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IGNACIO MARTIN GARCIA

SLUDGE FREE AND ENERGY NEUTRAL TREATMENT OF SEWAGE

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Supervisor: Dr. Bruce Jefferson

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ABSTRACT

Anaerobic biological processes have been recognised as the most suitable pathway towards sustainable wastewater treatment due to the lower energy required and the lower amounts of biosolids generated when compared to conventional aerobic technologies. The difficulties experienced with the implementation of anaerobic reactors for the treatment of low strength wastewater at low temperatures are related to the deterioration of treatment capacity and effluent quality due to inefficient removal of colloidal matter and biomass washout. Membrane technology can overcome the limitation of anaerobic bioreactors since they retain not only solids but also colloidal and high molecular weight organics. This thesis explores the potential of anaerobic membrane bioreactors as core technology for mainstream wastewater treatment. The impacts of seed sludge, temperatures and bioreactor configuration on treatment eficicency and membrane performance as well as nutrient removal using ion exchange resins are investigated.

Suspended and granular anaerbic membrane bioreactors produced compliant effluent BOD and COD concentrations for wastewater temperatures ranging from 6 to 25 ^{o}C. Prolonged start-up periods are required unless the bioreactors are preseeded. Seeding enabled compliant effluent to be provided within days of start up. Effluent organic concentrations were sufficiently low that downstream biological nutrient control becomes a less practical option and as such downstream processing of ammonia and phosphate is possible with modern adsorption resins enabling both very low effluent concentrations and recovery to be accomplished. Lower energy operation of the membrane is possible in the granular anaerobic MBR configuration which can operate with intermittent gas sparging to maintain sustainable fluxes due to a reduced colloid load on the membrane. Compared to a conventional activated sludge treatment scheme significant reductions in sludge and energy costs can be obtained by considering a flowsheet based on a granular anaerobic membrane bioreactor although the overall energy balance is strongly dependent of membrane operation.

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NOTATION AND ABBREVIATION

<	less than
>	greater than
%	per cent
=	equals
°C	degree Celsius
β	beta
α	gamma
$	dollar
±	plus or minus
£	pound sterling
°C	degree Celsius
μm	micrometre
AD	anaerobi digestion
AeMBRs	aerobic membrane bioreactors
Ae/F	aerobic flocculent sludge
Al	aluminium
AnMBRs	anaerobic membrane bioreactors
An/F	anaerobic flucculent sludge
An/G	anaerobic granular sludge
AS	activated sludge
ASP	activated sludge plant
BOD_5	biological oxygen demand after 5 days
BSA	bovine serum albumin
BV	bed volume
Ca	calcium
CF	critical flux
CFV	cross flow velocity
CH_4	methane
Cm	centimetre

cm^2	square centimetre
COD	chemical oxygen demand
COD_{EFF}	effluent chemical oxgen demand
COD_{IN}	influent chemical oxygen demand
COD_{WASTE}	waste sludge chemical oxygen demand
CSTR	continuous stirred tank reactor
d	day
d_{50}	volume weighted median particle size
DI	deionized water
dp	particle size
EBCT	empty bed contact time
Ecf	energy to control fouling
$E_{CF,G}$	energy to control fouling in sumberged configuration
$E_{CF,P}$	energy to control fouling with pumped crossflow configuration
eEPS	extracted extracellular polymeric substances
EGSB	expanded granular sludge blanket reactor
Ep	energy to permeate suction
E_{PER}	energy associated to permeate suction
EPS	extracellular polymeric substances
EPSc	carbohydrate fraction of extracellular polymeric ustances
EPSp	protein fraction of extracellulr polymeric substances
Eq	equation
eq	equivalents
et al.	and others
E_{tot}	total energy
EU	European Union
F:M	feed to microorganism ratio
Fe	iron
FS	flat sheet
g	gram
G-AnMBR	granular anaerobic membrane bioreactor
H	hour

H_2SO_4	sulphuric acid
HAIX	hybrid anion exchanger
HF	hollow fiber
HRT	hydraulic retention time
K	potassium
kDa	kilo Dalton
kPa	kilo Pascal
kWh	kilowatt hour
L	liter
LMH	liter per square meter per hour
m	meter
m^2	square meter
m^3	cubic meter
$m^3\ m^2\ h^{-1}$	cubic meter (gas) per hour and sqare meter of membrane
mbar	mili bar
mbar d^{-1}	mili bar per day
mbar min^{-1}	mili bar per minute
MBRs	membrane bioreactors
Meso	mesophillic
Mg	magnesium
mg	mili gram
mg g^{-1}	mili gram per gram
mg L^{-1}	mili gram per Litre
min	minutes
MLSS	mixed liquor suspended solids
MLVSS	mixed liquor volatile suspended solids
mm	mili metres
M_{O2}	mass of oxygen required for biological aeration
MT	multi tube membrane geometry
MW	molecular weight
N	nitrogen
Na	sodium

NaCl	sodium chloride
NaOCl	sodium hypochloride
NaOH	sodium hydroxide
NH_4^+	ammonium ion
NH_4-N	ammonia nitrogen
NO_3^-	nitrate
OLR	organic loading rate
OTE	oxygen transfer efficiency
P	phosphorus
PE	polyethylene
PO_3^{-4}	phosphate
PO_4-P	phosphourus in phosphate
PVC	polyvinylchloride
PVDF	polyvinyldifluoride
Px	sludge production
Q	influent flow
SADm	specific aeration demand normalized against membrane area
SEM	scanning electron micorscope
SF	sustainable flux
SGDm	specific gas demand normalized against membrane area
SMP	soluble microbial product
SMPc	carbohydrate content of soluble microbial product
SMPCOD	soluble microbial product measured as COD
SMPp	protein content of soluble microbial product
SOTE	specific oxygen transfer efficiency
SRT	sludge retention time
SS	suspended solids
Thermo	Thermophilic
TMP	transmembran pressure
TN	total nitrogen
TNeff	totoal nitrogen in effluent
TN_{INF}	Influent total nitrogen

TOC	Total organic carbon
TS	Total solids
UASB	upflow anaerobic sludge blanket reactor
UF	ultra filtration
Ug	gas velocity
UL	liquid velocity
VFA	volatile fatty acids
VSS	volatile suspended solids
WAS	waste activated sludge
WWT	wastewaeter treatment
Yobs	Observed yield
α	alpha factor

CHAPTER 1:
INTRODUCTION

CHAPTER 1. INTRODUCTION

1.1 PROJECT BACKGROUND

Aerobic biological processes such as the activated sludge system are the predominant wastewater treatment technology employed around the world. The activated sludge process comprises a reactor in which the biomass responsible for the biodegradation of influent organics is aerated and kept in suspension (Metcalf & Eddy, 2003). Its main feature is the formation of flocculent sludge that can be easily separated from the treated water by gravity sedimentation, allowing for low effluent suspended solids concentrations and as a result the possibility of applying different sludge and hydraulic retention times by recycling of the biomass back into the system. The main advantages of aerobic biological processes are the high treatment efficiency they are able to achieve, robustness against changes in environmental conditions (e.g. temperature) and the possibility of removing nitrogen and phosphorous when combined with anoxic and anaerobic processes respectively. However in aerobic waste water treatment (WWT) high energy demands and sludge productions are observed. Biological aeration and sludge treatment and disposal represent approximately 50-60 % of overall energy demands and operational costs of wastewater treatment plants (Metcalf & Eddy, 2003). The costs related to these assets are expected to increase due to the demand for higher wastewater treatment efficiencies and restrictions on disposal routes for treated water and excess sludge posing a significant technical challenge to wastewater treatment companies. Furthermore, these will also face additional charges based on regulations related to mitigation greenhouse gas emissions that penalize both energy consumption and biosolids production (Greenfield & Batstone, 2005). Therefore it is important to investigate the potential of alternative flow sheets which are able to deliver the required treatment capacity for both carbonaceous and nutrient pollutants whilst reducing the use of resources, energy demands and waste by products.

In this regards anaerobic biological processes have been recognised as the most suitable pathway towards sustainable wastewater treatment systems (Zeeman & Lettinga, 1999; Lettinga *et al.,* 2001b; Verstraete *et al.*, 2009). In anaerobic biological processes air is not required for removal of pollutants and only 10-20 % of the organic matter is assimilated into biomass (van Haandel & Lettinga, 1994) reducing thus both energy demands and sludge production as compared to aerobic wastewater treatment. Additionally, as a result of anaerobic digestion of organic matter, biogas which can be used as energy resource and recycled into the process is generated. Despite these advantages anaerobic bioreactors are more sensitive to environmental conditions such as low temperatures and biomass loss due to lower growth rates of anaerobic bacteria as compared to aerobic biomass. The feasibility of anaerobic technology as the core treatment step for mainstream wastewater treatment has been assessed in the last 20 years through the study of the high rate anaerobic reactors (Lettinga *et al.,* 2001a). In its various configurations, these systems rely on either the settlement of granular or flocculent biomass or the generation of a biofilm to enable high concentration of stable microbial populations to remain within the reactor. The difficulties experienced with the application of such technology to domestic wastewater treatment are related to the low physical removal of solids and colloids (Mahmoud *et al.,* 2003) rather than conversion of organics to methane (Rebac *et al.,* 1999) resulting in the deterioration of effluent quality and inefficient control of sludge retention time. Inefficient control of biomass retention is of special importance in the treatment of low strength wastewater at low temperatures, because the loss of biomass in the effluent is not compensated with the growth inside the reactor (Lettinga *et al.,* 2001a; Lant & Hartley, 2007).

Membrane technology can overcome the limitation of anaerobic bioreactors. Membrane bioreactors (MBRs) involve the use of membranes coupled to a biological reactor in order to achieve efficient separation of treated water from sludge generated. These two technologies have been regarded as complementary as the filtration process provides complete control over sludge retention time and rejects colloids and high molecular weight organics that are retained within the reactor until further mineralized and that otherwise would be lost in the effluent (Ince *et al.,* 2000). Although introducing a membrane might enhance the performance of existing anaerobic technologies, most of the research has been conducted with easily biodegradable synthetic wastewaters and at mesophilic temperatures (35-37°C) which depending on influent wastewater temperature can only be achieved at the expense of significant amounts of energy (Lettinga *et al.,* 2001a). Further research is therefore required to ascertain whether the combination of anaerobic treatment and membrane technology can provide enough effluent quality when treating low strength wastewaters and low temperatures.

The adoption of aerobic MBRs (AeMBRs) as a viable alternative for domestic wastewater treatment applications was driven by the introduction of turbulent aeration onto submerged membrane systems due to the significant decrease in energy demands associated to fouling control as compared to cross flow pumped operation (Le-Clech *et al.*, 2006). Although in recent years a number of studies have incorporated submerged membranes to anaerobic reactors, there have been limited attempts to optimize membrane operation through the investigation of the effect of gas sparging intensity (Imasaka *et al.*, 1989; Jeison & van Lier, 2006a) or backwashing (Vallero *et al.*, 2005; Jeison & van Lier, 2006b) on fouling amelioration. Additionally direct comparison of membrane configurations and geometries which is a issue of major interest in AeMBRs (Günder & Krauth, 1999; Le-Clech *et al.*, 2005; Guglielmi *et al.*, 2007; Guglielmi *et al.*, 2008) so as to ascertain the system that provides better hydraulic performance with lower energy, demands have been limited in anaerobic membrane bioreactors (Hu & Stuckey, 2006; Jeison & van Lier, 2007). Taking into account that the operational costs in submerged MBRs are mainly determined by the energy required for membrane operation, further research is required to determine whether the energy demanded to control fouling in anaerobic MBRs (AnMBRs) can be compensated by the energy generated by methane production or offset energy demands for aeration in aerobic systems.

Finally when considering a flow sheet built around AnMBRs as in the present study, downstream processing of effluent also has to be considered. Aerobic biological post treatment for removal of nitrogen and phosphorous is subjected to the carbonaceous removal efficiency of the AnMBR and to the concentration of easily biodegradable substrate in the effluent (Chernicharo, 2006). Additionally biological nutrient removal would increase both energy demands and sludge production of the overall flowsheet. Nitrification by autotrophic bacteria can represents a significant fraction of total aeration costs and although this energy is partially recovered by the denitrification step it requires additional carbon which is sourced either form influent solids or externally supplied with easily biodegradable substrate. In the case of phosphorous, as biological removal requires simple substrates which are not always available at the required concentration, chemical precipitation with metal salt such as Al, Fe or Lime is often practised (Metcalf & Eddy, 2003) resulting in higher costs arising from chemical addition and sludge production. Given that nitrogen and phosphorous in wastewater are present predominantly in the form ammonia and phosphate ions, physical means such as ion exchange resins have been considered not only for their simplicity and low effluent nutrient concentrations but also as an opportunity for their recovery, closing the natural cycles through production of fertilizers or other valuable materials (Zeeman & Lettinga, 1999; Verstraete *et al.,* 2009).

1.2 PROJECT DEVELOPMENT

The work presented in this document was commissioned by Severn Trent Water and Yorkshire Water to determine the potential for sludge free, energy neutral processes for wastewater treatment. The work was funded through support from the commissioning parties linked to a Marie Curie Host Fellowship for Early Stage Research Training on Process optimisation and fouling control in membrane bioreactors for wastewater and drinking water treatment (MBR-Train) [MEST-CT-2005-021050].

1.3 AIMS AND OBJECTIVES

It is hypothesised that the combination of anaerobic biological process and membrane filtration can enhance the performance of existing anaerobic technologies resulting in sludge free and energy neutral process for treatment of sewage. The aim of this thesis is therefore to investigate the potential of AnMBRs as core technology for mainstream wastewater treatment. Accordingly a series of objectives were identified:

1- To investigate the bioreactor and membrane operational parameters as well as sludge characteristics that have greatest impact on membrane fouling in anaerobic systems and to compare these relationships to those found in AeMBRs so as to ascertain whether knowledge transfer exist between them.
2- To investigate the potential of a self inoculated AnMBR and to compare biological and membrane performance of AnMBRs with more established AeMBR systems.
3- Characterise AnMBRs in terms of treatment performance under experimental conditions similar to real full scale applications.
4- To ascertain whether suspended or attached anaerobic bioreactor configurations present important advantages with respect to treatment efficiency and membrane fouling.

5- To investigate the most effective fouling control strategy in AnMBRs.
6- To determine the potential of nutrient removal from AnMBR effluents using ion exchange resins.

1.4 THESIS PLAN

The experimental work was completed Nacho Martin Garcia in collaboration with Maria Mokosch from Dresden University (Dresden, Germany), Victor Monsalvo from Universidad Autonoma de Madrid (Madrid, Spain) and Alberto Sanvia from Padova University (Padova, Italy) who contributed to the work presented in chapters 4 and 5, chapter 6 and chapters 5 and 7 respectively. All chapters were written by the primary author and edited by Dr. Bruce Jefferson (supervisor). This thesis is presented as a series of chapters which have been formatted for journal papers as follows:

Initially, a literature review was conducted investigating the interactions between bioreactor operational parameters and sludge properties as well as the influence of membrane operational parameters on fouling, so as to ascertain whether knowledge transfer between fouling in aerobic and anaerobic systems exists. *Chapter 2, Paper 1:* Martin Garcia, N., Pidou, M., Judd, S. J. and Jefferson, B. *Comparison of fouling characteristics in aerobic and anaerobic MBRs.*

In Chapter 3, literature data obtained in Chapter 2 was employed in order to assess the energy balance between aerobic and anaerobic MBRs through model calculations. The model components included membrane energy demands associated to both sidestream and submerged configurations as well as the energy required from biological aeration and recovered from methane production in aerobic and anaerobic systems respectively. This bench study allowed identification of the most promising membrane and bioreactor configuration which could be further investigated in the ensuing experimental work. *Chapter 3, Paper 2:* Martin Garcia, N., Pidou, M., Judd, S. J. and Jefferson, B. *Comparison of energy requirements in aerobic and anaerobic MBRs.*

In chapter 4, the potential of self inoculated AnMBRs with low strength domestic wastewater was examined and compared to the start up of unseeded AeMBRs and seeded AnMBRs.

Under steady state conditions the aerobic and anaerobic systems were compared in terms treatment efficiency, sludge production, and membrane fouling. *Chapter 4, Paper 3:* Martin Garcia, N., Mokosch, M., Pidou, M., Judd, S. J. and Jefferson, B. *Start-up and performance of aerobic and anaerobic MBRs treating domestic wastewater.*

Chapter 5 then compared long term operation of suspended and granular submerged AnMBRs with respect to treatment efficiency and membrane performance. The effect of gas sparging intensity on fouling amelioration in both systems was investigated using short and long term experiments. Fouling control by continuous and cyclic gas sparging together with backflushing was investigated so as to ascertain which bioreactor configuration required lower energy demands for fouling control and thus present the highest potential to deliver energy neutral treatment of sewage. *Chapter 5, Paper 4:* Martin Garcia, N., Mokosch, M., Pidou, M., Judd, S. J. and Jefferson, B. *A comparison between granular and suspended anaerobic MBRs.*

In Chapter 6, the effect of gas sparging rate on fouling in tubular and hollow fibre membranes was investigated and compared to sidestream crossflow operation. Comparison of filtration behaviour when challenged with sludges of different characteristics such as those from granular and suspended AnMBRs provided insights into the relative impact of different biomass characteristics on membrane fouling. *Chapter 6. Paper 5:* Martin Garcia, N., Monsalvo, V., Pidou, M., Judd, S. J. and Jefferson, B. *Comparison of fouling control strategies in aerobic and anaerobic MBRs.*

In the final result section, Chapter 7, equilibrium isotherms with model solutions and column trials with real effluent from the granular AnMBR system were conducted in order to evaluate capacity and bed life of two ion exchange resins removing phosphorous and ammonia in order to assess the potential to operate adsorption processes for downstream removal of non degraded nutrients. *Chapter 7. Paper 6 :* Martin Garcia, N., Sanavia, A., Pidou, M., Judd, S. J. and Jefferson, B. *Ammonia and phosphate removal from granular anaerobic membrane bioreactor effluent using ion exchange resins.*

The overall impact of the research is discussed in Chapter 8, in which an energy cost balance analysis of the proposed flow sheet is presented. The model calculations are based on the data obtained during the experimental work and compared to a conventional activated sludge flow sheet in terms of energy demands and operational costs. A summary of the thesis plan is detailed in Table 1.1.

Table 1.1 Summary of thesis plan

Chapter/Paper	Objective addressed	Summary of title	Intended for submission to
2/1	1	Fouling in anaerobic MBRs	Separation Science and Technology
3/2	1	Modelling the energy demands of aerobic and anaerobic MBRs	Environmental Technology
4/3	2,3	Start up and performance of aerobic and anaerobic MBR treating low strength sewage.	Water research
5/4	4	A comparison of granular and suspended anaerobic MBRs.	Water Research
6/5	5	Comparison fouling control strategies in aerobic and anaerobic MBRs	Journal of membrane Science
7/6	6	Nutrient removal from granular AnMBR using ion exchange membranes	Separation Science and Technology

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CHAPTER 2:
COMAPRISON OF FOULING CHARACTERISTICS IN AEROBIC AND ANAEROBIC MBRs

CHAPTER 2. COMPARISON OF FOULING CHARACTERISTICS IN ANAEROBIC AN ANAEROBIC MEMBRANE BIOREACTORS

Martin Garcia, N., Pidou, M., Judd, S. J. and Jefferson, B.
Centre for Water Science, Cranfield University, Cranfield, Beds, MK43 0AL, UK

ABSTRACT

Fouling is the main drawback in membrane bioreactors and has been extensively studied, mainly in aerobic systems. Research has principally focused on 1) identifying the characteristics of biological suspensions that determine fouling propensity 2) investigating the influence of environmental factors and bioreactor operational parameters on the chemical and physical properties of biological suspensions and 3) reducing and controlling fouling through optimisation of membrane operation. In the present chapter a literature research was undertaken in order to better understand the relationships between these factors in anaerobic systems and to ascertain whether knowledge transfer exist between fouling characteristics in aerobic and anaerobic MBRs. Analysis of literature data revealed that the levels of bound/extractable EPS (eEPS) are higher in aerobic MBRs than in anaerobic MBRs. Opposite to eEPS, higher levels of soluble microbial product (SMP) which have been widely reported to increase fouling propensity in aerobic systems, have been found in anaerobic systems. However even higher appears to be the difference when the colloidal fractions are considered as shown by a number of recent studies that have reported the presence fine solids in the range of 1 μm to 10-15 μm. This highly dispersed structure is likely to determine fouling characteristics in anaerobic systems and limit knowledge transfer from aerobic MBRs.

Keywords: Fouling, anaerobic, membrane bioreactors, biomass characteristics, SMP, EPS.

2.1 INTRODUCTION

Membrane technology and anaerobic biological processes are regarded as complementary when treating high and low strength particulate wastewaters (Liao *et al.*, 2006), where full-scale applications currently exist (Kanai *et al.*, 2010). Anaerobic operation allows both the energy demand of the MBR process and the sludge production to be significantly reduced due to the absence of aeration requirements and lower biomass yields of anaerobic microorganisms as compared to most commonly employed aerobic technologies. However, fouling represents a key limitation to MBRs decreasing permeate production and increasing the frequency of chemical cleaning and, possibly, membrane replacement (Chang *et al.*, 2002). Fouling depends on a number of factors that have been reviewed and classified by several authors (Chang *et al.*, 2002; Le-Clech *et al.*, 2006; Meng *et al.*, 2009) as relating generally to membrane characteristics, biomass properties and operational parameters. Biological suspensions have a high fouling potential attributable to bacterial flocs, colloidal species and dissolved organics and inorganic compounds all of which can increase the resistance to filtration (Chang *et al.*, 2002). The size distribution and concentration of organics in the anaerobic MBR (AnMBR) mixed liquor determines the characteristics and mechanisms of fouling, and thus informs of the most effective fouling control strategies, as compared with the more extensively studied aerobic MBRs (AeMBRs).

Anaerobic MBRs have been reviewed with respect to general applications, operating parameters and biological performance (Liao *et al.*, 2006) and impacts on control of membrane permeability specifically (Bérubé *et al.*, 2006). In both reviews the importance of fouling behaviour was acknowledged but not considered in depth. In order to complement the current knowledge, in the present review study the influence of bioreactor operational parameters on biomass characteristics of AnMBRs are reported and their impact on membrane fouling analysed and compared to those found in AeMBRs, so as to ascertain the main factors that determine differences in fouling behaviour and characteristics between these two systems. In addition, the current study updates with recent literature in which submerged membrane configuration has been employed in AnMBRs.

2.2 BIOMASS CHARACTERISTICS

Differences in characteristics between aerobic and anaerobic sludge are most readily attributed to the different mechanisms involved in the biological process. Aerobic biological suspensions mainly comprise microorganisms, decay products and influent solids forming microbial aggregates which are held together by high molecular weight polymers excreted by the own bacteria allowing them to exist at high population densities (Laspidou & Rittmann, 2002) in virtue of their high biomass yield and growth rates. It has been reported that the structure, morphology and surface properties of such flocs can be altered by changes in physiological state of the biomass induced by changes in bioreactor operational parameters such as sludge retention time (SRT) and feed to microorganisms ratio (F:M) (Liao *et al.*, 2001).

On the other hand anaerobic degradation of wastewater with dissolved, colloidal and particulate organic matter, involves several sequential steps such as hydrolysis, acidogenesis, acetogenesis and methanogenesis (Batstone, 2006). Hydrolysis is thought to be a surface based reaction that takes place on influent solids which are converted to simple monomers by extracellular enzymes excreted by hydrolytic and fermentative bacteria (Sanders *et al.*, 2000; Vavilin *et al.*, 2008). As a result and due to the low hydrolysis rates and biomass yield of anaerobic bacteria, the reactor solid inventory is considered to be mainly constituted by influent particulates (Soto *et al.*, 1993) that are of reduced particle size (Elmitwalli *et al.*, 2001) and density (Lant & Hartley, 2007). Therefore as opposed to aerobic systems sludge properties are probably more dependent on influent characteristics than on bioreactor operational parameters. Some physical characteristics such as particle charge, which affects colloidal interactions, have been reported to remain unchanged after digestion (Elmitwalli *et al.*, 2001). It has been reported by various studies (Wilén *et al.*, 2000) that aerobic sludge deflocculates under anaerobic conditions, due to the release of EPS from the biological matrix, leading to an increased supernatant turbidity and reduced filterability (Rasmussen, 1984).

As with aerobic systems, the key foulant characteristics are concentration and particle size. The latter is affected by diffusive and back transport enhanced by shear applied at the membrane-solution interface, which is less effective for smaller colloids and high molecular weight solutes than for the larger microbial flocs (Belfort *et al.,* 1994; Wisniewski *et al.,* 2000). According to the size of the foulant, fouling mechanisms are classified as cake filtration, product of deposition of larger solids on the membrane surface and complete, standard or intermediate blocking which reflects the accumulation of macromolecular and colloidal organics on the membrane pores. Additionally, the critical flux which sets the boundary between fast and slow fouling rates in MBRs is believed to represent the transition between fouling by sludge flocs and soluble/colloidal matter respectively (Pollice *et al.,* 2005; Le-Clech *et al.,* 2006). Since accumulation of the soluble colloidal fraction of the sludge is expected to be proportional to their concentration and to membrane throughput under constant flux operation (Guglielmi *et al.,* 2007b) its importance on membrane fouling is widely recognized in aerobic (Lesjean *et al.,* 2005; Pollice *et al.,* 2005; Fan *et al.,* 2006; Rosenberger *et al.,* 2006) and anaerobic systems (Harada *et al.,* 1994; Cho & Fane, 2002; Hu & Stuckey, 2006; van Voorthuizen *et al.,* 2008).

2.2.1 Mixed liquor suspended solids

Although fouling by sludge flocs is not considered as the main fouling mechanisms under low flux operation, mixed liquor suspended solid (MLSS) concentration have shown to negatively affect membrane fouling in AnMBRs. For instance Jeison (2006) found that changing biomass concentration by diluting with water and concentrating the mixed liquor by membrane filtration had a greater impact on the formation of a cake layer than varying gas sparging intensity. To illustrate, at a solid concentration of 25 gMLSS L^{-1} the critical flux increased from 15 LMH to around 21 LMH as the gas sparging rate increased from 1.2 to 2.4 m^3 m^{-2} h^{-1}. At the highest gas sparging rate an increase from 9 to 21 LMH was observed when the biomass concentration decreased from 50 gMLSS L^{-1} to 25 gMLSS L^{-1}. Le Clech (2003) also reported that the effect of MLSS was higher than gas sparging, although the higher critical fluxes were obtained for higher biomass concentration of 12 gMLSS L^{-1} as compared to the lowest of 4 gMLSS L^{-1}.

The contrasting influence that solid concentration has on membrane hydraulic performance in aerobic and anaerobic MBRs can be attributed to differences in the relationship between biomass and dissolved/colloidal compounds in the mixed liquor. While in aerobic MBRs it has been widely reported that higher levels of soluble microbial products are found at lower MLSS especially when short sludge retention times are applied (Lee *et al.*, 2003; Massé *et al.*, 2006; Liang *et al.*, 2007) in anaerobic systems SMP tend to accumulate together with biomass (Harada *et al.*, 1994) or at high sludge ages (see soluble/colloidal SMP section). For instance Ghyoot (1997) reported that fluxes at concentrations below 13 g L^{-1} total solids (TS) appeared to be higher than those recorded at 20 and 25 gTS L^{-1}. Analysis of the nutrients in the membrane digested primary sludge revealed that, on increasing the biomass concentration from 6 to 25 gTS L^{-1} the colloidal COD (8 μm filtered) increased from 69 to 716 mgCOD L^{-1}. Beaubien (1996) studied the impact of transmembrane pressure (TMP), crossflow velocity (CFV) and suspended solids (SS) concentration on membrane flux and observed that permeability decreased from 0.6 to 0.2 μm s^{-1} kPa^{-1} as suspended solid concentrations increased from 0.4 to 2.5 gSS L^{-1} remaining constant thereafter. Since permeability appeared to be independent of CFV at Reynolds numbers between 2000 and 15000, the author attributed this trend to higher concentrations of pore plugging particles rather than viscosity and concentration polarisation.

2.2.1 Particle size distribution

According to Lant and Hartley (2007) compared to aerobic flocs, anaerobic sludge presents particle sizes an order of magnitude lower, even though the range of particle sizes covers three orders of magnitude as opposed to only one for aerobic biomass. Additionally while aerobic effluents present low concentrations of suspended solids containing particle sizes much smaller than the associated sludge flocs, high concentrations of small particles of similar size to those found in the mixed liquor were present in anaerobic effluents. Although similar median particle sizes ranging from 5.2 to 80 μm and from 3 to 90 μm have been reported in aerobic and anaerobic MBRs respectively (Table 2.1), a significant difference between these systems is the presence of a population of fine colloidal matter in the latter which has been shown to negatively affect membrane performance. For instance Imasaka (1989) reported a bimodal particle size distribution for a methane fermentation broth, with peak values of 0.8 μm and 4 μm that were attributed to single microoganisms and their flocs respectively. Visual inspection of a fouled membrane by SEM analysis revealed that spherical particles of about 0.8 μm of diameter, assumed to be Methanosarcina, were deposited on the membrane surface. Similary, sludge morphology analyzed by light microscopy revealed the absence of sludge flocs in the mixed liquor of two thermophilic AnMBRs fed on different substrates where the sludges were comprised of abundant rod shaped microorganisms (Jeison *et al.,* 2009).

Table 2.1 Particle size distributions in aerobic and anaerobic MBRs

Reactor Type/ Configuration	Membrane Geometry/Configuration	Median dp (μm)	Reference
AnMBRs			
CSTR	MT/pumped	13	Elmaleh (1997)
CSTR	MT /pumped	3	Choo (1998)
UASB	MT /pumped	16	Bailey (1994)
UASB	MT /pumped	10	Cho(2002)
CSTR	FS-HF/gas sparged	60-65	Hu (2006)
CSTR	MT/gas sparged	70-90	Jeison (2006)
UASB	FS/gas sparged	50	Lin (2009)
AeMBRs			
CSTR	HF/sparged	5-7	Lee (2003)
CSTR	HF/sparged	80	Masse (2006)
CSTR	HF/sparged	15-30	Huang (2008)
CSTR	HF/sparged	20	Yu (2003)
CSTR	HF/sparged	15-60	Meng (2006)
CSTR	MT/pumped	50	Defrance (2000)

Analysis of particle size distributions in AnMBRs (Table 2.1) indicates that side stream systems yield average particle sizes between 0.8 μm (Imasaka *et al.,* 1989) and 16 μm (Bailey *et al.,* 1994) whilst immersed systems using gas sparging have particle sizes between 65 μm (Hu & Stuckey, 2006) and 90 μm (Jeison & van Lier, 2006). These results suggest that the reduction in particle size for sidestream MBRs, which is generally attributed to the floc breakage during pumping in crossflow operation, is not only responsible for the decreased biomass activity reported in some studies (Brockmann & Seyfried, 1997) but also for the deterioration of the filtration performance. Significantly lower critical fluxes were reported in an AnMBR operated in sidestream pumped configuration in comparison with a submerged system due to the lower particle size distribution induced by the higher shear at which the biomass was exposed (Jeison *et al.*, 2009). In another study Cho (1996) attributed the fast initial flux reduction from 60 to 15 LMH within 4 days to the decrease in average particle size from 16 to 6 μm. Comparison of flux evolution of a sludge in which particle size had been reduced to 7 μm prior to crossflow filtration at 1.25 m s^{-1} with the same sludge without such a particle size reduction revealed that in the first case a low, stable flux of 5 LMH was achieved whilst for the second the flux decreased from 18 to 5 LMH as particle size decreased from 16 to 6 μm.

A part from hydrodynamic conditions, reactor operating temperature has also been shown to influence biomass character. Comparison of mesophilic and thermophilic AnMBRs operated at the same MLSS concentration of 8 gMLSS L^{-1} showed that the latter contained a higher fraction of fine colloids between 1 and 15 μm and provided a 5 fold increase in cake layer resistance over that of the mesophilic system. Analysis of the cake layer revealed a higher compactness and lower porosity and moisture content for the thermophilic system (Lin *et al.*, 2009). Similarly Jeison and van Lier (2006; 2007) reported slightly higher fouling rates in thermophilic systems as compared to a mesophilic AnMBR although the latter was operated at fluxes three times higher than the former.

2.3 ORGANIC FOULING BY EPS

Extracellular polymeric substances (EPS) have been widely reported as being responsible for organic fouling in both aerobic (Lesjean *et al.*, 2005; Pollice *et al.*, 2005; Fan *et al.*, 2006; Rosenberger *et al.*, 2006) and anaerobic MBRs (Harada *et al.*, 1994;

Cho & Fane, 2002; Hu & Stuckey, 2006; van Voorthuizen *et al.,* 2008). These biopolymers, composed mainly of polysaccharides, proteins and lipids, have been fractionated according to whether they are found in the sludge supernatant as soluble microbial products (SMP) or bound to the sludge flocs and are thus extracted from the cell walls (eEPS). Although the term SMP implies that these substances are of bacterial origin, they may also be the result of recalcitrant or partially transformed influent organics, especially in anaerobic systems at lower temperatures where lower biodegradation rates apply than those found for aerobic biomass. However, independent of their origin, analysis of eEPS and SMP has contributed to further characterisation both of the solid and the colloidal/soluble fractions of biological suspensions respectively.

2.3.1 Bound/extractable EPS

The surface properties of the sludge are primarily determined by the eEPS, and the hydrophobicity and surface charge have been correlated with the total EPS concentration and the ratio of proteins to carbohydrates in both conventional activated sludge and MBR systems (Liao *et al.,* 2001; Lee *et al.,* 2003). For instance, increasing ratios of proteins to carbohydrates in activated sludge have been reported to enhance bioflocculation through the reduction of surface charge and increase in hydrophobicity (Liao *et al.,* 2001). On the other hand high proportions of carbohydrates in the eEPS are associated with a more dispersed sludge structure due to the greater repulsion between sludge particles and interaction with the aqueous phase resulting from the higher negative surface charge and reduced hydrophobicity (Liao *et al.,* 2001).

Literature data regarding surface properties of anaerobic sludge do not correlate as consistently as those from aerobic systems, and no conclusions can be drawn regarding comparative levels of eEPS and fractions thereof to allow comparison between aerobic and anaerobic sludges with respect to surface charge and hydrophobicity. For instance Morgan (1990) reported that aerobic sludges were more negatively charged, contained higher levels of total EPS and lower ratios of proteins to carbohydrates than anaerobic granules. These results support the findings of Daffonchio (1995) , who demonstrated that although most acidogenic microorganisms are hydrophilic, anaerobic granules are predominantly hydrophobic. Anaerobic digestion of primary (Houghton *et al.*, 2000) and activated sludge (Novak *et al.*, 2003) has also been reported to yielded a higher ratio of proteins to carbohydrates but the total eEPS concentration appeared to decrease and increase respectively. Comparative results by Liao (2001) and Jia (1996) using acetate as substrate show that under anaerobic conditions more EPS is generated but that the ratio of proteins to carbohydrates is higher in the aerobic systems.

However, analysis of literature data (Table 2.2) suggests that both eEPS and ratio of proteins to carbohydrates are higher in aerobic MBRs than in anaerobic systems. Expressed as percentage of total volatile suspended solids (VSS), the eEPS content of aerobic and anaerobic sludges ranges from 2.5 to 13.3 % and 2 to 5.7 % respectively. Comparison of anaerobic and aerobic MBRs operated with complete retention of solids and fed with settled sewage (Baek & Pagilla, 2006) showed levels of EPS to continuously decrease to a concentration of 26.6 mg $gVSS^{-1}$ and 32.6 mg $gVSS^{-1}$ respectively. Another study of AnMBRs (Lee *et al.*, 2008) attributed the fast fouling rate observed after a stable operation for 28 days to the sudden increase in eEPS from 30 to 235 mgTOC L^{-1}. A similar trend was reported by Fawehinmi (2004) who observed an increase in specific resistance to filtration as the EPS content of crushed granular sludge increased from 20 to 130 mg $gVSS^{-1}$. These concentrations of eEPS are amongst the highest found in AnMBRs, and are probably the result of the rupture of the granular structure. Indeed, the lowest levels of eEPS found in AnMBRs have been reported for an expanded granular sludge bed system (EGSB), in which granules provided 4.4-6.6 mg $gVSS^{-1}$ and 0.6-1.6 mg $gVSS^{-1}$ of eEPS as carbohydrates ($eEPS_C$) and proteins ($eEPS_P$) respectively (Chu *et al.*, 2005). Analysis of the cake layer revealed that the amount of eEPS on the solids deposited on the membrane surface was twice of that found in the granules and that the ratio of $eEPS_P$ to $eEPS_C$ had increased from 0.2 to 0.5. In contrast, Lin (2009) reported a decrease of 50 % in the ratio of proteins to polysaccharides when comparing sludge eEPS with fouling material deposited on the membrane of mesophilic and thermophilic AnMBRs. Regardless of the major fouling component, the discrepancies between biomass and cake layer eEPS composition found in these studies suggest that soluble or colloidal compounds are also responsible for the increase in membrane resistance in AnMBRs.

Table 2.2 Concentration and composition of eEPS from aerobic and anaerobic MBR sludge

System configuration	EPS tot (mg $gVSS^{-1}$)	EPSp (mg $gVSS^{-1}$)	EPSc (mg $gVSS^{-1}$)	Ratio EPSp:EPSc	MLSS (gMLSS L^{-1})	K (LMH bar^{-1})	Reference
AnMBR							
An/G	5-7.7	0.6-1.6	4.4-6.1	0.1-0.36	14-21	10	Chu (2005)
An/F	52	24	28	0.86	8	72	Lin (2009)
An/F	57	32	25	1.28	8	9.6	
An/F	24.9	18.8	6.1	3.01	9.4	25-125	Lee (2008)
An/F	75^{*}	16-50	1.5	8	6.5	-	Fawehimni (2004)
AeMBRs							
Ae/F	63-70	30-36	35-28	1	2.8-5.5	-	Lee (2003)
Ae/F	81-115	57-88	24-29	2.4	10-16	40-90	Trussell (2007)
Ae/F	133^{+}	81	52	1.6	18	-	Tek (2009)
Ae/F	40-70			2-4	1.9-6	-	Masse (2006)

*Measured as COD. +sum of proteins and carbohydrates. G:granular sludge. F: flocculent sludge

2.3.2 Soluble-colloidal EPS: SMP

It has been reported that the soluble organic matter in the effluent from the biological treatment processes is predominantly SMP which comprises the soluble cellular components released during cell lysis, lost during synthesis, or otherwise excreted for some purpose (Laspidou & Rittmann, 2002). They are classified according to their origin as products associated with biomass growth and are produced at a rate proportional to the rate of substrate metabolism and non-growth associated products related to cell lysis. It has been recognised for some time that, as with eEPS, the concentration of SMP normalized against influent COD is higher in aerobic (3.1 %) than in anaerobic systems (0.2 to 2.5%), as reported by Barker and Stuckey (1999) based on earlier reported work (Noguera *et al.,* 1994; Kuo *et al.,* 1996). This is due to the lower biomass uptake and decay rates of anaerobic microorganisms compared to aerobic biomass.

Comparative studies of MBR systems with conventional reactors have revealed the concentration of SMP to be higher in MBRs than for conventional biotreatment for both aerobic (Massé *et al.,* 2006) and anaerobic processes (Aquino & Stuckey, 2006). This arises because the high molecular weight organic fraction is retained by the membrane (Massé *et al.,* 2006) and because higher SMP production arises by endogenous decay and cell lysis as a consequence of long sludge age operation and high loadings (Harada *et al.,* 1994). However, converse to trends reported for conventional reactors, AnMBRs have slightly higher SMP levels than aerobic systems. Analysis of literature data shows that normalized levels SMP with respect to influent COD ranges from 10 to 22 % and from 10 % to 50 % for aerobic (Table 2.4) and anaerobic MBRs (Table 2.3) respectively. Direct comparison between aerobic and anaerobic MBR systems operated in parallel (Baek & Pagilla, 2006) has revealed residual COD concentrations in mixed liquor supernatant to be higher in the latter. However, colloidal matter appears to differ more significantly, with an order of magnitude difference in colloid concentration between aerobic and anaerobic systems being reported for only an 80% difference in soluble COD concentration (van Voorthuizen *et al.,* 2008b).

While the higher colloidal content of the AnMBR sludge compared with that of aerobic systems may reflect higher levels of free bacteria in the mixed liquor, the higher soluble organic concentration may result from lower biodegradation rates or SMP biodegradability under anaerobic conditions (Ince *et al.*, 2000). There is also evidence of high molecular weight (MW) polymeric matter of up to 1000 kDa being retained by the cake layer in AnMBRs, presumably decreasing its permeability and possibly increasing its rejection capability and thus accounting for the relatively high SMP levels found in the mixed liquor supernatant and the low MW (<1.5 kDa) of the permeate organic matter (Harada *et al.*, 1994). Analysis of SMP concentrations from AnMBRs (Table 2.3) seems to corroborate trends reported from conventional anaerobic chemostats, which indicate that higher levels of SMP are produced at higher wastewater strengths, sludge retention times (Noguera *et al.*, 1994; Kuo *et al.*, 1996) and lower temperatures (Schiener *et al.*, 1998). For instance, SMP concentrations of 150 mgCOD L^{-1} (Chu *et al.*, 2005; Hu & Stuckey, 2006) have been reported at SRTs of 145-150 days, while at a lower SRT of 30 and 60 days (Huang *et al.*, 2008) only 39 and 56 mgCOD L^{-1} were found respectively. Decreasing temperature from 25 °C by 15 °C has been shown to increase SMP_{COD} from 50 to 150 mgCOD L^{-1} (Ho & Sung, 2010).

The influence of organic loading on SMP production seems to depend on whether it responds to changes in hydraulic residence time or influent organic concentration. Hu and Stuckey (2006) showed that the mixed liquor soluble COD remained at a concentration of 150 mg COD·L^{-1} as HRT decreased from 48 and 3 hours (influent COD 460 mgCOD L^{-1}), supporting previous findings (Baek & Pagilla, 2006). Despite the number of variables involved in the production of SMP, comparative results shown in Table 2.3 seem to indicate that SMP production increases with wastewater strength. The low SMP to influent COD ration of 0.025-0.085 for wastewater strength of 10 gCOD L^{-1} reported by Lin (2009), compared to 1.2 gCOD.L^{-1} and 1.8 gCOD.L^{-1} reported by Harada (1994) and Akram and Stuckey (2008) for influent substrates concentrations of 5 gCOD L^{-1} and 4 gCOD L^{-1} respectively may be a result of employing a ethanol as influent substrate.

Table 2.3 Concentration and chemical composition of SMP in aerobic and anaerobic MBRs

SRT (d)	HRT (h)	Temp. (°C)	SMP^{*}_{COD} ($mg\ L^{-1}$)	SMPc (mg L)	SMPp (mg L)	MLSS ($g\ L^{-1}$)	K ($LMH\ bar^{-1}$)	Reference
AnMBRs								
-	19.7	35	250 (0.0025)	-	-	8	72	Lin (2009)
-	77	55	850 (0.0085)	-	-	8	9.6	
∞	15	35	1200 (0.24)	-	-	15	40-80	Harada (1994)
-	12	35	327 (0.28)	80	400	-	40	Voorthuizen (2008)
-	12	35	269 (0.23)	81	70	-	-	
∞	12	15	150 (0.3)	45	69	12	9-71	Ho and Sung (2010)
∞	12	25	50 (0.1)	-	-	7	9-71	
150	2.6	35	142 (0.3)	-	-	2.6	67	Aquino(2006)
> 250	15	35	1789 (0.45)	19	52	12.4	40	Akram (2008)
∞	24	30	51 (0.24)	-	-	7.1	10	Baek (2006)
30	12	25	39 (0.07)	-	-	5.5	-	Huang (2008)
60	12	25	56 (0.1)	-	-	5.7	-	
∞	8	12	180 (0.5)	-	-	6.5	-	Fawehimni (2004)
AeMBRs								
20-60	7.8	-	36-42 (0.12-0.14)	8	59	2.8-5.5	-	Lee (2003)
10-30	-	-	37-82 (0.1-0.21)	-	-	10-16	40-90	Trussell (2007)
10-53	16	-	45-80 (0.12-0.22)	-	-	1.9-6	-	Masse (2006)

[a] Normalized against influent soluble COD. [*] Values in brackets correspond to normalized SMP_{COD} with respect to influent COD.

2.4 FOULING CONTROL STRATEGIES

The higher operational cost of MBRs compared to conventional wastewater treatment systems are mainly due to the energy required to control fouling, which comprise control of hydrodynamic conditions, physical and/or chemical cleaning or simply flux reduction (Judd, 2008). Hydrodynamic control in immersed systems is through scouring of the membrane with air or biogas in the case of aerobic and anaerobic MBRs respectively. This is to be distinguished from sidestream systems where membrane scouring is by conventional liquid crossflow.

2.4.1 Submerged membrane configuration

The operational costs related to membrane operation in submerged MBRs are mainly determined by the relationship between the specific gas demand (SGD_m) and operating flux, the SGD_m being the gas flow rate per unit membrane area (the specific aeration demand, SAD_m, for aerobic systems). This reflects the relationship between the convective flow towards the membrane produced by permeate suction and the back transport induced by the gas sparging by tangential shear at the boundary layer (Liu *et al.,* 2003). In AeMBRs sustainable or critical fluxes have been reported to increase by increasing gas intensity up to a certain threshold value beyond which no further increase in flux is observed for flat sheet (Ueda *et al.,* 1997; Guglielmi *et al.,* 2008), hollow fiber (Guglielmi *et al.,* 2007a) and multitubular membranes (Le-Clech *et al.,* 2006). For instance, Yu (2003) reported an increase in critical flux from 7.3 LMH to 50.2 LMH as the specific gas demand increased from 0.08 to 0.68 m^3 m^{-2} h^{-1} in an AeMBR operated at a biomass concentration of 3 gMLSS L^{-1}. Analysis of full scale immersed AeMBRs (Verrecht *et al.,* 2008) indicated values of SAD_m between 0.21 and 0.88 m^3 m^{-2} h^{-1} for a selection of plants operating under optimal conditions, with fluxes between 24 and 31 LMH.

Increasing membrane flux, has been shown to increase fouling rates and decrease the duration of the slow fouling phase in both aerobic and anaerobic MBRs under conditions of sub-critical flux operation, prior to the widely reported "TMP jump" (Pollice *et al.,* 2005; Le-Clech *et al.,* 2006). For instance, Zhang (2006) reported increased fouling rates from 0.0016 to 0.12 kPa h^{-1} and decreased filtration time prior to the TMP jump from 280 hours to 48 hours when the flux was increased from 10 to 30 LMH. Under sub critical conditions a decrease in flux from 10 LMH to 2 LMH caused an exponential decrease in the fouling rate, from 19.8 to 0.46 kPa h^{-1} and prolonged the time before the TMP jump to up to 8 days (Brookes *et al.*, 2006). Results from a recent pilot scale study (Guglielmi *et al.*, 2007a) suggest that the duration before the TMP jump decreases linearly at fluxes close to the critical flux and that an asymptote exists at a certain flux below which operation can be extended to long filtration cycles.

Recent research into submerged AnMBRs (Table 2.4), suggests that while the flux range between 2.4 LMH (Lin *et al.*, 2009) and 12 LMH (Hu & Stuckey, 2006), the SGD_m widely varies between effectively no gas sparging (Wen *et al.*, 1999; Chu *et al.*, 2005) and 3 m^3 m^{-2} h^{-1} (Imasaka *et al.*, 1993; Hu & Stuckey, 2006; Lee *et al.*, 2008). A study of the impact of upflow velocity on fouling rates of a hollow fibre membrane/based AnMBR operated under intermittent cycles of 3 minutes filtration and 1.5 minutes relaxation showed that increasing upflow velocities from 3 m h^{-1} to 8 m h^{-1} produced insufficient shear to sustain the flux (Chu *et al.*, 2005). Although increasing gas sparging intensities appear to be effective in extending membrane operation, the increase of shear provided to the membrane surfaces does not enhance permeate flux or reduce permeability. For instance Hu and Stuckey (2006), employed 3 m^3 m^{-2} h^{-1} at a flux of 8 LMH resulting in a stable TMP of 0.4 bars during 90 days of operation. Stable permeability of 40 LMH bar^{-1} after 30 days of operation was also reported by (Imasaka *et al.*, 1993) for the same SGDm but with a liquid cross flow velocity across the membrane module of 0.2 m s^{-1} and periodic backwash of 30 second every half an hour. A stable TMP of 100 kPa was maintained for 20 days in a mesophilic AnMBR operated at a flux of 7.2 LMH with a gas sparging intensity of 1.5 m^3 m^{-2} h^{-1} (Lin *et al.*, 2009). Overall the permeability is much lower than for AeMBRs which for full scale domestic wastewater treatment plants is between 150-250 LMH bar^{-1} (Judd, 2006) even when the amount of gas provided to the membrane is up 4 times higher and fluxes between 2 and 3 times lower.

Table 2.4 Membrane performance of submerged anaerobic MBRs

Reactor type / sludge	**Membrane material/ Pore size (μm)**	**SGD ($m^3\ m^{-2}\ h^{-1}$)**	**Flux (LMH)**	**TMP (kPa)**	**Fouling rate ($kPa\ h^{-1}$)**	**t op (h)**	**Filtration cycle Filtration/relaxation**	**Reference**
UASB	PVDF/0.22	1.8	25		0.33-2.52		-	Wu (2009)
UASB	PET	0	5	<30	0.04-0.08		-	An (2009)
UASB	PE / 0.1	0	10.4	<100		480	3 min/1.5 min	Chu (2005)
CSTR	-	0.4	5.5	-	-	3600	8 min/2 min	Huang (2008)
CSTR	PE/250*	3	8-12	45	0	>2160	Continuous filtration	Hu (2006)
CSTR	PE/0.45	3	5	2-10	0.0083	720	7 min/3 min	Lee (2008)
UASB	PE/0.03	0	5	< 70	0.2	336	4 min /1 min	Wen (1999)
UASB		0	5	< 70	0.2	336		
Granular	PVDF/250*	0.27-0.54	10	-	-	-	8 min/ 1 min [b]	v.Voorthuizen (2008)
CSTR		1.35	8	-	-	-		
UASB (Meso)	PVDF/70*	1.5	7.2	10	0	450	-	Lin (2009)
UASB (Thermo)		1.5	2.4	25	0	250	-	

*Daltons, +VSS, [b] backwashing

2.4.2 Physical and chemical cleaning. Reversible, irreversible and irrecoverable fouling

As with aerobic systems, cake/gel layer fouling seems to dominate resistance to filtration in AnMBRs. Chu (2005) reported a total resistance to filtration of 10.65×10^{13} m^{-1} from which 90.0 % corresponded to cake resistance while only 8.9 % was internal fouling. Similarly, Lin (2009) reported a total resistance of 8.1×10^{13} m^{-1} and 1.53×10^{13} m^{-1} in a thermophilic and mesophilic AnMBR respectively with a contribution of cake resistance of 95.7 % and 89.2 % respectively. Lee (2008) divided the contribution of external fouling between cake and gel layer, the first one removed by flushing with water and the second one by removing the remaining material with a sponge. The results showed that gel layer resistance contributed to 69-75 % of the total fouling, while cake layer resistance only represented between 10 and 13 % of total fouling. Choo and Lee (1996) observed such an attachment of biosolids to the membrane that biomass concentration in the reactor volume decreased from 7 gMLSS L^{-1} to less than 1 gMLSS L^{-1} in 20 days, with a consequent decline in membrane flux of over 90 %. Although the reduction in solid concentration was attributed to cell lysis due to shear stress caused by membrane recirculation pump, fouling characterisation interestingly revealed that the major contributors to hydraulic resistance were concentration polarisation layer and cake layer accounting for 82 % and 16 % of total resistance which was 20.9×10^{13} m^{-1}.

Resistance to filtration due to membrane fouling can also be classified as reversible, irreversible or irrecoverable (Judd, 2006) depending on whether it can be removed physically during operation (by relaxation or backflushing), chemically or it if it remains after chemical cleaning. In submerged AeMBRs, physical cleaning procedures like relaxation and backwashing have shown to be effective in extending membrane operation compared to continuous filtration (Zhang *et al.*, 2005) reducing the chemical cleaning frequency. By applying physical cleaning procedures it is possible to apply fluxes which result in an increase of resistance to filtration as long as the cake layer deposited onto the membrane surface can be removed by relaxation or backflushing. For instance, operational cycles of 10 minutes of filtration followed by one minute relaxation at fluxes between 22.3 and 28.5 LMH, resulted in fouling rates during filtration cycles of 1.39 mbar.min^{-1} and 1.8 mbar.min^{-1} respectively, while the irreversible fouling rates were two orders of magnitude lower (Guglielmi *et al.*, 2007a).

Although reversible fouling has often been related to cake layer formation, they do not necessarily equate to each other. Reversible fouling refers to a fraction of the cake layer that is loosely bound and can be removed during operation whereas the irreversible component refers to the fraction of cake layer that is more strongly bound and is linked to internal fouling. For instance, Jeison and van Lier (2006) reported the development of a permanent cake layer that could not be removed using relaxation and backflushing in combination with gas sparging during long term operation but that could be removed to the same degree employing chemical cleaning and flushing the membrane surface with pressurized water. In an anaerobic immersed multitubular membrane system operated at a flux of 4.7 LMH, Vallero (2005) reported a decrease in residual fouling rate from 137 to 13 mbar d^{-1} employing continuous filtration and relaxation for 4 minutes every 12 minutes respectively. Implementation of backwash at double the permeate flux during two minutes, further decreased the fouling rate to 4.5 mbar d^{-1}. Another study (van Voorthuizen *et al.,* 2008) also showed that although resistance increased within 8 min of operation from $2x10^{11}$ to $1.6x10^{13}$ m^{-1} equating to an increase in TMP of 20 kPa at a flux of 8 LMH, the irreversible fraction was maintained below $10x10^{11}$ m^{1} by completing the filtration cycle with 1 min relaxation followed by 1 min backflusing at 30 LMH. A filtration cycle of 10 min followed by a backwash with compressed air for 5 seconds allowed operation during 50 days during which a flux decline of 19 % was observed, despite the increase in reversible fouling from $0.04x10^{13}$ m^{-1} up to $28.9x10^{13}$ m^{-1} within the filtration cycle (Lee *et al.,* 2001). Studies employing sidestream pumped membrane operation have also shown how once the cake layer is formed, it is difficult to remove it by increasing cross flow velocity (Imasaka *et al.,* 1993) or by flow stopping and depressurisation (Choo & Lee ,1996).

In anaerobic MBRs, different degrees of permeability recovery have been achieved after chemical cleaning. Chu (2005), reported that chemical cleaning with 0.03 % NaOCl was as efficient as using 0.03 % NaOCl followed by 0.3 % H_2SO_4, but that the permeability recovery was only complete after 70 hours. In the previous examples in which the membrane was operated for 50 days (Lee *et al.,* 2001) and 165 days (van Voorthuizen *et al.,* 2008) the residual fouling reduced membrane permeability by 11 % and 77 % respectively. Similarly, He (2005) found that after 2 or 3 chemical cleaning cycles of 1 hour with 0.5 % NaOH, the ratio between clean water flux before and after chemical cleaning stabilized at 40 %. However, relative flux reduction referred to the initial operative flux increased in four months of operation up to 70 %.

2.5 CONCLUSIONS

The present literature review reveals significant differences with respect to biomass characteristics and fouling behavior between aerobic and anaerobic MBRs which can be summarized as follows:

1. Both eEPS and ratio of proteins to carbohydrates are higher in AeMBRs than in anaerobic systems. Expressed as percentage of total VSS, the EPS content of aerobic and anaerobic sludges ranges from 2.5 to 13.3 % and 2 to 5.7 % respectively. However there is a lack of knowledge regarding the relationship between EPS concentration and composition and surface properties such as charge and hydrophobicity.
2. Although similar median particle sizes have been reported in aerobic and anaerobic MBRs, the presence of a population of fine solids with particle sizes ranging from 1 to 10 microns have been widely reported in the latter and associated to biomass of high fouling propensity.
3. Normalized levels SMP with respect to influent COD ranges from 10 % to 22 % and from 10 % to 50 % for aerobic and anaerobic MBRs respectively. However an order of magnitude in the difference in the concentration of SMP between aerobic and anaerobic sludges has been observed when the colloidal fraction is considered. The main operational parameters that have shown to enhance SMP production in AnMBRs are low temperatures, extended sludge ages and influent organic concentration.
4. The effect of turbulent gas sparging on membrane performance in immersed AnMBRs seems to be limited as compared to aerobic systems, indicating that fouling is more determined by sludge properties than by membrane operational conditions: Fluxes between 1/3 and1/2 and permeabilities below 50 % of those reported in AeMBRs have been reported in AnMBRs for specific gas demands between 50 % and 300 % higher.
5. Although as with aerobic systems the predominant fouling mechanism in AnMBRs has been reported to be cake filtration, contradictory results with respect to the effectiveness of membrane backwashing at reducing membrane fouling and permeability recovery after chemical cleaning have been reported and would require further research.

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CHAPTER 3 :
MODELLING THE ENERGY DEMANDS OF AEROBIC AND ANAEROBIC MBRs

CHAPTER 3. MODELLING THE ENERGY DEMANDS OF AEROBIC AND ANAEROBIC MBRs

Martin Garcia, N.[1], Pidou, M.[1], Judd, S. J.[1] and Jefferson, B.[1]

[1]Centre for Water Science, Cranfield University, Cranfield, Beds, MK43 0AL, UK

ABSTRACT

The current paper presents a model study in which the energy requirements in aerobic and anaerobic MBRs have been assessed so as to compare the energy balance between these water treatment technologies. The main components of the model included biological aeration (AeMBRs) and energy recovery from methane production (AnMBRs) as well as the energy demands of submerged and side stream membrane configurations. Aeration and membrane energy demands were estimated based on previously developed model studies populated with published literature data. Given the difference in sludge production between aerobic and anaerobic systems benchmarking of the model proceeded through comparison of energy demands of AnMBRs against AeMBRs operated at high sludge retention times or complete retention of solids. Analysis of biogas production in AnMBRs reveals that heat balance is only accomplished for influent wastewater strengths above 4-5 gCOD·L^{-1} when mesophilic temperatures are considered. The general trend of submerged configuration being less intensive than sidestream in aerobic systems in not observed in AnMBRs, mainly due to the wide variation in gas demands utilized in anaerobic systems. Compared to AeMBRs at high SRT or complete retention of solids for which energy requirements were estimated to approach 2 kWh·m^{-3}, the energy demands associated to fouling control in AnMBRs are lower although due to the low fluxes reported in literature capital costs associated to membrane material would be three times higher.

Keywords: Energy, membrane bioreactors, aerobic, anaerobic, submerged, crossflow.

3.1 INTRODUCTION

Aerobic membrane bioreactors represent a specific subset of bioreactor technology where the membrane replaces alternative means of solid liquid separation such as a gravity sedimentation tank. The impact of including a membrane results in the complete uncoupling of the hydraulic and sludge retention times providing greater operational flexibility and the potential to intensify the biological process. The advantages and disadvantages of MBRs are often quoted (Judd, 2006) but perhaps the key ones are improved effluent quality, lower sludge production (advantages) and membrane fouling with an associated high energy demands (disadvantages). Despite these disadvantages, several studies reveal that the membrane bioreactors market is expected to grow in industrial and municipal applications in both Europe (Lesjean & Huisjes, 2008) and North America (Yang *et al.,* 2006). The principle applications that are driving such market growth relate to either situations where tight effluent consents have to be meet, a small footprint is required, robust disinfection is required or the water is to be reused (Judd, 2006).

Anaerobic biological processes are mainly applied for high strength industrial wastewaters at high temperatures as an alternative to aerobic treatments because of the lower energy demands they require due to the absence of aeration, the possibility of recovering energy from the methane in the biogas produced and reduced biomass product and associated disposal costs. However, the main drawbacks of anaerobic technology are the lower effluent qualities they generate, especially when operated at low temperatures and wastewaters of low strength, the potential to generate odours and a need for downstream nutrient removal. Traditionally, anaerobic reactors have utilised granular sludge as a method of biomass retention although in more recent studies the potential of utilising membranes has been discussed (Ross & Strohwald, 1994). In particular benefit has been reported in relation to the high solids retention even at low temperatures and the rejection of high molecular weight organics which are further degraded and that otherwise would be lost in the effluent.

Translating the concepts to municipal sewage treatment indicate that adoption of AnMBRs will result in reductions in both energy use and sludge production. Given the current demands to reduce the energy and carbon footprint of sewage treatment consideration of AnMBRs appears timely. Recent reviews concerned with AnMBRs have looked at the impact of operational parameters on biological performance (Liao *et al.*, 2006) and parameters affecting membrane flux (Bérubé *et al.*, 2006). The main conclusions drawn from these reviews were the need to assess the feasibility of both side stream and submerged configuration to ascertain optimum fouling control strategies to minimise the overall energy demands.

The current paper addresses this point by comparing energy balances for AnMBR and AeMBRs for the treatment of low strength wastewater. The aim of the study is to establish the overall changes in energy that can be expected as well as identifying the critical components controlling the overall energy balance to indicate where future improvements may be possible.

3.2 MODEL DEVELOPMENT

The overall assessment will compare the energy balance across aerobic and anaerobic membrane bioreactors. The main components of the model include: biological aeration (AeMBRs), energy recovery from methane production (AnMBRs) and energy demands associated with operation of either a submerged (Figure 3.1, right) or a sidestream (Figure 3.1, left) systems. Aeration and membrane energy demands of submerged systems were estimate based on previously developed model studies (Fletcher *et al.*, 2007; Verrecht *et al.*, 2008). In the case of sidestream configuration energy demands for sludge pumping were estimated based on pressure loss calculations along the membrane modules using a previously validated rheological model (Chilton and Stainsby, 1998). The models have been populated with published operational data from the literature where possible and standard assumptions made where necessary.

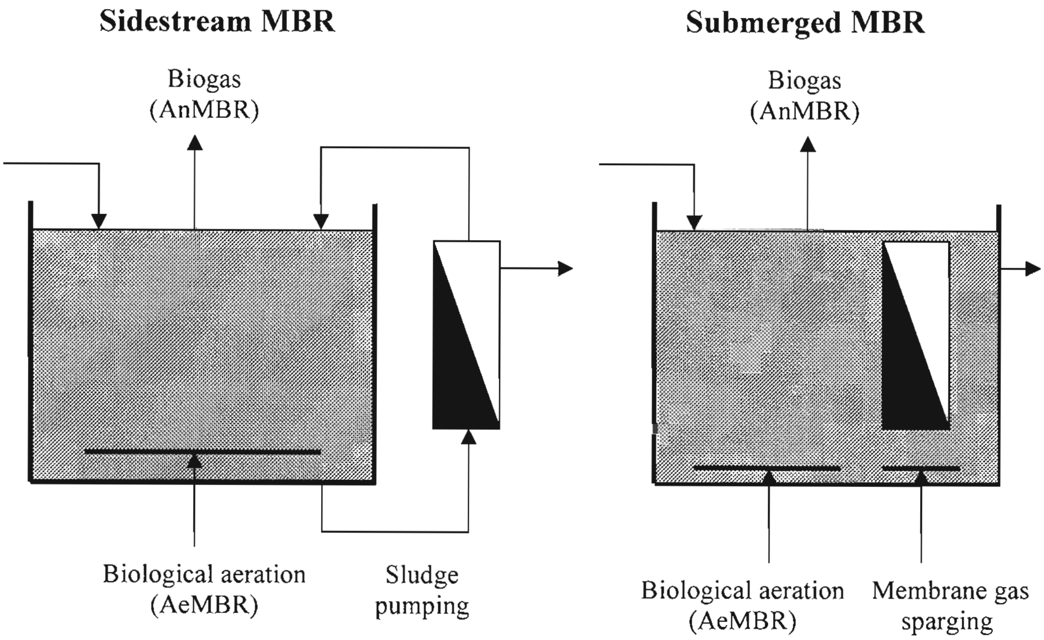

Figure 3.1 Schematic representation of crossflow (left) and submerged MBRs (right)

3.2.1 Biological aeration

Aeration demand for the AeMBR was based on the model of Verrecht (2008) adapted to use literature data as input parameters instead of direct kinetic modelling of the biological reactor. Aeration energy was calculated with respect to oxygen consumed M_{O2} by heterotrophic (organics) and autotrophic bacteria (nitrification) taking into account the reduction in oxygen due to denitrification in the anoxic zones of the bioreactor (Equation 3.1).

$$M_{O2} = Q(COD_{IN} - COD_{EFF}) + 4.33\,Q(TN_{IN} - TN_{EFF}) - 2.83QNO_{3,EFF}$$

[Eq. 3.1]

where Q represents the influent flow (L d^{-1}) and COD_{IN} and COD_{EFF} the influent and effluent COD concentrations respectively (mgCOD L^{-1}). The rest of the terms are related to the oxygen required for nitrification estimated from difference between influent and effluent total nitrogen (TN_{IN} and TN_{EFF} respectively) taking also into account the amount of oxygen saved by denitrification which was calculated with the effluent nitrate concentration, $NO_3^-{}_{EFF}$ (mg NO_3^--N L^{-1}). Sludge wastage was not included in the COD balance since AeMBRs operated at high SRT or with complete retention of solids were principally considered so as to provide a direct benchmark to the anaerobic systems (Table 3.1).

Table 3.1 AeMBR case studies and reported operational conditions

Parameter	Unit/Reference	Innocenti (2002)	Laera (2005)	Rosenberger (2002a)	Teck (2009)
Volume	L	1400	6	3500	20
HRT	h	14	8	14	8
SRT	d	∞	∞	∞	300
MLSS	$g\ L^{-1}$	16.6	22.9	16	18
MLVSS	$g\ L^{-1}$	8.7	17.2	11.2	16
COD_{in}	$mg\ L^{-1}$	300	400	790	1000
COD_{eff}	$mg\ L^{-1}$	19	57	10	5
TNin	$mg\ L^{-1}$	42.2	49.3	65.8	10.0
TNeff	$mg\ L^{-1}$	2	0.8	13	0.3
NO_3^-	$mg\ L^{-1}$	11.3	40.6	13	0.3

In the case of submerged MBRs, biological aeration demands $Q_{air,BIO}$ (Equation 3.2) were estimated considering the contribution of membrane gas scouring $M_{O2,MEM}$ (Equation 3.3) to the overall oxygen requirements M_{O2}, taking into account that the air flow required to control fouling $Q_{AIR,MEM}$ is provided by coarse bubble diffusers which are more effective in their scouring effect than fine bubble diffusers, but provide lower oxygen transfer efficiencies (Verrecht *et al.*, 2008).

$$Q_{air,bio} = \frac{M_{O2} - M_{O2,MEM}}{0.21 \cdot 1000 \cdot \alpha \cdot \beta \cdot \gamma \cdot OTE} \quad \text{[Eq. 3.2]}$$

$$M_{O2,MEM} = 0.21 \cdot 1000 \cdot \alpha \cdot \beta \cdot \gamma \cdot OTE_{MEM} \cdot Q_{Air,MEM} \quad \text{[Eq. 3.3]}$$

Where β and γ are oxygen transfer efficiency factors taken as 0.95 and 0.89 respectively. Oxygen transfer efficiencies (OTE) of 0.02 and 0.05 were assumed for membrane coarse bubble aeration and biological fine bubble diffusers while the α factor which accounts for the oxygen transfer efficiency relates to MLSS concentration according to Equation 3.4 (Günder & Krauth, 1999).

$$\alpha = e^{-0.082MLSS} \qquad \text{[Eq. 3.4]}$$

The power requirements associated to biological aeration were obtained from power consumption of a blower (Equation 3.4) delivering the correspondent air flow at the static pressure of the liquid column in the membrane tank h, which was considered to be 2 meters whenever it was not reported.

.

$$E_{Air,BIO} = \frac{108748 \cdot \lambda}{\xi \cdot (\lambda - 1)} \left[\left(\left(\frac{\rho \cdot g \cdot h + 101325}{101321} \right)^{(1-\frac{1}{\lambda})} - 1 \right) \right] \cdot Q_{air,BIO} \qquad \text{[Eq. 3.5]}$$

In Equation 3.4, ζ represents the blower efficiency (60 %) ρ the air density and λ is the heat capacity ratio which takes values of 1.4 for air and 1.3 for biogas.

3.2.2 Methane production

Under anaerobic conditions, biodegradation of organics takes place without the presence of oxygen or nitrate as an electron acceptor. The process end products are mainly methane and carbon dioxide which can be recovered as biogas or dissolved in the effluent. Energy associated with methane production in AnMBRs, was calculated with the biogas production data reported in literature studies assuming an energy content of 36500 kJ m^3 of biogas produced (Metcalf & Eddy, 2003). The highest methane yields reported in AnMBRs treating low strength wastewater range between 0.29 LCH_4 $gCOD^{-1}$ (Hu & Stuckey, 2006) and 0.33 LCH_4 $gCOD^{-1}$ (Cadi *et al.*, 1994) and correspond to those studies in which soluble and completely biodegradable substrate were employed as influent at mesophilic temperatures (35-37 °C). Lower methane yields of 0.12 LCH_4 $gCOD^{-1}$ (Chu *et al.*, 2005) and 0.08-0.09 LCH_4. $gCOD^{-1}$ (Huang *et al.*, 2008) have been reported with synthetic wastewaters but at temperatures ranging between 11-25 °C and 25-30 °C respectively, highlighting the importance of temperature in the production and recovery of biogas. Variable degrees of methanisation in AnMBRs treating real influents have ranged between 0.2-0.23 LCH_4 $gCOD^{-1}$ for screened sewage (Wen *et al.*, 1999), 0.27 LCH_4 $gCOD^{-1}$ for raw sewage (Saddoud *et al.*, 2006) and 0.09-0.12 LCH_4 $gCOD^{-1}$ for black water (van Voorthuizen *et al.*, 2008).

3.2.3 Membrane energy demands in submerged and sidestream configuration

Energy demands associated to membrane operation in MBRs were divided into components related to permeate pumping (E_P), and to fouling control (E_{FC}). In submerged MBRs (Figure 3.1, right) membranes are immersed in the mixed liquor and gas is sparged below the membrane module in the form of air or biogas in the case of aerobic and anaerobic MBRs respectively. In side stream pumped crossflow operation (Figure 3.1, left) the membrane module is located outside the bioreactor and the mixed liquor is pumped through the membrane module and recycled back to the bioreactor providing enough turbulence to enhance the back transport of foulants from its surface thus reducing membrane fouling. Although sidestream MBRs are usually operated at constant pressure with the pump generating both the liquid crossflow and the driving force for permeation (TMP), for direct comparison all systems were assumed to utilise permeate pumping irrespectively of system configuration. The energy required for permeate pumping was calculated as the product of permeate flow (Q_p) and the transmembrane pressure TMP (Equation 3.6).

$$E_{PER} = Q_P \, TMP \qquad \text{[Eq. 3.6]}$$

The energy required for fouling control E_{FC} was further subdivided into pumping $E_{FC,P}$ and gas sparging $E_{FC,G}$ for pumped crossflow and submerged configurations respectively. The power consumption required for fouling control in submerged configuration ($E_{FC,G}$) was obtained by replacing the biological aeration requirements ($Q_{AIR,BIO}$) in Equation 3.5 by the membrane gas demand $Q_{GAS,MEM}$ which is calculated according to:

$$Q_{GAS,MEM} = SGD_m \cdot A_m \qquad \text{[Eq. 3.7]}$$

where SGD_m denotes the specific gas demand normalized against membrane area ($m^3 \cdot m^{-2} \cdot h^{-1}$) which is equivalent to the specific aeration demand SAD_m employed in aerobic systems and A_m represents membrane filtration area. Although different relationships between flux and required SGD_m have been proposed in AeMBRs (Judd, 2006; Verrecht *et al.*, 2008), standard 50 % intermittent gas sparging intensities of 0.3 $m^3 \cdot m^{-2} \cdot h^{-1}$ and 0.5 $m^3 \cdot m^{-2} \cdot h^{-1}$ were employed for fluxes below and above 15 LMH respectively for the aerobic case studies shown in Table 3.1, all of which employed hollow fibre membranes. In the case of AnMBRs the SGDm reported in the different case studies were used to estimate the energy demands for fouling control in submerged configuration.

In the case of pumped sidestream configuration the energy associated to fouling control was calculated as the product of the tangential flow Q_{CFV} ($m^3\ s^{-1}$) through the membrane and the pressure loss ΔP (Pa) , considering a pump efficiency (ξ) of 60 % (Equation 3.8):

$$E_{CF,P} = \frac{Q_{CFV} \cdot \Delta P}{\xi} = \frac{CFV \cdot S_m \cdot \Delta P}{\xi} \qquad \text{[Eq. 3.8]}$$

where Q_{CFV} was obtained from the crossflow velocity (CFV, in m s^{-1}) and the cross sectional area S_m calculated from the geometric characteristics of the membrane modules reported in the different case studies. Pressure losses ΔP were estimated with Darcy –Weisbach equation (Equation 3.9) using the diameter (D) and length (L) of the membrane module and calculating Fanning friction factor (f) according to Colebrook's relationship (Equation 3.10). Chilton and Stainsby (1998) introduced a modified Reynolds number R' (Equation 3.11) in which the effective viscosity was calculated at the wall and which could be applied to the general Herschel Buckley model based on the previously defined generalized Reynolds number (Metzner & Reed, 1955). The parameter X obtained by solving Equation 3.11 represents the ratio between the yield stresses of the sludge τ_B and the shear stress at the membrane wall τ_w. Comparison of theoretical results based on rheological characterisation of sewage sludges and kaolin slurries predicted within a 15 % error of the experimentally measured pressure drop for crossflow velocities between 0.1 $m \cdot s^{-1}$ and 6 $m \cdot s^{-1}$ and so appear appropriate in the current case.

$$\Delta P = \frac{4\,\rho\, f\; CFV^2}{2\,D}\; L\; (\,turbulent\ conditions) \qquad [3.9]$$

$$f^{-0.5} = 4\, log_{10}(\,RE'\, f^{0.5}) - 0.4 \qquad [3.10]$$

$$RE = \frac{RE}{(1-X)^4} \qquad [3.11]$$

$$\tau_W = \mu_B\,(1-X)^{-1}\,(1-\tfrac{1}{3}X - \tfrac{1}{3}\,X^2 - \tfrac{1}{3}X^3)^{-1} \qquad [3.12]$$

The rheological parameters were obtained from the characterisation of aerobic and anaerobic MBR sludge given by Laera (2007) and Pevere (2007) respectively, in which the Bingham shear stress τ_B (Pa) and viscosity μ_B (Pa s^{-1}) were reported to increase with biomass concentration according to Equations 3.13 (Laera *et al.*, 2007) and 3.14 (Pevere *et al.*, 2007). A Bingham plastic rheological model was preferred to represent the behaviour of MBR sludge based on the constant viscosity presented in different studies at shear rates exceeding 100-500 s^{-1} (Rosenberger *et al.*, 2002b; Pollice *et al.*, 2006; Laera *et al.*, 2007). Additionally, adoption of the power law models which have been also been proposed (Rosenberger *et al.*, 2002b; Pollice *et al.*, 2006; Laera *et al.*, 2007; Pevere *et al.*, 2007), results in a decrease in frictional pressure losses with increasing solid concentrations due to the more prominent shear thinning behaviour that both aerobic and anaerobic sludges present when fitted to this rheological model.

$$\mu_B = 0.02894\, MLSS \qquad \tau_B = 0.001(0.233\, MLSS + 1) \qquad [Eq.\ 3.13]$$

$$\mu_B = 0.001e^{0.04\, MLSS} \qquad \tau_B = 0.067e^{0.07MLSS} \qquad [Eq.\ 3.14]$$

3.3 RESULTS AND DISCUSSION

3.3.1 Energy balance between aerobic and anaeobic MBRs

Analysis of the energy demands in AeMBRs with complete sludge retention show that irrespectively of organic load applied the total energy demand approaches 2 kWh m^{-3} for wastewater strength of 0.4 gCOD L^{-1} or above (Figure 3.2). Biological aeration represents as much as 88-93 % of total energy demands except for the lowest influent COD concentration in which membrane aeration provides 40 % of the biological oxygen requirements and accounts for 60 % of the total energy demands (1.2 kWh m^{-3}). Although operation of AeMBRs at high sludge ages or even complete retention of solids would avoid the high costs of sludge treatment and disposal which can represent between 40 and 60 % of total cost (Kennedy & Churchose, 2005), without having negative effects on biological performance (Muller *et al.,* 1995; Rosenberger *et al.,* 2002a; Laera *et al.,* 2005; Teck *et al.,* 2009), the high levels of MLSS accumulated in the system result in high energy requirements due to the reduction in aeration efficiency to below 20 % (Muller *et al.,* 1995; Günder & Krauth, 1999). As a result, in order to optimize operational costs, aerobic membrane bioreactors treating municipal sewage are usually operated at hydraulic retention times of less than 12 hours and variable sludge retention times between 15 and 50 days which aim to fix mixed liquor concentrations between 8-12 g MLSS L^{-1} (Judd, 2006). Different surveys have estimated their energy requirements to range between 0.6 and 1.2 kWh m^{-3} (Fatone *et al.,* 2007) distributed between membrane (60-70%) and biological aeration (30-40 %). Consequently an additional 0.8-1.4 kWh m^{-3} of energy is required to operate an AeMBR at low biomass production.

Results from model calculations reveal that the energy demands in submerged AnMBRs range from 0.03 to 5.7 kWh m^{-3} (Figure 3.3). Such variability in energy requirements for fouling control arises as a result of the wide range of gas demands reported in submerged configuration between effectively no gas sparging (Wen *et al.,* 1999; Chu *et al.,* 2005; An *et al.,* 2009) to 3 $m^3 \cdot m^{-2} \cdot h^{-1}$ (Imasaka *et al.,* 1993; Hu & Stuckey, 2006). In sidestream anaerobic MBRs energy demands range between from 0.23 to 16.52 kWh m^{-3} (Figure 3.4) with the variability being attributed to the impact that cross flow velocity and bioreactor MLSS have on flux and pressure losses. In contrast to aerobic MBRs, energy demands for submerged and sidestream anaerobic systems are within the same range due to the higher fluxes that have been reported in crossflow systems and to the uncertainty of appropriate gas sparging rates that result in sustainable membrane operation in submerged configuration which will be dealt with in the following section.

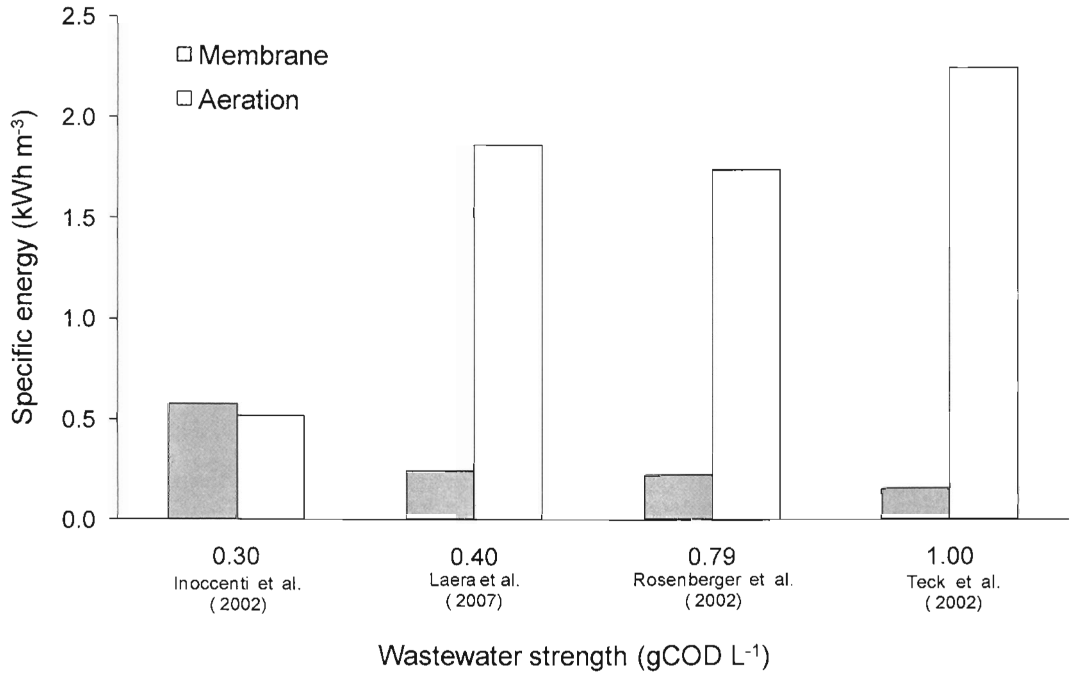

Figure 3.2 Aeration and membrane energy demands in AeMBRs

However literature data indicates that the available (electrical) energy produced ranges from 0.15 to 0.3 kWh m^{-3} as wastewater strength increases from 0.24 gCOD L^{-1} (Wen *et al.*, 1999) to 1.14 gCOD L^{-1} (van Voorthuizen *et al.*, 2008), showing that the amount of methane generated (expressed as heat in Figures 3.3 and 3.4) is sufficient to recover a significant proportion the total energy demand for sewage treatment and even offset the energy demands associated to fouling control. It is important to notice that the reported methane production in some studies (Cadi *et al.*, 1994; Hu & Stuckey, 2006; Saddoud *et al.*, 2006; van Voorthuizen *et al.*, 2008) were obtained at mesophilic conditions, and therefore lower biogas production can be expected at ambient wastewater temperatures given that biogas production from low strength wastewater is insufficient to compensate digester heat balance. This is illustrated with the horizontal lines depicted in Figures 3.3

and 3.4 which indicate the amount of energy required to heat the reactor up to 35°C for influent wastewater temperature of 15°C taking into account that 50 % of the energy could be recovered by heating the influent with the permeate. The heat recovered from biogas increases from 0.62 to 34.8 kWh.m^{-3} as wastewater strength increases for 0.24 gCOD L^{-1} (Chu *et al.*, 2005) to 10 gCOD L^{-1} (Lin *et al.*, 2009). Therefore, for domestic wastewater applications it is preferable energetically to operate without heating as the energy required to heat the reactor could only be compensated for influent COD concentrations above 5 gCOD L^{-1}.

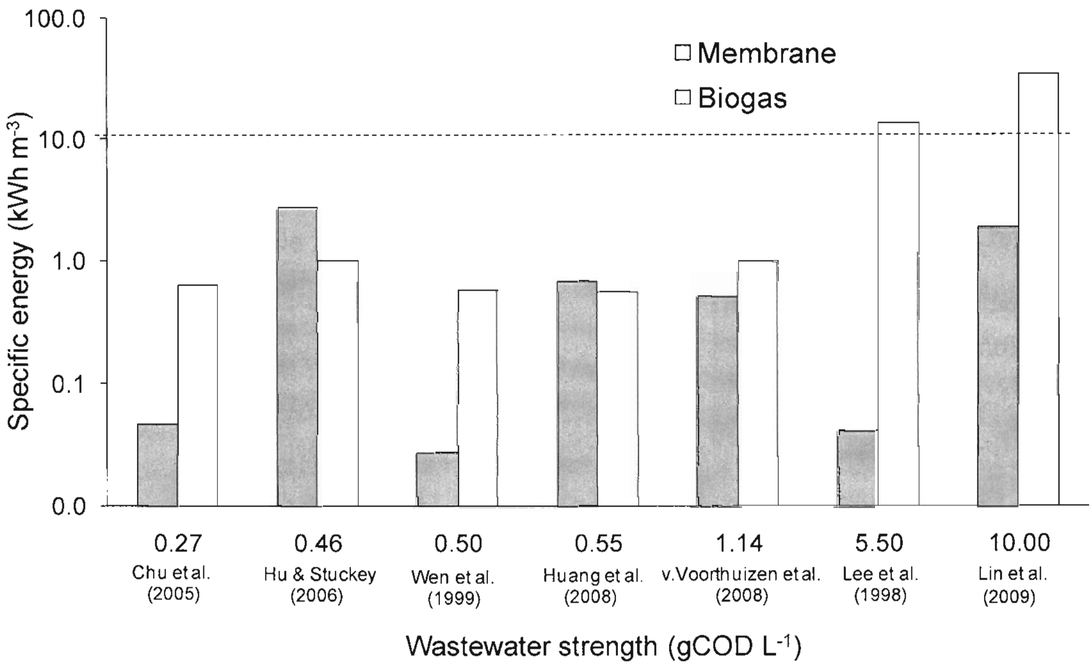

Figure 3.3 Energy balance in submerged AnMBRs

For high strength wastewaters the methane production not only covers the heat balance but can generate between 5-20 kWh m^{-3} of electrical power leading to energy export (Fuchs *et al.*, 2003). In fact commercial applications of AnMBR technology employing both sidestream and submerged membranes have been developed for different industrial and agricultural wastes. To illustrate the Biorek process developed by BIOSCAN A/S in Denmark (du Preez *et al.*, 2005) comprises an anaerobic digester coupled to sidestream crossflow membrane filtration to remove organic pollutants, which effluent is treated by reverse osmosis in order to recover clean water and a nutrient rich concentrate. More recently, Kubota has also commercialised a submerged anaerobic membrane

Table 3.2 Membrane energy demands in aerobic submerged MBRs

Area (m^2)	**SGD_m ($m^3\ m^{-2}\ h^{-1}$)**	**Flux (LMH)**	**TMP (kPa)**	**Ecf ($kWh\ m^{-3}$)**	**Ep ($kWh\ m^{-3}$)**	**Etot ($kWh\ m^{-3}$)**	**Reference**
0.21	0.73	88^{CF}	-	0.08	-	0.08	Le Clech (2006)
0.21	2.3	121^{CF}	-	0.17	-	0.17	
40	0.94	28^{SF}	7-15	0.38	$3.2\text{-}6.8 \cdot 10^{-3}$	0.38	Guglielmi (2008)
40	0.94	23^{SF}	6	0.48	$2.8\ 10^{-3}$	0.48	
69.6	0.5	10^{SF}	8	0.56	$3.7\ 10^{-3}$	0.57	Guglielmi (2007)
69.6	0.5	22^{SF}	17-25	0.26	$7.9\text{-}12 \cdot 10^{-3}$	0.26	
0.24	0.75	20^{SF}	7.5	0.42	$3.5\ 10^{-3}$	0.43	Zhang (2006)
0.5	0.3	18^{CF}	12	0.15	$5.6\ 10^{-3}$	0.16	Bouhabila (1998)
0.5	0.8	25^{CF}	15	0.29	$6.9\ 10^{-3}$	0.30	

CF: Critical flux. SF: Sustainable flux.

Table 3.3 Membrane energy demands in submerged anaerobic MBRs

Area (m^2)	**SGD_m** ($m^3\ m^{-2}\ h^{-1}$)	**Flux** (lmh)	**TMP** (kPa)	**Ecf** ($kWh\ m^{-3}$)	**Ep** ($kWh\ m^{-3}$)	**Etot** ($kWh\ m^{-3}$)	**Reference**
0.10	0	10.4	100	0.00	4.6E-02	0.05	Chu (2005)
0.30	0	5.0	60	0.00	2.8E-02	0.03	Wen (1999)
0.24	0.40	5.3	0.07	0.69	3.2E-03	0.69	Huang (2008)
0.10	3.00	8.0	38	3.41	1.8E-02	3.43	Hu & Stuckey (2006)
0.04	0.54	10.0	50	0.49	2.3E-02	0.51	van Voorthuizen (2008)
0.04	1.35	8.0	50	1.53	2.3E-02	1.56	
0.05	1.80	25.0	30	0.65	1.4E-02	0.67	Wu (2009)
0.03	1.50	7.2	10	1.89	4.6E-03	1.90	Lin (2009)
0.03	1.50	2.4	27	5.68	1.3E-02	5.70	
0.10	3.00	5.0	20	5.46	9.3E-03	5.47	Lee (2008)

Information pertaining to operationally significant sparging rates in submerged AnMBRs is currently less clear. Critical flux analysis has revealed that gas sparging is less effective at enhancing permeate flux compared to AeMBRs due to the presence of high concentration of colloidal matter and fine solids in anaerobic biomass (Jeison & van Lier, 2007; van Voorthuizen *et al.*, 2008; Lin *et al.*, 2009). Increasing specific gas demands from 0.78 to 3.76 m^3 m^{-2} h^{-1} increased critical flux from 6 to 10 LMH which is consistent with analysis of literature data (Table 3.5), which shows that while fluxes range only between 5 and 10 LMH, specific gas demands as high as 3 m^3 m^{-2} h^{-1} have been employed (Imasaka *et al.*, 1993; Hu & Stuckey, 2006). Although such high gas sparging intensity levels have shown prolong membrane operation they do not result in an increase in permeability. At the highest SGDm of 3 m^3 m^{-2} h^{-1} equivalent to an energy demand of 3.43 kWh m^3 stable permeability of 20 LMH bar^{-1} for over 90 days of operation was observed (Hu & Stuckey, 2006), while a higher permeability of 60-70 LMH bar^{-1} maintained during 450 hours was reported in a mesophilic AnMBR treating ethanol at a specific gas demand of 1.5 m^3 m^{-2} h^{-1} (Lin *et al.*, 2009) equating to an energy demand of 1.9 kWh m^3. A fouling resistance of 0.5 to 3% the membrane resistance was reported by Huang (2008) after 150 days of operation in two AnMBRs operated at a SRT of 30 to 60 days by applying a SGD of 0.4 m^3 m^{-2} h^{-1} which equates to an energy consumption of 0.69 kWh m^{-3}.

The lowest energy demands of 0.02-0.05 kWh m^{-3} reported for submerged AnMBRs (Table 3-5) result from permeate suction and correspond to those studies in which membrane filtration has been coupled to high rate anaerobic reactors where fouling was controlled without using gas sparging, just by applying intermittent filtration (Wen *et al.*, 1999; Chu *et al.*, 2005) or relying on the shear provided by upflow velocity (An *et al.*, 2009). Results have shown that although sustainable operation were not achieved, fouling rates between 10 mbar d^{-1} (An *et al.*, 2009) and 100 mbar d^{-1} (Chu *et al.*, 2005) were observed due to the low solid and colloidal load to the membrane (van Voorthuizen *et al.*, 2008). These fouling rates are in the same order of magnitude as residual fouling rates of 15-35 mbar d^{-1} and one-two orders of magnitude lower than 2000-2600 mbar d^{-1} reported for cake layer fouling within backwash cycles in aerobic systems (Guglielmi *et al.*, 2007). Therefore implementation of backwashing together with low gas sparging intensity in high rate anaerobic reactors coupled to membrane filtration could result in a efficient fouling control strategy for AnMBRs as suggested by a previous study of low solid anoxic membrane system (McAdam & Judd, 2008). Duration of filtration cycles of 10 min experimentally determined from analysis of the critical mass deposited on the membrane which leads to irreversible fouling, resulted in fluxes of 20 LMH maintained during 20 days equating to specific energy demands of 0.05 kWh m^{-3} for gas sparging.

3.3.3 Energy demands in sidestream aerobic and anaerobic MBRs

Analysis of the pressure drop per meter generated when pumping both aerobic and anaerobic sludges (Figure 3.4) reveals that aerobic biomass leads to higher frictional pressure losses than anaerobic biomass when compared at the same MLSS and CFV, reflecting the higher viscosity of the former. To illustrate, the curve corresponding to the aerobic MBR sludge at the lowest MLSS concentration of 5 gMLSS L^{-1} overlaps with the maximum concentration of 20 gMLSS L^{-1} for the anaerobic sludge. Similarly, the rate of increase of pressure loss with increasing CFV is higher in the aerobic systems as compared to the anaerobic MBRs. The effect of CFV on pressure drop (ΔP) is more prominent than the increase in MLSS in the range considered. Taking a reference point of 10 gMLSS L^{-1} and 3 m s^{-1} in the AeMBR, increasing CFV to 4 m s^{-1} results in a unitary pressure loss of 444 mbar m^{-1} while the resulting value for a biomass concentration of 15 gMLSS L^{-1} is 293 mbar m^{-1}. However, the rate of increase of pressure loss with solid concentration is more prominent for MLSS higher than 15 gMLSS L^{-1}.

Energy demands for fouling control in AeMBRs ranged from 0.67 to 7.42 kWh m^{-3} depending on crossflow velocity and MLSS concentration (Table 3.4). Increasing crossflow velocity and biomass concentration results in higher energy expenditures in sidestream systems due to a combination of higher volumetric flow and pressure losses in the first case and to the lower critical fluxes reported at higher MLSS in the second (Cicek *et al.*, 1998; Defrance *et al.*, 2000). For instance at a solid concentration of 10 gMLSS·L^{-1} (Defrance *et al.*, 2000) specific power requirements range from 0.67 to 4.25 kWh·m^{-3} with CFV varying from 1 to 4 m s^{-1} while for a higher MLSS of 15 gMLSS L^{-1} a more pronounced increase in energy demands from 0.38 to 5.14 kWh m^{-3} is obtained (Cicek *et al.*, 1998). To further illustrate the influence of biomass concentration, for a cross flow velocity of 3 m s^{-1}, energy demands increase from 0.67 to 3.1 kWh·m^{-3}as MLSS increased from 2.1 to 15 gMLSS L^{-1} due to a decrease in critical flux from 270 to 80 LMH (Cicek *et al.*, 1998).

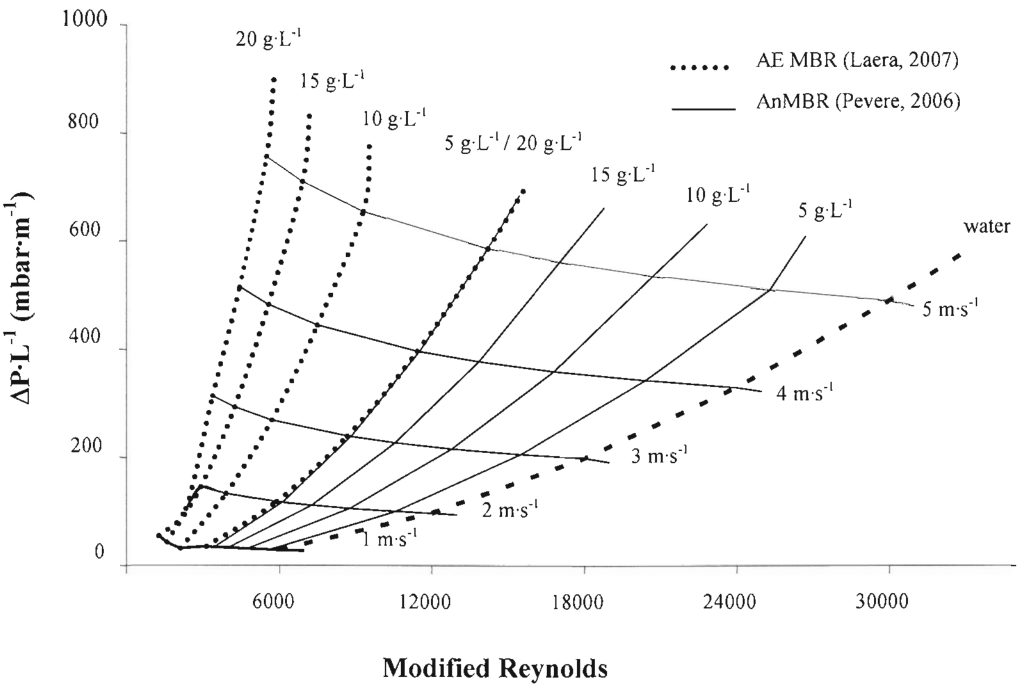

Figure 3.5 Influence of MLSS and CFV on unitary pressure drop for aerobic and anaerobic MBR sludges assuming 6 mm diameter lumen

In comparison the energy demands for fouling control in sidestream AnMBRs range between 0.23 and 16.5 kWh m^{-3} showing similar trends as in AeMBRs with respect to crossflow velocity and biomass concentration (Table 3.5). However, despite presenting lower fluxes than its aerobic counterpart, lower energy demands are predicted in AnMBRs when compared at the same biomass concentration and crossflow velocities due to the lower pressure losses resulting from its lower viscosity. For instance, at a solid concentration of 15 gMLSS L^{-1} varying CFV from 1.6 to 3.4 m s^{-1} results in an increase in energy demands from 0.88 to 3.01 kWh m^{-3} in an anaerobic system (Beaubien *et al.*, 1996) and from 0.38 to 5.14 kWh·m^{-3} in an aerobic MBRs operated at the same biomass concentration (Cicek *et al.*, 1998). The lowest energy demands in sidestream AnMBRs, correspond either to suspended growth reactors operated at low solid concentrations or when crossflow filtration has been coupled with attached growth systems (Cho & Fane, 2002) due to a combination of low frictional pressure losses and high fluxes. Fluxes of 252 LMH (Beaubien *et al.*, 1996) and 120 LMH (Elmaleh & Abdelmoumni, 1998) at corresponding crossflow velocities of 2.6 and 3.5 m s^{-1}, are amongst the highest reported in anaerobic MBRs and result in energy demands of 0.48 kWh $m^{-3}$and 1.45 kWh m^{-3} respectively although further reductions to 0.23 kWh m^{-3} are predicted for crossflow velocities in of 0.93 m s^{-1} (Cho & Fane, 2002).

Table 3.4 Membrane energy demands in sidestream aerobic MBRs

MLSS ($g\ L^{-1}$)	CFV ($m\ s^{-1}$)	Flux (LMH)	TMP (kPa)	ΔP (kPa)	$E_{CF,P}$ ($kWh\ m^{-3}$)	E_P ($kWh\ m^{-3}$)	E_{TOT} ($kWh\ m^{-3}$)	Reference
2.1		270	n.r	28.4	0.67	-	0.67	
4.6	3	180	180	31.0	1.10	0.08	1.18	Cicek (1998)
8.6		95	n.r	34.3	2.3	-	2.3	
15.4		80	n.r	38.9	3.10	-	3.1	
	1	20	10	6.3	0.67	< 0,01	0.67	
	2	40	25	11.0	1.30	0.01	1.32	
10	3	80	60	21.4	2.99	0.03	3.02	Defrance (2000)
	4	115	80	65.5	4.54	0.04	4.58	
	5	130	120	107.2	7.37	0.06	7.42	
1.8	1.6	10	1-3	12.5	3.3	<0.01	3.3	Ognier (2004)
8	4	100	60-90	24.7	4.59	0.04	4.63	Tardieu (1998)
10-50	1-5	40	20-30	-	-	0.01	$0.6\text{-}0.75^{+}$	Mueller (1995)
4.5-6	0.4	20-100	100	-	-	0.05	2^{+}	Zhang (2003)

$^{+}$ Reported by author

Table 3.5 Membrane energy demands in sidestream anaerobic MBRs

MLSS ($g \cdot L^{-1}$)	CFV ($m \cdot s^{-1}$)	Flux (LMH)	TMP (kPa)	ΔP (kPa)	Ecf ($kWh\ m^{-3}$)	Ep ($kWh\ m^{-3}$)	Etot ($kWh\ m^{-3}$)	Reference
0.4	2.6	252	90	21.5	0.44	0.04	0.48	Beaubien (1996)
7	2.6	108	80	23.1	1.33	0.03	1.36	
15	2.6	72	80	25.1	1.81	0.03	1.84	
10.8-14.5	2	15-18	40-45	15.2-15.7	3.51-4.04	0.02	3.53-4.06	Cadi (1994)
10	3	9	200	45.2	16.42	0.1	16.52	Saddoud (2006)*
0.18	3.5	120	50	10.4	1.43	0.02	1.45	Elmaleh (1997)
0.15-0.5	0.93	30	40	0.3	0.21	0.02	0.23	Cho & Fane (2002)

* Based on 8mm lumen diameter.

3.4 IMPLICATIONS FOR ANAEROBIC FLOWSHEETS.

Key findings from the modeling assessment of literature reported anaerobic MBR case studies leads to a number of concluding observations:

1. The energy required to heat the bioreactor up to mesophilic conditions can only be compensated at wastewater strengths above 4 or 5 gCOD L^{-1}. Therefore it is economically unfavorable to operate at mesophilic conditions when considering the treatment of municipal sewage with AnMBRs. Anaerobic treatment of sewage can only be feasible at low temperature.

2. A number of features about the energy associated with membrane operation are unique to AnMBR which prohibits direct knowledge transfer from AE MBRs. The general trend of submerged membranes systems being less energy intensive is not observed for AnMBRs due to the lower fluxes observed and the uncertainty of appropriate gas sparging rates required for sustainable operation.

3. From an energy point of view the most effective AnMBR configuration appears to be the combination of high rate anaerobic systems with membranes. Assessment of energy demands of systems in which biomass retention is not solely controlled by the membrane have show either very low energy demands and moderate fouling at low fluxes, or moderate energy demands together with high fluxes in sidestream configuration. Low solid membrane feed could be key to achieve low energy demands.

4. Comparison of energy demands of aerobic and anaerobic MBRs, considering complete retention of solids suggests that although the latter would present lower energy demands capital costs associated to membrane material would be three times higher.

3.5 CONCLUSIONS

Potential savings in sludge treatment and disposal costs can be achieved with both aerobic and anaerobic membrane bioreactors since operation under complete retention of solids is possible. Assessment of model calculations has shown that in aerobic MBRs low biomass production is attainable with energy inputs of 2 kWh m^{-3} due to the low oxygen transfer efficiencies at high biomass concentrations. In anaerobic MBRs fouling control is the determinant factor of the energy demand of the process and therefore sludge production and energy requirements are not so directly linked, although extended SRT are likely to influence biomass characteristics and thus membrane performance. A wide variation in energy demands in submerged AnMBRs ranging from 0.03 to 3.57 kWh m^{-3} has been found, highlighting the need to further investigate the gas intensities required to control fouling in order to ascertain the potential of such technology for mainstream wastewater treatment.

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CHAPTER 4:

START UP AND PERFORMANCE OF AEROBIC AND ANAEROBIC MBRs TREATING LOW STRENGTH WASTEWATER

CHAPTER 4. START UP AND PERFORMANCE OF AEROBIC AND ANAEROBIC MBRs TREATING LOW STRENGTH WASTEWATER

Martin Garcia, N.[1], Mokosch, M.[2], Pidou, M.[1], Judd, S. J.[1] and Jefferson, B.[1]

[1]Centre for Water Science, Cranfield University, Cranfield, Beds, MK43 0AL, UK

[2]Institute for Urban Water Management, Dresden University of Technology, Dresden, D-01062, Dresden, Germany

ABSTRACT

In this study the performance of aerobic and anaerobic membrane bioreactors operated in parallel was evaluated with respect to different start up strategies, treatment performance, sludge production and membrane fouling. Prolonged start up periods were observed in an anaerobic MBR (AnMBR) inoculated with wastewater compared to the aerobic system. However with a seeded AnMBR compliant effluent qualities were achieved within days without previous acclimatisation. Removal efficiencies ranged from 79 to 95 % in the AnMBR and from 85 to 99% in the aerobic system both run at 100 days SRT.Under these conditions similar sludge yields of 0.14-0.15 gVSS gCODrem^{-1} were observed in both systems although the AnMBR sludge presented higher COD suggesting that it was mainly composed of undigested influent solids. Anlysis of sludge supernatant revealed that the AnMBR presented seven times higher supernatant COD concentration than the AeMBR, the difference being mainly due to colloidal matter above 500 kDa. This difference was thought to be responsible for the lower influence of gas sparging on flux enhancement during critical flux tests conducted with the AnMBR as compared to the AeMBR. However sustainable membrane operation was demonstrated during long term filtration trials at a flux of 6 LMH and a specific gas demands of 0.38 m^3 m^{-2} h^{-1} equating to a specific energy demand of 0.58 kWh m^{-3} which is in the low end of those reported for AeMBRs.

Keywords: Star up, aerobic, anaerobic, membrane bioreactors, fouling, energy, sludge.

4.1 INTRODUCTION

The main feature of membrane bioreactors (MBRs) in comparison to traditional activated sludge systems is that sludge and hydraulic retention times are independent. In practice this has meant that the range of sludge retention times (SRT) at which MBRs are operated are higher than in conventional activated sludge systems, allowing for increased treatment capacity in smaller spaces and reduced sludge production. At its extreme this concept is manifested as near infinite sludge age where operation proceeds without sludge wastage. The consequence is a reduction in sludge production but an increase in aeration costs due to the decrease in oxygen mass transfer at higher levels of mixed liquor suspended solids and a greater biological respiration demand.

In anaerobic wastewater treatment, energy requirements and sludge production are not so directly linked as aeration is not required for biodegradation of organic matter. In fact when considering the treatment of low strength wastewater at low temperatures, conditions which significantly reduce biological activity of anaerobic biomass, high sludge retention times are required in order to avoid any viable biomass washout (Lettinga *et al.,* 2001). Currently this is achieved in modern granular processes such as UASB and EGSB reactors where biological communities are bound into large granules which can be operated without the risk of large scale washout. However, retention of biomass solids within the reactor is not always guaranteed as a number of factors such as temperature, hydraulic and organic loading rates, upflow velocity and influent wastewater characteristics can negatively affect the amount of solids lost in the effluent (Mahmoud *et al.,* 2003). It is anticipated that such issues will be heightened in the current application being considered in this study. The incorporation of membranes into anaerobic processes offsets this problem as in aerobic bioreactors, enabling independent control of sludge and hydraulic rates whilst not compromising the reliability of treatment.

Previous studies (Chapter 3) indicated that the reactors need to be operated under ambient temperature conditions to enable energy recovery. Given that the temperature will never approach mesophilic conditions a key question is whether there is a disadvantage utilising mesophilic seed materials which must then adapt to lower temperature compared biomass developed at low temperatures. Previous studies investigating start up without seeding the reactor in order to achieve the most adapted microbial community have found that whilst in aerobic MBR systems high treatment performances are achieved within weeks of operation (Rosenberger *et al.*, 2002; Pollice *et al.*, 2004) the anaerobic systems (UASB) require several months (Kalogo *et al.*, 2001; Álvarez *et al.*, 2006). It is postulated that the complete retention of solids, afforded by the membrane, provides the possibility of accelerated start-up in anaerobic systems.

The aim of the current study is to compare aerobic and anaerobic membrane bioreactors in terms of treatment capacity and energy demands. A series of experiments were conducted in order to investigate the start up of an AnMBR without seed sludge and compare its performance with one seeded with mesophilic digested sludge.

4.2 MATERIALS AND METHODS

4.2.1 Pilot plants

The aerobic and anaerobic MBR pilot plants were comprised of two identical tanks with a total volume of 1.5 m^3 divided between the biological (1 m^3) and membrane (0.5 m^3) compartments (Figure 4.1). The wastewater was introduced in the biological tank through a floating valve which controlled the level of sludge at a height of approximately 1.5 m, making a total working volume of 1.2 m^3. In the aerobic MBR biomass was pumped with a submersible centrifugal pump from the biological chamber into the membrane tank. The sludge was then recycled through an overflow located 25 cm above the level of the main compartment. Air was continuously supplied at a flow rate ranging between 50 L min^{-1} and 100 L min^{-1} through cylindrical fine bubble diffusers located at the bottom of the biological tank and providing oxygen for biomass respiration and mixing. The AnMBR pilot plant was isolated from the atmosphere by sealing the tank with a PVC lid (Figure 4.1). In this system no overflow was employed in order to avoid mixing of the headspace gas between the biological and membrane tanks. The biomass was cycled between both compartments through external pumps. An additional pump operated in cycles of 15 minutes ON and 15 minutes OFF acted to mix the reactor contents by recycling the biomass within the anaerobic bioreactor tank through venturi nozzles located at the base of this chamber.

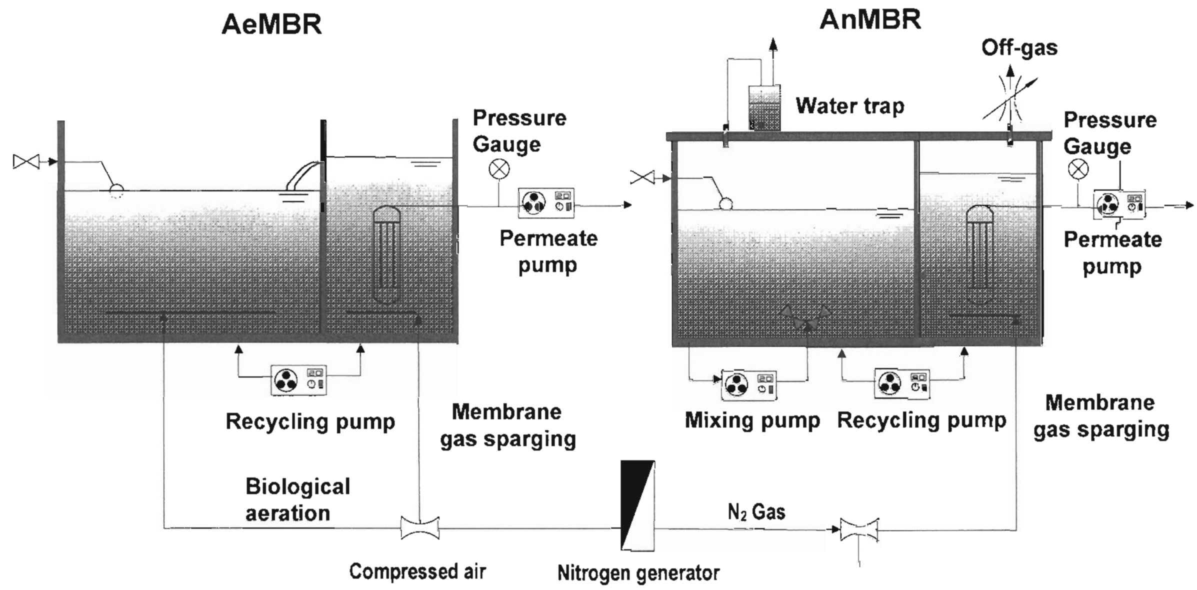

Figure 4.1 Schematic representation of AeMBR (right) and AnMBR (left)

Filtration elements consisting of PVDF hollow fibre membranes of 0.08 μm pore size and 12.5 m^2 surface area were employed in the aerobic and anaerobic MBRs. The permeate was extracted through peristaltic pumps (620 Du, Watson Marlow, Falmouth) in which membrane flux was set at 6 LMH. Membrane fouling was controlled by using crossflow gas sparging in both aerobic and anaerobic systems. In the aerobic MBR air was supplied from a compressed air line, while in the case of the anaerobic MBR nitrogen was supplied by a nitrogen generator. In both instances, a gas flow rate of 70-80 L min^{-1} equating to a specific gas demand normalized against membrane area of 0.34-0.38 m^3 m^{-2} h^{-1} was applied.

Table 4.1 Influent wastewater characteristics

Parameter	Unit	Average	Maximum	Minimum
COD	mg L^{-1}	350	553	197
% Soluble	%	52	76	39
BOD_5	mg L^{-1}	167	285	3
MLSS	mg L^{-1}	84	186	51
% MLVSS	%	92	99	83
NH-N	mg NH_4-N L^{-1}	35	48	15

4.2.2 Wastewater and Operating conditions

Partially settled primary wastewater was used as a feed source throughout the experiment and was supplied by a continuous ring main fed from the Cranfield University's sewage works. The wastewater presented characteristics of a low-medium strength sewage (Table 4.1), with average COD, SS and ammonia concentrations of 350 mgCOD L^{-1}, 35 $mgNH_4$-N L^{-1} and 84 mgSS L^{-1} respecitvely and COD to BOD_5 ratio of approximately 2.

A total of 4 long term experimental runs, two with the AeMBR and two other with the anaerobic systems, were conducted. In all of them hydraulic retention time was set at 16 hours (Table 4.2). During runs 1, 2 and 4 the pilot plants were started by introducing wastewater without any seed sludge. In the case of the AnMBR (run 4) no sludge was wasted and influent wastewater was heated resulting in temperatures varying between 18 °C and 23 °C, which are within the optimum range for psychrophilic development of anaerobic bacteria (Lettinga *et al.,* 2001). Two different start up conditions in which sludge retention times were set at 50 days (run 1) and 100 days (run 2) were conducted in the AeMBR. The SRT of 50 days represents the high end of sludge ages applied in full scale municipal MBRs whereas the longer SRT was applied to allow direct comparison at the same set of operational conditions as the experiment in which the AnMBR was inoculated (run 3). The seeded AnMBR was inoculated with 700 L of digested sludge of 2.5 % total solids (70 % volatiles) and 500 L of wastewater, making a total concentration of 10 gVSS L^{-1} in the reactor. Digested sludge was sourced from a municipal anaerobic digester treating a mixture of primary and secondary sludges. The seed sludge was pre screened through a 1 cm net prior to use and the experiment was started just after inoculation without heating or previous wastewater adaptation.

Table 4.2 Start-up experiments and operational conditions

	AeMBR		**AnMBR**	
Parameter/Experiment	**1**	**2**	**3**	**4**
Duration (d)	230	250	250	125
Start-up	Unseeded	Unseeded	Seeded	Unseeded
SRT (d)	50	100	100	∞
HRT (h)	16	16	16	16
Temperature (°C)	16-28	15-25	6-22	18-23

4.2.3 Sampling, monitoring and analyses

The AeMBR was continuously monitored for dissolved oxygen and temperature using a Insite IG dissolved oxygen and temperature analyzer (model 1000, InsiteIG Inc. Slidell, LA) while a DL2e logger (Delta-T Devices Ltd., Cambridge) was employed for monitoring temperature in the anaerobic system. Transmembrane pressure was measured in the permeate line of both aerobic and anaerobic systems using Druck pressure transducers (Druck Ltd, Leicester) and logged via an ADC16 digital analog converter (Pico Technology Ltd. , St Neots, Cambs) into a personal computer. Composite samples from the influent and membrane effluent were collected each 2-3 days by storing them in auxiliary tanks, together with grab sludge samples obtained from wasted sludge. Influent and effluent COD, NH_4-N, and NO_3^--N were analyzed with Merck vial test kits (VWR International adapted from Standard Methods, APHA, 1998). Mixed liquor suspended solids (MLSS) were measured according to Standard Methods (APHA, 1998) and sludge supernatant was obtained by centrifugation of the sludge at 10000 g for 15 minutes followed by filtration of decanted liquid with 1 μm membranes (Judd, 2006).

4.2.4 Molecular weight fractionation

Serial molecular weight fractionation of the supernatant was conducted using an Amicon 8400 dead-end stirred cell (Millipore, USA) with polyethersulphone UF membranes of nominal molecular weight of 10, 100, and 500 kDa. All experiments were conducted at room temperature and 2 bar pressure applied using the pure nitrogen gas supply.

4.2.5 Methanogenic activity

Methanogenic activity at 20 °C was measured once during the experiment for the self inoculated AnMBR. To 2 L of sludge with a solid concentration of 2 gMLVSS L^{-1} was added neutralized acetic acid (HAc) to make up a final concentration of 2 gHAc L^{-1}. Biogas volume was collected and quantified in an inverted measuring cylinder initially filled with sulphuric acid (2 %). Gas samples were analyzed by Servomex gas analyzer (Model 1440, Servomex Group Ltd, UK) in order to determine methane content.

4.2.6 Critical flux tests

The critical flux was determined by applying the flux step method (Le Clech *et al.*, 2003). Successive filtration steps with flux increments of 2-3 LMH were maintained during 15 minutes during which TMP was monitored. A PVDF hollow fibre membrane of 0.93 m^2 surface area and 0.04 μm pore size was submerged in 25 L of sludge contained in a cylindrical PVC tank. For each of the sludges a series of four critical flux experiments at different gas demands ranging from 0.38 $m^3\ h^{-1}\ m^{-2}$ to 1.16 $m^3\ h^{-1}\ m^{-2}$ were conducted.

4.3 RESULTS AND DISCUSSION

4.3.1 Treatment performance in aerobic and anaerobic MBRs

During the first two weeks of operation of the unseeded AnMBR COD removals ranged between 67 % and 93 % after which treatment efficiency gradually decreased, and averaged 49 ± 9 % during the last 90 days which correspond to effluent COD of 203 ± 45 mgCOD L^{-1}. Literature regarding start-up of self inoculated UASB at medium to low temperatures treating low strength wastewaters indicates that stabilisation of these systems is possible in 4 months of operation (Kalogo *et al.*, 2001; Álvarez *et al.*, 2006). However as the success of self inoculation relies on the input of microoganisms from the influent wastewater (Lettinga *et al.*, 2001) the lower hydraulic retention times between 4 and 6 hours applied in the literature studies (Barbosa & Sant'Anna Jr., 1989; Kalogo *et al.*, 2001) would be much more favourable than the 16 hours at which the AnMBR was operated. Despite the low COD removal efficiency presented by the self inoculated AnMBR methanogenic activity was measured at 0.085 $gCOD_{CH4}\ gVSS^{-1}$. Such levels correspond well with those reported in UASB systems operated at higher temperatures and exhibiting lower effluent qualities than in the present study (Barbosa & Sant'Anna Jr., 1989; Uemura & Harada, 2000; Kalogo *et al.*, 2001). Additionally the specific COD removal rate in the reactor of 0.1 $gCOD_{rem}\ gVSS^{-1}.d^{-1}$ compares well with the methanogenic activity indicating that there is no substrate limitation consistent with the situation that can be presented during start up when a low concentration of biomass is exposed to a excess of substrate (Van Haandel & Lettinga, 1994).

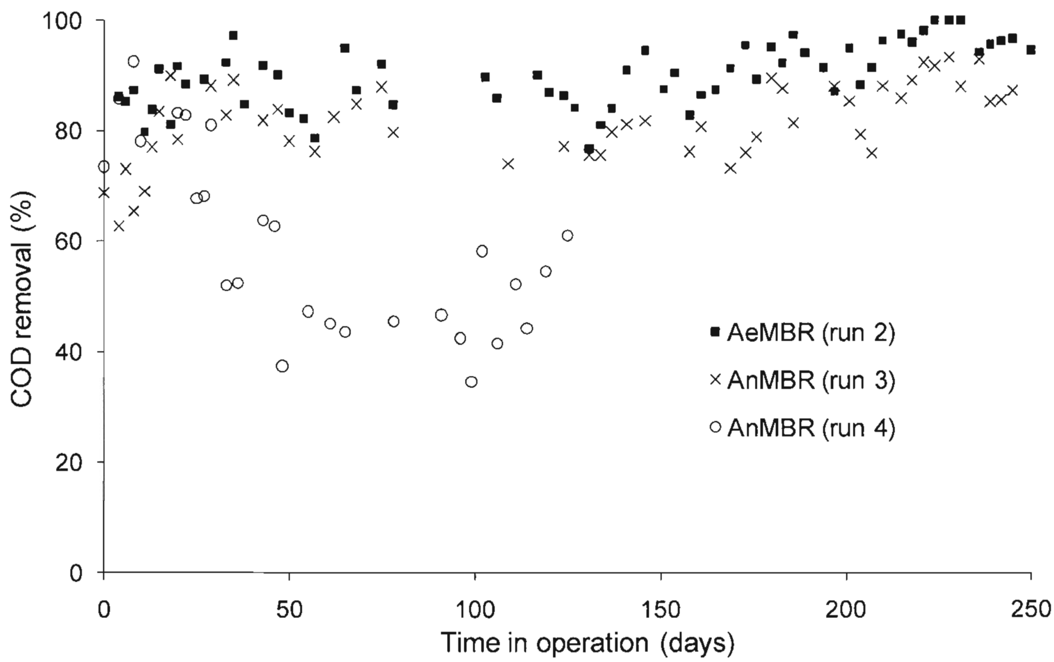

Figure 4.2 COD removal in seeded and unseeded AeMBRs and AnMBRs

The use of anaerobic municipal digested sludge as inoculum accelerated the start-up which in the first two months of operation generated effluent COD concentrations that ranged between 35 mgCOD L^{-1} and 139 mgCOD L^{-1} with an average value of 79 ± 35 mgCOD L^{-1} (Figure 4.2). The higher values corresponded to the first days of operation without previous acclimatisation to temperature or wastewater. It was also observed that higher effluent COD were obtained when the reactor was restarted after membrane chemical cleaning. This could be due to the absence of fouling layer during initial stages of filtration which reduces the effective pore size of the membrane thus increasing rejection of soluble materials. However the improvement in COD removal from 75-80 % to 90-95% observed during the last 150 days of operation in the seeded AnMBR can be attributed to the increase in temperatures from 8 °C to 22 °C respectively. Despite the lower temperatures in the seeded AnMBR with respect to the unseeded aerobic systems, COD removal during the 250 days of experimental period was very similar. The aerobic MBRs showed COD removals always exceeding 85 % during the first two months of operation which gradually improved reaching 95-99% towards the end the experiment (Figure 4.2) Nitrification of influent ammonia was complete in both 50 and 100 days SRT AeMBRs within less than one month. These results agree with those previously reported in other studies and highlight the fast development of the biological community in aerobic conditions when coupling a membrane to an activated sludge system

(Rosenberger *et al.*, 2002; Pollice *et al.*, 2004) as opposed to the self inoculated anaerobic system.

4.3.2 Sludge production and characteristics

Concentration of solids in the unseeded AnMBR increased from 0.2 gMLSS L^{-1} to 2 gMLSS L^{-1} during the first month of operation after which its accumulation rate decreased, reaching almost 3 gMLSS L^{-1} in the remaining 90 days (Figure 4.3). Taking into account the average suspended solids concentration of 0.08 gSS L^{-1} in the influent and that no sludge was wasted the rate of increase of solids should have been be around 144 mgMLSS d^{-1} resulting in a mixed liquor suspended solid concentration of 14.4 g L^{-1} by the end of the experiment. These results suggests that approximately 90 % of influent solids were solubilized, 40 % of which were permeable and contributed to the higher effluent COD concentration of 203 ± 45 mgCOD L^{-1} as compared to the 151 ± 57 mgCOD L^{-1} in the soluble fraction of the influent wastewater. Considering a solid mass balance within the reactor with first order kinetics to express the rate of solubilisation of particulates yields an hydrolysis constant of 0.056 d^{-1} which lies between 0.07 d^{-1} and 0.02 d^{-1} reported for anaerobic digestion of primary sludge at 25 °C and 15 °C respectively (Mahmoud *et al.*, 2004).

Despite the complete retention of solids in the unseeded AnMBR, the development of MLSS profiles in the AeMBR operated at 50 and 100 days SRT was faster and produced higher amounts of solids (Figure 4.3). During the first stages of the start up of the aerobic systems in which low amounts of biomass are exposed to large amounts of substrate, concentration of MLSS increased rapidly and at the same rate for both sludge ages. However, the growth rate declined after approximately 100 and 150 days of operation when biomass concetration reached 6 gMLSS L^{-1} and 8 gMLSS L^{-1} which corresponded to F:M ratios of 0.12 d^{-1} and 0.08 d^{-1}. In the seeded AnMBR no particular trend was observed with MLSS ranging between 6.6 gMLSS L^{-1} and 9.6 gMLSS L^{-1} and averaging 7.7 gMLSS L^{-1}. In the AeMBR operated at 100 and 50 days SRT equilibrium MLSS concentrations were 6.4 gMLSS $L^{-1}$and 8.7 gMLSS L^{-1}respectively (Table 4.3).

The sludge yields shown in Table 4.3, corresponding to the AeMBRs and the seeded anaerobic systems were calculated on the basis of the sludge production and COD removal at the end of the experiment. As no sludge was wasted from the self inoculated AnMBR, the sludge yield was estimated from the MLVSS accumulated in the reactor and the cumulative COD removed (Pollice *et al.*, 2004). The sludge yield decreased from 0.22 gMLVSS $gCOD_{rem}^{-1}$ to 0.15 gMLVSS $gCOD_{rem}^{-1}$ as the SRT increased from 50 to 100 days and this compares well with results reported by Massé (2006) under almost identical operating conditions and influent wastewater characteristics. Similarly in the AnMBRs the lower sludge yield of 0.08 gMLVSS $gCOD_{rem}^{-1}$ corresponded to the self inoculated system operated without sludge wastage, while the inoculated reactor presented a yield similar to that of the aerobic systems operated under the same conditions. Results reported by Baek (2006) in which the levels of MLSS in an aerobic and anaerobic MBRs operated in parallel with complete sludge retention appeared to be very similar would confirm the results shown here.

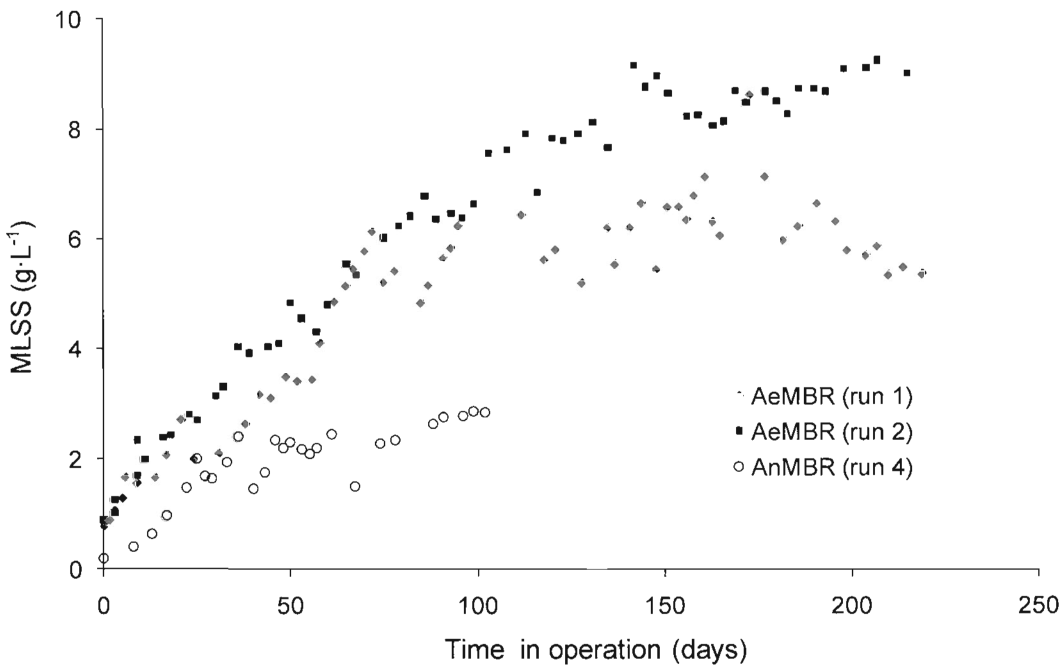

Figure 4.3 MLSS evolution in unseeded AeMBRs and AnMBRs

Whilst the sludge yields of 0.14-0.15 gMLVSS $gCOD_{rem}^{-1}$ of the aerobic and anaerobic MBRs operated at the same sludge age appear similar it is likely the source of the solids is considerably different. In the case of the aerobic system, the decrease in yield as sludge age increases indicated that the yield is controlled by a combination of the higher biomass decay rates or starvation conditions to which the biomass is subjected. In contrast, in the case of the anaerobic MBR it is likely that the sludge production occurs mainly from influent solids rather than unhydrolyzed products or biomass although the yields of anaerobic bacteria have been reported to increase with decreasing temperatures (Van Lier *et al.*, 1997). To characterise this difference, sludge wasted from the aerobic and anaerobic MBRs operated at 100 days SRT was monitored in relation to the ratio of COD to MLVSS in the wasted sludge over a period of 80 days. Solids from the anaerobic reactor presented a higher COD content of 1.72 ± 0.08 gCOD $gVSS^{-1}$ compared to 1.46 ± 0.12 gCOD $gVSS^{-1}$ for the solids in the aerobic system. Considering that the ratio of COD to MLVSS of 1.8 in the anaerobic reactor is comparable to influent sewage it can be considered that the solid inventory in the AnMBR is mainly composed of influent organics that were not biodegraded. On the other hand the ratio of 1.46 measured in the AeMBR agrees with the ratio 1.4-1.5 gCOD $gVSS^{-1}$ that is reported to represent the COD content of biomass (Metcalf & Eddy, 2003).

Table 4.3 Overall performance and sludge production in aerobic and anaerobic MBRs

		Aerobic MBRs		**Anaerobic MBRs**	
Parameter	**Unit**	**Run 1**	**Run 2**	**Run 3**	**Run 4**
COD removal	%	89	95	88	49
MLSS	$g.L^{-1}$	6.4	8.7	7.7	2.8
MLVSS	$g.L^{-1}$	5.6	7.7	6.6	2.4
Yobs	**gMLVSS $gCOD^{-1}$**	0.22	0.15	0.14	0.08*
Sludge COD	**gCOD $gMLVSS^{-1}$**	-	1.46	1.72	-
% COD_{eff}	%	11	5	12	51
% COD_{waste}	%	26^{x}	19	26	0

^{x} Estimated from the ratio 1.4 gCOD·$gVSS^{-1}$.* Estimated from sludge accumulation

The sludge yield is commonly employed to express the sludge production normalized against the COD removal and accounts for biomass growth, biomass decay and inert influent solids. However in anaerobic systems, it should also account for influent solids not biodegraded as hydrolysis is usually the limiting step of the process (Lettinga *et al.*, 2001; Mahmoud *et al.*, 2004). This means that while in AeMBR the levels of MLVSS are usually considered indicators of biomass concentration in anaerobic systems which present low biomass yield only a small fraction of the solid inventory can be considered as biomass (Soto *et al.*, 1993). In fact, studies in which completely soluble influent substrate have been employed have shown sludge yields decrease from 0.13 to 0.006 gMLVSS $gCOD_{rem}^{-1}$ as SRT increases from 30 days (Huang *et al.*, 2008) to 150 days (Hu & Stuckey, 2006).

Although high sludge ages have shown to result in similar sludge productions in aerobic and anaerobic systems, the implications for both energy and sludge disposal costs would be totally different. While accumulation of solids in the anaerobic MBR appeared to be mainly due to influent organics which can potentially be reduced using anaerobic digestion, not much benefit in terms of volatile solids reduction or biogas production can be expected from AeMBR sludge (Holbrook *et al.,* 2005) especially as sludge age is extended (Bolzonella *et al.,* 2005; Ekama *et al.,* 2007). However, recent reports have indicated that the use of primary clarification prior to biological treatment would result in economical benefits from lower biological aeration demands, reduced biological tank size and recovery of energy from biogas produced from primary solids (Amadeus final report, 2009) when compared to the more common flow sheet which treats raw screened sewage.

4.3.3 Supernatant characteristics

Analysis of sludge supernatant revealed that the concentration of soluble and colloidal organic matter measured as COD (SMP_{COD}) was between 6.5 and 7.5 times higher in the anaerobic systems as compared to the AeMBRs (Figure 4.4). Taking into account the last 100 days of operation in which MLSS were stabilized, both AeMBRs showed similar SMP_{COD} concentration which averaged 99 ± 37 mgSMP_{COD} L^{-1} and 99 ± 39 mg SMP_{COD} L^{-1} for the systems operated at 50 days and 100 days SRT respectively. However, comparison of supernatant and effluent COD levels, which difference has been assumed to correspond to the concentration of organic foulants affecting membrane performance in AeMBRs (Lesjean *et al.,* 2005; Meng *et al.,* 2006; Rosenberger *et al.,* 2006), showed that the concentration of rejected organics appeared to be 43 ± 31 mgCOD L^{-1} and 62 ± 40 mgCOD L^{-1} for the lower and higher sludge ages respectively. These results would suggest that that extending SRT results in an increased biodegradation of soluble organics whilst increasing the colloidal concentration of the supernatant due to higher dispersive growth and endogenous decay products arising from substrate limitation conditions imposed at lower F:M ratios (Massé *et al.,* 2006).

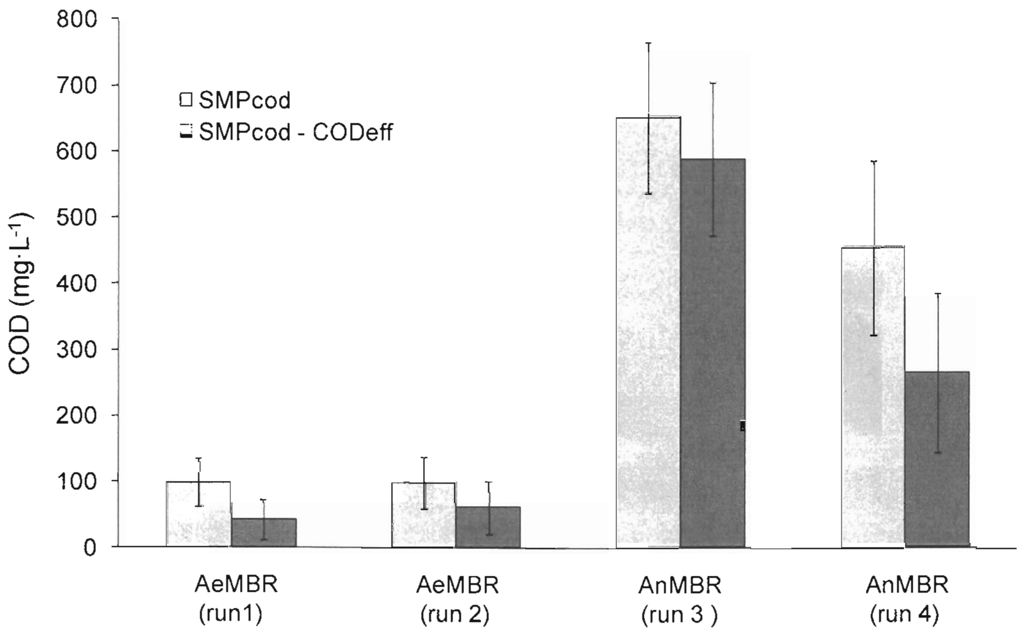

Figure 4.4 Supernatant COD concentrations

In contrast, in the AnMBRs SMP_{COD} concentration of 455 ± 133 mgSMP_{COD} L^{-1} and 652 ± 114 mgSMP_{COD} L^{-1} were measured in the unseeded and seeded systems respectively. The corresponding concentrations of rejected supernatant COD were 261 ± 121 mgCOD L^{-1} and 588 ±117 mgCOD L^{-1}, probably reflecting the higher conversion of soluble substrate to colloidal matter as a consequence of biomass growth (Elmitwalli *et al.,* 2001). Indeed, molecular weight fractionation of anaerobic supernatant determined during run 2 (Table 4.4) revealed that SMP was dominated by the colloidal fraction above 500 kDa which represented 87% of total COD and a 22 fold increase in concentration compared to the same fraction in the aerobic system. Although the fractions, below 100 kDa and 10 kDa only represented 4 % and 9 % of the total SMP_{COD} respectively, their combined contribution was three times higher than the same molecular weight fractions in the aerobic system. Similarly to the results obtained in the present study, direct comparison between aerobic and anaerobic MBRs, have shown that although the levels of soluble organics (<0.45 μm) in sludge supernatant range between 2 and 3 times higher than in its aerobic counterpart (Baek & Pagilla, 2006; van Voorthuizen *et al.,* 2008), the difference in colloidal content is over an order of magnitude higher (van Voorthuizen *et al.* 2008). While the higher colloidal content present in anaerobic MBR sludge with respect to aerobic system may reflect a higher degree of free bacteria present in the mixed liquor (Imasaka *et al.,* 1989; Elmaleh & Abdelmoumni, 1997; Ghyoot & Verstraete, 1997; Choo & Lee, 1998) or influent solids partially degraded or not adsorbed into sludge flocs (Lant & Hartley, 2007), the higher soluble concentration reflects the lower biodegradation rates or biodegradability of SMP under anaerobic conditions (Schiener *et al.*, 1998; Ince *et al.*, 2000).

Table 4.4 Molecular weight fractionation of AeMBR and AnMBR (run 2)

	AeMBR		AnMBR	
Fraction X (kDa)	**COD**	**% of total COD**	**COD**	**% of total COD**
X> 500	26 ± 11	43	572 ± 13	**87**
100 < X < 500	1 ± 11	2	0 ± 4	0
10 < X < 100	1 ± 0	2	30 ± 4	4
X< 10	33 ± 0	53	56 ± 1	9

4.3.4 Critical flux tests: Effect of gas sparging intensity

Fouling rate curves obtained from the critical flux test for the SGD of 0.38 $m^3\ m^{-2}\ h^{-1}$ and 0.77 $m^3\ m^{-2}\ h^{-1}$, (Figure 4.5) revealed that lower flux enhancements in critical fluxes were observed in the anaerobic sludges. For instance critical flux increased from 3.4 to 4.6 LMH and from 7.7 to 11.9 LMH in the seeded and unseeded systems respectively for gas sparging intensities of 0.4 and 0.88 $m^3\ m^{-2}\ h^{-1}$ (Figure 4.5). Corresponding critical fluxes at the higher gas sparging rates of 0.97 and 1.16 $m^3\ m^{-2}\ h^{-1}$ were 5 and 9 LMH for the seeded system, while in the unseeded AnMBR no further increase in critical flux was observed. The higher fluxes obtained in the unseeded AnMBR reflects the lower concentration of MLSS as this has been reported to have three times a greater impact on membrane fouling than gas sparging rate (Jeison & van Lier, 2006). However, the higher concentration of colloidal matter evidenced by the differences between supernatant and effluent COD concentrations could have also contributed to the higher fouling propensity as reported by several studies (Lesjean *et al.*, 2005; Fan *et al.*, 2006; Rosenberger *et al.*, 2006). Overall the critical flux obtained in the flux step method correspond well with the range of fluxes obtained in other AnMBRs employing submerged membranes which have varied between 2 and 10 LMH. As gas sparging rate is shown to have a limited effect on critical flux (Jeison & van Lier, 2007), fouling can be considered to be more dependent on sludge properties than membrane operating conditions for the range of specific gas demands studied. The higher flux enhancement obtained in the AeMBRs corresponded to the system operated at 50 days SRT which critical flux increased from 3LMH to 53 LMH for SGDm of and 0.77 m^3 m^{-2} h^{-1}.

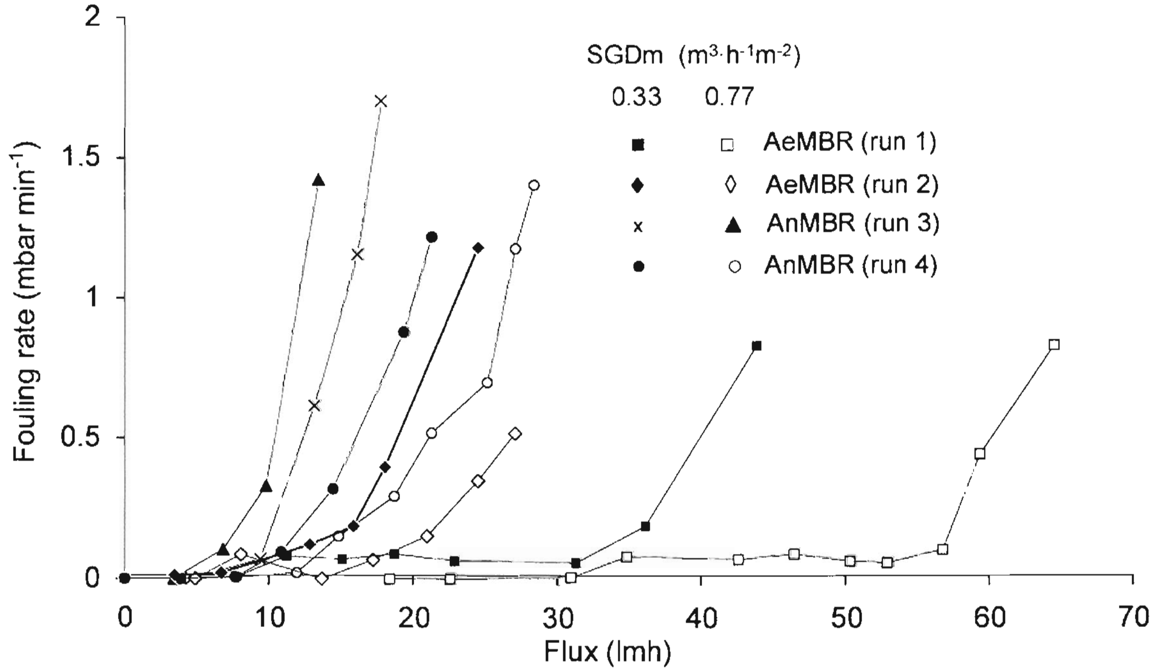

Figure 4.5 Fouling curves in AeMBRs and AnMBRs

The higher fouling propensity observed in the AeMBR operated at 100 days SRT was evidenced by the lower critical fluxes of 5 and 14 LMH. The higher flux enhancement obtained in the AeMBR operated at 50 days SRT would suggest that the main contributor to fouling would be sludge flocs, while in the system operated at higher SRT of 100 days, the soluble and colloidal fractions would be the main factor responsible for membrane fouling. Supporting this view, Han (2005), found that despite applying 30 % higher gas sparging intensities in an AeMBR operated at 100 days SRT, resistance to filtration was approximately double than at a lower SRT of 50 days. However, opposite results have also been reported (Zhang *et al.*, 2006; Ahmed *et al.*, 2007), as pointed out by Meng (2009) in a recent review on fouling, who suggested that depending on feed water quality and organic loading rate, the optimum SRT should lie between 20 and 50 days. Interestingly, reports that have shown a low or equal fouling propensity at increasing sludge retention times correspond to those cases in which high organic loading rates have been applied while those run at higher HRT and lower organic loading rates (Han *et al.*, 2005) have shown the opposite. Overall this confirms reports indicating the complexity of fouling and the lack of a clear consensus on the identification of the major foulants.

4.3.5 Long term membrane operation

Sustainable flux curves expressed as long term TMP transients for both aerobic (Figure 4.6 bottom) and anaerobic MBRs (Figure 4.6 top) were consistent with those obtained during the short term flux step experiments. New membrane modules with a permeability of 1000 LMH bar^{-1} were used during runs 1 and 4 with the self inoculated AnMBR and the AeMBR operated at 50 days SRT respectively. During the initial stages of filtration both systems presented a stabilized TMP between 30 mbar and 40 mbar equivalent to specific fluxes of 150-200 LMH bar^{-1}. However, while in the aerobic system this permeability was maintained during more than 150 days, consistent with the higher hydraulic capacity shown in the critical flux tests, in the AnMBR it slowly decreased to 60 LMH bar^{-1} by the end of the experimental run of 120 days equating to a fouling rate of 0.8 mbar d^{-1}. In run 2, after sequential cleaning with sodium hypochloride and citric acid, clean water permeability in the aerobic and anaerobic MBRs was reduced to 259 LMH bar^{-1} and 193 LMH bar^{-1} respectively, showing that a higher degree of irrecoverable fouling is accumulated in the anaerobic system. However, this difference was not translated during MBR operation as baseline TMP during run 2 for both the aerobic and anaerobic MBRs remained at 55-60 mbar even after subsequent chemical cleanings. The corresponding fouling propensity observed during run 2 in the aerobic and anaerobic MBRs was also consistent with the results obtained in the flux step experiments, as fouling rates appeared to be 0.4 mbar d^{-1} and 1.6 mbar d^{-1} respectively. Although lower fouling rates and prolonged stable operation have been reported in other AnMBR studies employing submerged membrane configuration, these have also shown extremely low membrane permeabilities (Hu & Stuckey, 2006). This supports the idea of the presence of a cake layer with high specific resistance due to a build up of fine particles and colloidal matter which preferentially deposits on the membrane surface under high shear conditions (Jeison & van Lier, 2006; Lin *et al.,* 2009).

The differences observed between the fouling behaviour in aerobic and anaerobic MBRs translated into the respective control variables to be considered for sustainable flux operation. In AeMBRs, in which gas sparging has been shown to increase critical flux until a threshold value above which no further increase is observed (Bouhabila *et al.,* 1998; Guglielmi *et al.,* 2007; Guglielmi *et al.*, 2008), flux is the main parameter to determine as it impacts directly on capital and energy costs.

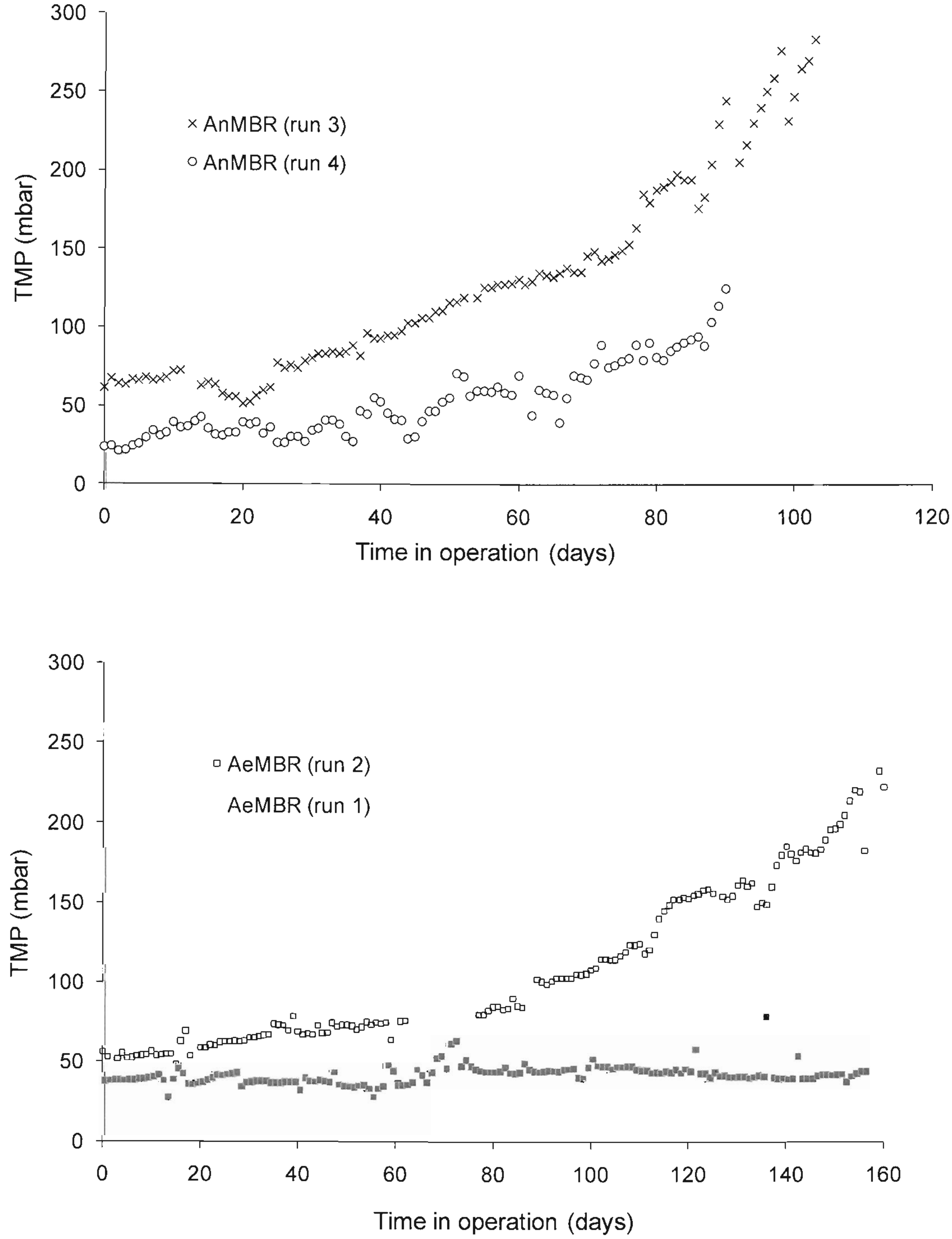

Figure 4.6 TMP transients in AeMBRs (bottom) and seeded and unseeded AnMBRs (top)

However in submerged AnMBRs in which fluxes appear to be more determined by sludge properties than from hydrodynamic conditions (Jeison & van Lier, 2007; van Voorthuizen *et al.,* 2008; Lin *et al.,* 2009) more attention should be paid to optimisation of gas sparging intensity in order to ascertain if the energy required to control fouling can offset aeration energy requirements in aerobic systems. Specific energy requirements in submerged membranes depend mainly on permeate flux, gas velocity employed to control fouling and membrane packing density which in turn depends both on specific density of the membrane and module length (Verrecht *et al.,* 2008). Assessment of energy demands based on membrane performance obtained with differently size membrane modules is often difficult as gas sparging is more effectively employed in larger systems than at lab and pilot scale (Pollice *et al.,* 2005; Kraume *et al.,* 2009). However considering the flux of 6 LMH and a SGD_m of 0.38 m^3 m^{-2} h^{-1} delivered at a hydrostatic head of 2 meters which corresponds to double the height of the membrane module employed in the present study and a blower efficiency of 60 %, a specific energy demand of 0.58 kWh m^{-3} would be required for fouling control. Although this is a considerable amount of energy when compared conventional activated sludge or high rate anaerobic reactors, different surveys have estimated the energy requirements in AeMBRs to range between 0.6 and 2 kWh m^{-3} (Cote & Thompson, 2000; Judd, 2006; Fatone *et al.,* 2007) distributed between membrane scouring aeration (60-70 %) and biological aeration (30-40 %).

4.4 CONCLUSIONS

The feasibility of applying AnMBR for mainstream wastewater treatment was evaluated by comparing the start up, treatment capacity and membrane performance of aerobic and anaerobic MBRs operated in parallel. While start-up of unseeded AeMBR proved to be fast, presenting COD and ammonia removals over 90 % within a few weeks of operation, the unseeded AnMBR presented effluent COD removal of 50 % after four months. The use of municipal anaerobic digested sludge as inoculum accelerated the start up in the AnMBR as COD removal ranging from 80 % to 95 % were observed through the study. The presence of higher concentrations of soluble and colloidal matter appeared to limit the efficacy of gas sparging at enhancing membrane flux in anaerobic systems as compared to AeMBRs. However, based on results obtained from long term filtration trials which showed that sustainable operation could be achieved at a flux of 6 LMH and a SGD of 0.38 m^3 h^{-1} m^{-2}, the energy demand required to control fouling was estimated to be 0.58 kWh m^{-3}. Although this is a considerable amount of energy when compared conventional activated sludge or high rate anaerobic reactors, it does compare favourably against AeMBRs and presents potential for further reduction.

4.5 ACKNOWLEDGEMENTS

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CHAPTER 5:

A COMPARISON BETWEEN GRANULAR AND SUSPENDED ANAEROBIC MBRs

CHAPTER 5. A COMPARISON BETWEEN GRANULAR AND SUSPENDED ANAEROBIC MBRs

Martin Garcia, N.[1], Mokosch, M.[2], Pidou, M.[1], Judd, S. J.[1] and Jefferson, B.[1]
[1]Centre for Water Science, Cranfield University, Cranfield, Bedfordshire MK43 0AL, UK
[2]Institute for Urban Water Management, Dresden University of Technology, Dresden, D-01062, Dresden, Germany

ABSTRACT

The treatment efficiency and membrane peformance of a granular and suspended growth anaerobic membrane bioreactor (G-AnMBR and AnMBR respectively) were compared and evaluated. Both anaerobic MBRs were operated in parallel during 250 days with low strength wastewater and under UK weather conditions. Both systems presented COD and BOD removal efficiencies of 80 - 95 % and > 90 % respectively. Effluent BOD remained between 5 and 15 mgBOD L^{-1} through the experimental period while effluent COD increased from 25 mg L^{-1} to 75 mg L^{-1} as temperature decreased from 25 °C to 10 °C respectively indicating the production of non biodegradable organics at lower temperatures. Although similar levels of low molecular weight organics were present in the sludge supernatant, recycling of the mixed liquor from the membrane tank to the bioreactor at a low upflow velocity enhanced interception of solids in the sludge bed of the G-AnMBR limiting the solid and colloidal load to the membrane as compared to the suspended system. Results from flux step test showed that critical flux increased from 4 to 13 LMH and from 3 to 5 LMH with gas sparging intensities varying from 0.007 m s^{-1} to 0.041. Additional long term trials in which the effect of gas sparging rate and backwashing efficiency were assesed confirmed the lower fouling propensity of the G-AnMBR.

Keywords: Anaerobic, membrane bioreactors, fouling, granular, SMP, backwashing.

5.1 INTRODUCTION

There is great interest in applying anaerobic processes for mainstream wastewater treatment due to the lower energy demands associated with the absence of aeration, the reduction in sludge production and the possibility of recovering part of the energy from biogas production. Current preference is to consider granular or biofilm based high rate reactors such as upflow anaerobic sludge blanket reactor (UASB) or expanded granular sludge blanket reactor (EGSB) as they are able to retain high amounts of biomass by relying either on the settlement of biomass or the generation of a biofilm. The mechanisms of removal of organic compounds in both cases are a combination of physical and biological pathways (Mahmoud *et al.*, 2003). Previous trials have identified the limitation associated with the application of such technology is related to the physical removal of colloids (Mahmoud *et al.*, 2003) rather than biological conversion of soluble organics (Lettinga *et al.*, 2001). Low physical removal of colloids and solids results in the deterioration of effluent quality and inefficient control of sludge retention time which is of special importance in the treatment of low strength wastewater at low temperatures, because the loss of biomass in the effluent is not compensated with the growth inside the reactor.

Membrane bioreactors represent a configuration suited to overcome such issues whereby a membrane retains all solids and a large percentage of colloids and high molecular weight compounds which can be further degraded and that otherwise would be lost in the effluent (Ross & Strohwald, 1994). Consequently, anaerobic MBRs (AnMBRs) are increasingly viewed as a suitable technology option for low temperature low feed strength applications such as municipal wastewater treatment (Liao *et al.*, 2006). The specific benefit of incorporating membranes in anaerobic processes depends on the application. For high strength wastewater introducing a membrane allows a reduction in the reactor size while producing an effluent free of solids and enough biogas to balance the heating and membrane energy demands of the process (Ross & Strohwald, 1994; Fuchs *et al.*, 2003; Kanai *et al.*, 2010). In the treatment of low strength wastewaters in which operation is restricted to ambient temperatures introducing a membrane avoids biomass washout and enables operation at high SRT which is necessary to compensate for the decrease in biological activity of slow growing anaerobic bacteria at low temperature. Despite the importance of temperature on the biological performance of anaerobic systems only a few studies have focused on the operation of AnMBRs at temperatures below 15 °C (Wen *et al.*, 1999; Chu *et al.*, 2005).

As with all membrane processes, fouling is the main factor limiting the widespread adoption of MBRs, as it negatively affects membrane performance resulting in increasing maintenance and operative costs (Chang *et al.*, 2002) associated to chemical cleaning and fouling control respectively. The use of gas sparging in submerged membrane systems has become established as the most energy efficient fouling control strategy in aerobic MBRs (AeMBRs) and is now a commercially viable option to other conventional treatment technologies for municipal wastewater applications whenever high effluent quality or robust disinfection are required (Judd, 2006). Although less studied than in AeMBRs the use of submerged configuration in anaerobic systems has also been increasingly investigated in the recent years (Hu & Stuckey, 2006; Jeison & van Lier, 2006; Huang *et al.*, 2008; Lin *et al.*, 2009). Reflecting the higher fouling propensity of anaerobic biomass, literature data has shown that applicable fluxes are lower than in aerobic systems, ranging between 5 LMH and 12 LMH. However, no concluding results with respect to the levels of energy associated to fouling control can be drawn as gas sparging intensities have varied widely between effectively no gas sparging (Wen *et al.*, 1999; Chu *et al.*, 2005) and 3 m^3 h^{-1} m^{-2} (Imasaka *et al.*, 1989; Hu & Stuckey, 2006; Lee *et al.*, 2008).

The application of membrane processes into anaerobic reactors offers an additional consideration as it is feasible to utilise suspended or granular biomass which may present additional advantages. In terms of membrane performance uncoupling the biological and filtration processes reduces the potential effect of suspended solids on membrane fouling (Lee *et al.*, 2001). Previous research has demonstrated that decreasing biomass concentration in AnMBRs improves membrane performance resulting in higher permeabilities, fluxes and lower gas sparging intensities (Beaubien *et al.*, 1996; Jeison & van Lier, 2006; van Voorthuizen *et al.*, 2008). A comparison between suspended and attached growth AeMBRs (Lee *et al.*, 2001) showed that fouling proceeded faster in the latter. Despite showing similar levels of soluble compounds, the author attributed the lower fouling rates obtained with the suspended growth system due to the formation of a protective cake layer which acted as a prefilter to smaller colloids and high molecular weight compounds. However, a high specific gas demand (2 $m^3 \cdot h^{-1} \cdot m^{-2}$) was employed, imposing hydrodynamic conditions which would minimize deposition of solids on the membrane surface.

When considering the application of anaerobic reactors for main flow wastewater treatment in temperate climates two key questions require consideration: (1) can an acceptable level of treatment be achieved in the reactor and (2) does the energy required to control fouling generate net increases in operating energy demands over conventional systems. To address these questions the performance of a suspended and granular anaerobic membrane bioreactors are presented and compared so as to ascertain which bioreactor configuration presents higher treatment efficiency and lower fouling propensity. Granular and suspended submerged anaerobic membrane bioreactors were operated in parallel with settled sewage for more than 250 days without temperature control and in UK weather conditions. The impact of bioreactor configuration on membrane fouling was evaluated using both short term flux step method and long term filtration experiments at different gas sparging intensities.

5.2 MATERIALS AND METHODS

5.2.1 Pilot plants and operating conditions

The granular and suspended AnMBR pilot plants had a total volume of 125 L and 1200 L respectively, distributed between a biological (75%) and membrane tank (25 %) (Figure 5.1). In the suspended system the wastewater was introduced in the biological tank through a floating valve which kept the reactor volume constant. The biomass was cycled between the biological and membrane tanks through external pumps in order to homogenise reactor contents between both chambers. An additional pump operated in cycles of 15 minutes ON and 15 minutes OFF acted to mix the reactor contents by recycling the biomass within the anaerobic bioreactor tank through venturi nozzles located at the base of this chamber. The AnMBR was inoculated with 700 litres of municipal digested sludge of 2.5 % total solids (70 % volatiles) and 500 L of wastewater, making a total concentration of 10 gVSS L^{-1} in the reactor. Digested sludge was sourced from a municipal anaerobic digester treating a mixture of primary and secondary sludges.

The G-AnMBR comprised two Perspex columns of 25 cm and 20 cm of internal diameter for the biological and membrane tanks respectively. Influent wastewater was introduced through the bottom of the biological tank via a peristaltic pump which was controlled with a level sensor setting the water level to a height of 1.5 m from the base of the column. The effluent from the biological tank overflowed into the membrane tank through a floating valve and was then recycled back using another peristaltic pump. Internal recirculation is typical of EGSB systems in which high upflow velocities are employed in order to enhance substrate biomass contact when low biogas production is expected (Lettinga *et al.* 2001) and prevent accumulation of influent solids in the sludge bed. However in the present experiments a low upflow velocity of 0.7-1 m h^{-1} was applied so as to reduce the accumulation of solids in the membrane tank. As a result the 40 L of granular sludge sourced from a sugar processing factory that were used as seed material in the G-AnMBR conformed a highly dense packed sludge bed with biomass concentration of 70 gVSS L^{-1}.

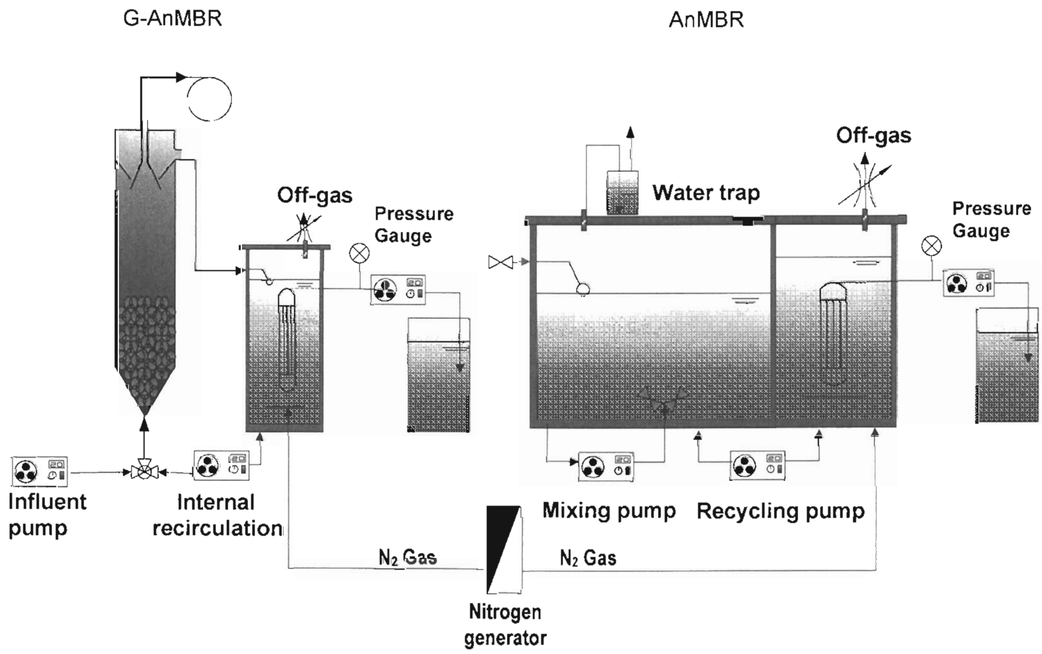

Figure 5.1 Schematic representation of G-AnMBR (left) and suspended AnMBR (right)

Membrane modules with a pore size of 0.08 μm and 0.04 μm and surface area of 0.93 m^2 and 12.5 m^2 were used in the granular and suspended AnMBRs respectively (Table 5.1). Nitrogen gas provided by a nitrogen generator was employed for membrane cross flow gas sparging. Gas velocities ranging from 0.02 m s^{-1} to 0.078 m s^{-1} and from 0.02 m s^{-1} to 0.057 m s^{-1} were applied during normal operation in the suspended and granular anaerobic systems and calculated based on the gas sparging rate and the free cross sectional area of the membrane modules (Table 5.1). The permeate was extracted through peristaltic pumps in which the flux was set at 6 LMH equating to a hydraulic retention time of 16 hours. The suspended system was operated at a sludge retention time of 100 days, while in the G-AnMBR system no biomass was withdrawn throughout the experiment, except for 1-2 Litres sampled each week from the membrane tank for analysis.

Table 5.1 Bioreactor characteristics and operational parameters

Parameter/System	Units	G-AnMBR	AnMBR
Bioreactor Characteristics			
Biological tank volume	L	100	1000
Membrane tank volume	L	25	200
HRT	h	16	16
SRT	d	-	100
Membrane Characteristics			
Flux	LMH	6	6
Area	m^2	0.93	12.5
Material	-	PVDF	PVDF
Pore size	μm	0.04	0.08
Module length	m	0.7	1
Cross sectional area	cm^2	73.6	176.7
Packing density	$m^2\ m^{-3}$	300	710

5.2.2 Sampling, monitoring and analyses

Partially settled primary wastewater was used as a feed source throughout and was taken from a continuous ring main fed from Cranfield University's sewage works. Temperature and pH were not controlled during the entire experimental period. However temperature was continuously monitored online using a DL2e logger (Delta-T Devices Ltd., Cambridge) while pH was measured in the effluent samples analysed in the laboratory according to standard methods (APHA, 1998). Transmembrane pressure was measured in the permeate line of both suspended and granular systems using a Druck pressure transducer (Druck Ltd, Leicester) and logged via an ADC16 digital analog converter (Pico Technology Ltd. , St Neots, Cambs.) into a personal computer. Integrated samples from the influent and membrane effluent were collected each 2-3 days by storing them in auxiliary tanks, together with grab sludge samples obtained from wasted sludge. Influent and effluent COD, NH_4-N, and BOD_5 were analyzed with Merck vial test kits (VWR International adapted from Standard Methods, APHA, 1998). Mixed liquor suspended solids (MLSS) were measured according to Standard Methods

(APHA, 1998) and sludge supernatant was obtained by centrifugation of the sludge at 10000 g for 15 minutes followed by filtration of decanted liquid with 1 μm membranes (Judd, 2006). Protein concentration was determined with the phenol sulphuric method according to Dubois (1956) with modifications usin bovine serum albumin (BSA) as a standard while carbohydrates were quantified according to Lowry (1951) with modifications using glucose as calibration reference.

5.2.3 Molecular weight fractionation

Molecular weight fractionation of the sludge supernatant was determined using an Amicon 8400 dead-end stirred cell (Millipore, USA) by sequential filtration with polyethersulphone UF membranes of a nominal molecular weight of 10, 100 and 500 kDa. All experiments were conducted at room temperature and 2 bar pressure applied using the pure nitrogen gas supply.

5.2.4 Critical flux tests

The critical flux was determined by applying the flux step method (Le Clech *et al.*, 2003) in which successive filtration steps with flux increments of 2-3 LMH were maintained during 15 minutes. The trials were conducted in batches of 25 L of sludge placed in a cylindrical tank, with a membrane module with the same characteristics as the one employed in the G-AnMBR during continuous operation. For each of the sludges a series of four critical flux experiments at gas velocities ranging from 0.07 m s^{-1} to 0.57 m s^{-1} were conducted.

5.3 RESULTS AND DISCUSSION

5.3.1 Treatment performance

The granular and suspended AnMBRs showed similar treatment performance in terms of COD and BOD removal. Effluent COD concentrations ranged between 115 mgCOD L^{-1} and 8 mgCOD L^{-1} in the G-AnMBR and between 139 mgCOD L^{-1} and 13 mgCOD L^{-1} in the AnMBR throughout the 250 days of operation. The highest values corresponded to the start up phase just after inoculation of the reactors without previous acclimatisation. Taking into account the last 100 days of operation in which the influent COD averaged of 338 mgCOD L^{-1} (Table 5.2), both anaerobic systems presented COD removals of 84-86 %, which agrees with the anaerobic biodegradability of sewage (Elmitwalli *et al.*, 2001).

Table 5.2 Influent characteristics and performance in the AnMBR and G-AnMBR

Parameter	Unit	Influent	AnMBR*	G-AnMBR*
COD	mg L^{-1}	338 (74)	-	-
Soluble COD	mg L^{-1}	183 (50)	54 (20)	47 (24)
BOD_5	mg L^{-1}	155 (46)	10 (4)	11 (5)
TSS/MLSS	mg L^{-1}	0.084	7.7	0.1-0.6
%VSS/MLVSS	%	92	86	87
NH_4-N	mg L^{-1}	35 (8)	40 (12)	39 (8)
pH	-	7.5 (0.2)	8.1 (0.3)	8.1 (0.2)

*Standard deviations in parenthesis

However, treatment performance was affected by temperatures as both granular and suspended MBRs showed effluent COD concentrations which increased from 25 mgCOD L^{-1} to 75 mgCOD·L^{-1} as temperature decreased from 20 °C to 10 °C (Figure 5.2). The decrease in removal efficiency from 92 % to 78 % compares well with results obtained by Wen (1999) and Chu (2005) who reported a decrease in COD removal from 95 % to 80 % and from 97 % to 88 % as temperature decreased from 25 °C to 11 °C in UASB and EGSB reactors coupled to membrane filtration. The increase in effluent COD with decreasing temperatures appeared to be due mainly to non biodegradable organic matter as the BOD remained between 5 mgBOD L^{-1} and 15 mgBOD L^{-1} irrespective of the operating temperature and reactor configuration (Figure 5.2). The low levels of effluent BOD found throughout the study indicate that VFAs were not accumulated in the reactors and that methanonogenesis was not the rate limiting step of the digestion process.

It is unlikely that the inert COD measured in the AnMBR originates directly from influent organics not degraded at lower temperatures as the feed wastewater was aerobically degradable as shown by the BOD_5 to COD ratio of 0.5. Therefore two possible reasons for the generation of inert material within the AnMBRs at lower temperatures can be considered. The first one is related to the effect of temperature on the fraction of VFA-yield which represents the amount of solubilised matter converted to VFA (Ucisik & Henze, 2008). Although several studies have demonstrated that the formation of non VFA soluble organics from primary sludge as temperature decrease is insignificant (Maharaj & Elefsiniotis, 2001), the results showed in the present study agree with those which report that increasing amount of non degradable soluble COD is generated at lower temperatures (Ferreiro & Soto, 2003; Cokgor *et al.,* 2008). However it is more likely that the higher amounts of non biodegradable permeate COD represent an increased production of SMP or decreased SMP biodegradation rates at lower temperatures (Schiener *et al.,* 1998; Barker & Stuckey, 1999). In contrast to the result obtained in the present study residual COD originated from effluents of different anaerobic reactors was easier to degrade aerobically than anaerobically (Schiener *et al.,* 1998). This will imply the introducing a membrane results in the rejection of high molecular weight organics which are retained in the system until they are completely mineralized. Supporting this view, Ince (2000) showed that accumulation of soluble compounds in the supernatant of an anaerobic MBR corresponded mainly to aerobically inert compounds which were linked to metabolic products as the influent wastewater was completely degradable under aerobic conditions.

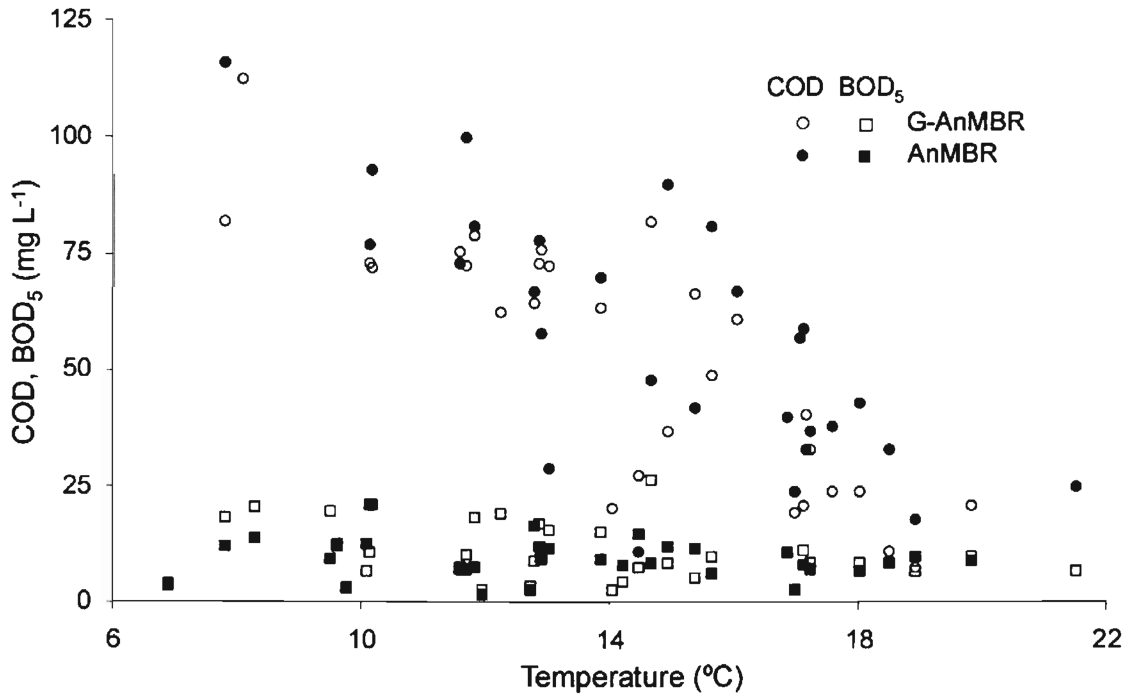

Figure 5.2 Effect of temperature on permeate COD and BOD_5 concentration in G-AnMBR and AnMBR

Although plug flow reactors are more effective than mixed systems due to the higher removal rates at the start of the sludge bed where higher substrate concentrations exist (Batstone,

2006) results from the present study have shown not only that granular and suspended systems present comparable effluent qualities, but also that their response to lower temperatures is very similar with respect to production of inert organics. Therefore it is likely that the advantage of up flow reactors is only relevant for high strength influent wastewaters and that their hydrodynamic behaviour approaches a completely mixed regime (Hulshoff Pol *et al.*, 2004), especially if as in the present study a short sludge bed is employed. Overall, the low levels of effluent BOD found through the study indicate that VFA were not accumulated in the reactor highlighting that stable methanogenisis can be achieved at low temperatures (Lettinga *et al.*, 2001) and that it wasn't the rate limiting step of the digestion process. These results have important implications as the lower effluent quality obtained at lower temperatures is mainly due to biologically inert organics and thus bioavailability for nutrient and carbonaceous aerobic polishing would be limited.

5.3.2 Suspended solid concentration and supernatant characteristics

Recycling of the mixed liquor from the membrane tank to the bioreactor at a low upflow velocity of 0.7-1 m h^{-1} enhanced interception of solids in the sludge bed limiting their accumulation in the granular system. The MLSS concentration in the membrane tank increased slowly from the levels in the influent wastewater (Table 5.1) up to 0.6 gMLSS L^{-1} in 150 days of operation. Therefore most of the influent solids were entrapped in the sludge bed. On the other hand, the levels of MLSS in the AnMBR ranged between 6.6gMLSS L^{-1} and 9.6 gMLSS L^{-1} and in the last 100 days of operation averaged 7.7 gMLSS L^{-1} of which 6.6 gMLSS L^{-1} were volatiles. Higher concentrations of colloidal matter were encountered in the supernatant of the suspended growth AnMBR (Figure 5.3). Average concentrations of SMP measured as COD, proteins and carbohydrates in sludge supernatant were 598 $mgSMP_{COD}$ L^{-1}, 108 $mgSMP_P$ L^{-1}, and 47 $mgSMP_C$ L^{-1} for the suspended growth AnMBR and 198 $mgSMP_{COD}$ L^{-1}, 50 $mgSMP_P$ L^{-1}, 18 $mgSMP_C$ L^{-1} for the granular system.

However, molecular weight fractionation of both supernatants revealed that the difference in concentration was due to the higher colloidal fraction between 1.2 μm and 500 kDa which represented 76 % and 86 % of the total COD in the granular and suspended AnMBRs respectively (Table 5.3). Similar results were recently reported by van Voorthuizen (2008), who attributed the higher fouling propensity of an anaerobic MBR as compared to an UASB reactor system coupled to membrane due to the higher concentration of colloidal matter.

Fractionation revealed that while the levels of SMP_{COD} were 269 $mgSMP_{COD}$ L^{-1} and 161 $mgSMP_{COD}$ L^{-1} in an anaerobic and aerobic MBR respectively, colloidal COD obtained with filter paper was 1270 mgCOD L^{-1} and 123 mgCOD L^{-1} respectively. The higher concentration of high molecular weight compounds found in the suspended systems can be attributed to the production of colloidal particles arising from reduction in particle size of influent solids (Lant & Hartley, 2007), dispersive growth of anaerobic bacteria (Imasaka *et al.,* 1989; Elmitwalli *et al.,* 2001) or cell decay products (Boero *et al.,* 1996; Hu & Stuckey, 2006) which could have been physically adsorbed and retained in the sludge bed of the granular system (Mahmoud *et al.,* 2003). Although the molecular weight fraction below 10 kDa represented 21 % and 8% of the SMP_{COD} in the G-AnMBR and AnMBR respectively, similar concentrations of 60.5 mgCOD L^{-1} and 56.1 mgCOD L^{-1} were found in both systems which is consistent with the comparable concentrations of effluent COD found through the study. This fraction represents 17% of influent total COD, which is between the 25-30 % reported for SMP concentration in AnMBR studies with higher sludge retention times (Harada *et al.,* 1994; Aquino *et al.,* 2006; Baek & Pagilla, 2006) and 7-10% reported at lower sludge ages of 30-60 days (Huang *et al.,* 2008).

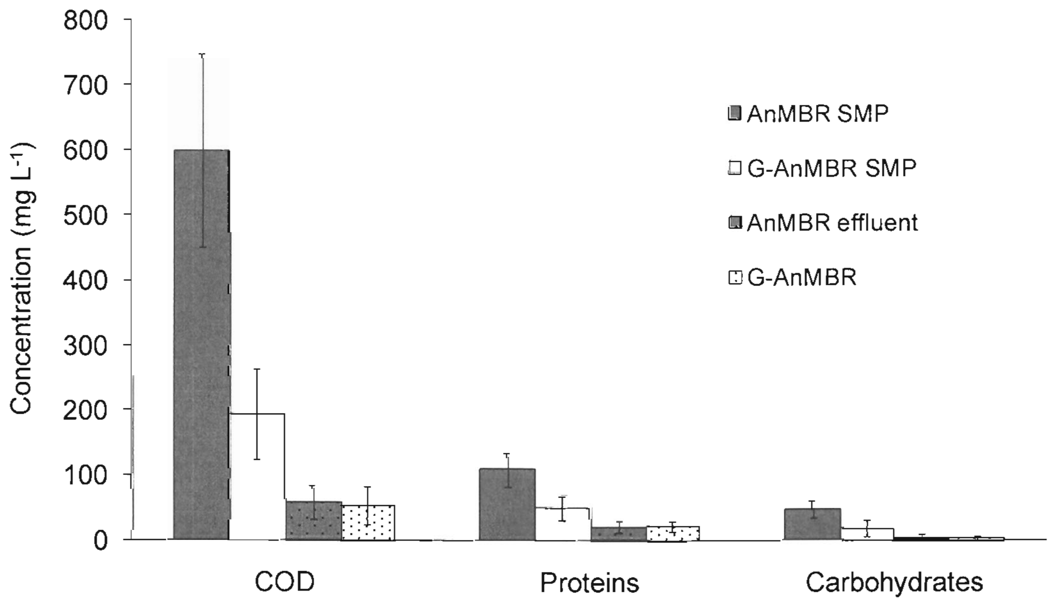

Figure 5.3 Supernatant and effluent COD, proteins and carbohydrate concentrations in G-AnMBR and AnMBR

The chemical composition of the SMP was dominated by proteins whose concentration averaged 108 ± 27 mgSMP$_P$ L^{-1} and 50 ± 18 mgSMP$_P$ L^{-1} in the dispersed and granular systems respectively. The correspondent levels of carbohydrates were 47 ± 14 mgSMP$_C$ L^{-1} and 18 ± 13 mgSMP$_C$ L^{-1}, resulting in a ratio of SMP$_P$:SMP$_C$ of 2.7 and 2.3 in the granular and suspended AnMBRs respectively. In AnMBR studies treating synthetic wastewaters in which it is considered that influent non degraded products do not contribute to the SMP higher concentrations of proteins resulting in SMPp:SMPc ratios between 2 and 7 have also been reported (Harada *et al.*, 1994; Aquino *et al.*, 2006). However, the higher concentration of proteins reflects in part their lower anaerobic biodegradability over that of carbohydrates: biodegradability of proteins is around 30-40 % compared to 70-90 % for carbohydrates (Miron *et al.*, 2000). Considering that degradation of 1 g of proteins results in 0.16 g of ammonia nitrogen the difference of 4-5 mgNH$_4$-N L^{-1} between influent and effluent inorganic nitrogen (Table 5.2) would imply that approximately between 25 mg L^{-1} and 30 mg L^{-1} of proteins were degraded which accounts to 37 %-44 % removal and so fits within the overall picture.

Table 5.3 SMP$_{COD}$ fractionation in AnMBR and G-AnMBR

Fraction X (kDa)	G-AnMBR	AnMBR
Total	287 ± 12	657.7 ±12.7
X> 500	225 ± 2.0	571.7 ±12.7
100 < X < 500	0 ± 1.6	0 ± 4.3
10 < X < 100	1.5 ± 3.6	29.9 ± 4.2
< 10 kDa	60.5 ± 3.5	56.1 ± 0.4

All units are in mgCOD L^{-1}. Standard deviations in parenthesis

5.3.3 Critical flux tests: effect of gas sparging intensity

Increasing gas sparging intensities in the suspended AnMBR from 0.007 m s^{-1} to 0.041 m s^{-1} resulted in an increase in critical flux from 1.9 to 5.4 LMH which increased further to 9.7 LMH when the gas velocity was further increased to 0.057 m s^{-1} (Figure 5.4 top). The limited effect of gas sparging in flux enhancement was also reported by Jeison (2007) with gas velocities ranging from 0.029 m s^{-1} and 0.138 m s^{-1} suggesting that fouling is dominated by deposition of colloidal matter as these experience lower backtransport velocities generated by shear in membrane liquid interface (Belfort *et al.,* 1994; Tardieu *et al.,* 1998). Also supporting the importance of colloidal fouling in AnMBRs, is the observation that external fouling such as cake layer or gel layer is the major contributor to resistance to filtration (Chu *et al.,* 2005; Jeison & van Lier, 2006).

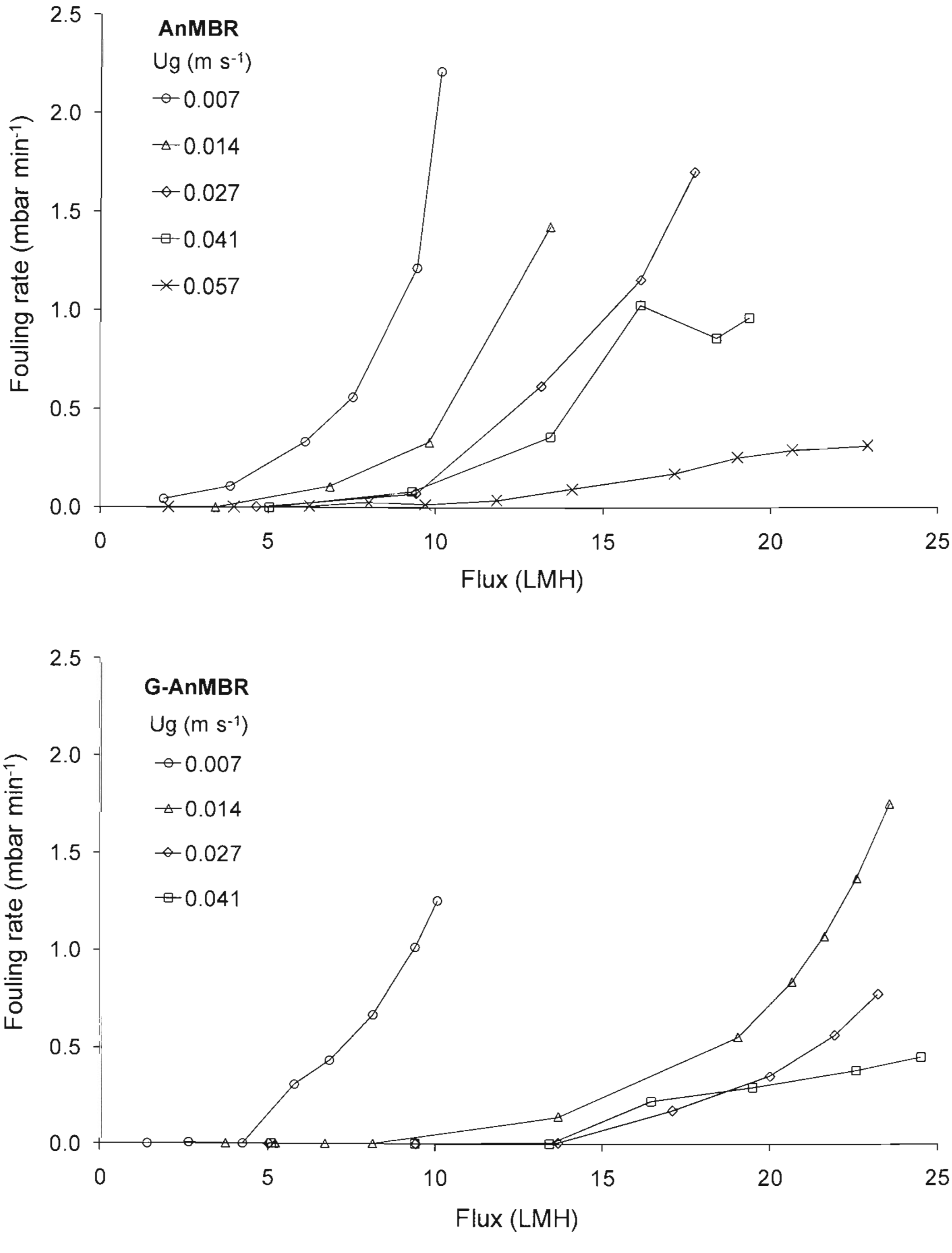

Figure 5.4 Effect of gas sparging velocity and flux on fouling rates in AnMBR (top) and G-AnMBR (bottom)

The critical fluxes in the G-AnMBR were 4.3, 8.1, 13.7 and 13.4 LMH for gas sparging intensities of 0.007, 0,014, 0,027 and 0,041 m s^{-1} respectively (Figure 5.4 bottom). Although ostensibly higher critical fluxes were not achieved in the G-AnMBR compared to the suspended system for a given sparging intensity, higher fouling rates and lower critical fluxes are obtained in the suspended system. These results are of practical importance as they suggest that the granular system would require lower gas sparging intensity and therefore lower energy requirements for fouling control. Supporting this view, van Voorthuizen (2008) found that filtration of a suspended growth AnMBR sludge exhibited a continuously increasing resistance as opposed to filtration of UASB effluent which remained constant despite employing one fourth of the gas demand used in the suspended system.

5.3.4 Long term continuous filtration

Sustainability of MBR operation under sub critical conditions is evaluated according to the fouling rate and the duration of the slow fouling phase obtained in long term filtration trials (Pollice *et al.,* 2005) . Previous studies that have evaluated flux sustainability have focused on increasing flux below the critical flux previously obtained with some form of the flux step method. Although results don't usually correlate well across different studies, it is has generally been observed that while fouling rates increase, the duration of the slow fouling rate decrease when fluxes aproach the critical flux (Cho & Fane, 2002; Guglielmi *et al.,* 2007; Guglielmi *et al.,* 2008). In the current case this was applied to anaerobic biomass so that the effect of gas sparging in long term trials could be employed to ascertain which bioreactor configuration required lower energy for fouling control. During the course of the experiments both AnMBR and G-AnMBR were operated at a constant flux of 6 LMH while gas sparging intensities calculated as the flow of nitrogen normalised against module cross sectional area (see Table 5.1 for module characteristics) varied between 0.077 and 0.017 m s^{-1} and between 0.057 and 0 m s^{-1} respectively. The effect of gas velocity on the TMP transients of the AnMBR and the G-AnMBR are shown in Figures 5.5 and 5.6 respectively. It was observed that TMP profile in the suspended AnMBR followed the two phase mechanism previously reported in aerobic and anaerobic MBRs studies (Figure 5.5). The initial slow TMP rise phase yielded fouling rates of 0.5, 0.6 and 3.7 mbar d^{-1} which were maintained during 85, 30 and 6 days at gas velocities of 0.077, 0.038 and 0.028 m s^{-1} respectively before a sharp increase in TMP occurred. A further decrease of gas velocity to 0.017 m.s^{-1} resulted in a fast TMP increase from 60 mbar to 300 mbar in less than 20 hours of filtration (results not shown).

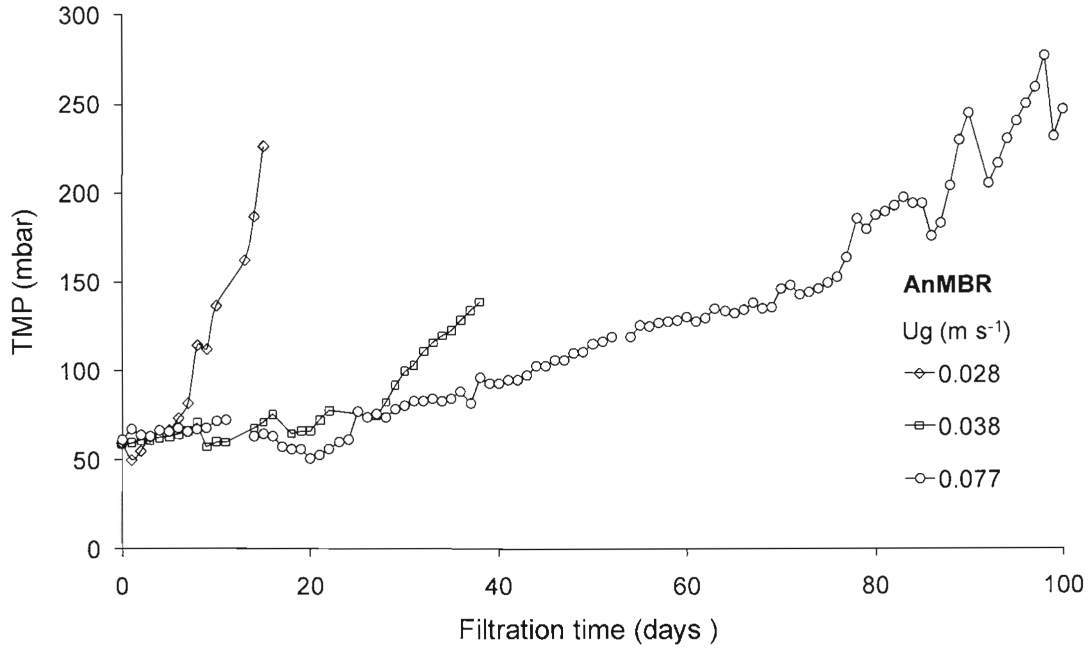

Figure 5.5 Effect of gas sparging rate on TMP transients of suspended AnMBR

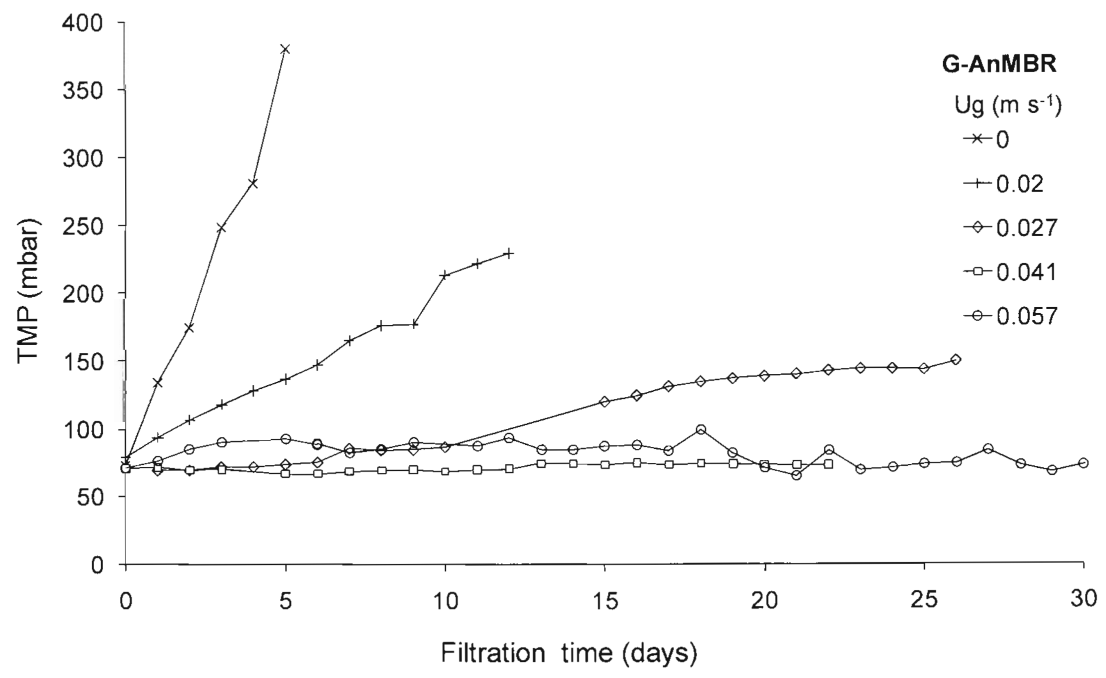

Figure 5.6 Influence of gas sparging on TMP transients in G-AnMBR

This could be considered the critical gas velocity which marks the transition between fast and slow fouling rate. Interestingly above this threshold, increasing gas sparging intensities prolongs the duration of filtration but does not affect the residual fouling rate in the range of gas sparging rates studied. This behaviour is consistent with the fouling mechanism proposed by several authors (Cho & Fane, 2002; Ognier *et al.*, 2004; Guglielmi *et al.*, 2007). Under subcritical conditions in which fouling is caused by accumulation of colloidal and soluble matter, varying gas intensities do not significantly affect residual fouling rate because higher shear only increase backtransport velocity of larger foulants. Additionally, although the effective membrane area is reduced at the same rate, the TMP jump is reached before because the critical flux, representing fouling caused by suspended solids is reduced with decreasing gas sparging intensity.

The difference between fast and slow fouling rates was more evident for the suspended MBR than for the granular system. In the latter a stable TMP of 85 mbar during 80 days was observed for the highest gas velocity of 0.057 m s^{-1}, while for 0.041 and 0.027 m s^{-1} fouling rates increased from 0.27 to 1.8 mbar d^{-1} (Figure 5.6). Below 0.27 m.s^{-1}, which was the gas sparging rate that marked the transition between slow and fast fouling in the suspended system, filtration without gas sparging and 0.2 m s^{-1} resulted in fouling rates of 58 mbar d^{-1} and 13 mbar d^{-1} respectively. Similar fouling rates were found by Wen (1999) and Chu (2005) who studied the effect of upflow velocity combined with intermittent filtration cycles in granular AnMBRs without gas sparging. Although stable TMP was not achieved fouling rates of 48 mbar $d^{-1}$and 120 mbar d^{-1} allowed continuous operation for over 140 and 70 hours respectively. These fouling rates lie between residual and fast reversible fouling rates reported in AeMBR studies using relaxation and backwash in combination with air sparging (Guglielmi *et al.*, 2008) and suggest that fouling could be controlled by applying low gas intensities together with physical membrane cleaning procedures such as relaxation and backwashing providing that fouling is reversible.

To illustrate this further, a series of shorter term experiments in which cyclic gas sparging was combined with backflushing were performed. Gas sparging intensities of 0.077 and 0.041 m s^{-1} were intermittently supplied in cycles of 10 seconds on and 10 seconds off in the suspended and granular AnMBRs respectively resulting in a 50 % reduction of gas demand with respect to the corresponding continuous filtration experiments. Accordingly, membrane flux was increased to 7 LMH and backflushing was applied every 10 minutes for one minute at that same flux in order to operate at a net flux of 6 LMH. In Figure 5.7 the maximum TMP monitored at the end of each filtration cycle has been represented for the granular and suspended anaerobic MBRs operated under the above described conditions. The TMP profile in the suspended AnMBR revealed that although backwashing was effective in removing fouling within filtration cycles, the TMP reached 300 mbar within 3 hours and then increased at a rate of 4.3 mbar d^{-1} before a sudden TMP rise was observed after 170 hours of operation. Compared to the continuous filtration experiment at the same net gas demand (0.038 m s^{-1}) permeability, fouling rate and duration of slow fouling phase increased by a factor of 4, 7 and 4 respectively. In the granular system TMP increased between 5 and 10 mbar within cycles and residual fouling rate was limited to approximately 1 mbar d^{-1}, less than one tenth of that obtained under equivalent gas demands in continuous filtration trials (0.02 m s^{-1}). Figure 5.7 also shows the TMP transient of another experiment in the granular system in which gas sparging was employed during one minute each 10 minutes. During filtration phase, without gas sparging TMP increased in average 40 mbar, while residual fouling was 3 mbar d^{-1}. Overall these results indicate that while the suspended system requires continuous gas sparging in order to prolong membrane operation, in the granular system gas demands can be reduced by employing intermittent gas sparging in combination with backflushing.

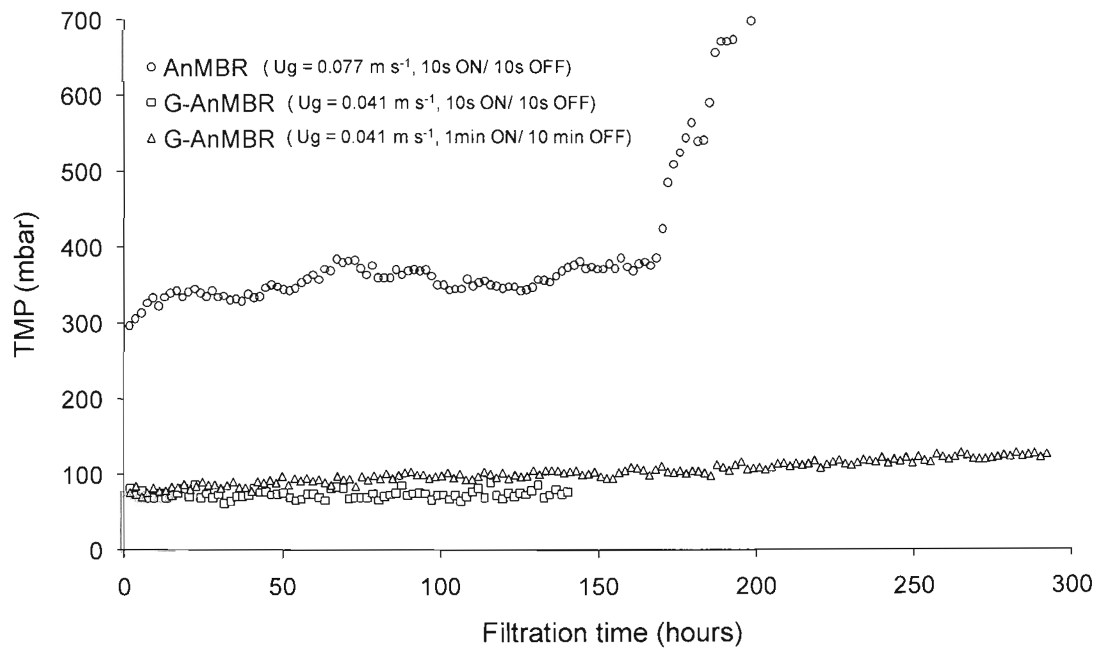

Figure 5.7 TMP transients in G-AnMBR and AnMBR operated with intermittent gas sparging

5.4 CONCLUSIONS

Attached and suspended submerged anaerobic membrane bioreactors were operated in parallel with settled sewage at low-medium temperatures for more than 250 days in order to investigate differences in terms of biological and membrane performance associated to the different reactor configurations and sludge inoculum. No significant differences were observed between granular and dispersed systems with respect to COD and BOD removal. In fact it was found that temperature had the same effect on both systems. While effluent COD concentrations varied between 100 mgCOD L^{-1} and 25 mgCOD L^{-1} as temperature increased from 8 °C to 22 °C, BOD_5 remained constant at an average concentration of 5-15 mgBOD L^{-1}. As a result of uncoupling biological and filtration steps both MLSS and colloidal organic matter concentration were reduced by a factor of 10 and 3 in the granular AnMBR with respect to the suspended system thus reducing the organic loading to the membrane. Although critical flux steps revealed that increasing gas sparging intensities results in similar enhancement of permeate flux the results suggested that lower gas sparging intensities were required in the attached system. This was further corroborated in long term filtration trials which revealed that difference in fouling rates between suspended and attached systems increase as gas sparging intensities are reduced. Therefore it was demonstrated that the attached AnMBR exhibited greater potential in reducing energy demands especially when backwashing was implemented.

5.5 ACKNOWLEDGEMENTS

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CHAPTER 6:

COMPARISON OF FOULING CONTROL STRATEGIES IN AEROBIC AND ANAEROBIC MBRs

CHAPTER 6. COMPARISON OF FOULING CONTROL STRATEGIES IN AEROBIC AND ANAEROBIC MBRs

Martin Garcia, N.[1], Monsalvo, V.[2], Pidou, M.[1], Judd, S. J.[1] and Jefferson, B.[1]
[1]Centre for Water Science, Cranfield University, Cranfield, Beds, MK43 0AL, UK
[2] Dept. Ingeniería Química, Universidad Autónoma de Madrid, Madrid, 28049, Spain

ABSTRACT

In this study different fouling control strategies were investiagted in a granular and suspended anaerobic MBR sludges and compared with biomass sourced from an aerobic MBR. Fouling propensity was evaluated with the flux step method at varying gas sparging rates in tubular and hollow fibre membranes. The influence of crossflow velocity in sidestream configuration was also examined. The different sludges were characterised in terms of particle size distribution and soluble microbial products (SMP) concentrations and chemical composition in order to ascertain the relative impact of different biomass characteristics on fouling propensity. After 200 days of operation both granular and suspended growth AnMBRs were characterized by the presence of a population of colloidal particles while the aerobic system showed an unimodal particle size distribution with a d_{50} of 20 μm. The suspended growth AnMBR presented the lowest critical fluxes under all experimental conditions and membrane configurations. The G-AnMBR with double the SMP_{COD} and a 15 fold lower biomass concentration in relation to the AeMBR presented a critical flux of 14 LMH with hollow fibre membranes at the maximum gas sparging rate tested compared to 16 LMH in the AeMBR. A more prominent increase in the critical flux was observed for the G-AnMBR as compared to the AeMBR in sidestream pumped configuration indicating that the higher fouling propensity anticipated in the former due to the higher colloidal organic concentration was compensated by the higher shear resulting from its lower viscosity.

Keywords: Fouling, anaerobic, membrane bioreactor, critical flux, submerged, sidestream.

6.1 INTRODUCTION

Membrane bioreactors consist of the use of membranes coupled to a biological process in order to achieve efficient separation of treated water from sludge generated. The advantages of MBRs when coupled to aerobic biological process are 1) improved effluent quality due to the rejection not only of solids but also of high molecular and colloidal organics, 2) lower footprint as high biomass concentration can be achieved in reduced space 3) complete control over sludge retention time as solid-liquid separation is not limited by biomass concentration and 4) ease to retrofit existing activated sludge plants. These advantages, specially 1) and 3) are perhaps more crucial for anaerobic systems than for aerobic ones as they present lower effluent qualities and are more sensitive to biomass losses due to the lower growth rates of anaerobic microorganisms. Considering anaerobic MBRs would therefore overcome some of the limitations of anaerobic technology at the same time as potentially reducing the energy demand of the process and minimise the costs arising from sludge treatment due to the absence of biological aeration and the lower biomass productions they present.

However, fouling remains one of the main disadvantages in MBRs as it negatively affects membrane hydraulic performance, resulting in an increase in operational costs with respect to other conventional treatment technologies due to the energy required to control fouling. Application of turbulent hydrodynamic conditions in the liquid membrane interface either through gas sparging in submerged membranes or by liquid pumping in sidestream operation are together with flux reduction, relaxation, backwashing and chemical cleaning the most common fouling control strategies (Judd, 2008). In submerged MBRs membranes are immersed in the bioreactor itself and gas is sparged below the membrane module in the form of air or biogas in the case of aerobic and anaerobic MBRs respectively, in order to scour the membrane surface and prevent clogging and deposition of large particles. On the other hand, in side stream pumped crossflow operation the membrane module is located outside the bioreactor and the mixed liquor is pumped through the membrane module and recycled back to the bioreactor providing enough turbulence to enhance the back transport of foulants from its surface. Although it has been extensively documented that pumping biomass across membrane surface provides better hydrodynamic conditions than gas sparging in immersed configuration, turbulent aeration onto hollow fibre and flat sheet membrane geometries is established as the most cost effective means of fouling control for domestic wastewater treatment applications due to the lower energy they require (Le-Clech *et al.*, 2006). Although much less common in municipal applications than hollow fibre and flat sheet geometries, gas sparging in tubular membranes has been shown to be a very efficient means of achieving high fluxes with relatively low gas demands (Le-Clech *et al.*, 2003); In gas lift operation the gas is injected into the lower end of the membrane module, with the lift effect driving the liquid flow upwards, tangentially across the membrane surface. Hydrodynamics within the membrane module are characterized by slug flow in which succession of liquid and gas slugs induces two alternate shear stresses near the membrane wall of opposite direction exerting a cleaning effect on the membrane surface (Cui *et al.*, 2003).

Recent studies in which AnMBRs have been investigated have employed gas sparging using tubular (Kayawake *et al.*, 1991; Imasaka *et al.*, 1993; Jeison & van Lier, 2007a; Huang *et al.*, 2008; van Voorthuizen *et al.*, 2008) flat sheets (Hu & Stuckey, 2006; Lin *et al.*, 2009) and hollow fibre membranes (Wen *et al.*, 1999; Chu *et al.*, 2005; Hu & Stuckey, 2006). However, while a considerable number of studies have investigated the effect of crossflow velocity on filtration performance in anaerobic MBRs (Beaubien *et al.*, 1996; Choo & Lee, 1996; Elmaleh & Abdelmoumni, 1997; Ghyoot & Verstraete, 1997), the impact of gas sparging rates in immersed configuration has been limited and mainly focused on tubular geometry (Kayawake *et al.*, 1991; Imasaka *et al.*, 1993; Jeison & van Lier, 2007b). Additionally direct comparison of membrane configurations and geometries which is a issue of major interest in AeMBRs (Günder & Krauth, 1999; Le-Clech *et al.*, 2005; Guglielmi *et al.*, 2007; Guglielmi *et al.*, 2008) so as to determine the system that provides better hydraulic performance with lower energy demands have been limited in anaerobic systems. For instance Jesion (2007b) compared liquid pumping and gas sparging in tubular membranes and Hu & Stuckey (2006) operated two AnMBRs in parallel one with flat sheet and another one with hollow fibre membranes.

The aim of the present study is to investigate the effect of gas sparging rate in tubular and hollow fibre membranes and to compare it with the performance of sidestream crossflow configuration. The flux step method was employed to evaluate fouling propensity of suspended and granular anaerobic membrane bioreactor and compared to that of an aerobic MBR using the different fouling control strategies. All sludges were characterized in terms of particle size distribution and SMP concentrations and chemistry in order to provide insights into the relative impact of different biomass characteristics on membrane fouling.

6.2 MATERIALS AND METHODS

6.2.1 Experimental methods

Suspended growth aerobic and anaerobic membrane bioreactors were operated in parallel with a G-AnMBR during 250 days. The characteristics of the different pilot plants and operational conditions have been described previously (Chapters 4 and 5). Fouling propensity of the sludges sourced from the different reactors was evaluated by comparing fouling rates and critical fluxes obtained from the flux-step method (Le Clech *et al.,* 2003) , in which membrane flux is increased in a stepwise manner and the change in transmembrane pressure monitored. During the experiments flux was increased in steps of 2-3 LMH with step duration of 15 minutes. For each biomass the effect of gas sparging on critical flux was evaluated using hollow fibre and tubular membranes which characteristics are depicted in Table 6.1. In order to allow direct comparison between both membrane geometries the same specific gas demands ranging from 0.2 to 1.16 m^3 m^{-2} h^{-1} were applied. Another set of experiments in which the tubular membrane was operated in pumped crossflow configuration were conducted in order to compare the influence of crossflow velocity on critical flux. For each sludge 5 different crossflow velocities ranging from 0.4 to 2 m s^{-1} were investigated. Between each critical flux test, membranes were cleaned overnight with 1 % hypochlorite and clean water permeability was measured before a new trial was conducted.

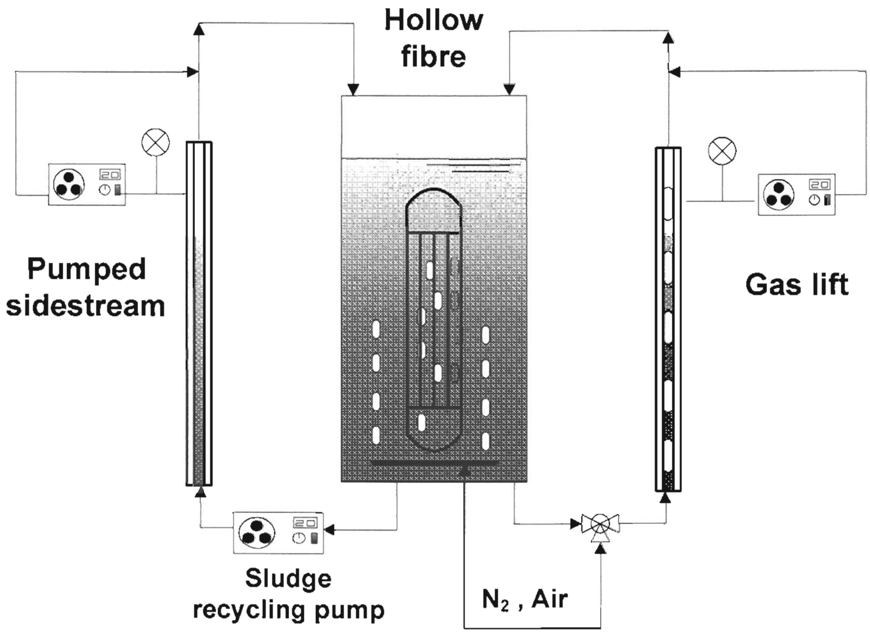


Figure 6.1 Schematic of experimental set up employed for critical flux trials

6.2.2 Experimental set up

Flux step experiments with the AeMBR and AnMBR sludges were conducted using a cylindrical tank in which fresh batches of 25 L of the slurries were placed (Figure 6.1). In the case of the G-AnMBR system, trials were performed directly in the membrane tank of the pilot plant by replacing the hollow fibre membrane used during normal operation with the one employed for the critical flux tests. Nitrogen and air supplied by a nitrogen generator and compressed air line were employed for membrane scouring and biomass mixing of the anaerobic and aerobic sludges respectively. For gas lift experiments the membrane and the tank were connected through a T junction placed at the bottom of the membrane module to allow gas injection. In pumped cross flow operation a centrifugal pump fed from the reservoir tank was employed to generate the cross flow velocities which were set with a valve placed upstream of the membrane module. For both gas lift and pumped crossflow experiments the mixed liquor passing through the membrane module were recycled into the reservoir tank. Similarly, membrane permeate was also recycled back in order to maintain a constant MLSS concentration during the trials.

Table 6.1 Characteristics and tubular and hollow fibre membranes

Parameter/Membrane	Unit	Tubular	Hollow fibre
Filtration Area	m^2	0.93	0.022
Material	-	PVDF	PVDF
Pore size	μm	0.04	0.03
Module length	(m)	0.7	1
Fibre/ lumen diameter	mm	2	8

6.2.3 Analytical procedures

Mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS) and BOD_5 were measured according to Standard Methods (APHA, 1998). Similarly COD, BOD_5, and NH_4-N, were analyzed with Merck vial test kits (VWR International adapted from Standard Methods, APHA, 1998). Sludge supernatant was obtained by centrifugation of the biomass at 10000 g for 15 minutes followed by filtration of decanted liquid with 1 μm membranes (Judd, 2006). Protein concentration was determined with the phenol sulphuric method according to Dubois (1956) with modifications. Bovine serum albumin (BSA) was used as a standard. Carbohydrates were quantified according to Lowry (1951) using glucose as calibration reference. Particle size distribution was determined with a Malvern Mastersizer 2000 particle analyser (Malvern Instrument Ltd.Worcestershire, UK).

6.3 RESULTS AND DISCUSSION

6.3.1 Overall bioreactor performance

All bioreactors were operated in parallel at hydraulic and sludge retention times of 16 hours and 100 days respectively (Table 6.2). In the G-AnMBR no granular biomass was wasted though the experimental period of 250 days. Influent wastewater consisted of settled sewage with an average COD, BOD_5 , SS and ammonia concentrations of 338 mgCOD L^{-1}, 155 mgBOD L^{-1}, 84 mgSS L^{-1} and 35 mgNH_4-N respectively. In the

AeMBR which was started without external seed material, COD removal improved from 85 % to over 95 % as MLSS accumulated from 1 gMLSS L^{-1} to the final average concentration of 8.7 gMLSS L^{-1}, while BOD removal was always over 95 %. Both anaerobic systems presented similar behaviour in terms of treatment performance with COD removal increasing from 80 % after inoculation without previous wastewater or temperature acclimatisation, up to 90 % by the end of the experiment. Effluent BOD ranged between 5 mgBOD L^{-1} and 15 mgBOD L^{-1}, yielding a stable removal of 90-95 %. In the G-AnMBR upflow velocity was kept below 1 m h^{-1} so as to reduce the concentration of solids in the overflow of the biological tank and hence in the membrane feed. As a result solid concentration measured in the membrane tank gradually increased from the levels of the influent wastewater up to 0 6 gMLSS L^{-1}. In the seeded AnMBR, although levels of MLSS ranged between 6.6 gMLSS L^{-1} and 9.6 gMLSS L^{-1} the MLSS stabilized around 7.7 gMLSS L^{-1}.

Table 6.2 Operational parameters and overall bioreactor performance

Parameter	Unit (mg·L^{-1})	AeMBR	AnMBR	G-An MBR
SRT	d	100	100	-
HRT	h	16	16	16
COD removal	%	95	84	86
BOD removal	%	97	93	95
MLSS	g·L^{-1}	8.7	7.7	0.1-0.6

6.3.2 Evolution of particle size distribution

Analysis of particle size distribution measured after 30 days of operation showed that both aerobic and anaerobic MBRs presented unimodal distributions with most frequent particles sizes of 46 μm and 69 μm respectively and corresponding d_{50} values of 32 μm and 60 μm (Figure 6.2 top and Figure 6.2 middle). On the other hand, the G-AnMBR initially showed a bimodal distribution very similar to the one of the influent wastewater with a main fraction comprised of particles of 138 μm and a secondary smaller one in the range of 1 μm (Figure 6.2 bottom). However, floc structure evolved differently in the aerobic and anaerobic systems. While in the AeMBR the normal distribution shifted from a particle size of 44 μm to 20 μm, a bimodal distribution comprising of particles of 11.5 μm and 1.7 μm appeared in the suspended AnMBR after 200 days of operation. Interestingly, the smaller fraction in the AnMBR corresponds well with the one shown in the G-AnMBR and that became predominant at the end of the experiment accounting for more than 5 % by volume of the overall population, while the remaining was comprised of larger particles with a wide range of size distribution between 7 μm and 2200 μm.

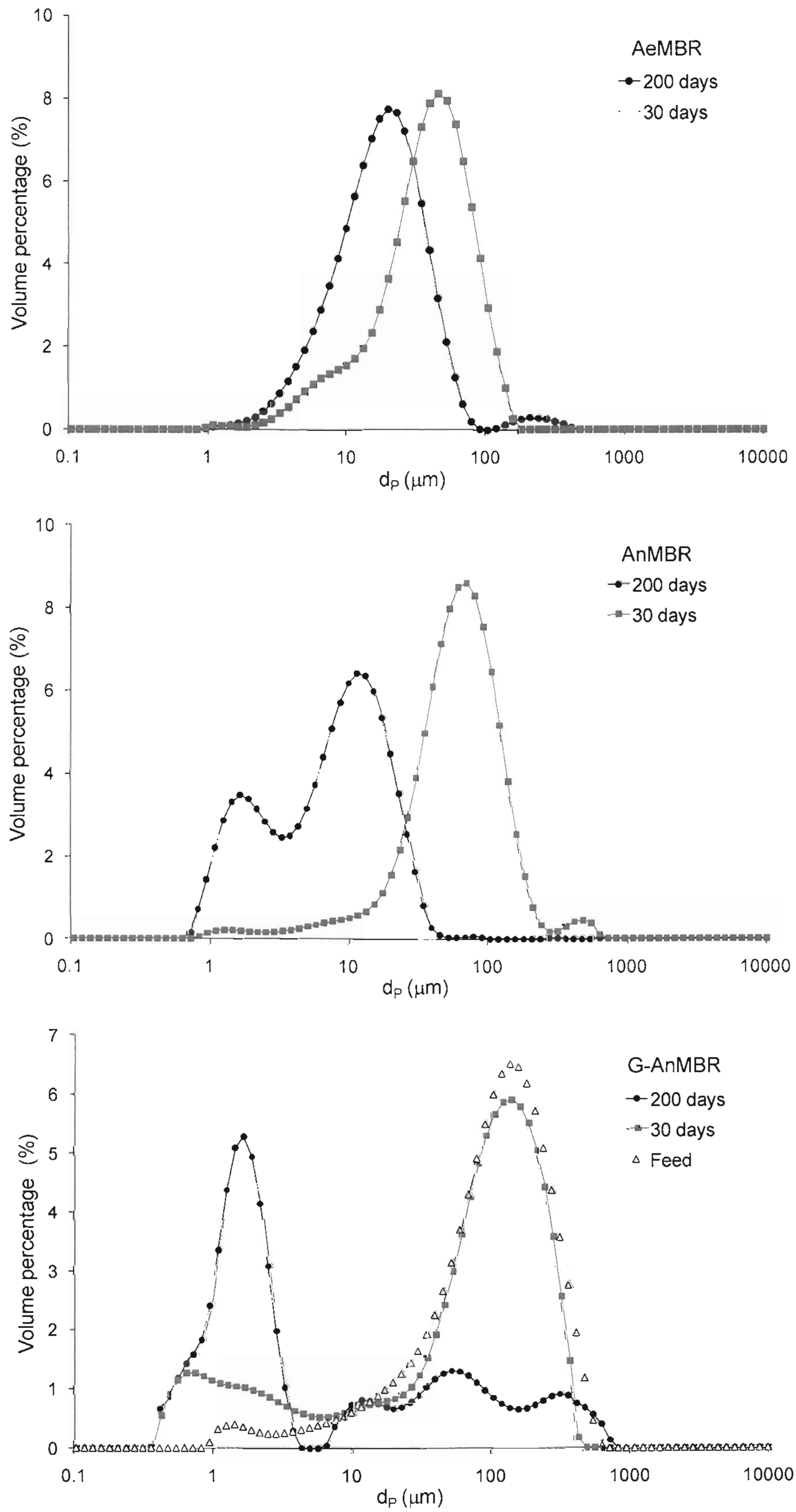


Figure 6.2 Evolution of particle size distribution in AeMBR, AnMBR and G-AnMBR

The higher colloidal content in the anaerobic MBRs as compared to the aerobic system is a result of a higher degree of dispersive growth (Imasaka *et al.*, 1989; Jeison & van Lier, 2007b) and has been associated to high fouling propensities (Jeison & van Lier, 2007b; McAdam *et al.*, 2007; Lin *et al.*, 2009). To illustrate a recent study (Jeison & van Lier, 2007a) in which the performance of a VFA and glucose/VFA anaerobic MBR were compared, showed that the latter presented higher fouling propensity as a result of the high extent of single cell growth of acidogenic bacteria which was evidenced by a reduction in particle size and supported by light microscopy observations.

6.3.3 SMP concentration and composition

Concentrations of SMP measured as COD, proteins and carbohydrates were routinely monitored through the entire experimental period of the different MBRs (Table 6.3). The concentrations of SMP_{COD}, SMP_P and SMP_C in the AeMBR averaged 99 $mgSMP_{COD}\ L^{-1}$, 33 $mgSMP_P\ L^{-1}$ and 9 $mgSMP_C\ L^{-1}$ respectively. The SMP_{COD} corresponds well with results reported by Spérandio (2005) in an AeMBR operated under similar operational conditions and with similar wastewater characteristics. Corresponding values for the same parameters in the suspended growth anaerobic system were 598 $mgSMP_{COD}\ L^{-1}$, 108 $mgSMP_P\ L^{-1}$ and 47 $mgSMP_C\ L^{-1}$ while for the G-AnMBR values of 198 $mgSMP_{COD}\ L^{-1}$, $mgSMP_P\ L^{-1}$ and 18 $mgSMP_C\ L^{-1}$ were observed. These values are higher than the 145-150 mg $SMP_{COD}{\cdot}L^{-1}$ reported by Hu & Stuckey (2006) and Chu (2005) in the treatment of diluted wastewaters in AnMBRs operated at a high sludge ages of 140-150 days. However since these studies obtained sludge supernatant by filtration with 0.45 μm membranes the concentration of SMP may be lower than the ones obtained in the present work in which 1 μm filters were employed specially given the presence of a high content of submicron sized colloids as shown by particle size distribution analysis of both anaerobic systems (Figure 6.2). Additionally the particle size distribution agrees with supernatant analysis of molecular weight fractionation which indicated that whilst in the anaerobic systems more than 80 % the SMP were between 1 μm and 500 kDa (Table 5.3, Chapter 5), in the AeMBR only 40 % were present in that fraction (Table 4.4, Chapter 4).

Table 6.3 Average concentrations of SMP_{COD}, SMPc and SMPp in AeMBR, AnMBR and G-AnMBR

	SMP_{COD} ($mg \cdot L^{-1}$)	SMPc (mg glucose eq$\cdot L^{-1}$)	SMPp (mg BSA eq$\cdot L^{-1}$)
AE MBR	99 ± 28	18 ± 9	18 ± 13
AN MBR	598 ± 150	47 ± 14	108 ± 27
G-An MBR	198 ± 73	18 ± 12	50 ± 21

6.3.4 Critical fluxes in gas lift membrane configuration.

The injection ratios obtained by measuring the amount of liquid displaced by the gas lift effect for the different gas sparging rates (Table 6.4) passed through a minimum value of 0.31 and 0.35 for the aerobic and anaerobic MBR sludges respectively after which they increased up to 0.43 and 0.52 for the gas velocity of 0.21 m s^{-1}. Injection ratios in the range 0.2-0.9 correspond to "slug flow". However, in the G-AnMBR in which the liquid flow induced by the gas lift was approximately three times higher as compared to the aerobic and anaerobic suspended systems injection ratios continuously decreased from 0.345 to 0.125 which correspond to the less favourable bubbly flow conditions for membrane fouling.

Analysis of fouling rates obtained during flux step experiments with the different sludges and for gas velocities ranging between 0.02 to 0.21 m s^{-1} revealed that in gas lift operation with tubular membranes fouling is more influenced by the type of sludge than by gas sparging intensities since irrespectively of gas velocity critical fluxes in the aerobic, anaerobic and granular MBRs remained at 8, 4 and 4 LMH respectively (Figure 6.3). The only noticeable improvement was recorded in the suspended AnMBR in which the fouling rate for a flux of 8 LMH, decreased from 11.8 mbar min^{-1} to 0.26 mbar min^{-1} as gas velocity increased from 0.02 to 0.21 m.s^{-1}. The low efficiency of gas sparging at increasing critical flux could suggest that colloidal and soluble organics with lower backtransport velocities than larger sludge flocs are the main contributors to membrane fouling. Supporting this view, Cui (1997) observed that permeate flux was only dependent on applied pressure within an order of magnitude of change in gas velocity (0.001-0.01 $m.s^{-1}$) during ultrafiltration of Dextran solutions with nominal molecular weight of 260 kDa. Similarly, Chang & Judd (2002) showed that gas sparging had no effect on the hydraulic performance of an air lift AeMBR due to the higher contribution of internal fouling (77 %) as compared to cake layer formation (23 %). Within the same range of gas velocities as the ones used in the present study, Le Clech (2005) observed an increase in critical flux from 16 to 26 LMH in an aerobic system run on settled sewage at a biomass concentration of 3 gMLSS L^{-1}. The discrepancies may be explained by the lower concentration of colloidal and soluble matter which expressed as SMP carbohydrates ranged between 0 and 2.4 $mgSMP_C\ L^{-1}$, which are considerably lower than 32 $mgSMP_C\ L^{-1}$ obtained in the present study for the aerobic system.

Table 6.4 Influence of gas velocity on liquid velocity and injection ratios

	AeMBR		**AnMBR**		**G-AnMBR**	
Ug ($m\ s^{-1}$)	**U_L ($m\ s^{-1}$)**	**ε**	**U_L ($m\ s^{-1}$)**	**ε**	**U_L ($m\ s^{-1}$)**	**ε**
0.02	0.07	0.26	0.09	0.47	0.26	0.35
0.05	0.13	0.27	0.12	0.40	0.36	0.25
0.09	0.20	0.31	0.17	0.35	0.50	0.17
0.14	0.21	0.39	0.21	0.36	0.62	0.15
0.18	0.25	0.42	0.24	0.34	0.71	0.13
0.21	0.28	0.43	0.12	0.52	0.35	0.31

Although previous research has demonstrated that decreasing biomass concentration in anaerobic MBRs improves membrane performance resulting in higher fluxes and reduced gas sparging intensities (Jeison & van Lier, 2006; van Voorthuizen *et al.*, 2008) in the present study neither the lower MLSS or SMP concentration in the G-AnMBR was translated into higher critical fluxes as compared to the suspended system. However due to the lower MLSS concentration, fouling rates for fluxes over the critical value were significantly lower in G-AnMBR. For instance for a flux of 11-12 LMH the fouling rate in the AnMBR varied between 8 mbar min^{-1} and 25 mbar min^{-1} compared to 1-2 mbar min^{-1} in the G-AnMBR. Similar critical flux values of 8 and 4 LMH were reported by Jeison (2007b) in an AnMBR fed with a mixture of VFA and glucose respectively, even though gas velocity inside the tubular membrane ranged beyond the current study. Further experiments corroborated the colloidal nature of fouling as within the same gas velocity range, replacement of sludge supernatant with tap water resulted in a increase in critical flux from 10 to 20 LMH, while diluting sludge by 50 % yielded the same results as with the original sample. This argument is consistent with the fact that the highest fluxes were obtained in the AeMBR, with the lowest concentration of soluble and colloidal organics, while the anaerobic systems presenting the highest SMP concentration showed the lowest critical flux.

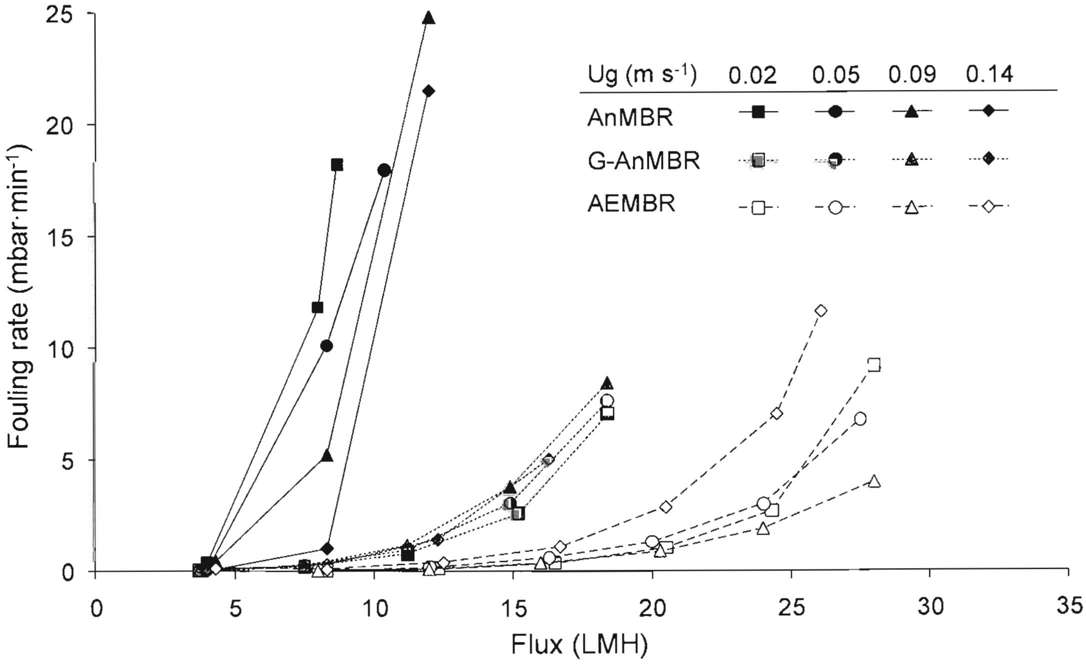
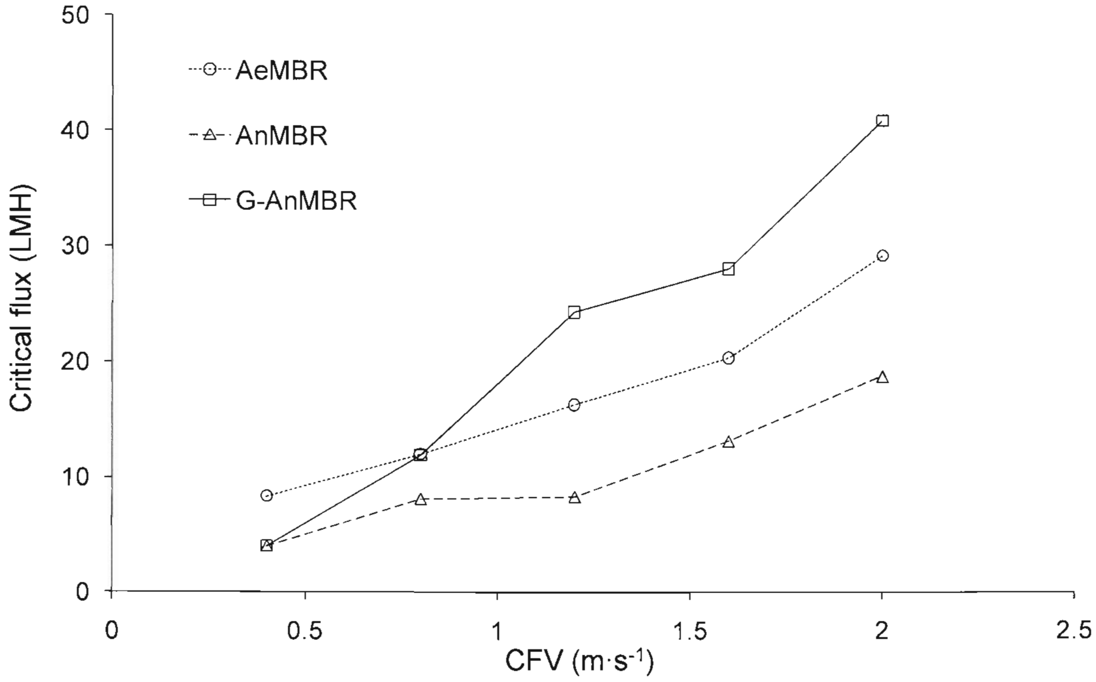


Figure 6.3 Effect of gas velocity on fouling rates in gas lift membrane configuration

However significant flux improvements with increasing gas sparging intensities with tubular membranes have also been reported in anaerobic MBRs. Early studies in which gas lift tubular membranes were coupled to anaerobic bioreactors have also shown a positive effect of gas velocity on membrane fouling (Imasaka *et al.*, 1989; Kayawake *et al.*, 1991). For instance Imasaka (1989), reported an increase in flux from 25 to 100 LMH with increased gas velocity from 0.2 m s^{-1} to 8.9 m s^{-1}, although at higher gas sparging intensities an appreciable decline in flux was observed, which was attributed to the absence of a protective cake layer, allowing small particles to deposit on the membrane surface (Lee *et al.*, 2001). In a later study long term membrane operation of methane fermentation broth was also demonstrated, as a stable flux of 25 LMH was maintained for 50 days by gas sparging inside 3.5 mm tubular membranes at a gas velocity of 1.4 m s^{-1} (Imasaka *et al.*, 1993). However these studies employed ceramic membranes, which are known to provide a higher resistance against organic fouling than the polymeric materials employed in the present study (Ghyoot & Verstraete, 1997; Kang *et al.*, 2002).

6.3.5 Critical flux in tubular pumped configuration

Critical flux in the sidestream pumped configuration increased from 4 to 41 LMH and from 4 to 19 LMH in the G-AnMBR and the suspended AnMBR respectively for cross flow velocities ranging from 0.4 m s^{-1} to 2 m s^{-1} (Figure 6.4 top). Flux enhancement in the AeMBR appeared to be between the ones obtained for the anaerobic systems as a maximum critical flux of 29 LMH was obtained at the highest crossflow velocity of 2 m s^{-1}. Comparison of these results with those previously described for the gas lift experiments indicate that increasing sludge cross flow velocity has a higher impact on attainable fluxes than gas sparging as pumping biomass across membrane surface provides better hydrodynamic conditions than gas sparging in immersed configuration due to complete control over liquid cross flow velocity. However, in the aerobic system the critical flux of 12 LMH obtained for a cross flow velocity of 0.8 m s^{-1} in pumped configuration was matched for the lowest gas velocity of 0.02 m s^{-1} in gas lift operation, suggesting that although limited, gas sparging had a positive effect in increasing critical flux in the AeMBR. Similar results were reported by LeClech (2005), although in contrast to the trend obtained in the present study increasing gas velocity was demonstrated to reduce membrane fouling as critical flux reached a maximum flux of 32 LMH for a gas velocity of 0.2 m s^{-1}. On the other hand both anaerobic sludges presented critical fluxes equivalent to those obtained in gas lift operation for the lowest cross flow velocity of 0.4 m s^{-1}.



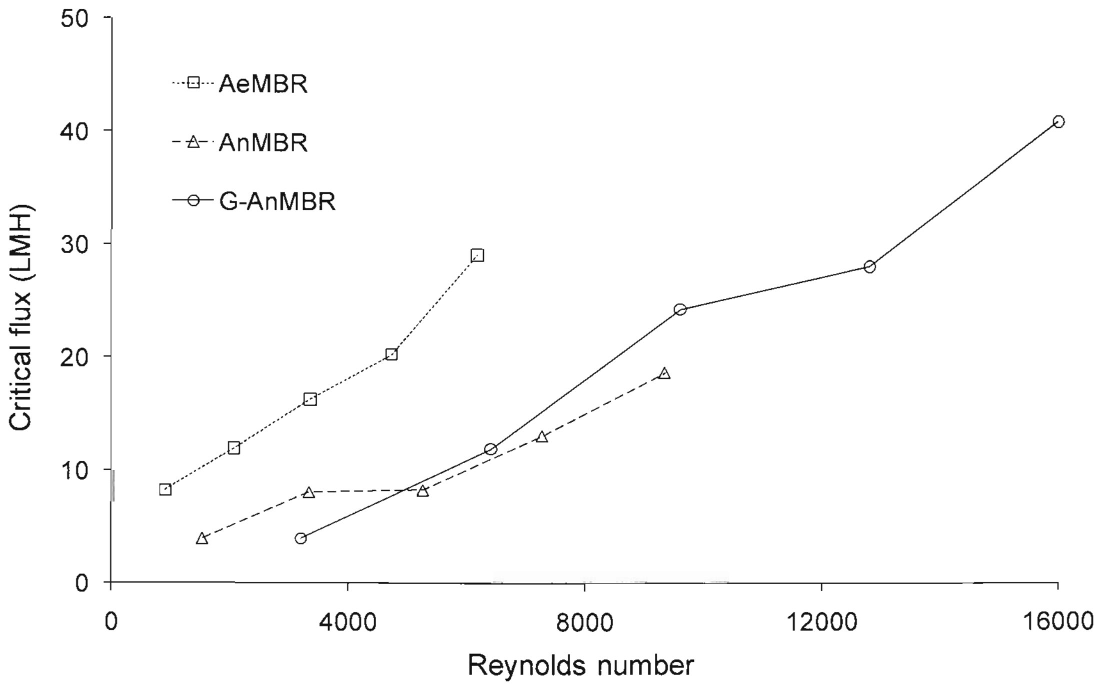


Figure 6.4 Influence of crossflow velocity (top) and Reynolds number (bottom) on critical flux

The more prominent increase in critical flux obtained in the G-AnMBR as compared to the suspended systems suggests that the MLSS concentration is main factor determining critical flux in pumped sidestream configuration. It is likely that the higher concentration of colloidal foulants in the G-AnMBR with respect to the AeMBR was compensated by the higher shear resulting from its lower viscosity. Comparison of filtration performance between a granular and suspended anaerobic bioreactor operated at low concentrations of MLSS reported by Elmaleh (1997) and Cho (2002) showed that for a crossflow velocity of 1 m s^{-1} similar fluxes of 50 LMH were obtained. Considering a viscosity of 1 mPa s^{-1} for the G-AnMBR supernatant, the variation in crossflow velocities from 0.4 to 2 m s^{-1} would result in Reynolds numbers ranging from 3200 to 16000. In the suspended systems corresponding Reynolds number would range between 900 and 9000, according to the rheological parameters given by Laera (2007) and Pevere (2007) for the average MLSS concentration in the suspended aerobic and anaerobic systems respectively. When compared against Reynolds number, higher critical fluxes are obtained in the AeMBR than the G-AnMBR (Figure 6.4 bottom). To illustrate increasing Re from 3200-3350 to 6170-6400 resulted in critical fluxes of 4 and 12 LMH in the G-AnMBR and 16 and 29 LMH in the AeMBR (Figure 6.4 bottom).

6.3.6 Critical flux in submerged membrane configuration

Flux enhancement in hollow fibre membranes proceeded differently in the AeMBR and G-AnMBR as compared to the suspended AnMBR. Increasing gas velocity from 0.007 to 0.027 m s^{-1} resulted in a similar improvement of critical flux from 3-4 LMH to approximately 14 LMH in the AeMBR and G-AnMBR. As reported by several other studies (Bouhabila *et al.,* 1998; Guglielmi *et al.,* 2007; Guglielmi *et al.,* 2008), a threshold gas sparging rate after which no further improvement in flux is obtained was observed at gas velocities of 0.027 and 0.041 m.s^{-1}, corresponding to fluxes of 14 LMH and 17 LMH in the aerobic and granular anaerobic MBRs respectively. On the other hand, in the suspended anaerobic MBR, an increase in gas velocity from 0.007 to 0.041 m s^{-1} produced a small increase in critical flux value from 2 LMH to 5 LMH, while further increase to 0.057 m.s^{-1} yielded a flux of 12 LMH close to the maximum obtained in the granular system. These results indicate that the granular system requires approximately 50 % lower gas demand than the suspended anaerobic MBR. Similar conclusions were reported by van Voorthuizen (2008) in a comparison between suspended and granular anaerobic MBR treating black water, based on the lower concentration colloidal matter present in the latter. However it is also possible, that similarly to the results shown in the tubular gas lift and pumped experiments the velocity of the sludge induced by gas sparging was higher for the less viscous G-AnMBR sludge than in the suspended aerobic and anaerobic MBRs with MLSS concentrations of 8.7 gMLSS L^{-1} and 7.7 gMLSS L^{-1} respectively. The velocity gradients associated to membrane scouring range from 75 to 200 s^{-1} and from 87 to 208 s^{-1} in the aerobic and anaerobic MBRs respectively, while for the same difference in gas sparging rate (0.007 $m.s^{-1}$ - 0.041 $m.s^{-1}$) shear stress varied between 230 s^{-1} and 670 s^{-1} in the granular system.

When normalised against the membrane area, the gas flows applied in the tubular and hollow fibre geometries yielded specific gas demands ranging from 0.2 to 1.6 $m^3\ m^{-2}\ h^{-1}$. Comparison of results obtained with these two membrane geometries show that for the lowest specific gas demands of 0.2 and 0.4 $m^3\ m^{-2}\ h^{-1}$ tubular geometries produced the higher fluxes of 4 and 8 LMH obtained in the suspended aerobic and anaerobic MBR respectively (Figure 6.5). However, above this threshold gas sparging rate the critical fluxes with the hollow fibre are higher. These results indicate that gas sparging in tubular membranes is more effective than in hollow fibre systems when low gas velocities are applied, since under these conditions fouling by sludge flocs prevails over soluble/colloidal fouling. In contrast, in the G-AnMBR with low suspended solids concentration, the hollow fibre membrane yielded higher critical fluxes over the entire range of gas velocities tested corroborating the previous argument. According to Cui (1997) the low effect of increasing gas sparging in tubular membranes can be due to increasing frequency of gas slugs which reduces the gap between them resulting in the suppression of the effect of primary wakes in the bubble tail reducing turbulence on the membrane surface. On the other hand the greater effect of gas sparging on critical flux obtained with hollow fibre membranes suggests that mechanisms such as fibre movement which physically removes mass transfer boundary layer and induces liquid flows transverse to the fibres (Cui *et al.*, 2003) may be more determinant in removing colloidal fouling than the shear induced in the liquid membrane interface by gas bubbling inside the tubular membrane.

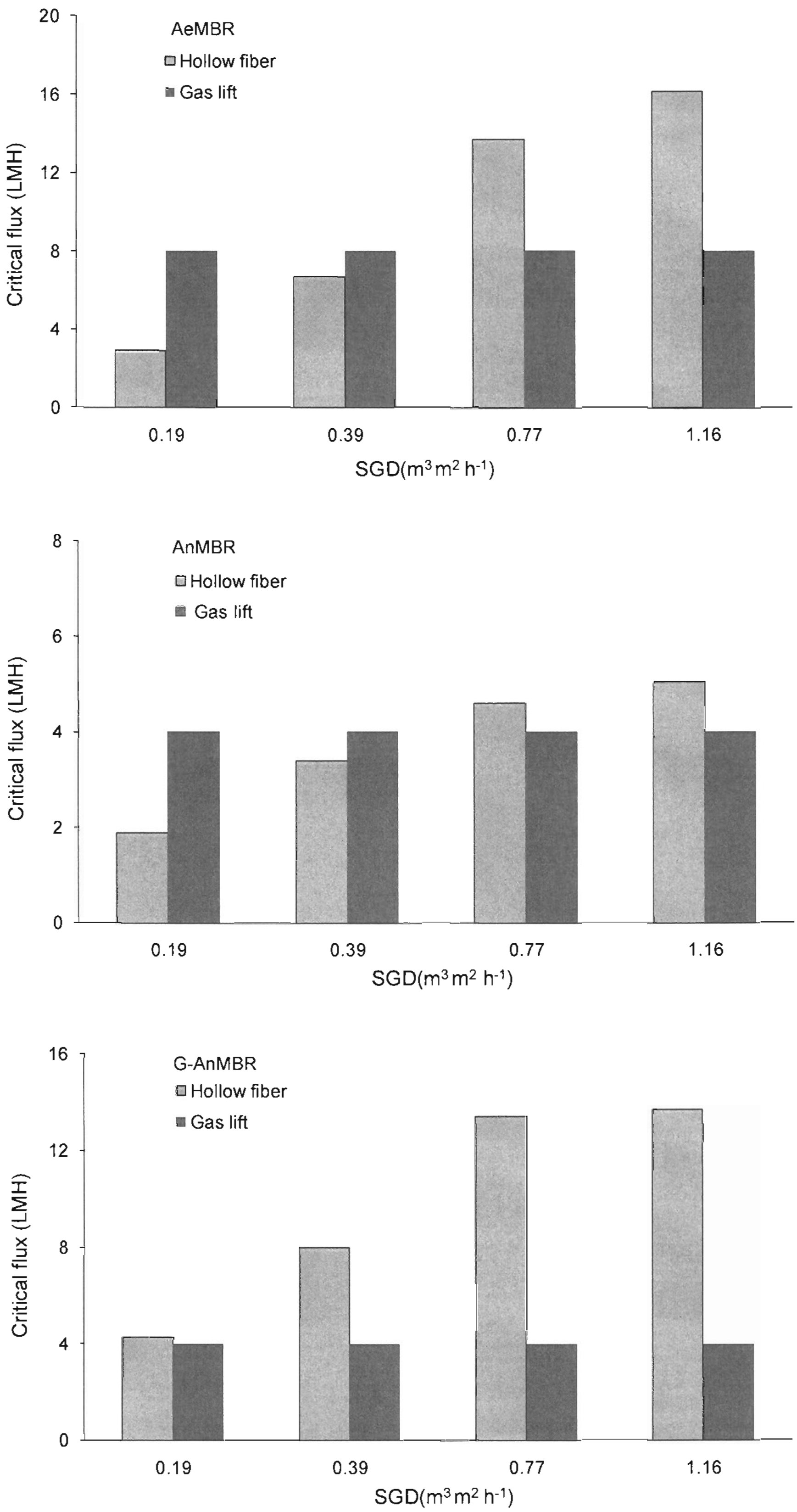


Figure 6.5 Influence of SGD on critical flux in hollow fibre and gas lift membrane configurations in the AeMBR (top), AnMBR (middle) and G-AnMBR (bottom)

6.4 CONCLUSIONS

The filtration performance of aerobic and anaerobic MBR sludges was evaluated using the flux step method with gas sparging in tubular and hollow fibre membranes as well as pumped cross flow operation. The suspended anaerobic MBR with similar MLSS concentration as the aerobic system but with a 5 fold higher concentration of colloidal organics presented the lowest critical fluxes for all membrane configurations and operational conditions. On the other hand the granular system, despite presenting double the concentration of SMP yielded similar fluxes compared to the aerobic MBR with hollow fibre membranes and a more pronounced increase in critical fluxes in pumped sidestream configuration. These results, indicate that the effect of applying turbulence by gas sparging or sludge pumping in immersed and pumped crossflow membrane configuration are most effective in the granular system probably due to the higher shear resulting from its lower viscosity.

6.5 ACKNOWLEDGEMENTS

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CHAPTER 7:

AMMONIA AND PHOSPHATE REMOVAL FROM GRANULAR ANMBR EFFLUENT USION ION EXCHANGE RESINS

CHAPTER 7. AMMONIA AND PHOSPHATE REMOVAL FROM GRANULAR AN MBR EFFLUENT USING ION EXCHANGE RESINS

Martin Garcia, N.[1], Sanavia, A.[2], Pidou, M.[1], Judd, S. J.[1] and Jefferson, B.[1]

[1] Centre for Water Science, Cranfield University, Cranfield, Beds, MK43 0AL, UK

[2] Facoltà di Ingegneria ,Università degli Studi di Padova, Padova, 35131, Italy

ABSTRACT

In the current study the potential for nutrient removal using ion exchange processes downstream of granular anaerobic MBR has been evaluated. Batch isotherm experiments with model solutions and real effluent as well as standard column trials have been conducted in order to evaluate capacity, impact of competing species and bed life. Results showed that both ammonia and phosphate equilibrium isotherms could be adequately represented by Langmuir model with maximum capacities of 71 gNH_4-N Kg_{media}^{-1} and 9 gP Kg_{media}^{-1}. The phosphate resin appeared to be more affected by competitive adsorption than the ammonia resin since reductions in capacity of 21 % and 42 % respectively were observed with respect to model solutions. These trends were confirmed by column trials which revealed capacities of 17.7 gNH_4-N Kg_{media}^{-1} and 5-6 gP Kg_{media}^{-1} at the effluent standards of 5 mg NH_4-N$\cdot L^{-1}$ and 1 mg $PO_4^{-3}\cdot L^{-1}$.

Keywords: Ammonia, phosphate, ion exchange, anaerobic, membrane bioreactor.

7.1 INTRODUCTION

Traditional wastewater treatment is designed to remove organics, nutrients and pathogens to render the effluent safe to discharge. Typical consents depend on the flow to be treated but range between 1-2 mg PO_4^{-3} L^{-1} phosphate and up to 7 mg NH_4-N L^{-1} ammonia. A more restrictive consents are typical at large works used whereby a total N consent is required which can be as low as 10 mgTN L^{-1}. Traditional approaches to this focus on the adoption of advanced configurations of the activated sludge process for total nitrogen removal coupled with either biological or chemical phosphorus removal depending on the organic strength available. A switch to anaerobic bioreactors such as the G-AnMBR alters this balance as the organics are reduced to below the compliance standard but the nutrients are unaffected and in the case of ammonia, increased slightly. The removal of organics does not inhibit the nitrification process but does limit biological denitrification and phosphorus removal and as such neither would be possible without considerable import of organic substrates or an alternative removal technology.

The argument can be taken further in that the adoption of anaerobic processes within wastewater flowsheets is discussed in terms of a need to reduce or neutralise the energy demand associated with wastewater treatment. Given that nitrification represents potentially as much as 30% of the total energy demand, the need for alternative non biological approaches appears critical. Since both phosphate and ammonia exists principally as ions in sewage an alternative route to removal and recovery is the use of ion exchange systems. Consideration in the past has concluded that such approaches are non viable due to concerns over limitations in capacities and issues around solids in the effluent clogging the beds. The use of AnMBRs removes the problems associated with solids and new media development has resulted in much higher capacities suggesting that it may be technically viable. In the case of phosphate, the development of hybrid ion exchanges (HAIX) has transformed the potential for P removal. HAIX media utilised embedded nanoparticles of transition metals to provide very selective uptake of phosphate due to a preferential free energy of adsorption (Martin *et al.,* 2009). The result is a highly selective resin that can be easily regenerated through simple pH adjustment. The current preferred embodiment of the concept is with hydrated ferric oxide nanoparticles (Phos-X- SolmeteX, USA) with competing systems manufactured in Australia and Japan. In the case of ammonia, a number of previous studies have investigated a range of natural zeolites with most focus on the mineral clinoptilolite (Lahav & Green, 1998; Šiljeg *et al.,* 2010). However, recent studies on sludge liquor treatment have shown that a manufactured material, MesoLite, has a much greater capacity (Thornton *et al.,* 2007b). MesoLite is produced by chemical modification of clay minerals and other aluminium-bearing materials to produce a predominately tetrahedrally coordinated Al^{3+} ion enhancing the number of exchange sites.

In the current study the use of both PhosX and Mesolite is considered to assess the potential to operate ion exchange processes downstream of the G-AnMBR for nutrient control. Experimental investigations into the efficacy of the contact resins were conducted through a combination of batch isotherm determinations and standard column trials to evaluate bed life and capacity.

7.2 MATERIALS AND METHODS

7.2.1 Batch equilibrium experiments

Adsorption isotherms were determined in a modified batch column experiment whereby a solution of the test liquid was recycled through a column containing 1 g of contact resin using a peristaltic pump. Halves a Litre of synthetic solutions (N and P) were used during isotherm equilibrium trials. The synthetic solutions contained either combinations of ammonium dihydrogen phosphate with diammonium hydrogen phosphate (for the ammonia solution) and potassium dihydrogen phosphate with dipotassium hydrogen phosphate (for the phosphorus solution) dissolved in DI water and buffered to neutral pH with NaOH or HCl as required. Model solutions concentrations ranged up to 1500 $mgNH_4\text{-}N\ L^{-1}$ in the case of the ammonia resin and up to 500 $mgPO^{3-}_4\ L^{-1}$ for the phosphate trials. These values were chosen to cover the range of expected concentrations after equilibration during fixed bed column trials with real effluent from the G-AnMBR and to compare at high concentrations to the maximum capacities reported in previous studies with different contact medias. In order to ascertain the impact of competing species on capacity, equivalent experiments to the ones conducted with model solutions were done with effluent sourced from a pilot granular anaerobic membrane bioreactor (G-AnMBR) for both ammonia and phosphate contact medias. Physical characteristics, operational conditions and performance of the G-AnMBR are explained in detail in Chapter 5. One day composite samples of the G-AnMBR effluent were collected and stored at 4°C until used. All experiments were conducted at 21 °C with run times of 18 hours for the P contact resin and 56 hours for the ammonia contact resin as previous work had shown these to be in excess of the time required to reach equilibrium. Following equilibration the concentration of ammonia and phosphate in the remaining solutions was determined and used to calculate the capacity of the corresponding ion exchange resins using the following mass balance (Equation 7.1):

$$Q_e = \frac{(C_0 - C_e)\,V}{M} \qquad \text{[Eq. 7.1]}$$

where V is the volume of the solution (0.5 L), M the mass of ion exchanger resin (1 g) and C_o the initial concentration of ammonia or phosphorous (mg L^{-1}). The equilibrium capacity Q_e (g $Kg^{-1}{}_{resin}$) represents the maximum amount of ammonia or phosphate adsorbed by the ion exchanger and C_e (mg L^{-1}) the corresponding equilibrium concentration in solution. Equilibrium data was then fitted to Langmuir (Equation 7.2) and Freundlich (Equation 7.3) isotherm models to derive standard adsorption parameters K ,b and n.

$$\text{Langmuir:} \qquad Q_e = \frac{K\,b\,C_e}{1 + K\,C_e} \qquad \text{[Eq 7.2]}$$

$$\text{Freundlich:} \qquad Q_e = K\,C_e^{\frac{1}{n}} \qquad \text{[Eq. 7.3]}$$

In the Langmuir model b (g $Kg^{-1}{}_{resin}$) represents the maximum exchange capacity of the media while K (m^3 g^{-1}) is the energy constant which indicates the degree of affinity between the adsorbate and the resin. Similarly in Freundlich model the parameter K is related to the capacity of the resin while the adsorption intensity 1/n reflects favourable adsorption when it adopts a value lower than unity.

7.2.2 Fixed bed column experiments

Fixed bed column experiments were conducted in a similar set up to the equilibrium tests with the exception that the flow was single pass. A controllable peristaltic pump (model 401U/D1 ,Watson Marlow , Falmouth, Cornwall) was employed to set the EBCT at 12 and 25 minutes in the case of the ammonia resin based on a previous study that showed that 42 minutes was the optimum for digested sludge liquors with ammonia concetrations above 500 $mgNH_4\text{-}N\ L^{-1}$ (Thornton *et al.,* 2007a). Similarly three different EBCT of 2, 4 and 8 minutes were chosen for the phosphate media in order to investigate the performance of the resin at contact times around the 4 minutes recommended by the manufacturer. Glass chromatographic columns of 25 mm internal diameter and 250 mm length (Omnifit, Kinesis, St Neots) containing 7.6 g of PhosX and 10 g of Mesolite (23 g for EBCT of 12 minutes) were fed with the effluent of the G-AnMBR from the top of the column. At regular intervals the effluent from the column was sampled for analysis of either ammonia or phosphate and the volume collected up to that instant measured in order to calculate media capacity. The operating removal capacity was determined by means of graphical integration of the area above the breakthrough curve fixed either to a pre set effluent limit of 1 mg $PO_4^{-3}\ L^{-1}$ and 5 mg $NH_4\text{-}N\ L^{-1}$ or until bed exhaustion was attained (Equation 7.4):

$$q_e = \frac{1}{M}\int_0^V (C_0 - C_{eff})\, dV \qquad \text{[Eq. 7.4]}$$

Where q_e is the mass adsorbed per unit mass of media (M) or capacity, C_0 is the influent ion concentration, C_{eff} the effluent concentration as the volume of flow pass V progresses.

7.2.3 Analytical methods

The concentration of COD, total phosphorous (TP), phosphates ($PO^{3-}{}_4$), total nitrogen (TN), ammonium ($mgNH_4\text{-}N$) and sulphates (SO_4^{-2}) was measured according to Standard Methods (APHA, 1998) with Merck vial test kits (VWR International, Lutterworth, Leicestershire). Inorganic carbon was analysed using a total organic carbon analyser (TOC-5000, Shimadzu). UV absorbance was measured against a blank at 254 nm using a Jenway Spectrophotometer (model 6505, Jenway). Analysis of pH and BOD_5 were also determined according to standard methods (APHA, 1998).

7.3 RESULTS AND DISCUSSION

7.3.1 G-AnMBR performance: Nutrient characterisation

The average concentrations of total phosphorous and nitrogen in the influent wastewater were 7.0 ± 1.7 mgTP L^{-1} and 54.9 ± 12.6 mgTN·L^{-1} with 67 % and 65 % of total present in soluble form respectively. The G-AnMBR pilot plant generated an effluent quality with average values of 14.3 ± 4.2 mgPO^{3-}_4 L^{-1} and 43.9 ± 9.3 mgNH_4-N L^{-1} showing that while phosphate levels remained unaffected ammonia increased by 8 mgNH_4-N L^{-1} with respect to the influent wastewater concentration (Figure 7.1). These results supports previous findings which have shown a high conservation of N and P due to the low nutrient requirements associated to biomass assimilation during anaerobic digestion, especially when treating low-medium strength wastewaters (Baek & Pagilla, 2006; van Voorthuizen *et al.*, 2008). However unlike nitrogen and phosphorous, COD removal ranged between 78 % and 94 % with an average concentration of 47 ± 24 mgCOD·L^{-1} during steady state operation. Considering only the ammonia concentration, these results correspond to a COD/N ratio of approximately 1 which is 25 % of the minimum required in order to achieve complete biological N removal (Henze, 1991; Bolzonella *et al.*, 2001). To illustrate batch denitrification trials conducted with effluent from a suspended growth anaerobic MBR operated in parallel to the G-AnMBR and with similar effluent characteristics showed a low denitrification potential of 2 mgN L^{-1} and a specific denitrification rate of 0.09 mg$NO_{x\cdot}$ gVSS^{-1}·h^{-1} (Eusebi et al, in preparation). These results indicate the limited applicability of the effluent of the G-AnMBR for biological nutrient removal due to the low biodegradable organic fraction.

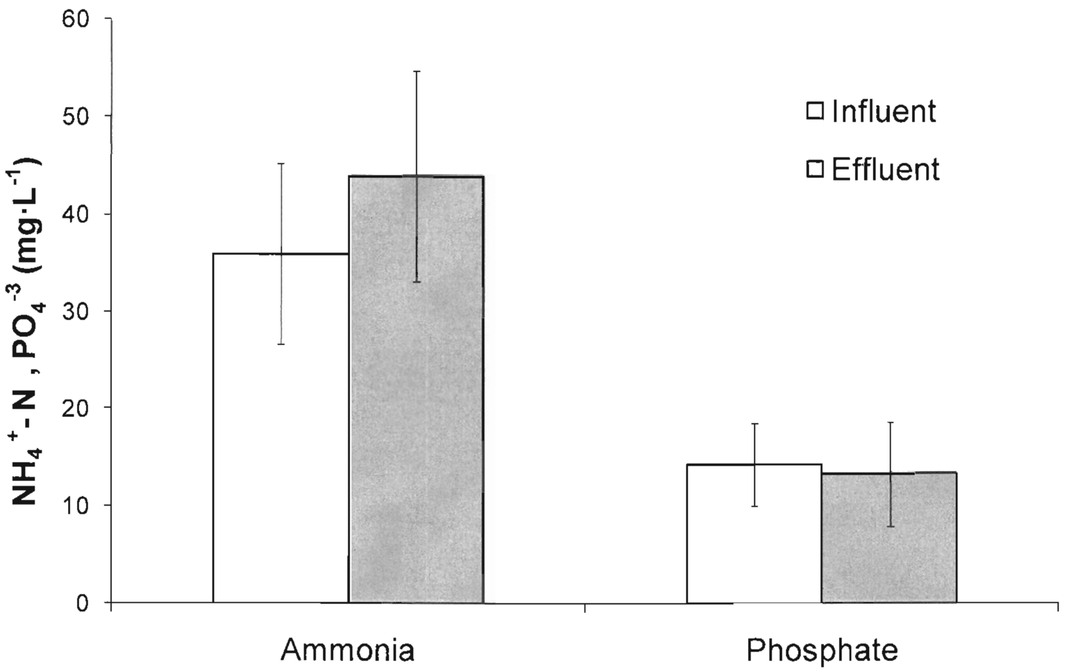

Figure 7.1 Ammonia and phosphate concentration in the influent and effluent of the G-AnMBR

7.3.2 Batch isotherm trials with model solutions

Media capacity increased with initial nutrient concentrations for both ammonia and phosphate during batch isotherm trials. In the case of Mesolite an increase in capacity from 2 gNH_4-N Kg_{media}^{-1} to 55.3 gNH_4-N Kg_{media}^{-1} was observed as ammonia concentration in equilibrium varied from 0.5 to 285 $mgNH_4$-N L^{-1} remaining constant at 65-68 gNH_4-N Kg_{media}^{-1} for concentrations above 700 $mgNH_4$-N L^{-1} (Figure 7.2 top). A similar trend was observed for the Phosphate media in which capacity increased from 11.1 to 22.8 gPO_4^{-3} Kg_{media}^{-1} as equilibrium concentration varied from 1.3 to 54.4 $mgPO_4^{-3}$ L^{-1} and reached a maximum capacity of 25-28 $mgPO_4^{-3}$ Kg_{media}^{-1} for phosphate concentrations exceeding 191 $mgPO_4^{-3}$ L^{-1} (Figure 7.2 bottom). Analysis of batch isotherm experiments conducted with Mesolite revealed that the Langmuir isotherm model provided a more consistent fit than the Freundlich isotherm since equilibrium capacities were predicted more closely at low ammonia concentrations and a limit in capacity was observed at higher levels. In contrast results obtained with the phosphate media indicated reasonable fits to both Langmuir and Freundlich isotherms although the former provided higher correlation coefficients of 0.9944 as compared to 0.9781 for the latter. Therefore since equilibrium capacity appeared to reach a plateau for both ammonia and phosphate contact resins the parameters obtained with Langmuir model were further examined and compared to those obtained with other medias reported in literature as a mean of comparison of adsorption characteristics.

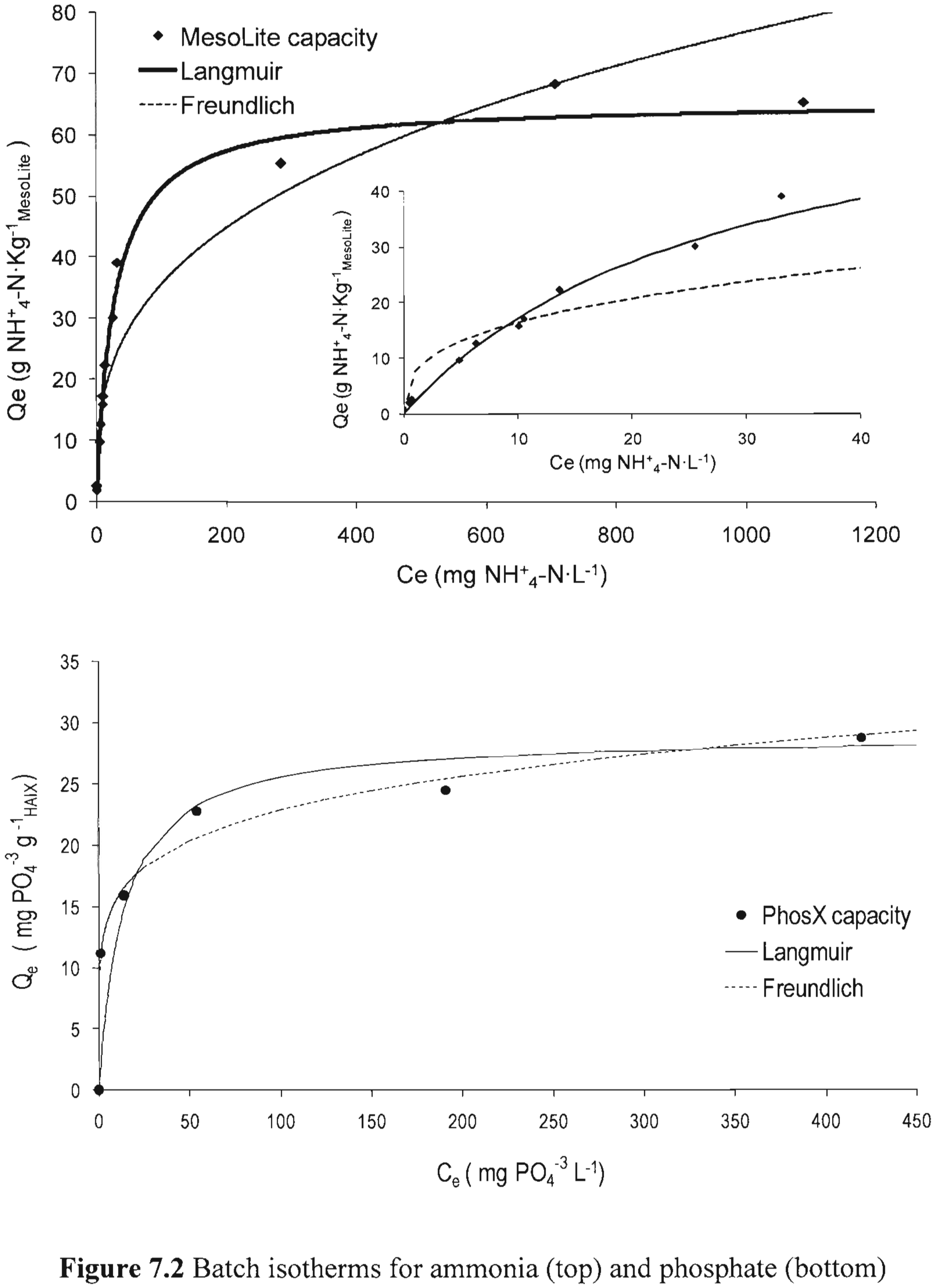

Figure 7.2 Batch isotherms for ammonia (top) and phosphate (bottom)

The Langmuir capacity constant b representing the saturation conditions was 71 gNH_4-N Kg_{media}^{-1}, which agrees with previous trials where values of 72 gNH_4-N Kg_{media}^{-1} (Thornton *et al.*, 2007b) and 69 gNH_4-N Kg_{media}^{-1} (Mackinnon *et al.*, 2003) were reported. Comparison to other trials and media reveals Mesolite to exhibit much higher capacities than other commercially available ion exchangers and natural zeolites. For instance, reported capacities for Dowex 50wx8 and Pirolite MN500 are 27 gNH_4-N Kg_{media}^{-1} and 19.5 gNH_4-N Kg_{media}^{-1} (Jorgensen & Weatherley, 2003) while for the widely studied clinoptinolite maximum values of 21.5 gNH_4-N Kg_{media}^{-1} have been observed (Sprynskyy *et al.*, 2005). On the other hand a value of 0.036 $m^3 \cdot g^{-1}$ was obtained for the Langmuir energy constant which is in the low range of 0.027 $m^3 \cdot g^{-1}$ to 1.69 $m^3 \cdot g^{-1}$ reported for Turkish clinoptinolite (Karadag *et al.*, 2006) and chabazite (Lahav & Green, 1998) respectively, indicating that despite the higher capacity of Mesolite with respect to other media, the affinity of its exchange sites towards ammonium is less favourable (Table 7.1). High values of K correspond to high affinities and result in an equilibrium curve in which the maximum capacity is attained at lower adsorbent concentrations. Employing the same resin and mass loading as in the present study but with ground media instead of resin beads, Thornton (2007b) reported a more favourable energy constant of 0.049 m^3 g^{-1}, indicating that the lower affinity can be partially attributed to the external mass transfer limitations or the establishment of a concentration gradient across the resin beads as a result of intraparticle diffusion which would result in higher equilibrium concentrations with respect to grounded media. To illustrate the maximum capacity was attained at an equilibrium concentration of approximately 700 $mgNH_4$-N L^1 for the resin beads, while for powdered Mesolite 400 $mgNH_4$-N L^{-1} was required.

Equivalent experiments with the phosphate adsorbing media indicate a Langmuir capacity of 28.98 gPO_4 Kg_{media}^{-1} which equates to 8.6 gP Kg_{media}^{-1} (Figure 7.2 bottom). Column trials with model phosphate solutions with the same media and a concentration of 500 $mgPO_4^{-3}$ L^{-1} resulted in column exchange capacities of 23.6 gPO_4 Kg_{media}^{-1} (Martin *et al.*, 2009) . However, capacities reached up to 35-50 gPO_4 Kg_{media}^{-1} (11-16 gP Kg_{media}^{-1}) after several regeneration cycles indicating that resin conditioning is required for optimum results. According to a classification provided by Cucarella (2009) in a recent review of phosphorous filter media, the capacity shown by HAIX would fall in the group of very high P sorption, together with other commercial products such as Filtra P and Polonite which have shown capacities of 19.4 and 7.4 gP Kg_{media}^{-1} respectively (Gustafsson *et al.*, 2008) or natural materials such as dolomite with 20 gP Kg_{media}^{-1} (Roques *et al.*, 1991) or ochre presenting 30 gP Kg_{media}^{-1} (Heal *et al.*, 2005). Unlike the Mesolite the Langmuir affinity constant value of 0.075 m^3 g^{-1} (0.236 as P) measured for the PhosX reflects a favourable adsorption (Table 7.1). However higher affinity towards phosphate have been reported for other medias based on ferric oxides and hydroxides (Table 7.1). To illustrate Zeng (2004) reported a Langmuir affinity constant of 0.44 m^3 g^{-1} for iron oxide tailings (30 % iron oxide content by mass) while for granular ferric hydroxide (GFH) value of 2.81 m^3 g^{-1} was shown (Genz *et al.*, 2004).

Equivalent batch experiments with effluent from the G-AnMBR revealed that reduction in capacity with the phosphate resin was twice as much as with the ammonia media. To illustrate at ammonia and phosphate equilibrium concentrations of 24.3 mg NH_4-N L^{-1} and 11 mgPO_4^{-3} L^{-1} corresponding capacities were 23.9 gNH_4-N Kg_{media}^{-1} and 7.6 gPO_4 Kg_{media}^{-1} while according to the experimentally determined Langmuir model they should have reached 30.5 gNH_4-N Kg_{media}^{-1} and 13.1 gPO_4 Kg_{media}^{-1} respectively. A similar reduction in capacity from 72 gNH_4-N Kg_{media}^{-1} to 49 gNH_4-N Kg_{media}^{-1} obtained both with model solutions but prepared with DI and tap water respectively have been previously reported (Thornton *et al.*, 2007b), indicating that competitive adsorption can be attributed to cations present in the influent water rather than released during the anaerobic degradation in the G-AnMBR. In the case of the PhosX the reduction in capacity cannot be solely attributed to the presence of competing ions since previous work has demonstrated that in model solution both the iron nanoparticles and the ion exchange resin uptake phosphate whereas in real systems the ion exchange component is retarded through competitive adsorption (Martin *et al.*, 2009).

Table 7.1 Langmuir and Freundlich model parameters for adsorption of model solutions of NH_4^+ and PO_4^{-3}

Resin/Parameter	**Langmuir**		**Freundlich**		**Reference**
	b ($g \cdot Kg^{-1}$)	**K ($m^3 g^{-1}$)**	**K**	**n**	
NH_4^+ contact medias[1,2]					
Clinoptinolite	6.59	1.493	2.27	2.97	Weatherley (2004)
Mordeite	12.88	0.139	1.26	1.85	Weatherley (2004)
Sulfonated polyester silica gel	8.82	0.213	2.79	3.85	Helminen (2006)
Clinoptinolite	2.47	0.167	0.43	2.02	Wang (2006)
Mesolite (grounded)	57.80	0.048	6.0	2.55	Thornton (2007b)
MesoLite (beads)	65.40	0.036	4.8	2.23	this study
PO_4^{-3} contact medias[3]					
Blast furnace slag	18.94	4.328	10.90	6.30	Kostura (2005)
Iron oxide tailings	8.21	0.444	3.59	5.30	Zeng (2004)
Dolomite	-	-	1.19	2.80	Roques (1991)
Akaganeite (β-FeOOH)	16.9	2.810	-	-	Genz (2004)
HAIX	9.27	0.236	4.205	6.02	this study

[1] Adapted from (Thornton *et al.*, 2007b).[2] b in g $NH_4\text{-}N \cdot Kg^{-1}_{resin}$. [3] b in g $P \cdot Kg^{-1}_{resin}$

7.3.3 Ammonia Kinetic experiments

Kinetic experiments on G-AnMBR effluent run under a feed concentration of 46.1 $mgNH_4$-N L^{-1} and EBCT of 12 minutes showed that ammonia breakthrough at 5 $mgNH_4$-N L^{-1} occurred at approximately 100 BV for both Clinoptinolite and Mesolite, with exhaustion occurring at 200 and 600 bed volumes respectively. Integration of breakthrough curves revealed the column exchange capacity of MesoLite to be 18.5 gNH_4-N Kg_{media}^{-1} which compared favourably to 2.2 gNH_4-N.Kg_{media}^{-1} for Clinoptilolite (Figure 7.3). However, from the faster mass transfer zone established in the Clinoptinolite it can be inferred the despite its lower capacity it presents a higher affinity towards ammonium supporting the low affinity constant observed in the batch trials with Mesolite. Comparison with equilibrium experiments also revealed a reduction in ammonium exchange capacity of approximately 25 % with respect to saturation conditions corroborating the results obtained during batch trials with effluent from the G-AnMBR. These results also support previous findings which showed that in the absence of competing ions a maximum capacity 72 gNH_4-N Kg_{media}^{-1} (Thornton *et al.,* 2007b) was attained while with anaerobic digestion liquors ammonium capacity decreased to 47-51 gNH_4-N Kg_{media}^{-1} (Thornton *et al.,* 2007a) due to the presence of cations in solution such as Ca, Mg, Na or K.

The shape of the breakthrough curve, in which ammonia leakage occurred from the beginning of the trial (Figure 7.3) suggested that ion exchange could be limited either by external mass transfer mechanisms (Ghorai & Pant, 2004) or by free ammonia present at the pH of the G-AnMBR effluent (Thornton *et al.,* 2007b). Subsequently two additional experimental runs in which EBCT was increased to 25 minutes and pH was maintained at the normal pH of the G-AnMBR effluent and decreased to a value of 7 were conducted (Figure 7.4). Thornton (2007b) reported that optimum Mesolite maximum capacity of 49 gNH_4-N Kg_{media}^{-1} was attained at a pH between 6-7, while it readily decreased to 37 and 29 gNH_4-N Kg_{media}^{-1} for pH values of 8 and 10 respectively. It was argued that reduced capacity obtained at higher pH values was due to the displacement of ammonia equilibrium from positively charged ammonium towards uncharged ammonia thus reducing the Coulombic interaction with ion exchange sites.

However alteration of the pH of the G-AnMBR effluent from 8 to 7 had no discernable impact on either capacity or bed life suggesting that free ammonia leakage was not a major factor (Figure 7.4). In fact, despite being fed with different influent pH, the effluent from the columns presented the same evolution in pH which decreased from 10.3-10.5 and stabilized at 7.8-8.1 after 1000 bed volumes indicating that in continuous operation the pH of the resin is determined by ion exchange process rather than by influent characteristics in these dilute conditions.

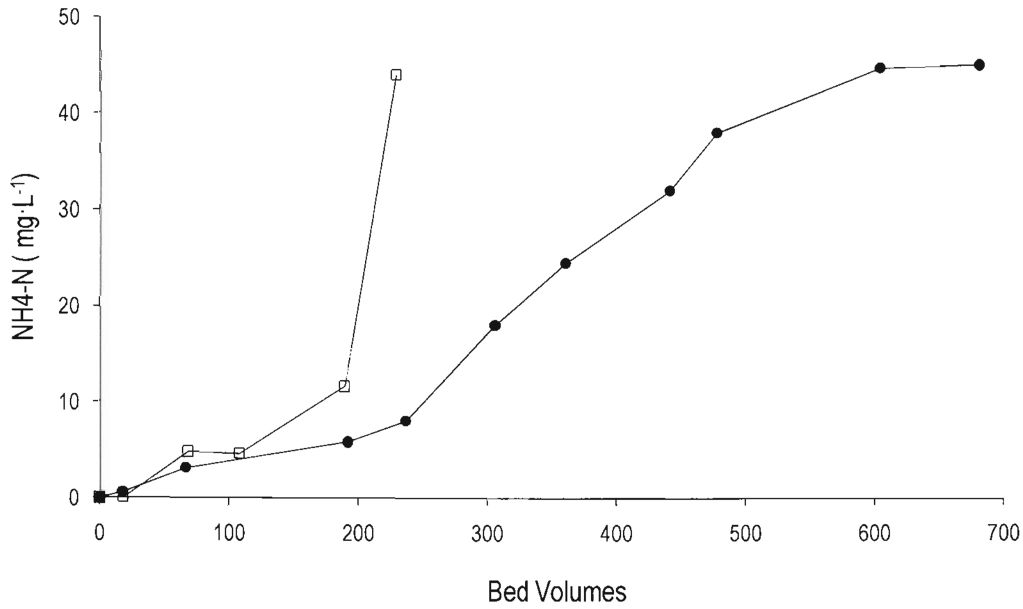

Figure 7.3 Comparison of Clinoptinolite and Mesolite for ammonia removal

In contrast, an increase in hydraulic retention time from 12 minutes to 25 minutes EBCT significantly improved the performance of the contact resin increasing capacity at 5 mgNH_4-N L^{-1} effluent standard from 9.5 gNH_4-N Kg_{media}^{-1} to 17.7 gNH_4-N Kg_{media}^{-1} and bed life from 100 BV to 350 BV (Figure 7.4). Batch kinetic trials showed that for an initial concentration of approximately 50 mgNH_4-N L^{-1}, 70 % removal was attained within 10-15 minutes when powdered Mesolite was employed (Thornton *et al.,* 2007b). Therefore an EBCT of 12 minutes was probably a too short contact time in order to allow for enough ammonia to diffuse into the resin beads. Although breakthrough profiles at the EBCT of 25 minutes suggest that ion exchange was still mass transfer controlled, media capacity increased to 32 gNH_4-N Kg_{media}^{-1}, representing a 78 %

increase with respect to the trial at 12 minutes EBCT. Such levels are also 20 % below those obtained during equilibrium experiments with model solutions at the correspondent influent concentration of 40 mgNH4-N L^{-1} and further corroborate the results obtained during batch trials with effluent from the G-AnMBR.

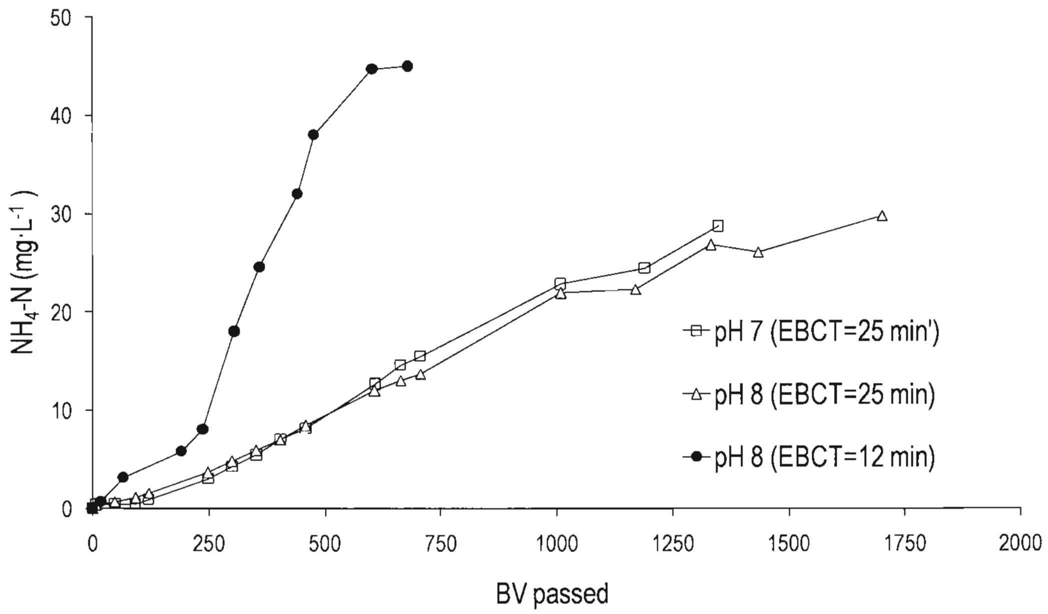

Figure 7.4 Influence of pH and contact time on breakthrough curves with MesoLite

7.3.4 Phosphate kinetic experiments

In the case of PhosX, kinetic experiments focused on the influence of both EBCT and influent phosphorus concentration on bed life and capacity. Results showed that there was no clear relationship between EBCT and media performance since increasing EBCT from 2 to 4 minutes decreased the bed life from 760 BV to 674 BV, whilst the highest capacity of 809 BV was attained at 8 minutes EBCT (Figure 7.5). A different order was observed for breakthrough at 1 $mgPO_4^{-3}$ L^{-1} effluent standard which was exceeded at 350, 250 and 300 bed volumes for EBCT of 2, 4 and 8 minutes respectively. Although these results indicate similar bed life to the ammonia contact resin, a distinct mass transfer zone was established such that low effluent quality could be maintained for extended periods, suggesting that mass transfer may not be the limiting factor of phosphate uptake under present experimental conditions. On the other hand comparison of breakthrough curves at an EBCT of 8 minutes and influent phosphate concentration of 4.6 $mgPO_4^{-3}\ L^{-1}$ and 15 $mgPO_4^{-3}\ L^{-1}$ revealed that with the lower concentration, the breakthrough at 1 $mgPO_4^{-3}\ L^{-1}$ effluent standard was extended from 300 to 1012 bed volumes whereas the capacity decreased from 7.2 $gPO_4^{-3}\ Kg_{media}^{-1}$ to 5.1 $gPO_4^{-3}\ Kg_{media}^{-1}$ (Figure 7.5). Previous findings also showed that decreasing phosphate concentrations increased bed life at the expense of decreasing capacity due to the establishment of a faster mass transfer zone (Martin *et al.*, 2009) suggesting that a higher rate of adsorption occurs when the media is exposed to higher phosphate concentrations.

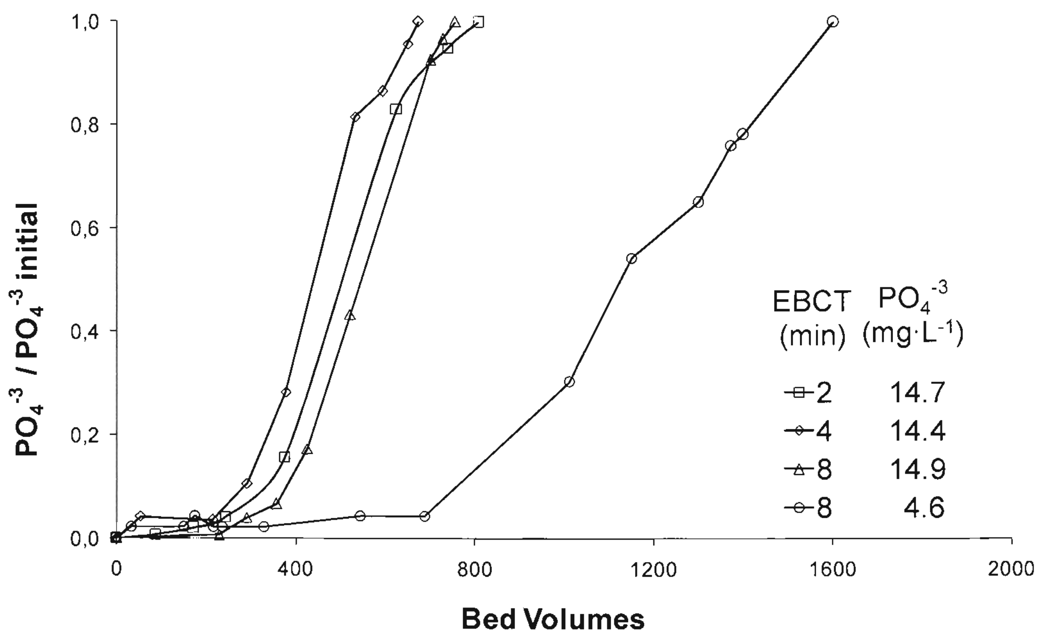

Figure 7.5 Influence of EBCT and influent concentration on phosphate removal

Overall capacities at saturation for the different EBCT ranged between 8.3-10.8 $gPO_4^{-3}\ Kg_{media}^{-1}$ which represents a reduction of 36-45 % compared to the equilibrium capacity supporting the results obtained during batch equilibrium trials with real effluent. At the lower influent phosphate concentration of 4.6 $mgPO_4^{-3}\cdot L^{-1}$, the capacity was reduced to 6.8 $gPO_4^{-3}\ Kg_{media}^{-1}$ but a similar decrease of 51 % with respect to its corresponding equilibrium capacity was observed. Comparative results reported for trickling filter effluent under similar influent phosphate concentration of 15.2 $mgPO_4^{-3}\cdot L^{-1}$ and EBCT of 4 minutes (Martin *et al.*, 2009), showed a saturation capacity of 23.6 $gPO_4^{-3}\ Kg_{media}^{-1}$, representing a 2.1-2.8 fold increase with respect to the effluent of the G-AnMBR. At the levels of competing ions such as sulphate, chloride, nitrate and carbonate/bicarbonate reported in the trickling filter and G-AnMBR effluents only the former has shown to exert a significant impact on the performance ferric oxide hybrid ion exchange resin, reducing capacity by 55 % at a concentration of 50 mgSO_4^{-2} L^{-1} (Pan *et al.,* 2009). However sulphate concentrations in the aerobic and anaerobic effluents were actually in the same range and therefore cannot justify the lower capacity shown with the G-AnMBR effluent. A higher degree of competitive adsorption of organic matter with the anaerobic effluent could provide a possible explanation for the difference in capacity since it was observed that Phos-X improved the effluent quality of the G-AnMBR removing colour and 40 % of UV_{254} from the beginning of the loading up to the end of the trial even after phosphate saturation had been attained (Figure 7.6). These results support a number studies that have shown that iron oxides and hydroxides present a high affinity towards organic matter as these can also form ligand exchange complexes with ferric oxide nanoparticles reducing phosphate capacity through competitive adsorption (Gu *et al.,* 1995; Korshin *et al.,* 1997; Teermann & Jekel, 1999; Genz *et al.,* 2008).

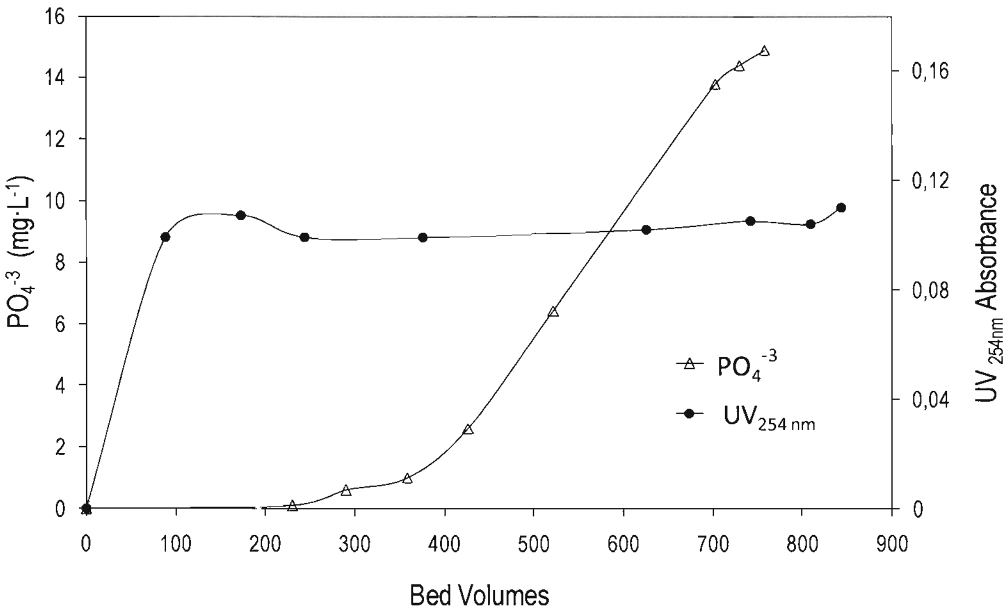

Figure 7.6 Breakthrough curves for PO_4^{-3} and UV_{254} ($PO_4^{-3}$$_{influent}$ = 14.9 mg NH4-N·L^{-1}, UV_{254} Absorbance $_{influent}$ = 0.175, EBCT = 8 min)

7.3.5 Implication for anaerobic based flowsheets

Results from previous investigations have shown that the combination of a high rate anaerobic reactor and membrane filtration operated on low-medium strength domestic sewage and at temperatures between 8 °C and 22 °C can be effectively employed for COD and BOD removal (Chapter 5). However as phosphate and ammonia effluent levels remained unchanged with respect to influent wastewater, biodegradable organic concentrations were sufficiently low that downstream biological nutrient control becomes a less practical option. Results from the present study have shown that consideration of an ion exchange processes as an alternative, enables efficient nutrient removal to be achieved without significantly increasing energy requirements of the overall process, as it can be assumed that energy expenditures would be similar to those of depth filters. For instance considering a headloss of 0.5 barg an energy demand of 0.037 kWh m^{-3} would be required for pumping the G-AnMBR effluent through the ion exchange columns. Additionally adoption of ion exchange processes, results in the separation and concentration of both ammonia and phsophate which makes their removal or recovery more economically viable as compared to the original diluted anaerobic effluent. However the economic feasibility related to the ion exchange process would be difficult to assess since it depends on the capacity and nutrient recovery yields as much as on the efficiency of the process employed for recovery and reuse of concentrated solutions.

For instance, a recent study investigating ammonia removal with clinoptinolte has demonstrated recovery efficiencies from regeneration brines of over 90 % by employing air stripping at pH = 11 when ammonia concentrations exceeded 300 mgNH_4-N L^{-1} (Rahmani *et al.,* 2009). The main parameter affecting the economic balance of ammonia air stripping are influent pH and temperature as well as influent and effluent ammonia concentration (Saracco & Genon, 1994; Zeng *et al.,* 2006). Alkaline conditions favours desorption up to a pH of 11 at which all ammonia is assumed to be as dissolved gas. The air requirements for ammonia stripping which determines the energy demands associated to gas blowers are inversely related to temperature according to Henry's solubility constant which increases from 1 atm at 30 °C to 9 atm at 90 °C (Saracco & Genon, 1994). Another option is steam stripping which is usually considered for high ammonia concentrations resulting in an enhanced recovery rate as well as the production of a highly concentrated ammonia solution which enables ammonia recovery as ammonium sulphate by addition of sulphuric acid after vapour condensation. An economic balance between air and steam stripping processes based on results obtained from pilot plant experiments showed that for an ammonia concentration around 500 $mgNH_4\text{-}N{\cdot}L^{-1}$ steam stripping presented a higher unitary cost of 9 $\${\cdot}Kg^{-1}$ NH_4-N removed compared to 7 $\${\cdot}Kg^{-1}$ NH_4-N removed of air stripping while for concentrations exceeding 1800 $mgNH_4\text{-}N{\cdot}L^{-1}$ both presented a similar cost of 5 $\${\cdot}kg^{-1}$ NH_4-N-removed (Janus & Van Der Roest, 1997). At the expected concentration of 600 $mgNH_4\text{-}N{\cdot}L^{-1}$ obtained with the 20 BV required for Mesolite regeneration (Thornton *et al.,* 2007a) normalised cost against treated water for steam and air stripping would be 0.26 $\${\cdot}m^3_{flow}$ and 0.19 $\${\cdot}m^3_{flow}$ respectively although further reductions could be expected in both cases if a heat source is available.

In the case of phosphate, recovery of the concentrated brines would involve chemical precipitation of concentrated solution. To illustrate, Liberti (2001) proposed a flowsheet for downstream nutrient removal from sewage based on combinations of anionic and cationic ion exchange resins in which all the phosphate was removed and an equimolar amount of ammonium adsorbed by processing only the correspondent fraction of the flow in the cationic media. The assessment of the feasibility of the process revealed total operational and capital costs of 0.048 $euros{\cdot}m^3_{flow}$ when regeneration brines were reused by removing phosphate by precipitation in the form struvite which revenue accounted for 40 % of running costs. However in contrast to ammonia in which preconcentration from wastewater is required before removal or recovery is practiced, phosphate control in a low investment flowsheet based on a UASB reactor has been previously demonstrated with similar costs of 0.05 $euros{\cdot}m^3_{flow}$ (Aiyuk *et al.,* 2004). Effluent phosphate concentrations of 0.8 $mgTP{\cdot}L^{-1}$ were achieved during long term operation by applying coagulation-flocculation with 50 $mg{\cdot}L^{-1}$ ferric chloride and 10 $mg{\cdot}L^{-1}$ polymeric flocculant upstream of a UASB which effluent then percolated through ammonia adsorbing zeolite that was then biologically regenerated to produce nitrate rich stream. Addition of the polymeric flocculant which accounted for 80 % of the chemical expenditure was justified by the production of a highly concentrated primary sludge stream and a lower solid load into the UASB system which allowed the application of an HRT of 5-6 hours whilst preventing accumulation of inactive solids in the reactor.

7.4 CONCLUSIONS

The results from the downstream nutrient trials have demonstrated that non biological processes are applicable to the treatment of ammonia and phosphate from the effluent of an anaerobic reactor. Trials with model solutions revealed that both ammonia and phosphate equilibrium can be adequately represented by Langmuir isotherm model with maximum capacities of 71 gNH_4-N Kg_{media}^{-1} and 9 gTP Kg_{media}^{-1}. Comparison to other medias revealed that despite the higher capacities both ammonia and phosphate resins presented low affinity constants. In the case of Mesolite this was partially attributed to mass transfer limitations, as corroborated by the breakthrough curve profiles obtained during fixed bed column experiments with effluent from the G-AnMBR. Unlike with the ammonia resin, the uptake of phosphate was not mass transfer limited as varying EBCT between 2 and 8 minutes had no effect on capacity and extended periods of low effluent quality were observed. However capacity was reduced 33-51 % with respect to equilibrium experiments with model solutions. Overall comparisons between the media reveals that the capacity of the phosphate resin is approximately double that of the ammonia media. To achieve effluent ammonia of 5 $mgNH_4$-N L^{-1} the bed life was 350 BV with a corresponding capacity of 17.7 gNH_4-N Kg_{media}^{-1}. At the reported ammonia concentration in the effluent of the G-AnMBR this equates to 0.54 m^3_{flow} Kg_{media}^{-1} before regeneration would be required. On the other hand averaged across the experimental conditions the operational capacity of PhosX is between 5-6 gP Kg_{media}^{-1} which equates to approximately 1 m^3_{flow} Kg_{media}^{-1}.

7.5 ACKNOWLEDGEMENTS

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CHAPTER 8:

GENERAL DISCUSSION: IMPLICATIONS FOR NEW FLOWSHEETS

CHAPTER 8. GENERAL DISCUSSION: IMPLICATIONS FOR NEW FLOWSHEETS

The work outlined in this report has identified a number of key features that can be combined to consider potential flowsheets to meet the overall aim of the project:

1. AN-MBRs (either configuration) were able to produce an effluent BOD and COD concentration that was compliant at below a COD of 125 $mg.L^{-1}$ and a BOD_5 of 20 $mg.L^{-1}$. Prolonged start up periods were observed compared to traditional aerobic systems but with seeded anaerobic reactors stabilised compliant effluent qualities were obtainable within days of operation.

2. Effluent organic concentrations were sufficiently low that downstream biological nutrient control becomes a less practical option and as such alternative methods were considered. Downstream processing of ammonia and phosphate is possible with modern adsorption resins enabling both very low effluent concentrations and recovery to be accomplished. Analysis of the initial trials suggests that resin capacity to meet 5 $mg.L^{-1}$ ammonia and a 1 $mg.L^{-1}$ phosphate standard are 0.5 $m^3\ Kg_{resin}^{-1}$ and 1 $m^3.Kg_{resin}^{-1}$ respectively.

3. Although both granular and suspended systems were configured in two chambers in order to avoid dilution of biogas with nitrogen supplied for fouling control, no biogas production was observed during the experimental period. An EGSB reactor operated in parallel to the granular and suspended anaerobic MBRs presented a methane yield of 0.13 L CH_4 g COD^{-1} which represented 51 % of the overall production based on methane solubility at the operating temperature of 15 C (McAdam et al, in preparation). However in the present study it is likely saturation of the liquid with biogas was never attained and that the dissolved methane was stripped out of the liquid phase with the nitrogen employed for membrane gas sparging. In practice gas recycling would be used as a source of sparge gas leading to a saturated liquid effluent.

4. Sludge production in the suspended growth anaerobic MBR system was equivalent to aerobic MBR run under 100 days sludge retention time with observed yields of 0.14 g VSS g COD^{-1}. Operation of the G-AnMBR resulted in a further reduction in yield to 0.02 g VSS g COD^{-1} although this only accounts for the gradual build up of solids in the membrane tank. In the future, bed operation needs to be tailored in order to ascertain the wasting and backwash requirements of the granular bed.

5. Based on the specific gas demand of 0.38 m^3 h^{-1} m^{-2} and the flux of 6 LMH, an energy demand of 0.58 kWh m^{-3} for fouling control would be required in the suspended growth anaerobic MBR. Lower energy operation of the membrane is possible in the granular anaerobic MBR configuration which can operate with intermittent gas sparging to maintain sustainable fluxes due to a reduced colloid load on the membrane. Stable performance was observed with 1 minute on, 10 minute off gas sparging sequence at gas crossflow velocity of 0.041 m s^{-1} equating to an energy demand of 0.18 kWh.m^{-3}. Consideration of the current study indicates that translation to a crossflow velocity of 0.027 m s^{-1} is appropriate since it resulted in low fouling rate during continuous gas sparging experiements. At this level the energy demand for fouling control with 10 % intermittent gas sparging would be 0.12 kWh.m^{-3}. Although further validation is required to optimize the gas velocity required to assist backwashing at reducing fouling, it is likely that low energy operation will only be possible through adoption of G-AnMBRs.

To illustrate the potential of using G-AnMBR in comparison to conventional ASP, a mass and energy balance of the two flowsheets is provided based around a previous Cranfield University study looking into a range of possible new flow sheets for low energy operation (MacAdam *et al.,* 2010) but incorporating the performance and operating data reported from this project. The case study is based around a 10000 m^3.d^{-1} plant meeting a TN and TP consents of 10 mg TN L^{-1} and 1 mg.TP L^{-1} respectively (Table 8.1) and will compare treatment costs related to energy requirements, sludge treatment and disposal and use of chemicals for nutrient removal between a G-AnMBR and a predenitrification ASP plant (Figure 8.1). In the ASP the addition of ferric chloride at a ratio of 2:1 with respect to influent phosphate concentration before primary sedimentation was assumed to produce compliant TP effluent concentrations.

In the ASP a sludge retention time of 15 days was set so as to ensure enough nitrification at the wastewater temperature of 15 °C, while hydraulic retention time and biological tank volumes were calculated from the MLSS which was set at 3 g MLSS L^{-1}. A standard model for aeration demand was utilised based on a SOTE of 4.5 % and a water depth of 4.5 m. Parasitic energy demands related to mechanical pre treatment, pumping of return activated sludge and internal recycling were obtained from Nowak (2003) while energy for mixing in the anoxic tank and anaerobic digesters was considered to be 10 W m^{-3} (Metcalf & Eddy, 2003): Energy demand for the contact resin plant is based on the parasitic energy demand for pumping assuming a headloss of 0.5 barg.

Table 8.1 Wastewater and flow characterisation

Parameter	Units	Value
Influent flow	Q, m^3 d^{-1}	10000
Chemical oxygen demand (COD)	mg L^{-1}	420
Biochemical oxygen demand (BOD)	mg L^{-1}	190
Fraction of unbiodegradable particulate COD	gCOD gCOD $total^{-1}$	0.2
Total suspended solids (TSS)	mg L^{-1}	290
NH_4^+-N	mg L^{-1}	35
Fraction of soluble unbiodegradable total Kjeldahl nitrogen (TKN)	gN $gTKN^{-1}$	0.01
Crude wastewater temperature	°C	16
Proposed treatment standard	COD/BOD, mg L^{-1}	125/25
	TN, mg L^{-1}	10
	TP, mg L^{-1}	1

Although primary and secondary sludges have been reported to present different degradation kinetics and methane yields under anaerobic conditions it has been assumed that in both instances an SRT of 20 days is sufficient to achieve 50 % VS reduction and that 0.75 m^3 $Kg_{VS\ destroyed}$ of biogas is produced. The amount of sludge and methane produced by the G-AnMBR was estimated based on the sludge yield of 0.14 gVSS $gCOD^{-1}$ shown by the suspended growth system and the methane yield of 0.13 LCH_4 g

COD^{-1} reported in an EGSB operated in parallel to the G-AnMBR assuming biogas would be recycled for sparging (McAdam *et al*, 2010). Operation of the G-AnMBR is based around standard energy demand for a UASB with an additional demand due to membrane fouling control. This is based around the intermittent gas sparging rates described in chapter 6 of 1 minute on, 10 minutes off.

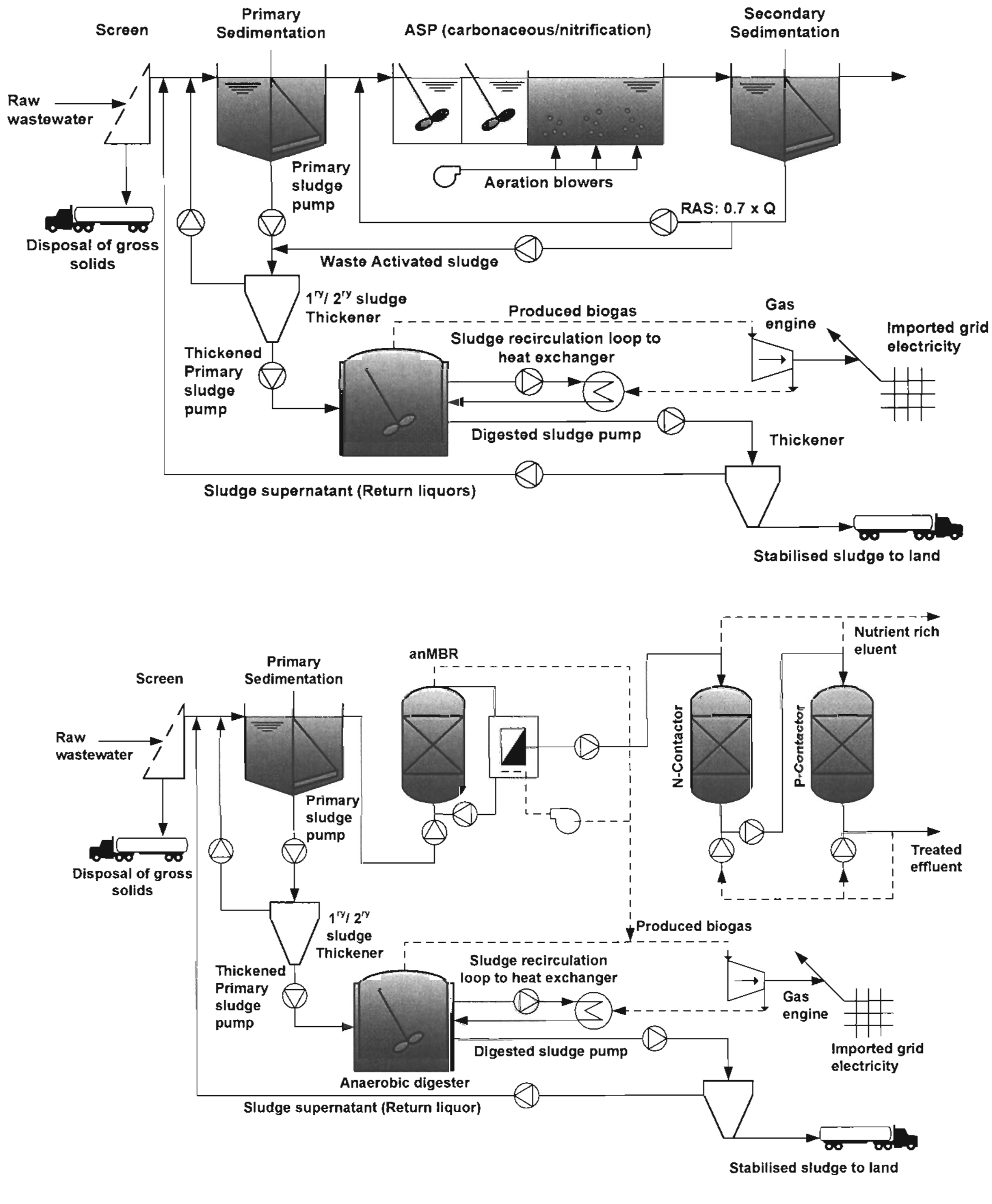

Figure 8.1 Schematic diagram of conventional (above) and proposed (below) flow sheet

Annual energy demand for the conventional ASP flow sheet was estimated at 1525700 kWh.year^{-1} equating to an energy demand of 0.418 kWh m^{-3} with 48 % corresponding to aeration (Table 8.2). This energy demand is partially offset from the biogas from the anaerobic digestion of the sludge yielding a net energy demand of 1197200 kWh year^{-1} which equates to 0.33 kWh m^{-3} costing £26, 735 year^{-1}. Switching to the G-AnMBR flow sheet results in a 40 % reduction of the total energy demand of the plant to 928925 kWh year^{-1} which equates to 0.26 kWh m^{-3} from which 47 % corresponds to the energy demand related to fouling control. However in the case of the G-AnMBR, electricity production from both AD and primary effluent generated 611667 kWh.year^{-1} leading to a net consumption of electricity of 317258 kWh.year^{-1} or 0.09 kWh.m^{-3} which represents a reduction of 73 % with respect to the aerobic flowsheet. In terms of sludge production the G-AnMBR flowsheet generated 0.181 Kg $_{DS}$ m$^{-3}{}_{flow}$ which represents a 26 % reduction with respect to the conventional ASP that produced 0.245 Kg $_{DS}$ m$^{-3}{}_{flow}$ for disposal. The difference in sludge production accounts mainly for the lower sludge yield of 0.14 gVSS gCOD^{-1} in the G-AnMBR as compared to 0.29 gVSS gCOD^{-1} in the ASP. This clearly illustrates that primary solids are the main contributor to the overall sludge production and that higher reductions with respect to conventional aerobic treatment could be only attained in a flowsheet in which the full flow is treated in the anaerobic reactor. However, such flowsheet will also alter the overall energy balance since higher fouling propensity and lower overall methane productions would be expected as compared to the treatment scheme proposed herein. Converted into economic factors the reductions related to energy and sludge disposal by switching to the G-AnMBR flow sheet represents a reduction of 47 % of the overall costs. However this does not take into account the cost associated for chemical regeneration of the ion exchange resins proposed for nutrient removal.

Table 8.2 Summary of flow sheet comparison between ASP and G-AnMBR

	ASP	**G-AnMBR**
Energy used kWh m^{-3} (kWh y^{-1})		
Mechanical treatment	0.033 (120450)	0.033 (120450)
Aeration	0.202 (737300)	-
Anoxic mixing	0.042 (153300)	-
Internal recycling	0.050 (182500)	0.025 (91250)
Return activated sludge	0.039 (142350)	-
Sludge	0.052 (189800)	0.040 (146000)
G-AnMBR	-	0.12 (438000)
Nutrient plant	-	0.037 (133225)
Total energy used kWh m^{-3} (kWh y^{-1})	**0.418 (1525700)**	**0.255 (928925)**
Energy produced kWh m^{-3} (kWh y^{-1})		
AD (primary and secondary solids)	0.110 (401500)	0.096 (350400)
G-AnMBR (primary effluent)	-	0.072 (261267)
Total energy produced kWh m^{-3} (kWh y^{-1})	**0.11 (328500)**	**0.17 (611667)**
Net energy kWh m^{-3} (kWh y^{-1})	**0.31 (1197200)**	**0.09 (317258)**
Sludge produced KgDS m^{-3} (Ton y^{-1})		
Primary treatment	0.302 (1100.81)	0.257 (936.83)
Secondary treatment	0.097 (353.12)	0.058 (209.94)
Sludge disposed	0.245 (894.73)	0.181 (661.68)
Economic £ m^{-3} (£ y^{-1})		
Energy	**0.023 (83804)**	**0.006 (22208)**
Sludge disposal	**0.029 (107368)**	**0.022 (79401)**
Total Costs £ m^{-3} (£ y^{-1})	0.051 (186062)	0.028 (101609)

The above analysis highlights a number of features related to low energy flow sheets that are worth considering:

1. The energy balance of the AnMBR flow sheet is heavily dependent on the energy used for membrane fouling control (Figure 8.2). The application of intermittent gas sparging reduces the sensitivity of the membrane operation on the overall energy balance. In the current case of G-AnMBR with 10 % intermittent gas sparging and a SGD_m of 0.8 m^3 h^{-1} m^2 energy demands for the overall flowsheet could be reduced to 0.09 kWh m^{-3} without methane recovery. However, if the suspended growth AnMBR were adopted, the best case fouling control would results in an increase in energy demand of the individual process unit to 2117000 kWh.year^{-1} and would render the overall flow sheet as a net energy user at a rate of 0.55 kWh.m^{-3}. Clearly the success of using AnMBRs is largely dependent upon development of low energy fouling control strategies as suggested within this report.

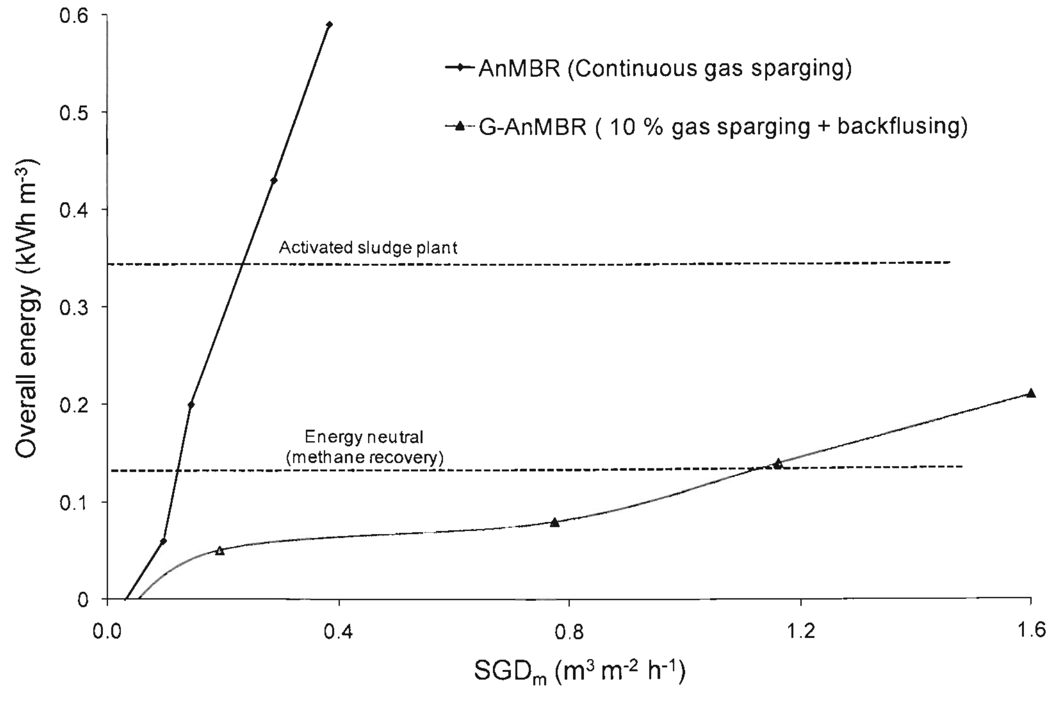

Figure 8.2 Influence of gas sparging rate and frequency on energy balance

2. Adoption of an anaerobic reactor for organics removal results in increased biogas production of around 40% as a fraction of the total biogas produced compared to the ASP. This excludes liquid phase methane which can represent up to 50% of the total produced from the anaerobic reactor. As such with appropriate methane recovery, biogas production from the G-AnMBR will exceed that from the AD and compensate the power consumption for fouling control in the G-AnMBR shifting the overall flowsheet energy balance from 0.09 to 0.02 kWh m^{-3}.

3. In terms of sludge treatment, given that primary sludge represents the major fraction of the sludge generated, the reduction in overall sludge production by considering the treatment of primary effluent with an AnMBR is limited to 30 % despite presenting a sludge yield 50 % lower than the ASP. This would lead to the consideration of a flowsheet in which the full flow is treated in the AnMBR for which a suspended growth reactor would be more suitable than a granular system due to the higher influent solid concentration.

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CHAPTER 9:

CONCLUSIONS AND FUTURE WORK

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9.1 CONCLUSIONS

This project has investigated the potential of anaerobic MBRs as a core treatment step of low strength domestic wastewater. The main aspects investigated were related to start up strategies, treatment performance, sludge production, membrane fouling control and post treatment of AnMBR effluents for nutrient removal. With regards to these points a series of conclusions can be drawn from the experimental work carried out through the project.

1. Compared to a conventional activated sludge treatment scheme significant reductions in sludge and energy costs can be obtained by considering a flowsheet based on a G-AnMBR. The overall energy balance is strongly dependent of membrane operation.

2. The complete retention of solids attained by introducing a membrane does not accelerate the start up of unseeded anaerobic reactors at low temperatures under the experimental conditions shown in the present study. Prolonged starts up periods are required unless the bioreactors are preseeded. Seeding enables compliant effluent to be provided within days of start up.

3. Adoption of AnMBRs reduces the organics to such a level that downstream biological nutrient removal is not practical. Consequently non biological methods are required such as with modern adsorption resins enabling both very low effluent concentrations and recovery to be accomplished.

4. Appropiate, energy sustainable fouling control is possible in a G-AnMBR due to the predominantely colloidal nature of the foulants. In such cases, intermittent gas spargign is effective thus providing a route to low energy fouling control.

9.2 FUTURE WORK AND RECOMENDATIONS

The research conducted in this project has shown that anaerobic membrane bioreactors in which the biological treatment and membrane filtration processes are separated are a potential technology for mainstream wastewater treatment as compliant effluent concentrations were achieved even at low temperatures and low energy demand associated for fouling control were required. Although in Chapter 8 it was shown that energy demands could be reduced with respect to the commonly utilised conventional activated sludge system, extremely high investment costs would be necessary due to the low membrane fluxes applied. Therefore further research that comes out of this project has to focus on the optimisation of the filtration process as well as the bioreactor operational conditions that reduce fouling propensity of the biological matrix.

1. Long term trials in which higher fluxes than those reported in the present study are required in combination with different chemical cleaning protocols in order to balance energy demands and capital costs are required. Additionally the impact of implementing different backwashing and gas sparging frequencies should be assed in order to minimize residual fouling rates.

2. Further research is required to ascertain whether the granular structure could be maintained through bioreactor operation and further granulation would occur or if suspended growth biomass would dominate as a result of introducing a membrane filter. Additionally as granular sludge is difficult to source future investigations could examine the potential of a high rate anaerobic membrane bioreactor in which the sludge bed is formed by flocculent sludge.

3. In either case accumulation of solids in the sludge bed needs to be accounted for as well as the impact of periodically wasting bioreactor effluent as a mean of limiting the load of colloidal matter onto the membrane.

4. Characterisation of anaerobic MBR sludge in terms of surface charge and hydrophobicity might provide insights into the interactions between membrane and the biological suspension which could then be used to select the membrane materials that minimise fouling.

5. Further research is required in order to ascertain the concentration of dissolved methane on the bioreactor effluent, in order to avoid environmental issues related to its release to the atmosphere and achieve further energy recovery.

6. A better understanding of the mechanisms involved in biodegradation of organics including identification of dominant microbial communities at low temperatures, relative rates of different steps of anaerobic digestion would be helpful in order to be able to predict behaviour and set design parameters for anaerobic MBRs.

7. Further investigations have to be conducted to explain the production of soluble inert organics at lower temperatures in order to verify whether they originate from influent solids or microbial products.