Effect of sparging rate on permeate quality in a submerged anaerobic membrane bioreactor (SAMBR) treating leachate from the organic fraction of municipal solid waste (OFMSW)

Short communication

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Abstract

This paper focuses on the treatment of leachate from the organic fraction of municipal solid waste (OFMSW) in a submerged anaerobic membrane bioreactor (SAMBR). Operation of the SAMBR for this type of high strength wastewater was shown to be feasible at 5 days hydraulic retention time (HRT), 10 $L.min^{-1}$ (LPM) biogas sparging rate and membrane fluxes in the range of 3-7 $L.m^{-2}.hr^{-1}$ (LMH). Under these conditions, more than 90% COD removal was achieved during 4 months of operation without chemical cleaning the membrane. When the sparging rate was reduced to 2 LPM, the transmembrane pressure increased dramatically and the bulk soluble COD concentration increased due to a thicker fouling layer, while permeate soluble COD remained constant. Permeate soluble COD concentration increased by 20% when the sparging rate increased to 10 LPM.

Keywords : anaerobic digestion, landfill leachate, submerged anaerobic membrane bioreactor, sparging rate, effluent quality.

1. Introduction

The main advantages of membrane bioreactors (MBR) include rapid start-up and a higher loading rate than classical technologies (Stephenson et al., 2000), combining in one unit the removal of COD, solids and nutrients, thus resulting in a small footprint and a very high quality permeate with no suspended solids. Anaerobic MBRs have the added advantage of producing energy in the form of biogas, and generating very little excess sludge, thereby reducing the burden of sludge disposal. In a submerged anaerobic membrane bioreactor (SAMBR) the membrane is submerged within the reactor, and membrane cleaning is accomplished by recirculating the biogas; the coarse bubbles produced underneath the membrane scour it and reduce biofouling to manageable levels, i.e. low transmembrane pressure (TMP) drops. Several researchers have observed fouling minimization by gas sparging (Hong et al., 2002; Li et al., 2005) and other turbulence promoting techniques such as gas/liquid slug flow (Mercier-Bonin et al., 2001) or polymeric particles (Imasaka et al., 1989).

In sidestream membrane bioreactors the membrane module is external to the bioreactor. Sidestream membranes usually operate at higher crossflow velocities (1 - 5 $m.s^{-1}$), transmembrane pressures (TMP = 2-7 bars) and permeate flux (70 - 100 LMH) compared to the SAMBR, but they generate more shear (Berube et al., 2006). This can lead to more cell lysis and extracellular polymer production, which also causes biofouling; sidestream operation in a MBR can lead to a 50% decrease in sludge activity after circulating the sludge 20 times, and a 90% loss within 100 cycles (Brockmann and Seyfried, 1997). Despite being costly (Al-Malack, 2006), the main advantage of crossflow filtration is the limitation of cake build-up at the membrane surface due to the shear stress caused by the tangential flow. In a sidestream configuration it has been shown by several researchers that a higher crossflow velocity has a beneficial influence on the flux as it increases the critical flux and reduce cake formation (Chen et al., 1997; Defrance and Jaffrin, 1999) by decreasing the resistance associated with the polarization layer (Choo and Lee, 1998): concentration polarisation (CP) is the tendency of solutes to accumulate on the membrane surface within a concentration boundary layer, and this liquid film is stagnant since the liquid velocity at the membrane itself is zero. This implies that the only mode of transport is diffusion, and the solute concentration near the membrane increases exponentially with increasing flux.

Furthermore, due to CP, permeation of the reactor solutes and colloids through the membrane decreases depending on the thickness of the layer; however, this thickness decreases when turbulence in the reactor is increased. At high fluxes, significant flux decline is observed for any membrane/wastewater combination (Amy, 2008), and this can be attributed to a thicker CP layer, manifesting itself in the aggregation of soluble microbial products (SMPs), humic substances, organic colloids and suspended matter, and calcium carbonate precipitates (Boussu et al., 2006; Mahvi and Razavi, 2005).

The submerged configuration is usually preferred because of low operating costs, gentle mixing and high COD removal efficiency. With a synthetic low strength wastewater feed for an anaerobic submerged membrane bioreactor, Hu (2004) showed that biogas should be sparged as soon as there is a flux applied through a Kubota membrane with a 0.4 micron pore size. Otherwise, if the flux is too high, the cake will consolidate and gas sparging will be inefficient to remove the cake once it has formed. He also showed that the TMP was minimal (0.1 bar) with the highest gas flowrate (15 LPM). It turned out that this flowrate caused a cake to form with bigger particles than at lower flowrates, indicating that the smallest particles produced at lower flowrates were responsible for the fouling and thus the increase in TMP. However, gas sparging is only effective up to a limit, i.e. there are some forms of fouling that are resistant to gas sparging (Hong et al., 2002; Hu, 2004; Li et al., 2005).

Stephenson et al. (2000) stated that most studies in the literature showed that the concentration of soluble COD was consistently two or three times higher in the reactor than that observed in the effluent due to the rejection of soluble organics (COD) by the membrane. Akram and Stuckey (2008) reported ratios of COD reactor/COD permeate as high as 12 in SAMBR and that ratio was 1-5 when activated carbon was added. Considerably lower COD concentrations in the permeate compared to the bulk are due to filtration by the fouling layer and narrowed pores (Choi and Ng, 2008; Hu, 2004). Furthermore, the membrane rejects most of the high molecular weight and slowly degradable compounds (Trzcinski and Stuckey, 2009a).

This indicated that a large amount of dissolved COD was retained by the thin polarization layer on the membrane surface, thus enhancing the effluent quality substantially. Interestingly, several researchers found that the cake layer acted as a "dynamic" membrane on top of the actual membrane, and also led to a greater rejection of volatile fatty acids (Choo and Lee, 1996b; Hu, 2004) and viruses (Fox and Stuckey, 2015a). These authors observed that virus rejection increased at low sparging rate due to membrane fouling which demonstrated that fouling can also be beneficial for effluent quality. This has important practical applications as the costs associated with tertiary treatment (activated carbon, sand filters and chlorination/ozonation) could significantly decrease if the SAMBR permeate quality can be fine-tuned using the sparging rate, but there is a lack of information regarding its feasibility and its impact on maintainable flux. Based on the available information in the literature it was hypothesized that effluent quality could be improved further due to concentration polarization and membrane rejection, i.e. by reducing the sparging rate, the cake layer should become thicker and permeate quality should improve. The aim of this paper was, therefore, to study the effect of sparging rate on effluent quality and membrane flux.

2. Materials and methods

2.1. High-strength leachate wastewater

The leachate used in this study was produced in a continuous bench scale hydrolytic reactor (20L) fed real components of municipal solid waste: 41.3% kitchen wastes, 10.8% garden wastes and 47.9% paper wastes on a wet basis according to a previous study (Trzcinski and Stuckey, 2009b). The leachate had the following properties: pH: 6.7-7.7, soluble chemical oxygen demand (SCOD-filtered through a 0.45 microns Sartorius filter) and ~~soluble COD~~: 530-2840 mg/L (average: 1,410 mg/L), total chemical oxygen demand (TCOD)~~total COD~~: 1.3-11.8 g/L (average: 7.3 g/L), volatile fatty acids: 30-980 mg/L as COD (average: 390 mg/L), ammonia-nitrogen: 7-140 mg N/L (average: 44 mg N/L), phosphorus: 3.9-24 mg P/L as orthophosphates (average: 11 mg/L).

2.2. Reactors and start-up

Two submerged anaerobic membrane bioreactors (SAMBRs) were fed in parallel with the OFMSW leachate at 5 days hydraulic retention time (HRT) and 300 days solid retention time (SRT). The two SAMBRs were three liter reactors fitted with a Kubota polyethylene flat sheet membrane with 0.1 m^2 of total surface and a pore size of 0.4 microns. A detailed description of the reactor can be found elsewhere (Trzcinski and Stuckey, 2009b). One pump was used to set a constant flux, and some of the permeate was recycled back to the SAMBR with a separate pump in order to control the HRT. Both SAMBRs were maintained at 35 ± 1°C. The biogas sparging rate was initially set at 5 $L.min^{-1}$ (LPM) to minimize cake formation on the membrane until steady-state in terms of SCOD concentration was achieved.

SAMBR1 was inoculated with 0.5 L of seed from a SAMBR fed on the same leachate at 5 days HRT. The volume was adjusted to 3 L with the anaerobic biomedium defined in Owen et al. (1979) so that the initial mixed liquor total suspended solids (MLTSS) and mixed liquor volatile suspended solids (MLVSS) were 3.3 and 2.5 g/L, respectively. SAMBR2 was inoculated with biomass from a 4 litre chemostat batch-fed (once a week) on a 8 g COD/L synthetic feed (Nachaiyasit and Stuckey, 1995) to assess the effect of culture on COD removal. The supernatant was discarded and the settled solids were used to inoculate SAMBR2. The volume was adjusted to 3L with the anaerobic biomedium defined in Owen et al. (1979) so that the initial MLTSS and MLVSS were 2.6 and 1.78 g/L, respectively.

2.3. Analytical and statistical methods

The measurement of pH (Jenway 3020 pH Meter) was accurate to within ±0.02 units. The mixed liquor total suspended solids (MLTSS), volatile suspended solids (MLVSS), soluble chemical oxygen demand (SCOD-filtered through a 0.45 microns Sartorius filter) and total chemical oxygen demand (TCOD) were measured as described in standard methods (APHA, 1999). Their coefficient of variation (COV) for ten identical samples was ±4%, 3.1%, 2.6% and 9.9%, respectively. Volatile fatty acids (VFAs) were measured using a Shimadzu gas chromatograph with a flame-ionized detector and a SGE capillary column (12mx0.53mm ID-BP21 0.5μm). The COV was ±3% for ten identical samples. Ammonia-nitrogen was measured using the nesslerization method by reading absorbance at 425 nm, and the COV was equal to ±6.6% for 10 identical samples. The measurement of orthophosphates was carried out according to the vanadomolybdophosphoric acid colorimetric method described in standard methods (APHA, 1999). The absorbance was read on a spectrophotometer at 470 nm, and the coefficient of variance for ten identical samples was ± 0.6%.

3. Results and discussion

3.1. Reactor start-up

The pH was in the range 6.9-7.2 in both SAMBRs throughout the study, and the only difference was in the different inoculum used to seed the SAMBRs. SAMBR2 was inoculated with an inoculum almost free of colloids from OFMSW leachate, whereas SAMBR1 was inoculated with bacteria acclimatised to the leachate medium but containing colloids that are known to be detrimental for the flux. The SCOD in the bulk and permeate of SAMBR1 and SAMBR2 are shown in Figures 1 and 2, respectively. COD removal was generally over 90%, except on a few days when the leachate had a low COD concentration. Although both reactors were started up at similar MLTSS of 3.3 and 2.6 g/L, the initial flux in SAMBR2 was 21.6 LMH and only 8.6 LMH in SAMBR1 (Figure 3); presumably this was due to the inoculum used for SAMBR2 containing less colloids. Nonetheless, as SAMBR2 was fed on the same leachate containing colloids, the transmembrane pressure (TMP) started to rise rapidly and the flux had to be reduced manually (Figure 3); to stabilize the TMP between 150 and 200 mbar, the flux was reduced to 10 LMH. This approach was followed by other researchers in order to avoid serious fouling of the membrane (Fox and Stuckey, 2015b; Howell et al., 2004). Fox and Stuckey (2015b) determined that the critical flux for the SAMBR operating with low strength synthetic wastewater at a 6 LPM sparging rate was 11.8 LMH, which is in line with our results. In only 30 days, the flux in SAMBR2 decreased to the same flux as in SAMBR1 meaning that the advantage of a ”colloid free” inoculum was effective only for a short period of time.

3.2. Effect of the Biogas Sparging Rate on permeate quality and COD Rejection

Previous work on the SAMBR has shown that the presence of a fouling layer is responsible for a cleaner effluent (Akram, 2006; Choo and Lee, 1996a; Harada et al., 1995). Both SAMBRs were fed on the same leachate at the same HRT of 5 days. That HRT was chosen to keep a relatively constant MLTSS in the SAMBR. The objective was to see whether the fouling layer results in an enhanced rejection, i.e. higher bulk COD, lower permeate COD, both together or only a higher bulk COD with no change in permeate COD. A lower permeate COD would suggest that bacteria attached to the membrane can degrade further the organics as they pass through the fouling layer. This would demonstrate if there is an active biofilm on the membrane which can further polish the wastewater, but there is currently no clear consensus about this among the scientific community.

As both SAMBRs were at steady-state, the sparging rate was set at 2 LPM on day 31 (Figures 1 and 2, top). It was observed that the permeate SCOD remained stable at around 350-360 mg/L in both reactors. In contrast, the TMP rose to circa 580 and 380 mbar, in SAMBR1 and SAMBR2, respectively (Figures 1 and 2, bottom). This indicated that the flux became greater than the critical flux as soon as the sparging rate was reduced, and that a cake layer formed very quickly above the critical flux; fouling was worse in SAMBR1. The flux was manually reduced to 6.1 and 7 LMH in SAMBR1 and SAMBR2, respectively, to avoid operation at high TMP. The TMP went back to 150 and 100 mbar in SAMBR1 and SAMBR2, respectively, but the permeate SCOD did not decrease. Although the flux had been manually reduced to 6.1 LMH in SAMBR1, the TMP slowly increased to 650 mbar at which point the flux was only 5 LMH (measured on day 49). Thus a cake layer had been formed at 6.1 LMH, but at a much lower rate than at 8.3 LMH. The rate of increase in TMP indicates the degree of compaction of the fouling layer which can translate to a denser and less permeable fouling layer.

The higher the rate of increase in TMP, the more compact the fouling layer, and the thicker the layer will probably be. Also the degree of compaction will probably determine the ease and the rate at which the fouling layer can be removed if the sparging rate is increased. At very high TMPs, Elmaleh and Abdelmoumni (1997) also reported a decrease in permeate flux with an increase in TMP which was attributed to the compaction of the foulant layer. Li et al. (2003) explained that cakes formed at higher flux and TMP are much more consolidated than cakes formed marginally above the critical flux.

In SAMBR2, although the flux had been manually reduced to 7 LMH, the TMP rose slowly to 400 mbar before day 42. On day 42, SAMBR 2 was set at a high sparging rate (10 LPM), while the sparging rate in SAMBR1 remained 2 LPM. As a result, the TMP in SAMBR2 dropped to virtually zero, while the permeate SCOD in SAMBR2 increased to about 440 mg/L on day 45. The increase in the sparging rate did not allow us to re establish the initial flux in SAMBR2, although the TMP dropped back to zero. Operation at 2 LPM led to a thicker fouling layer caused by CP, and an increase in bulk SCOD due to rejection by the CP layer. On the other hand, when the sparging rate was increased to 10 LPM, the permeate COD increased typically from 360 to 440 mg/L due to a better scouring of the membrane, which resulted in a thinner fouling layer and a better mass transfer of solutes, leading to higher SCOD concentration in the permeate.

The permeate SCOD in SAMBR1 remained constant below 400 mg/L even though the bulk SCOD was increasing due to a thick fouling layer generated by low turbulence at 2 LPM. This thick fouling layer led to enhanced rejection (Figure 4), which increased from 20% to 50% over time. As humic acids accumulated in the boundary layer they adsorbed to particulate matter in the fouling layer which resulted in a more compact and dense layer. Nghiem et al. (2006) observed that the highest TOC rejection coincided with the highest level of fouling which is consistent with our observations.

On day 52, the sparging rate in SAMBR1 was also set at 10 LPM and the TMP immediately dropped to 100 mbar. However, the effect on the permeate SCOD was not immediate. As SAMBR1 was kept for a long time at 2 LPM, the fouling layer was more consolidated and increasing the sparging flowrate to 10 LPM could not remove enough of the fouling layer to return to very low TMPs (0 mbar), as was the case in SAMBR2. Choo and Lee (1996b) also concluded that cake compaction over time had more impact on SCOD rejection by the membrane. It was only on day 59 that the permeate SCOD also rose to values around 440 mg COD/L as in SAMBR2. Thus seven days (day 59-52) were necessary to see the effect of a sparging rate increase in SAMBR1 which had initially used a low sparging rate for 21 days, while it only took 3 days in SAMBR2 that had initially used a low sparging rate for only 11 days. This highlighted the effect of a denser cake layer for which an increase in sparging rate was not immediately followed by an increase in permeate SCOD concentration.

The effect of the low sparging rate on the flux was immediate and detrimental. As can be seen from Figure 3, the flux drop was 4.8 (8.3 to 3.5 LMH) and 3 LMH (10 to 7 LMH) for SAMBR 1 and 2, respectively. The effect of low sparging rate was to increase the thickness of the fouling layer while the membrane could still let the low molecular weight solutes pass through its pores resulting in a constant permeate COD. Leachate from OFMSW contains a wide range of molecular weight organics and the higher molecular weight cannot pass through the fouling layer and remain in the bulk which causes the membrane rejection to increase. This size exclusion phenomenon is however not sufficient to make the permeate COD decrease. The fact that the permeate COD concentration did not decrease also suggests that biological degradation on the membrane did not play a significant role.

As a result of thicker fouling layer and pore blocking mechanisms, fewer molecules could pass which caused a significant increase in the bulk SCOD of both SAMBRs. Furthermore, the increase in sparging rate to 10 LPM did not allow for the initial flux to be recovered, showing that irrecoverable fouling occurred in both SAMBRs. The fouling was, however, worse for SAMBR1 because of the greater degree of compaction due to the longer period at the low sparging rate. Moreover, this increase in COD was faster for SAMBR2 showing that part of the cake layer could easily be scoured as indicated by the TMP returning to zero after the high sparging rate was applied. In contrast, in SAMBR1 the low sparging rate mode was kept for 21 days, and due to the greater compaction of the cake, removal of the reversible fouling layer was impossible, or much slower, as indicated by a persistently high TMP, and as a result the effect of a higher sparging rate took longer to take effect.

Li et al. (2003) also concluded that the removal of the cake strongly depends on its age; if the cake existed for a short period of time, the lift forces due to surface shear can break some of the enmeshed bonding between the bacteria, allowing for the removal of the cake in the form of flocs. On the other hand, if the cake has been built up for a longer period, the possibility of breaking the enmeshment is reduced. The aggregation of the cake will also depend on ionic strength: at high ionic strength, which is likely to be the case in landfill leachate, the electrical double layer of bacteria is compressed resulting in more interaction and binding with molecules which allows for a dense biofilm (Li et al., 2003).

Therefore, this experiment performed in duplicate in separate SAMBRs showed similar results; when the sparging was set at a low rate (2 LPM), no significant change was noticed in the permeate COD concentration suggesting that bacteria attached on the membrane do not play a significant role. The direct effect of 2 LPM was to increase the bulk SCOD due to a thicker fouling layer and low turbulence, while the permeate SCOD concentration remained constant. Once a higher bulk SCOD is established due to enhanced rejection, the high sparging rate suddenly decreased concentration polarization which allowed more solutes to permeate, which in turn led to an increase in permeate SCOD; this was delayed in the case of SAMBR1 due to a denser fouling layer. Nevertheless, the change in COD (about 80 mg/L or a 20% increase) was relatively small compared to the total COD concentration when changing the sparging rate from 2 to 10 LPM. When the bulk SCOD decreased due to biodegradation (see after day 70), then the permeate SCOD decreased accordingly, but not lower than previous concentrations.
These results are consistent with Choo and Lee (1996b) who, after the initial fouling of the membrane, observed a decrease in the SCOD in the bulk and the permeate due to acclimation of the inoculum, but afterwards the permeate SCOD remained relatively constant for the rest of the experiment and did not decrease any further although the flux was decreasing due to internal fouling and cake layer compaction. This suggests that there is a lower limit for the permeate SCOD concentration because low molecular weight recalcitrants will still be able to pass through the membrane pores. This study, therefore, showed that permeate COD cannot be lowered by manipulating the sparging rate. The common approach in membrane filtration at large scale is to

minimize the formation of the fouling layer by using a high sparging rate, and if for any reason the sparging pump loses its intensity over time, or failed, it would result in an increase in TMP, rejection of solutes and most likely constant permeate SCOD. Upon reactivating the sparging pump, this study has shown that a slightly higher COD can be expected temporarily in the permeate. This is relevant for plant operators as it may affect tertiary treatment (ozone dosage, activated carbon, sand filters, chlorination, or reverse osmosis).

Overall, it was demonstrated that the treatment of high-strength OFMSW leachate in the SAMBR was feasible at 5 days HRT, a MLTSS around 2-3 g/L, 10 LPM and under these conditions COD removal greater than 90% and fluxes of 3-7 LMH are possible at TMP levels lower than 200 mbar for 4 months without chemical cleaning of the membrane. Conventional anaerobic reactors such as continuously stirred tank reactors (CSTR), would typically require 30 days or longer to treat such complex high-strength wastewater which means that a considerably larger reactor (6x) would be required. The SAMBR can achieve good COD removal at relatively low HRTs due to the membrane which retains the slow growing methanogens and uncouples the HRT from the SRT. For large scale application where space is a constraint, the anaerobic membrane bioreactor can offer substantial advantages over conventional anaerobic treatment. This study emphasizes the importance of a high sparging rate in the case of anaerobic membrane bioreactors treating complex high strength wastewater such as landfill leachate. Practical recommendations for operation at larger scale include : maintaining a high sparging rate and its accurate monitoring, automatic permeate pump shut down in case of sparging pump failure so that filtration is not conducted without gas sparging, and a backup sparging pump to allow for easy maintenance and replacement.

4. Conclusion

This work has shown that the permeate COD of SAMBRs cannot be lowered by decreasing the sparging rate. The main effect of low sparging rate (2 LPM) was the enhanced rejection of the membrane and the significant flux drop while the permeate COD remained constant within the experimental timeframe. On the other hand, permeate COD increased due to higher bulk SCOD when the sparging rate was increased to 10 LPM, in which case the permeate COD increased from 360 to 440 mg/L due to a better scouring of the membrane, which resulted in a thinner fouling layer and a better diffusion through the biofilm. The time required to observe the increase in permeate COD depended on the compaction of the cake layer. The increase in sparging rate to 10 LPM allowed the reactor TMP to recover to 0 mbar, but not the initial flux.

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