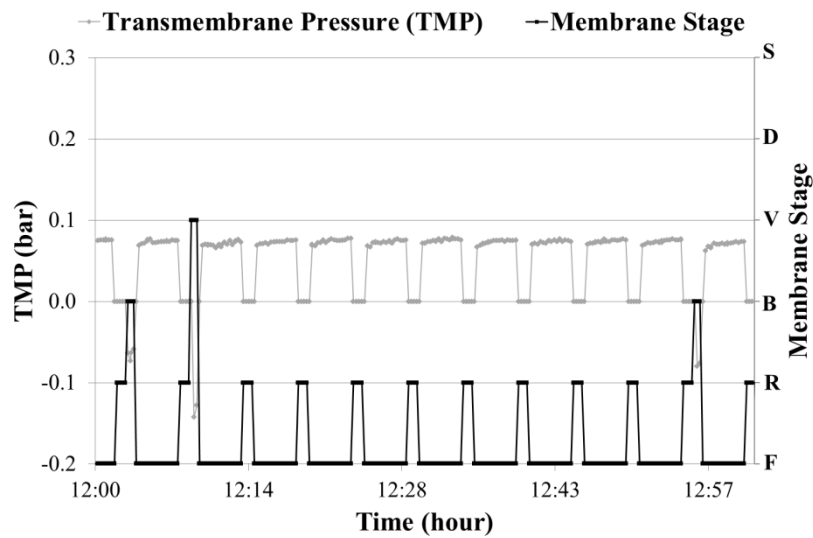
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Table 1. Oligonucleotide sequences employed in this study

Probe	Sequence (5'→3')	Specificity	% FA	Reference
EUB338	GCTGCCTCCCGTAGGAGT	*Eubacteria*	0-50	Amann *et al*. (1990)
EUB338 II	GCAGCCACCCGTAGGTGT	*Planctomycetales*	0-50	Daims *et al*. (1999)
EUB338 III	GCTGCCACCCGTAGGTGT	*Verrucomicrobiales*	0-50	Daims *et al*. (1999)
ARCH915	GTGCTCCCCCGCCAATTCCT	*Archaea*	35	Stahl & Amann (1991)
Sulphate Reducing Bacteria				
SRB385	CGGCGTCGCTGCGTCAGG	most *Desulfovibrionales*	35	Amann *et al*. (1990)
SRB385Db	CGGCGTTGCTGCGTCAGG	*Desulfobacteraceae*	30	Rabus *et al*. (1996)
DBB660	GAATTCCACTTT CCCCTCTG	some *Desulfobulbus*	60	Devereux *et al*. (1992)
Dtm230	TAATGGGACGCGGACCCA	*Desulfotomaculum* cluster I	10	Hristova *et al*. (2000)
Methanogenic Archaea				
MSMX860	GGCTCGCTTCACGGCTTCCCT	*Methanosarcinales*	45	Raskin *et al*. (1994)
MG1200b	CRGATAATTCGGGGCATGCTG	*Methanomicrobiales*	20	Crocetti *et al*. (2006)
MB311	ACCTTGTCTCAGGTTCCATCTCC	*Methanobacteriales*	30	Crocetti *et al*. (2006)
MC1109	GCAACATAGGGCACGGGTCT	*Methanococcales*	45	Raskin *et al*. (1994)

Table 2. Average influent wastewater characteristics

Parameter	Unit	Mean ± SD
TSS	mgTSS L^{-1}	186 ± 61
VSS	mgVSS L^{-1}	150 ± 54
Total COD	mgCOD L^{-1}	445 ± 95
Soluble COD	mgCOD L^{-1}	73 ± 25
VFA	mgCOD L^{-1}	11 ± 7
SO_4-S	mgS L^{-1}	99 ± 18
NH_4-N	mgN L^{-1}	27.0 ± 8.1
PO_4-P	mgP L^{-1}	2.7 ± 0.9
Alk	mgCaCO$_3$ L^{-1}	292.5 ± 37.2

Table 3. Mean values of the main operational variables from the period between day 75 and day 135.

Parameter	Unit	Sample location	Mean ± SD
pH		Reactor	6.72 ± 0.08
ORP	mV	Reactor	-488 ± 0.08
T	℃	Reactor	33.3 ± 0.2
MLTS	gTS L^{-1}	Reactor	19 ± 2
MLVS	gVS L^{-1}	Reactor	12.5 ± 0.2
COD	mgCOD L^{-1}	Effluent	77 ± 33
VFA	mgCOD L^{-1}	Effluent	12 ± 7
NH_4-N	mgN L^{-1}	Effluent	33.4 ± 8.2
PO_4-P	mgP L^{-1}	Effluent	3.1 ± 0.9
SO_4-S	mgS L^{-1}	Effluent	1.7 ± 2.4
S^{2-}	mgS L^{-1}	Effluent	94.7 ± 11.4
CH_4	%	Biogas	55 ± 10

Table 4. Average treatment efficiency values from the period between day 75 and day 135.

Parameter	Unit	Mean ± SD
COD Removal	%	86.9 ± 3.4
Observed Methane Yield	LCH$_4$ g^{-1}COD	0.069 ± 0.022
Methane Yield	LCH$_4$ g^{-1}COD	0.294 ± 0.04