DEVELOPMENT OF SPECIFIC GRANULAR GROWTH ANAEROBIC MEMBRANE BIOREACTORS FOR DOMESTIC WASTEWATER TREATMENT

Cheng Chen

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CERTIFICATION OF ORIGINAL AUTHORSHIP

I certify that the work in this thesis has not previously been submitted for a degree nor has it been submitted as part of requirements for a degree except as part of the collaborative doctoral degree and/or fully acknowledged within the text.

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis. This research is supported by an Australian Government Research Training Program Scholarship.

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LIST OF ABBREVIATIONS

ABR	Anaerobic baffled reactor
ADUF	Anaerobic digestion ultrafiltration
AF	Anaerobic filter
AFBR	Anaerobic fluidized bed reactor
AFMBR	Anaerobic fluidized bed membrane bioreactor
AG-MBR	Aerobic granular-membrane bioreactor
AG-MMBR	Aerobic granule-mesh filter MBR
AHR	Anaerobic hybrid reactor
AMBR	Anaerobic migrating blanket reactor
AMBBR	Anaerobic moving bed biofilm reactor
AMBRs	Aerobic membrane bioreactors
Anammox	Anaerobic ammonium oxidization
AnAOB	Anaerobic ammonia-oxidizing bacteria
AnBEMRs	Anaerobic bio-entrapped membrane bioreactors
AnDMBRs	Anaerobic dynamic membrane bioreactors
AnGBR	Anaerobic granular bioreactor
AnMBRs	Anaerobic membrane bioreactors
AnMDBRs	Anaerobic membrane distillation bioreactors
AnMSBRs	Anaerobic membrane sponge bioreactors
AnOMBRs	Anaerobic osmotic membrane bioreactors
ARMBR	Anaerobic rotary disk membrane bioreactor
ASBR	Anaerobic sequencing batch reactor
ATU	Allylthiourea
BOD	Biological oxygen demand
BPC	Biopolymer clusters
C-AnMBRs	Conventional anaerobic membrane bioreactors
CANON	Completely autotrophic nitrogen removal over nitrite
CDOC	Chromatographic dissolved organic carbon
CFMBR	Continuous flow membrane bioreactor
CFV	Crossflow velocity

CG-AnMBR	Conventional granular anaerobic membrane bioreactor
CGSFDMBR	Continuous-flow bioreactor with aerobic granular sludge and self-forming dynamic membrane
COD	Chemical oxygen demand
DCE	Dichloroethylene
DM	Dynamic membrane
DO	Dissolved oxygen
DOC	Dissolved oxygen demand
DS	Draw solution
EEM	Excitation emission matrix
EG-AnMBR	External granular anaerobic membrane bioreactor
EGSB	Expanded granular sludge bed reactor
EGSB-ZBF	Expanded granular sludge bed reactor-Zeolite bed filtration
EPS	Extracellular polymeric substances
EPS_C	Polysaccharides concentration of extracellular polymeric substances
EPS_P	Protein concentration of extracellular polymeric substances
Ex/Em	Exitation/Emission
FA	Free ammonia
FAN	Free ammonium nitrogen
FB	Fluidized bed
F/M ratio	Food-to-microorganisms ratio
FO	Forward osmosis
FOG	Fat, Oil, and Grease
FS	Feed solution
GAC	Granular activated carbon
G-AnMBR	Granular anaerobic membrane bioreactor
GHG	Greenhouse gas
Gl-AnMBRs	Gas-lifting AnMBRs
GLS	Gas/liquid/solid
GWP	Global warming potential
H/D	Height to diameter ratio
HOC	Hydrophobic organic carbon
HPI	Hydrophilic organics

HPO	Hydrophobic organics
HRARs	High rate anaerobic reactors
HRT	Hydraulic retention time
HSP	Hydraulic selection pressure
IAFMBR	Integrated anaerobic fluidized-bed membrane bioreactor
ID	Internal diameter
JFAB	Jet flow anaerobic bioreactor
KSAMBR	Kubota Submerged Anaerobic Membrane Bioreactor
LCFAs	Long chain fatty acids
LC-OCD	Liquid chromatography-organic carbon detection
LMW	Low molecular weight
MARS	Membrane anaerobic reactor system
MBR	Membrane bioreactor
MD	Membrane distillation
MFSBR	Conventional floc based membrane bioreactor
MF/UF	Microfiltration/Ultrafiltration
MGSBR	Membrane bioreactor with aerobic granular sludge
MLSS	Mixed liquor suspended solids
MLVSS	Mixed liquor volatile suspended solids
MPB	Methane producing bacteria
MS	Mass spectrometer
MTBE	Methyl-tert-butyl ether
O&G	Oil and grease
ORP	Oxidation and reduction potential
PAOs	Phosphate accumulating microorganisms
PCP	Pentachlorophenol
PES	Polyethersulfone
PP	Polypropylene
PSD	Particle size distribution
PTFE	Polytetrafluoroethylene
PUS	Polyester-urethane sponge
PVC	Poly vinyl chloride
PVDF	Polyvinylidene fluoride

SAAMB-AnGMBR	Sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor
SAAMBR	Sponge-assisted aerobic moving bed reactor
SAF-MBR	Staged anaerobic fluidized membrane bioreactor
SA-GAnMBR	Sponge assisted-granular anaerobic membrane bioreactor
SAMA	Specific acetoclastic methanogenic activity
SAnMBR	Submerged anaerobic membrane bioreactor
S-AnMBR	Suspended growth AnMBR
SBR	Sequencing batch reactor
SCOD	Soluble chemical oxygen demand
SDR	Specific denitrification rate
SG-AnMBR	Submerged granular anaerobic membrane bioreactor
SIM	Selected ion monitoring
SMA	Specific methanogenic activity
SMP	Soluble microbial products
SMP_C	Polysaccharides concentration of soluble microbial products
SMP_P	Protein concentration of soluble microbial products
SND	Simultaneous nitrification and denitrification
SNR	Specific nitrification rate
SOLR	Specific organic loading rate
SRB	Sulphate reducing bacteria
SRT	Solid retention time
SS	Suspended solids
SS-AnGMBR	Submerged sponge-assisted anaerobic granular sludge membrane bioreactor
STP	0° C Standard Temperature and 1 atm Pressure
SVI	Sludge volume index
TCE	Trichloroethylene
TCOD	Total chemical oxygen demand
TMP	Transmembrane pressure
TN	Total nitrogen
TOC	Total organic carbon
TPAD	Two-phase anaerobic digestion

TPAnMBRs	Two-phase AnMBRs
TS	Total solids
TSS	Total suspended solids
OLR	Organic loading rate
UAGB	Upflow anaerobic granular bioreactor
UASB	Upflow ananerobic sludge bed
UASB-AFF	Upflow-anaerobic fixing filter
US-AnMBR	Ultrasonic anaerobic membrane bioreactor
UTS	University of Technology Sydney
V-AnMBRs	Vibrating AnMBRs
VC	Vinyl chloride
VFAs	Volatile fatty acids
VTS	Volatile total solids
WAS	Waste activated sludge
WHO	World Health Organization Guidelines
WWTPs	Wastewater treatment plants
ZVI-UASB	Zero valence iron packed UASB

LIST OF SYMBOLS

B - A	The number of days that the system is operated
C_2	Acetic acid
C_3	Propionic acid
C_4	Butyric acid
C_5	Valeric acid
Ca	Calcium
$CaCl_2 \cdot 2H_2O$	Calcium chloride
CH_4	Methane
$C_6H_{12}O_6$	Glucose
Co	Cobalt
$CoCl_2$	Cobalt chloride
CO_2	Carbon dioxide
$CuSO_4 \cdot 5H_2O$	Cupric sulphate
Fe	Iron
Fe^{2+}	Ironized ferrous
$FeCl_3$	Ferric chloride anhydrous
$FeSO_4$	Ferrous sulfate
H_2	Hydrogen
H_2S	Hydrogen sulfide
H_2SO_4	Hydrogen sulfate
i-C_4	Iso-butyric acid
i-C_5	Iso-valeric acid
IC_{50}	50% activity inhibitory concentration
J	Permeation flux
KH_2PO_4	Potassium phosphate
Mg	Magnesium
$MgSO_4 \cdot 7H_2O$	Magnesium sulphate
$MLSS_A$	MLSS concentration of granular sludge when operation is started
$MLSS_{A1}$	MLSS concentration of sponge-attached biosolids when operation is initiated

$MLSS_B$	MLSS concentration of granular sludge when operation is terminated
$MLSS_{B1}$	MLSS concentration of biosolids attached on the sponge when operation is terminated
$MnCl_2 \cdot 7H_2O$	Manganese chloride
N_2	Nitrogen gas
Na^+	Ionized sodium
NaCl	Sodium chloride
NaClO	Sodium hypochlorite
$NaHCO_3$	Sodium biocarbonate
$Na_2MoO_4 \cdot 2H_2O$	Sodium molybdate dehydrate
NaOH	Sodium hydroxide
$(NH_4)_2SO_4$	Ammonium sulphate
NH_3	Free ammonia
NH_4^+	Ionized ammonia
NH_4-N	Ammonia nitrogen
Ni	Nickel
$NiSO_4$	Nickel sulphate
NO_2-N	Nitrite nitrogen
NO_3-N	Nitrate nitrogen
PO_4-P	Orthophosphate
R_C	Cake layer resistance
R_M	Intrinsic membrane resistance
R_P	Pore blocking resistance
R_T	Total resistance
SO_4-S	Sulfate sulfur
SO_4^{2-}	Ionized sulfate
Sw	The weight of sponge
TMP_A	The transmembrane pressure value (kPa) when operation is started
TMP_B	The transmembrane pressure value (kPa) when operation is terminated
μ	Dynamic viscosity of the permeate
U_v	Upflow velocity
V_T	Total volatile fatty acids

$ZnSO_4 \cdot 7H_2O$	Zinc sulphate
$\Delta MLSS/\Delta t$	Biomass growth rate
$\Delta MLSS/Sw$	Attached biomass growth rate
ΔP	Transmembrane pressure
$\Delta TMP/\Delta t$	Membrane fouling rate

PhD DISSERTATION ABSTRACT

Author: Cheng Chen

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Faculty: FEIT

School: Civil and Environmental Engineering

Supervisors: Dr. Wenshan Guo (Principal supervisor)

Dr. Huu Hao Ngo (Alternative supervisor)

Abstract

In recent years, water scarcity has brought global health and environmental concerns. To overcome this issue, anaerobic granular membrane bioreactors (G-AnMBRs) have been widely used for reclaiming domestic wastewater. However, there were still critical issues associated with G-AnMBRs, such as membrane fouling and granule fragmentation.

This thesis focused on developing a novel sponge-based G-AnMBR for domestic wastewater treatment. Results showed that the G-AnMBR start-up could be successfully accomplished using flocculent aerobic sludge as the inoculum. Hydraulic retention time (HRT) of 12 h permitted better organic removal and superior granular sludge quality. The external G-AnMBR (EG-AnMBR) served as a better G-AnMBR configuration due to less fouling propensity and superior granule quality. Membrane direct incorporation into the submerged G-AnMBR (SG-AnMBR) significantly enhanced microbial products (e.g. soluble microbial products (SMP) and extracellular polymeric substances (EPS)) in the mixed liquor and cake layer, and reduced granules EPS content and settleability. The EG-AnMBR demonstrated less SMP and EPS in the mixed liquor and cake layer, which might reduce the cake layer resistance and lower fouling rate.

The sponge assisted-granular anaerobic membrane bioreactor (SA-GAnMBR) showed enhanced treatment performance than the converntional G-AnMBR (CG-AnMBR). Granular sludge from the SA-GAnMBR had superior quality with better settleability, larger particle size, higher EPS content and more granule abundance. The SA-GAnMBR also exhibited slower fouling development with 50.7% lower total filtration resistance than those of the CG-AnMBR. Sponge addition effectively reduced the concentration of microbial products in the cake layer and settling zone mixed liquor, and lowered the concentrations of major foulant organics, thus alleviating the fouling propensity.

The new hybrid sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor (SAAMB-AnGMBR) showed organic removal efficiencies over 94% at all COD/N (C/N) ratio conditions. Nutrient (nitrogen and phosphate) removal was over 91% at C/N ratio of 100/5 but was negatively affected when decreasing C/N ratio to 100/10. At lower C/N ratio (100/10), more noticeable membrane fouling was caused by aggravated cake formation and pore clogging, and EPS accumulation in the mixed liquor and sludge cake as a result of deteriorated granular quality. Significant difference existed in the foulant organic compositions under different C/N ratios. The performance of the hybrid system was found to recover when gradually increasing C/N ratio from 100/10 to 100/5. This work aimed to offers a useful performance enhancement and fouling control strategy for G-AnMBR operation, and provide a solid platform for advances in novel G-AnMBR applications for domestic wastewater treatment.

Keywords: Granular anaerobic membrane bioreactor; Membrane fouling; Methane yield; Biogas; Soluble microbial products; Sponge; Nutrient removal

CHAPTER 1

INTRODUCTION

1.1 Research background

With ever-increasing consumption and pollution of our limited water resources, water scarcity has brought increasing levels of public concern on a global scale. Nevertheless, this worsening situation could be relieved via water reuse schemes using suitable wastewater treatment technologies to treat and reclaim wastewater. Currently, conventional wastewater treatment schemes are typically characterized by its energy intensive and centralised feature and its failure to recover the potential valuable resources available in wastewater including nutrients and bioenergy (Balkema et al., 2002; Smith et al., 2012). Due to the increasing interest in the concept of sustainability within wastewater management, the focus on future sustainable development has driven innovations in anaerobic membrane bioreactors (AnMBRs), which were considered as a more promising alternative to aerobic bioprocesses and conventional anaerobic processes. With the complete retention of anaerobic microorganisms at a decentralized onsite treatment scope, AnMBRs can not only achieve high effluent quality, small footprint and very low sludge production, but also significantly contribute to nutrient recovery for irrigation use and renewable bioenergy generation for the substitution of fossil fuel in power and heat production (Stuckey et al., 2012; Lin et al., 2013; Pretel et al., 2013; Chen et al., 2016a).

Currently, conventional AnMBRs (C-AnMBRs) are dominated in the form of continuous stirred tank reactor (CSTR) configuration with either internal or external membrane separation devices, and they are operated based on suspended growth pattern (Lin et al., 2013). AnMBRs studies by far were predominantly C-AnMBRs (Huang et al., 2011; Martinez-Sosa et al., 2011; Martinez-Sosa et al., 2012), because of the ease of use and construction. However, critical obstacles including membrane fouling, low flux and high operational costs still exist and limit the wider application of C-AnMBR. Firstly, C-AnMBR was usually operated at a lower biomass concentration compared to high rate anaerobic reactors (HRARs) mainly due to fouling issues, corresponding to a lower organic loading rate (OLR) (<10 kg chemical oxygen demand (COD)/$m^3 \cdot d$) (Lin et al., 2013). Secondly, to maintain well-mixed flow regime and sufficient mass transfer, rigorous mechanic mixing is required, which is energy intensive. Thirdly, the membrane unit is directly exposed to the bulk sludge, and the high suspended solid concentration subjected to MF/UF membrane filtration exacerbate cake deposition in CSTR configuration, either with pressure-driven or vacuum-driven membranes. More rapid and dense cake layer build-up results in heavy membrane fouling and low flux, as effluent solids concentration of CSTRs is equal to the bulk solids concentration. To resolve this issue, frequent physical and chemical cleaning, membrane replacement, interval operation, and likely sub-critical flux operation to sustain the flux should be in place, which increases operation costs (Lew et al., 2009; Yoo et al., 2012). Lastly, sludge recirculation through the membrane feed pump, especially for external cross-flow membranes, results in a substantial decrease in the mean particle size, bringing in severe membrane fouling (Ozgun et al., 2013).

Incorporating AnMBR with granular biotechnology to form granular anaerobic membrane bioreactor (G-AnMBR) offers a viable countermeasure to the high fouling propensity and energy consumption due to the superior quality of anaerobic granules such as its settleability, mechanical strength, diverse microbial populations, porosity, and substrate permeability. Unlike C-AnMBRs predominantly relied on the suspended growth, granular biomass retention is achieved by the spontaneous formation of granular sludge in G-AnMBRs. The anaerobic granular bed is usually featured with total biomass concentrations ranging from 20 to 40 g/L whereas significantly lower effluent total solids (TS) concentration at 50 mg/L was possible (An et al., 2009). All the biological reactions occurred within the dense sludge bed at the bottom of the upflow anaerobic granular bioreactor (UAGB). When combining UAGB with membrane filtration, the entrapment of most particulate organics by adsorption and biodegradation in the granular sludge bed allowed membrane module only being challenged by the supernatant of the granular sludge bed, thus reducing the organic loading to the membrane (Martin-Garcia et al., 2011; Ozgun et al., 2015). Hence, less apparent formation of dense cake layer and its consolidation occurs as compared to C-AnMBR (Ozgun et al., 2015). Furthermore, G-AnMBR required much lower gas sparging intensity, resulting in lower energy demand for fouling control (Martin-Garcia et al., 2013). Last but not least, the natural occurring turbulence caused by the rising biogas bubbles and liquid upflow force, which buoy the granular sludge, provides sufficient substrate and microorganism contact so as mechanical mixing is no longer required and relevant operational cost would be greatly decreased (Chong et al., 2012).

1.2 Research objectives

This thesis focused on the development of novel G-AnMBRs for municipal wastewater treatment. The impact of various factors (i.e. inoculum, hydraulic retention time (HRT), membrane addition method, sponge addition, COD/N (C/N) ratio) on G-AnMBR performance was investigated. The long-term goal of the research is to investigate and develop a novel G-AnMBR for domestic wastewater treatment and reuse, and to optimize such G-AnMBR in terms of treatment performance, renewable energy production, granular sludge quality and membrane fouling through various strategies. The specific objectives of this study are as follows:

- To present a comprehensive review of current studies on AnMBRs and granular biotechnology;
- To investigate the feasibility of using aerobic inoculum to develop anaerobic granules;
- To evaluate the effects of HRT on the G-AnMBR performance;
- To assess the effects of reactor configuration on the overall performance of the G-AnMBR;
- To develop and appraise a sponge assisted-granular anaerobic membrane bioreactor (SA-GAnMBR) in terms of treatment performance, granular sludge properties, membrane fouling behaviour and biogas production; and
- To propose a new hybrid sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor (SAAMB-AnGMBR), and evaluate the effect of C/N ratio on the performance of such system in municipal wastewater treatment.

1.3 Scope of the study

(1) Preliminary Research on G-AnMBR

Laboratory experiments were conducted at Environmental Engineering Lab at University of Technology Sydney (UTS) with the experimental conditions at different inoculums and HRT. These experiments were conducted to investigate the feasibility of using aerobic sludge as seed for G-AnMBR, and to determine the optimum HRT for G-AnMBR operation.

(2) Study on effect of reactor configuration on the performance of G-AnMBR

A detailed experimental study was conducted to investigate the effect of reactor configuration on the performance of G-AnMBR. For this purpose, a direct comparison of an external and a submerged granular anaerobic membrane bioreactor (EG-AnMBR and SG-AnMBR) was conducted. The comprehensive evaluation of the two G-AnMBRs included the investigation of treatment efficiencies, granules properties (eg. particle size, settling velocity and extracellular polymeric substances (EPS)), membrane fouling behaviour (transmembrane pressure (TMP), potential foulants, fouling resistance analysis), and renewable energy recovery (methane yield).

(3) Study on SA-GAnMBR system

A new SA-GAnMBR was developed at UTS for the enhancement of organic and nutrient removal and membrane fouling mitigation. To this aim, the overall treatment performance of a sponge assisted-granular anaerobic membrane bioreactor (SA-GAnMBR) and a conventional G-AnMBR (CG-AnMBR) was evaluated. Granule properties (e.g. particle size distribution (PSD), soluble microbial products (SMP) and EPS, sludge volume index (SVI)), fouling propensity (e.g. TMP, SMP and EPS of the mixed liquor and cake layer, and foulant organics) and biogas production were also assessed.

(4) Study on SAAMB-AnGMBR system

A laboratory scale SAAMB-AnGMBR was developed for the enhancement of nutrient removal when treating domestic wastewater. The impact of C/N ratios on the treatment performance and membrane filtration performance of SAAMB-AnGMBR was evaluated. The system recovery after the overloaded nitrogen event was also investigated in this study.

1.4 Organization of the report

The thesis is composed of 8 chapters and is structured as follows: Chapter 1 proposes the rationale of the study. Chapter 2 - 7 addresses the objectives of the research specifically. Chapter 8 draws conclusions and recommendations in relation to G-AnMBR operation for municipal wastewater treatment.

Chapter 1 briefly presents an overview of existing AnMBR applications and highlights the need for developing the next generation of technologies (i.e. G-AnMBR). The research scope and objectives are highlighted afterwards.

Chapter 2 exhibits fundamental aspects of AnMBRs (e.g. their evolution, mechanism, current applications and membrane fouling). It also critically reviews the current challenges in biogas production from AnMBRs. Most importantly, the background and the rationale for developing G-AnMBR for domestic wastewater were presented as the main part of this chapter.

Chapter 3 demonstrates the detailed materials and methods in this study. More specifically, domestic wastewater composition, membrane and sponge specifications, experimental setup and operation conditions, and analytical methods are explicitly included.

Chapter 4 identifies the potential for the development of anaerobic granules using aerobic seeds, and evaluates the impact of HRT on G-AnMBR performance in terms of treatment performance and granular properties.

Chapter 5 compares the overall biological performance, sludge properties and membrane fouling of an EG-AnMBR and a SG-AnMBR for municipal wastewater treatment, with the aim to investigate the impact of reactor configuration on G-AnMBR operation.

Chapter 6 develops a new SA-GAnMBR for enhancing treatment performance, granular quality and biogas production, as well as reducing membrane fouling.

Chapter 7 proposes a novel SAAMB-AnGMBR for improving nutrient removal during municipal wastewater treatment. The effects of C/N ratio on the performance of such hybrid system are also investigated.

Chapter 8 summarizes the major results of studies in this thesis, draws conclusions, and addresses future perspectives.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter provides an extensive review of recent AnMBR development with specific reference to its evolution, configurations, applications, fouling and renewable energy production. The current progress of granular membrane bioreactors, particularly for G-AnMBR in the treatment of wastewater is also critically elaborated.

2.2 Anaerobic membrane bioreactors (AnMBR)

2.2.1 Evolution of AnMBRs

The very first concept of AnMBRs was originated from the 1980s when Grethlein (1978) used two different external cross-flow membrane modules to treat the septic tank effluent. Increased biomass concentration was reported and this first AnMBR application achieved the reduction of Escherichia *coli*, turbidity, biological oxygen demand (BOD), nitrate and orthophosphate by 100%, 100%, 85 - 95%, 72 - 75% and 24 - 85%, respectively. More importantly, the practical flux was the highest among the available membrane related treatment practice, with the flux (400 to 600 $L/m^2 \cdot d$) UF module for over 900 pump hours or 1500 hours of real time without frequent cleaning of the membrane. With the recognition on AnMBRs' potential, both private and governmental sectors have made substantial financial commitment to the promotion of AnMBRs innovation over the 3 decades. In 1985, the first commercially available AnMBR, known as the membrane anaerobic reactor system (MARS), was considered as a notable milestone of the AnMBRs' revolution (Li et al., 1985). MARS was developed for treating dairy and wheat starch wastewater and was composed of a suspended growth anaerobic reactor followed by an external ultrafiltration membrane for solid-liquid separation. 95.1 - 99.2% COD removal and 90.7 - 99.9% total suspended solids (TSS) removal were reported in the various MARS pilot studies when treating different types of wastewaters including sweet whey, sweet whey permeate, acid whey permeate as well as wheat starch wastewater. Methane yield rate was also investigated and reported from 0.28 to 0.34 L CH_4/g $COD_{removed}$.

In 1987, independent pilot-scale research utilizing locally manufactured UF membranes for solid-liquid separation in the anaerobic treatment was launched. As a new generation process for treating industrial organic wastes, anaerobic digestion ultrafiltration (ADUF) was regarded as the major outcome of this research because it effectively eliminated the sludge concentration and retention dilemma, which were encountered by conventional systems (Ross et al., 1990). It was reported that 93% COD removal and almost 100% TSS reduction were achieved in the pilot-scale studies treating wine distillery waste. For the digester, no strict solid retention time (SRT) control and complete mixing were required. Sludge could be withdrawn from the digester and returned to the external UF unit at different levels. ADUF was also highlighted as a high rate process with high space load rates (>10 kg COD/m^3·d) and this led to a significant reduction in digester volume and capital cost. The use of compact locally produced membrane for phase separation instead of imported technology was deemed to increase its economic attractiveness and eliminated the necessity of large settling tanks and packing material such as fixed bed systems. Based on ADUF process, pilot- and full-scale BIOREK® system has been developed by BIOSCAN Engineering A/S, Denmark to treat pig manure. The system was composed of six unit operations (pre-separation, the ADUF process, ammonia stripping process, reverse osmosis (RO), gas purification and power generation). More than 80% of water was recovered from the slurry and more than 90% COD was removed through the mesophilic anaerobic digestion reaction at HRT of 6 days. Results from full-scale plant showed that it was possible to produce energy from biogas recovery, fertilizer from potassium and phosphate recovery, as well as potable demineralized water.

By 2000s, the focus on AnMBRs has started to shift from external configuration to submerged module. Kubota Submerged Anaerobic Membrane Bioreactor (KSAMBR) process, developed by Kubota Corporation, has been successfully employed in numerous food and beverage industries (Kanai et al., 2010). Up to 92% COD removal rate was reported in the case study because methane fermentation inhibitors such as ammonia were filtered out, enabling stable methane fermentation process. One of the main advantages of KSAMBR was that membranes retained the methanogenic bacteria while dissolved methane fermentation inhibitors (e.g. ammonia) were filtered out with the permeate. The digester volumes could be scaled down to 1/3 to 1/5 of the conventional digesters due to concentrated biomass. Similarly, ADI-AnMBR, developed by ADI Systems, Inc. (ADI), has been specifically employed in food wastewater treatment (McMahon, 2010). The largest ADI-AnMBR was completed by ADI in Ken's Foods of Marlborough MA, USA for upgrading one of its three WWTPs in order to maximize biogas production and treat high-strength organic content wastewater, containing high levels of fat, oil and grease. The performance was exceptional by any industry standards with 99.4% COD reduction and 100% suspended solids removal, allowing 100,000 gallon per day of wastewater to be easily discharged into the municipal system. As a part of the system upgrade, the AnMBR has been operated together with previously installed low-rate anaerobic reactor. The combined system produced approximately 5660 - 8500 m^2 biogas every day, which could not only satisfy 100% heating requirements of the WWTPs, but also provide more than 50% of power supply for the company's manufacturing facility.

2.2.2 Mechanisms of anaerobic digestion

Anaerobic digesters perform the most of the biodegradation of organic materials into usable energy in the form of methane at wastewater treatment plants, using the locally available residual biomass from various sources (animal waste, domestic sewage, industrial wastewater, and agricultural waste). The anaerobic digestion process is a complex bioprocess, which occurs in four main steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis (Figure 2.1). Hydrolysis acts as the first attack on the insoluble compounds such as particulate and colloidal wastes. These wastes are often higher molecular mass compounds such as polysaccharides, fat, and protein. In this stage, the enzyme-mediated transformation is accomplished to split such polymers into compounds suitable for the use as source of energy and cell carbon such as monosaccharaides, amino acids, acetate and varying amounts of volatile fatty acids (VFAs). Acetate and hydrogen, which can be utilized by methanogens directly, can also be produced as ending products in the first stage. Hydrolytic bacteria or facultative anaerobes and anaerobes are the main groups which perform the splitting of the compounds with water during the hydrolysis process.

In the second step, the acidogenic bacteria or acid-formers such as *Clostridium* further break down the simple monomers (simple sugars, amino acids, and fatty acids) into VFAs and alcohols along with ammonia, carbon dioxide, hydrogen sulphide as well as other byproducts (Yadvika et al., 2004; Weiland, 2010; Wikipedia, 2017). For the third step of anaerobic digestion, namely, acetogenesis, acetogens further digest the simple molecules formed in the acidogenesis stage. At this stage, many of the acids and alcohols are converted into largely acetic acid as well as carbon dioxide and hydrogen that can be directly used up by methane-forming bacteria. Nevertheless, the maintenance of extremely low partial pressure of hydrogen is required for the well-being of the acetogenic and hydrogen-producing bacteria, because the accumulation of hydrogen can inhibit the metabolism of the acetogenic microbes (Weiland, 2010).

In the final phase, methanogens convert intermediate products of the preceding stages including acetate, hydrogen and carbon dioxide into methane, carbon dioxide and water. This stage contributes to the most methane production, with about 75% of methane production from decarboxylation of acetate and the rest from CO_2 and H_2 (McCarty and Smith, 1986). The availability of hydrogen is believed to be a limiting factor for methanogens and this is based on the fact that the addition of hydrogen-producing bacteria can increase the biogas yield (Bagi et al., 2007). As the final product, biogas from anaerobic digestion is mainly composed of methane, which can be considered an alternative source of renewable energy (Zhao, 2011).

A balanced methane fermentation process requires individual degradation phases to be carried out by distinct consortia of bacteria, namely fermentative bacteria, syntrophic acetogens, homoacetogens, hydrogenetrophic methanogens and aceticlastic methanogens (Figure 2.1). The symbiotic relationship among these microorganisms contributes to efficient anaerobic digestion and biogas production (Angelidaki et al., 1993; Weiland, 2010; Visvanathan and Abeynayaka, 2012). The most common upset and failure of anaerobic digesters occurs mostly because of methanogenic inhibition. This is due to the fact that methane-forming stage is the most sensitive and rate limiting step in the process since methane-forming bacteria have a much slower growth rate compared to acid-forming bacteria, and are sensitive to inhibitors such as ammonia, temperature, pH and other operation conditions. It is, therefore, imperative to retain sufficient slow-growing methanogenic bacteria and prevent active biomass from being washed out from the fermenter, and to reduce inhibitory levels.

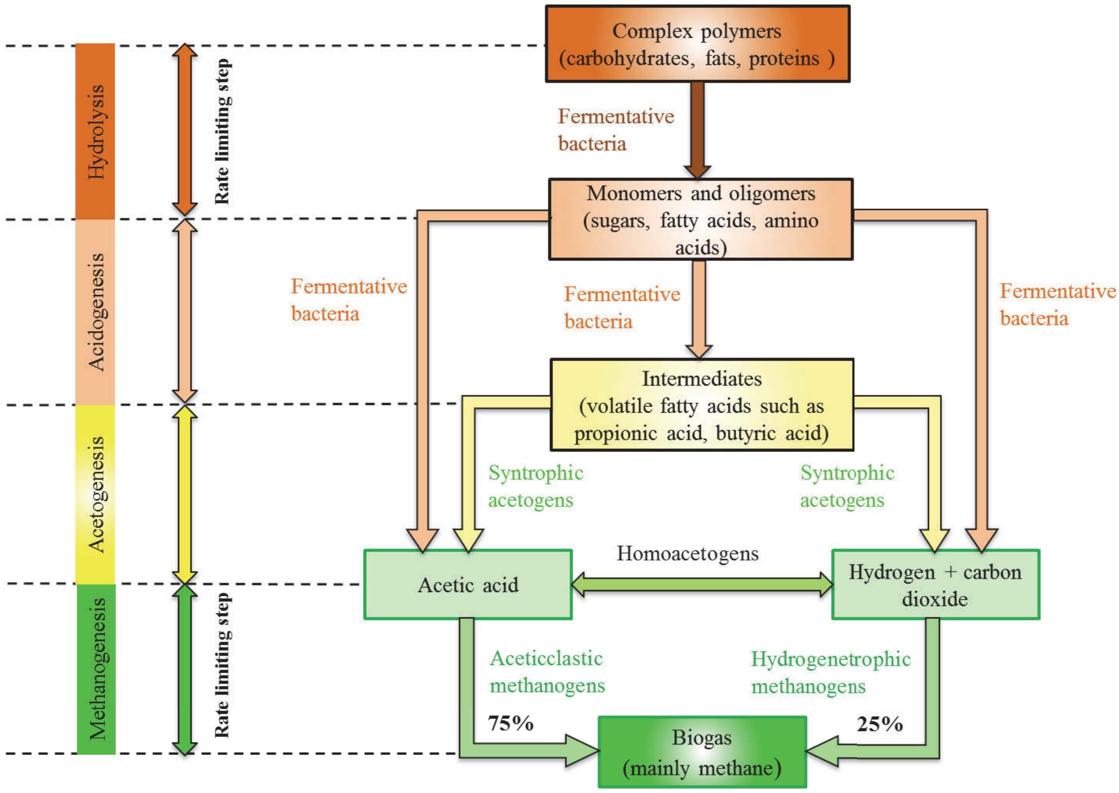


Figure 2.1 A schematic diagram showing the comprehensive processes of anaerobic digestion

2.2.3 Current status of AnMBR applications

(1) Conventional single-phased AnMBRs

Traditionally, an AnMBR consists of an anaerobic reactor and a membrane separation unit. This means that the main steps of methane fermentation including hydrolysis, acidogenesis and methanogenesis have to be completed in the one reactor. Currently, available anaerobic reactors could be generally categorized into two major groups depending upon whether there is biomass retention. CSTR is a classic example of reactor design that is unable to provide biomass retention, but it is the most frequently used reactor configuration in AnMBRs. Upflow ananerobic sludge bed (UASB), anaerobic filter (AF), fluidized bed (FB) are the most common designs that offer biomass retention (Liao et al., 2006). Anaerobic processes have been historically less employed for treating domestic wastewater, which is typically characterized by its low organic strength and high-suspended solids concentration. The kinetic limitations of anaerobic metabolism and poor biomass retention serve as the two main reasons impeding the development of conventional anaerobic processes (Liao et al., 2006; Herrera-Robledo et al., 2010; Lin et al., 2013). The advent of AnMBRs has resolved the dilemma associated with conventional anaerobic digestion. It was well documented that such a novel technology could contribute to a sustainable municipal wastewater treatment with complete biomass retention, excellent effluent quality, renewable source of energy (methane production), lower sludge production and no extra energy consumption associated with aeration as compared with aerobic membrane bioreactors (Lin et al., 2013).

Martinez-Sosa et al. (2011) proposed a pilot scale submerged AnMBR (SAnMBR), which was composed of a CSTR followed by an external flat sheet ultrafiltration unit, for domestic wastewater treatment under both mesophilic and psychrophilic temperature conditions. 90% COD removal efficiency was reported in the study under both temperature conditions. The author explained the higher COD removal rate by emphasizing the fact that the membrane could retain all particulate matter and a major fraction of soluble chemical oxygen demand (SCOD) inside of the reactor. Apart from that, the influent COD contained high amount of readily biodegraded COD because glucose was added to increase the total COD in the influent. This partially contributed to the high COD removal efficiency. Effluent quality in terms of COD, BOD and pathogen was exceptional, but nutrient removal in this case could be neglected. Therefore, the effluent could be readily reused for irrigation purposes in agriculture. The author also suggested that the fouling rate increased when temperature was dropped to psychrophilic temperatures and it was due to the accumulation of TSS and COD in the reactor. In terms of biogas yield, an average value of 0.27 L CH_4/g $COD_{removed}$ was reported under mesophilic conditions (at 35 °C), and a decreased methane yield was observed at 0.23 L CH_4/g $COD_{removed}$ under psychrophilic conditions (at 20 °C). Decreased methane content was also reported when temperature was decreased. Moreover, methane loss represented a big issue for this single stage AnMBR not only for economic considerations but also for environmental concerns. The global warming potential (GWP) of methane is about 21 times higher than that of carbon dioxide, and so methane leakage should be minimized through further system upgrades (Martinez-Sosa et al., 2011; Martinez-Sosa et al., 2012).

Saddoud et al. (2007a) investigated an anaerobic cross-flow ultrafiltration membrane bioreactor for municipal wastewater treatment. The single stage AnMBR system was composed of a jet flow anaerobic bioreactor and an external membrane module. The removal efficiency of such a system was considered excellent, with the removal rates of TSS, soluble COD and BOD at 100%, 90% and 88%, respectively. The quality of effluent was well compliant with World Health Organization Guidelines (WHO) for the microbiological quality of treated sewage unrestrictedly used in agriculture. However, further aerobic post-treatment should be added to reduce its contents in residual nitrogen and phosphate if the effluent is to be reclaimed in the hydraulic public domain. The methane production was at an average of 0.27 L CH_4/g $COD_{removed}$ due to the insignificant production of VFAs and stable methanogenic process. The author also observed that for the long-term operation, a sever flux decline was occurred due to pore clogging, and therefore routine cleaning was required. Saddoud et al. (2009) applied a single stage AnMBR for treating domestic water from the local wastewater treatment plant. The results showed that such a conventional AnMBR system was inefficient in terms of unstable performance such as considerable variations in the biogas yield rate and the methane composition in the biogas. The results confirmed that this single-phase process was unable to accommodate the great fluctuations in the domestic wastewater composition and the presence of toxic compounds originated from the industrial activities.

Originally, AnMBRs were innovated to deal with high strength industrial wastewater rather than municipal wastewater. This was because anaerobic digestion had a strong capability of biodegrading high strength influent, which was typically characterized by its high organic strength and extreme physical-chemical conditions in terms of pH, temperature and salinity. In general, AnMBRs have been widely investigated and used in food industrial wastewater treatment, and applications were not only at bench scale but also at full scale. On the other hand, the treatment of non-food processing effluents such as pulp and paper, textile, chemical, pharmaceutical and petroleum wastewaters by

AnMBRs applications was generally restricted at laboratory and pilot scale (Liao et al., 2006; Dereli et al., 2012; Lin et al., 2013).

Saddoud and Sayadi (2007) initiated a laboratory scale AnMBR study with the focus on treating slaughterhouse wastewater. The AnMBR system was a one phase anaerobic process similar to the one they used in Saddoud et al. (2007a). In this study, COD removal efficiency could be maintained at 93.7% although OLR was increased from 4.37 to 8.23 kg COD/m^3·d. The permeate quality was considered good with suspended solids, fats and protein completely removed. The VFA were also monitored with reported low concentrations ranging from 62 to 378 mg/L. However, a drastic reduction of the performance (53.6% COD removal efficiency) was observed when OLR was increased to 16.32 kg COD/m^3·d. The increase of the OLR was believed to overload the system and exert a devastating influence on the anaerobic biological processes, jeopardizing the balance of the microbial populations. More than 3000 mg/L VFA were found in the reactor, and this led to the acidification of the reactor, thus inhibiting the activity of methanogenic microbes. As an important indicator of treatment efficiency, methane yield was measured. The gradual decrease of the methane production from 0.31 L CH_4/g $COD_{removed}$ at OLR of 4.37 kg COD/m^3·d to 0.13 L CH_4/g $COD_{removed}$ at OLR of 13.27 kg COD/m^3·d showed the deteriorated treatment performance.

Casu et al. (2012) investigated a bench scale single-phase SAnMBR for the treatment of cheese whey, with the purpose of identifying the main operating conditions for the SAnMBR operation and its treatment efficiency in terms of COD removal and biogas production. OLRs varying between 1.5 and 13 kg COD/m^3·d were applied to observe the capability of the system in treating different level of OLRs. The lab scale SAnMBR demonstrated very good COD removal with an average value of 94% when OLRs of 1.5 - 10 kg COD/m^3·d were applied. However, a further increase in loading rate to 13 kg COD/m^3·d appeared to overload the system, causing a significant drop in COD removal to the minimum value of 33%. Therefore, the author concluded that the maximum applicable OLR was 10 kg COD/m^3·d. The system failure in this case was due to the VFA accumulation in the mixed liquor. It was measured that acetic acid surged to the highest value of 10.03 g/L while n-butyric acid showed a quite gentle increase to the maximum value of 3.15 g/L since the system overloading began. However, all the VFA reduced to normal levels once the OLRs were decreased. Biogas production also showed a significant reduction when the system was compromised by the overload. Varying biogas compositions were observed with methane content ranging from 49 to 67%, indicating unstable AnMBR performance.

(2) Hybrid AnMBRs

It is widely accepted that compared to traditional anaerobic digestion, conventional single-phase AnMBRs have the capacities to achieve higher removal efficiency, increased biogas production and better fuel quality of biogas due to the supplementary merits of incorporated membrane separation technology. However, the single-reactor design of AnMBRs has its own inherent weaknesses. Firstly of all, the single reactor configuration could easily fail to provide enough buffer capacity to neutralise any eventual VFA accumulations and loading shocks (Ke et al., 2005). Secondly, it rarely performed successfully to accommodate great fluctuations of wastewater composition (Saddoud et al., 2009) and dilute the toxic compounds below the toxic level. VFA accumulation, methanogenic inhibition, high toxic level, low loading capacity, reduced removal efficiency, serious membrane fouling, low biogas production and methane content were the classic manifestations of the problems with the single phased AnMBRs. Instability or failure of single-phase methanogenic AnMBRs has been reported several times, when they were used under high loading conditions. More and more researchers have begun to explore the possibility of innovating the hybrid AnMBR systems with the aim to promote the process stability, and enhance effluent quality, bioenergy production, and treatment capacity (Trzcinski and Stuckey, 2009).

Two-phase AnMBRs (TPAnMBRs) are classic examples of the hybrid AnMBRs. The idea of developing two-phase anaerobic digestion originates from the view that there are two different major sets of microbial activities involved in the whole anaerobic digestion process, namely acidogenic and methanogenic activities (Saddoud et al., 2007b). Each anaerobic microbial species have their own optimum microbiological environments, which are distinctive to the others (Ke et al., 2005). For example, both hydrolysis and acidogenic bacteria need the pH range from 5 to 6 to perform efficiently, while methanogenic bacteria require strictly the optimal pH between 6.5 and 7.8 (Zhao, 2011). In theory, the optimum pH in the reactor for methane production should be in the range from 6.5 to 8, which suits acetogens and methanogens (Antonopoulou et al., 2008). Therefore, two stages should be separated and operated at different pH values so that acidogenesis and methanogenesis processes proceed in their optimal conditions (Ke et al., 2005). Moreover, due to the differences in the kinetic rates of acidogenesis and methanogenesis, acidogenic species are found to grow relatively faster and are less sensitive to pH variations in comparison with methanogenic bacteria (Saddoud and Sayadi, 2007). The production of acids during acidogenesis can reduce the pH of the bioreactor and even cause acid inhibition in the single reactor. The slow-growing methanogenic bacteria, which normally grow in a strictly defined pH range, would be adversely affected in this case. Consequently, the single reactor system could be easily compromised, leading to the depression of the methane yield and the stability, and subsequent low treatment efficiency (Chelliapan et al., 2006). Unfortunately, this issue has been largely neglected or misunderstood by most AnMBRs researchers. As a result, the majority configurations of existing AnMBRs are inefficient single-stage reactor designs. Optimization of the environmental conditions for the different anaerobic process, i.e. hydrolysis and acidogenesis in the first phase and acetogenesis and methanogenesis in the second phase has made phased AnMBRs a promising option in the future. Each reactor can be operated at its optimal condition, and therefore it could improve treatment efficiency, energy production and process stability of anaerobic systems (Ke et al., 2005; Saddoud and Sayadi, 2007).

Saddoud and Sayadi (2007) reported the process failure of a traditional single-phase AnMBR when the system was exposed to the high OLR at 16.32 kg COD/m^3·d. They believed that VFA accumulation was the main cause resulting in the significantly declined AnMBR performance. To fix the problem associated with VFA inhibition, they developed an innovation two-phase anaerobic digestion (TPAD) system combining anaerobic fixed bed reactor for optimized acidogenesis with the AnMBR for methanogenesis step. The rational of using the fixed bed reactor refered to the recognition of its simplicity, process stabilities at higher loading rates and safety of process operation. The researcher reported that at high OLR, the integrated system showed a significantly improved AnMBR performance. VFA concentration in the AnMBR system was reduced to an average of 262 mg/L and averaged COD reduction of 98.75% was reported at OLR of 12.7 kg COD/m^3·d. In terms of biogas production, the hybrid system demonstrated an excellent biogas conversion rate as compared with the conventional single stage AnMBR they operated before. With the increase of the OLR, biogas production gradually increased to the maximum value of 215 L/d and the methane yield raised to a maximum average value of 0.31 L CH_4/g $COD_{removed}$. In addition, such a system was highlighted by its total removal of all tested pathogens, and so the microbiological quality of treated wastewater was largely conform to the microbial WHO standards. The results proved that this hybrid AnMBR system had better performance than that of corresponding single-phase AnMBR system, and phased AnMBRs were capable of dealing with high loading conditions, loading shock and acid inhibition.

Kim et al. (2011) applied a laboratory-scale hybrid staged anaerobic fluidized membrane bioreactor (SAF-MBR) system for treating synthetic wastewater (COD = 513 mg/L) at HRT of 5.0 h at 35 °C. The main purpose of the study was to reduce the energy costs for membrane fouling control. The AnMBR system consisted of an anaerobic fluidized bed reactor (AFBR) and an anaerobic fluidized bed membrane bioreactor (AFMBR) used for effluent polishing. Both reactors were filled with fluidized granular activated carbon (GAC). The researchers reported that the treatment efficiency was excellent with COD removal of 99% and TSS reduction of 100%. No membrane fouling control was needed except the scouring action of GAC, and this contributed significantly to the energy savings for fouling mitigation. The potential energy efficiency of the combined AFBR/AFMBR was apparent. Total system energy consumption for the combined two reactors system was 0.058 kWh/m^3, which was much lower than the reported 0.25 - 1.0 kWh/m^3 energy required in other AnMBRs where gas sparging was adopted to prevent membrane fouling (Liao et al., 2006). Moreover, the authors highlighted that the overall energy expenditure could be fully satisfied by using only 30% gaseous methane energy produced. Yoo et al. (2012) studied a similar SAF-MBR system for treating real municipal wastewater at HRT of 2.3 h at 25 °C. The lab study demonstrated the excellent potential of the SAF-MBR as a much less energy intensive, but cost-effective system for domestic wastewater treatment. The system had strong capabilities of generating high quality effluent (84% COD removal, 92% BOD removal) with low energy consumption and low sludge production rate. Therefore, the energy and biosoilds disposal costs were significantly reduced. Membrane fouling control was successfully implemented through the scouring effect of fluidized GAC particles. The renewable bioenergy by using the methane produced was considered more than sufficient to cover the electrical energy required for the SAF-MBR system (0.047 kWh/m^3), indicating such a system was a net energy producer.

Saddoud et al. (2007b) proposed a two stage AnMBR to treat cheese whey. The anaerobic system consisted of a completely stirred reactor for hydrolysis and acidogenesis and a continuously stirred reactor coupled to a membrane module for methane conversion. The methanogenic reactor was operated under applied OLR up to 19.78 kg COD/m^3·d, corresponding to HRT of 4 days. The experiment showed an excellent treatment performance when treating the high loading cheese whey wastewater, with the removal efficiencies of the COD, BOD and TSS at 98.5%, 99% and 100%, respectively. With regards to the methane yield, an average of 0.3 L CH_4/g $COD_{removed}$ was reported and the methane productivity appeared to increase with the increasing organic loading rate. The daily biogas yield was considered substantial, exceeding 10 times the volume of the reactor. The methane content was reported more than 70%, indicating a good performance of this hybrid AnMBR. The results implied that the hybrid AnMBR was capable of dealing with high loading protein wastewater and efficiently transforming organic wastes into renewable bioenergy. Santos et al. (2017) reported that the two-stage AnMBR was biologically stable and effective in vinasse treatment and such system demonstrated a high capacity of organic removal and biogas production regardless of reductions in HRT.

Liao et al. (2006) pointed out the location of membrane separation unit within a two-phase anaerobic system could exert a significant influence on treatment performance of AnMBRs. Yushina and Hasegawa (1994) reported 78% COD removal efficiency when using a membrane after the methanogenic reactor, compared to 92% when a membrane was placed after the acidogenic reactor. Therefore, they supported the use of a membrane coupled acidgenic reactor followed by a methane-phase reactor for wastewaters containing organic suspended solids. However, slightly higher concentration of the effluent TSS with 4 mg/L was reported with a membrane fixed after the acid-phase reactor while 0 mg/L was measured with the membrane after the methanogenic stage. Additionally, there were many cases of hybrid AnMBRs coupled with other processes for the purpose of nitrogen removal, enhancement of membrane filtration, flux reduction control and fouling mitigation. Rezania et al. (2006) studied a hybrid AnMBR system for the production of reusable water from municipal final effluent containing nitrate. The hybrid system was composed of a SAnMBR and an innovative hydrogen delivery system for hydrogenotrophic denitrification. The results showed that the novel process was efficient for effective hydrogen delivery and nitrate removal. Nitrate was reduced below the detectable levels due to the complete hydrogen transfer, and no accumulation of nitrite was observed during the experiment. The authors reported that the quality of treated effluent fitted largely with drinking water guidelines in terms of nitrate, hardness, turbidity and total coliforms. However, color and organic carbon concentration failed to meet the US EPA guidelines for potable water, and hence post-treatment was required when the effluent was intended for water reuse.

Xu et al. (2011) applied a novel hybrid AnMBR coupled with an online ultrasonic equipment (US-AnMBR) for the long-term digestion of waste activated sludge. The results showed that the introduction of ultrasound application to the AnMBR systems could effectively control membrane fouling and successfully facilitate fouling mitigation when treating high mixed liquor suspended solids (MLSS) wastes. The reasons mainly refer to the positive effects of ultrasound on the enhancement of membrane filtration, sludge properties and methanogenic activities.

2.2.4 Membrane fouling and control

Membrane fouling represents the most critical impediment for the more generalized AnMBR applications in wastewater treatment because it negatively affects system operation in spite of the selection of process designs. The adverse effects of fouling in AnMBRs are very well documented. First of all, fouling is reported to reduce membrane flux and increase effective membrane area per reactor volume, which obviously leads to more capital costs. Secondly, membrane fouling reduces permeate flux and system productivity, and increases system downtime. Thirdly, more operational energy consumption is required to increase gas scouring or sludge recirculation to reduce fouling propensity. Last but not least, frequent physical and chemical cleaning for fouling control can reduce the lifespan of membrane and increase membrane replacement costs (Guo et al., 2012; Stuckey et al., 2012; Lin et al., 2013).

(1) Mechanisms of membrane fouling

In general, the mechanisms of membrane fouling in AnMBRs are similar to those in aerobic MBRs, mainly resulting from the interactions between the membrane material and the components of sludge suspension (Lin et al., 2013). Nevertheless, anaerobic processes are faced with other unique challenges. Fouling in AnMBRs is much more intense than that in aerobic MBRs, because AnMBRs are usually experienced with lower sludge filterabilities (Skouteris et al., 2012). Membrane fouling in AnMBRs is composite fouling, and it is deemed to be a much more complex phenomenon and caused by a combination of different mechanisms such as pore clogging, cake formation, and cake layer consolidation (Guo et al., 2012; Lin et al., 2013). Liao et al. (2006) summarized mechanisms of membrane fouling into three major categories (Figure 2.2) namely biofouling, organic fouling and inorganic fouling.

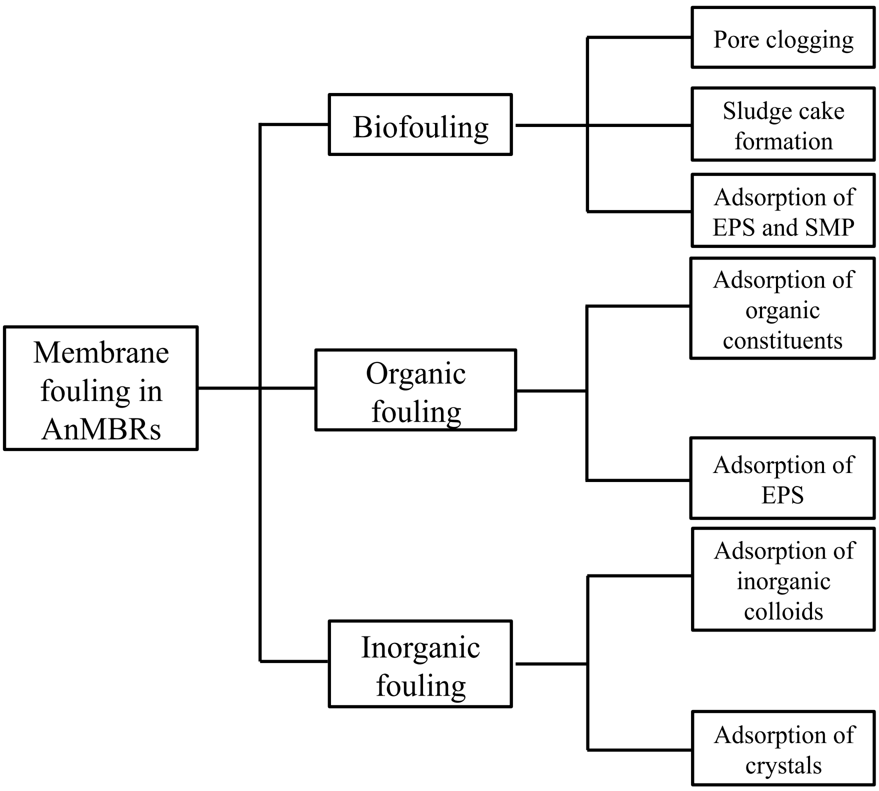


Figure 2.2 A typical classification of membrane fouling in AnMBRs

They suggested that all three mechanisms usually occurred simultaneously. Biofouling is defined as the membrane fouling resulted from interaction between membrane surfaces and constituents of the biological treatment broth. The definition of organic fouling refers to fouling associated with the accumulation and adsorption of organic components on the membrane surfaces. As effluent COD concentrations are relatively higher in AnMBR systems as compared with those in aerobic MBR systems, organic fouling is much more serious in AnMBR processes. Inorganic fouling is due to the accumulation and adsorption of inorganic colloids and crystals onto membrane and pore surface. Anaerobic MBRs are believed to be more prone to inorganic fouling than aerobic MBRs. This is partly due to the fact that anaerobic digestion tends to have relatively higher concentration of carbonate and biocarbonate, and produce higher concentration of ammonia and phosphate concentrations (You et al., 2005; Liao et al., 2006; Guo et al., 2012; Lin et al., 2013).

(2) Factors affecting AnMBR fouling

Membrane fouling in AnMBRs is originated from the interaction between sludge suspension and membrane surface and pores. In this regard, parameters that can exert influences on membrane and sludge suspension would affect AnMBR fouling propensity either in a microcosmic or in a macrocosmic manner. These parameters can be generally classified into four groups: feed water characteristics (influent water chemistry, influent variability and toxic shock), membrane properties (pore size, surface morphology and hydrophobicity), sludge properties (MLSS, SMP and EPS) and operating conditions (HRT, SRT and temperature). Parameters such as SMP, EPS, shear force and gas sparging can exert direct impacts on membrane fouling, and therefore they are regarded as the principal parameters affecting membrane fouling. On the other hand, the others including HRT, SRT and OLR indirectly influence membrane fouling through altering the broth characteristics and biomass properties (Guo et al., 2012; Stuckey, 2012; Lin et al., 2013). Table 2.1 summarizes the effects of some major parameters on membrane fouling based on the most recent researches relating to AnMBR applications.

- **Sludge properties**

Sludge properties such as MLSS, SMP and EPS tend to significantly affect membrane fouling in AnMBRs in a microcosmic manner. These parameters are also closely linked to each other. For example, concentrations of both protein and carbohydrate fractions of EPS and SMP increase when the MLSS concentration increases (Yigit et al., 2008). MLSS refers to the total mass suspended solids, which mainly consist of microorganisms and non-biodegradable suspended matter. It is a vital parameter to maintain the sufficient amount of active biomass available to consume the applied quantity of organic pollutant in the anaerobic digestion. Rosenberger et al. (2005) pointed out in their case studies that the effects of varying MLSS on fouling were different depending on the applied MLSS concentrations in the operation. For example, the fouling potential could be reduced with an increase in MLSS when at very low MLSS concentrations (< 6 g/L), while severe fouling would be observed as the result of increased MLSS when at very high MLSS concentrations (> 15 g/L). Surprisingly, there was no impact on membrane fouling at medium MLSS concentrations (8 - 12 g/L). Ho and Sung (2009a) studied the effects of solid content on flux decline of an AnMBR treating diluted anaerobic sludge.

Lin et al. (2013) interpreted the flux and fouling tests results gained by Ho and Sung (2009a), and stated that initial flux and stabilized flux decreased dramatically with the increase in MLSS. The optimal MLSS for the AnMBR operation in terms of stabilized flux and fouling mitigation were supposed to be approximately 15 - 18 g/L. Baek et al. (2010) launched a lab-scale study on an AnMBR for dilute municipal wastewater treatment under different MLSS concentrations. In the study, different levels of MLSS concentrations were maintained by altering HRTs and SRTs. The results showed that deposition rates of the suspended solids on the membrane surface and EPS content in the MLSS generally decreased as the result of decreasing MLSS. This meant that lowering the level of MLSS concentration in anaerobic reactors could reduce the level of accumulation of the suspended solids on the membrane surface, and hence mitigate fouling in AnMBRs. Stuckey and Hu (2003) initiated a food industry wastewater treatment process by using a SAnMBR. As an indicator of membrane fouling, TMP was used in this case to evaluate the level of membrane fouling. They found that under constant flux condition, TMP rose incrementally with the increase in MLSS to compensate for the fouling. Therefore, it could be concluded that MLSS had a positive correlation with membrane fouling rate in AnMBRs.

EPS and SMP are the two important sludge characteristic parameters, which were heavily researched recently in relation to fouling propensity in AnMBRs. EPS refer to polymeric material bound to cell surface, and are the construction materials for microbial aggregates including biofilm and activated sludge flocs. EPS can be subdivided into bound EPS and soluble EPS, and they are extracted by using different chemical and physical methods. Meng and Yang (2007) and Khor et al. (2007) mentioned in their studies that total EPS content positively coincided to the membrane fouling rate.

Gao et al. (2010a) emphasized in the fouling studies that EPS were predominately proteinaceous, and EPS and microbial cells of the foulant layer were the key contributors to the membrane fouling. They also investigated the bound EPS proteins (EPS_P) and polysaccharides (EPS_C) and EPS_P/EPS_C ratio in EPS, and found that EPS_P/EPS_C ratio was negatively correlated to the membrane fouling rate. This correlation could be explained by other researchers' findings that the decline in the ratio could contribute to poorer bioflocculation and result in reduced flocs' size causing serious fouling (Liao et al., 2001; Masse et al., 2006). Therefore, EPS_P/EPS_C ratio is deemed to be a vital parameter, which governs the size of flocs in the SAnMBR system, and hence influences the membrane permeability and fouling. By contrast, Lin et al. (2009) argued that the higher EPS_P/EPS_C ratio of bulk sludge in the thermophilic SAnMBR would increase fouling propensity in the system. This was because sludge with higher value of EPS_P/EPS_C ratio in bound EPS seemed to have higher stickiness, hence favouring the development of cake formation. It is also worth mentioning that the increase in EPS correlates the increase of cake resistance during the treatment of acidified wastewater (Jeong et al., 2010a). However, Lee et al. (2003)'s research supported the view that it is EPS_P/EPS_C ratio rather than the quantity of total EPS that could significantly affect the fouling resistance.

The definition of SMP is regarded as the pool of microbial products that are released into the bulk solution from the cell lysis, the hydrolysis of EPS and the interaction of the microorganism with its surroundings (Guo et al., 2012; Skouteris et al., 2012; Lin et al., 2013). SMP generally are identified as humic and fulvic acids, polysaccharides, proteins, structural components of cells and products of energy metabolism (Barker and Stuckey, 1999). Generally, anaerobic SMP were believed to be more complex than the aerobic ones, and would lead to much severe membrane fouling. For example, Martin-Martin-Garcia et al. (2011) conducted a comparable study between aerobic MBRs and anaerobic MBRs. They found that SMP in the flocculated AnMBR supernatant were 500% higher than those in aerobic MBR supernatant, and therefore more attention should be paid to fouling control in AnMBRs. Santos et al. (2017) observed that SMP had the highest concentrations as the biopolymer causing fouling, and the protein fraction was the most representative in SMP. Chen et al. (2017c) attributed the serious fouling in the AnMBR to SMP-induced pore blockages and EPS-induced fast-growth and compact cake layer on the membrane surface.

Akram and Stuckey (2008) conducted three parallel SAnMBR studies in treating a sucrose-meat extract based medium strength wastewater, and they attributed the low flux to the high quantities of SMP. Aquino et al. (2006) characterized dissolved compounds in the AnMBRs responsible for membrane fouling, and they found that soluble high molecule weight protein and carbohydrate-like SMP made great contributions to greater internal membrane fouling. Lin et al. (2009) studied sludge properties and their effects on membrane fouling in SAnMBRs treating kraft evaporator condensate. They concluded that biomass properties such as SMP, floc size, biopolymer clusters (BPC) and bound EPS played an important role in governing sludge cake formation and membrane fouling in SAnMBR systems. Apart from that, they also mentioned that SMP were positively correlated to filtration resistance and demonstrated considerable influence on membrane fouling. Other parameters that were reported to influence membrane fouling were PSD, sludge hydrophobicity or sludge viscosity (Guo et al., 2012; Lin et al., 2013).

- **Temperature and pH**

Both Temperature and pH significantly affect membrane fouling behavior in AnMBRs in an indirect and direct manner, through having profound impacts on microbial metabolism, sludge properties (SMP, EPS and PSD) and membrane characteristics such as surface chemistry (Lin et al., 2009; Gao et al., 2010a; Lin et al., 2013). In terms of temperature's effects on membrane fouling, many researches have reported that thermophilic AnMBRs were more likely to experience serious membrane fouling. Lin et al. (2009) compared two identical SAnMBRs (thermophilic vs. mesophilic) under similar hydrodynamic conditions to investigate temperature's effects on membrane fouling. The thermophilic SAnMBR was reported to experience a filtration resistance of about 5-10 times higher than that of the mesophilic AnMBR. The analysis of sludge properties also revealed that a higher temperature could induce the production of more SMP and BPC, more EPS release, a larger portion of fine flocs (<15μm), and increased EPS_P/EPS_C ratio in bound EPS, which would lead to the significant increase of filtration resistance in the thermophilic SAnMBR. Additionally, the evidences of higher amount of both organic and inorganic foulants, smaller particle sizes, and in particular a denser, more compact and less porous sludge cake structure were found in the cake layer formed in the thermophilic SAnMBR, which further proved severe membrane fouling in the thermophilic condition.

He et al. (2005) suggested that AnMBRs should be operated within mesophilic range approximately at 37 °C since the maximum operating temperature for most organic membranes was 45 °C. The study of an anaerobic membrane bioreactor in treating high concentration food wastewater treatment indicated that the AnMBR experienced a significant increase in the permeate flux by 44.3% when the temperature was increased from 33 °C to 39 °C. Therefore, they believed that high temperature was beneficial to AnMBR operation in terms of the membrane flux. This conclusion was supported by the theory that operation at higher temperatures could result in the accelerated decomposition of organic pollutants, thus reducing the viscosity of anaerobic mixed liquid and increasing membrane filtration flux. Trzcinski and Stuckey (2010) proposed that too low temperatures were closely linked with higher bulk SCOD, which led to the serious flux decline. But this effect was reported partly reversible once the SCOD was degraded. Similarly, Martinez-Sosa et al. (2010) and Martinez-Sosa et al. (2011) concluded in their studies that an increase in the fouling rate was associated with an accumulation of TSS, a higher viscosity, the decreased particle size of the anaerobic sludge, and increased bulk SCOD, when the temperature was reduced from 35 °C to 20 °C. They believed that this was because that the biological reactions proceeded much slower at lower temperatures and the degradation of soluble compounds was therefore usually limited under the psychrophilc conditions. Berube et al. (2006)' review also pointed out that a higher content of SMP might be presented in the mixed liquor when AnMBRs were operated at relatively low operating temperatures, and this could lead to a reduced membrane permeate fluxes.

pH is regarded as a vital parameter which strongly impacts membrane fouling in AnMBRs. On the one hand, a change in pH value can induce noticeable effects on the sludge properties such as PSD, SMP, EPS, and MLSS in the anaerobic reactor, and thus exert profound influences on membrane fouling in AnMBRs. On the other hand, pH shocks are likely to affect membrane fouling directly by the way of varying membrane morphology, surface charge, pore structure and surface chemistry of the membranes. Therefore, the changes in membrane fouling propensity were the result of a combined effect of pH variation on sludge and cake properties and membrane function. (Gao et al., 2010a; Nanda, 2010; Stuckey, 2012).

Gao et al. (2010a) investigated the impact of elevated pH shocks (pH 8.0, 9.1 and 10.0) on the anaerobic sludge properties and membrane fouling behavior in a SAnMBR system. A slight rise in fouling rate after a pH 8.0 shock, and a significant increase of fouling rate after pH 9.1 and 10.0 shocks were observed in the study. The severe membrane fouling in the SAnMBR system was closely correlated to the dispersion of sludge flocs and smaller size particles induced by the elevated pH shocks. Furthermore, the accumulation of colloids and solutes or biopolymers in the sludge suspension was discovered after pH shock, and these substances exerted a significant influence on membrane fouling resistances. They also reported that the high supernatant COD and BPC after pH 10.0 shock, were responsible for the facilitation of colloidal or smaller size particle deposition on the membrane surface. This could give rise to a production of a denser and more compact cake layer with higher specific resistances, resulting in higher membrane fouling rates. Lin et al. (2013) also reported that under similar hydrodynamic conditions, extreme pH could promote the extent of reduced PSD of sludge liquor, which would in turn adversely influence membrane fouling.

- **SRT and HRT**

As controllable bioreactor operating parameters, HRT and SRT are regarded as two major factors affecting treatment performance and biomass properties. Therefore, they would inevitably influence membrane fouling in AnMBRs in a macroscopic manner (Hanndel and Lettinga, 1994; Liao et al., 2006). Too high or too low values of SRTs and HRTs are generally considered inappropriate for AnMBR applications, and this is because the extreme values can inevitably result in the deterioration of membrane fouling in AnMBRs (Guo et al., 2012).

Huang et al. (2008) observed a severe internal irreversible membrane fouling when they increased applied SRTs of 30 to 60 days when treating synthetic low-strength wastewater. They reported that prolonging SRTs could induce higher concentrations of non-flocculation flocs. Due to the breakage of sludge flocs, cell lysis and microbial metabolism, the generated fine particles would increase membrane fouling propensity by easily depositing on the membrane surface or in membrane internal pore, resulting in deteriorated internal pore blocking. Huang et al. (2011) further investigated the mechanism of membrane fouling in terms of effects on sludge properties such as MLSS, SMP and EPS at different HRTs and SRTs. They reported that elongated SRTs tended to lower metabolism rates of microorganisms. The weak sludge activity could give rise to higher MLSS, higher production of SMP, higher carbohydrate and protein concentrations in SMP, thus accelerating particle deposition and biocake development. Moreover, lower EPS at prolonged SRTs could enable less flocculation of particulates and subsequent finer particle sizes, further exacerbating membrane fouling in SAnMBRs. With regard to HRT, Huang et al. (2011) summarized that with the reduction of HRT, an increase in OLR could enhance biomass concentration, increase SMP and SMP_P/SMP_C ratio in the SMP composition (proteins have higher hydrophobicity and tend to adhere membrane surface with ease, thus more easily contributing to membrane fouling.

Similarly, Jeong et al. (2010a) and Salazar-Peláez et al. (2011a) pointed out that shortened HRTs could enhance the production of SMP and release of EPS, causing higher cake resistance and fouling potential in SAnMBRs. Additionally, Salazar-Peláez et al. (2011a) reported that particle fragmentation and the washout of smaller sized particles occurred partly because of the higher shear forces and higher upflow velocities occurring inside the reactor at lower HRTs. Decreased PSD could pose a threat to the membrane performance, and particularly, finer particles would lead to a severe deposition of solids on the membrane surface. Continuing deposition of such small solids could eventually result in greater membrane resistances against flux. Furthermore, fouling cake layer would be denser and more compact, and specific fouling cake resistance would be much higher (Choo and lee, 1998).

Table 2.1 Effects of major parameters on membrane fouling in AnMBRs

Fouling factors	Effects on AnMBR fouling	Reference
MLSS	(1) Initial flux and stabilized flux decreased dramatically when MLSS increased. The optimal MLSS for the AnMBR operation were approximately 15 - 18 g/L for optimized flux and fouling mitigation.	Ho and Sung (2009a)
	(2) Low MLSS concentration reduced the suspended solids accumulation on the membrane surface, thereby mitigating fouling in AnMBRs.	Baek et al. (2010)
	(3) Under constant flux condition, TMP increased incrementally with the increase in MLSS to compensate for the fouling, and MLSS had a positive correlation with membrane fouling rate in AnMBRs.	Stuckey and Hu (2003)
EPS	(1) EPS was predominately proteinaceous. EPS and microbial cells of the foulant layer mainly contributed to membrane fouling. EPS_P/EPS_C negatively correlated with membrane fouling rate.	Gao et al. (2010a)
	(2) The decreased EPS_P/EPS_C ratio could result in poorer bioflocculation and reduced flocs' size, causing severe fouling propensity.	Liao et al. (2001); Masse et al. (2006)
	(3) Sludge with higher EPS_P/EPS_C ratio had higher level of stickiness, hence favoring cake formation.	Lin et al. (2009)
	(4) The increase in EPS concentration correlated with the increase of cake resistance.	Jeong et al. (2010a)
	(5) EPS_P/EPS_C ratio other than total EPS significantly affected the fouling resistance.	Lee et al. (2003)
	(6) Sludge properties including bound EPS, SMP, floc size and BPC governed the sludge cake formation and membrane fouling in SAnMBR systems.	Lin et al. (2009)
SMP	(1) Anaerobic SMP were more complex than the aerobic ones, causing much worse membrane fouling in MBR operations.	Martin-Garcia et al. (2011)
	(2) The large quantities of SMP were responsible for the internal fouling and the decline in flux.	Akram and Stuckey (2008)
	(3) Soluble high molecule weight protein and carbohydrate-like SMP make great contributions to serious internal membrane fouling.	Aquino et al. (2006)

	(4) SMP positively correlated with filtration resistance and exerted considerable influences on membrane fouling.	Lin et al. (2009)
Temperature	(1) Increase in temperature induced more SMP, BPC and EPS release, increased EPS_P/EPS_C ratio and a larger portion of fine flocs (< 15μm).	Lin et al. (2009)
	(2) Higher temperature accelerated biodegradation of organics, reduced the viscosity of anaerobic mixed liquid, hence increasing the membrane filtration flux.	He et al. (2005)
	(3) Too low temperature resulted in higher bulk SCOD, and subsequent flux decline.	Trzcinski and Stuckey (2010)
	(4) Relatively low operating temperatures lead to a higher SMP in the mixed liquor, and reduced membrane permeate flux.	Berube et al. (2006)
pH	(1) Extreme pH promoted the reduced PSD of sludge liquor, exacerbating fouling.	Lin et al. (2013)
	(2) Elevated pH shocks induced sludge flocs dispersion, caused the accumulation of colloids and solutes or biopolymers in the sludge suspension, thus resulting in serious membrane fouling.	Gao et al. (2010a)
SRT	(1) Too long SRT induced higher concentration of non-flocculated flocs, thereby accelerating fouling.	Huang et al. (2008)
	(2) Elongated SRT lead to lower metabolism rates of microorganisms, higher MLSS and SMP, and therefore resulted in more particle deposition and cake formation. Lower EPS at prolonged SRTs reduced flocculation of particulates and promoted finer particle sizes, further exacerbating membrane fouling.	Huang et al. (2011)
HRT	(1) Reduced HRT enhanced MLSS concentration and SMP and EPS production, and increased SMP_P/SMP_C ratio, thus worsening fouling propensity.	Jeong et al. (2010a); Huang et al. (2011)
	(2) The higher shear forces at lower HRT promoted particle fragmentation and the washout of smaller sized particles, and therefore lead to greater membrane fouling	Salazar-Peláez et al. (2011a)

(3) AnMBR fouling control

Since membrane fouling, the major impediment for widespread applications, is governed by feed wastewater characteristics, membrane properties, sludge characteristics, and hydrodynamic and bioreactor operation conditions, different strategies have been adopted by researchers to control fouling development from these areas. Fouling control procedure could be generally proceeded in the two approaches (Figure 2.3), including reducing the rate of fouling and cleaning the fouled membrane (Liao et al., 2006).

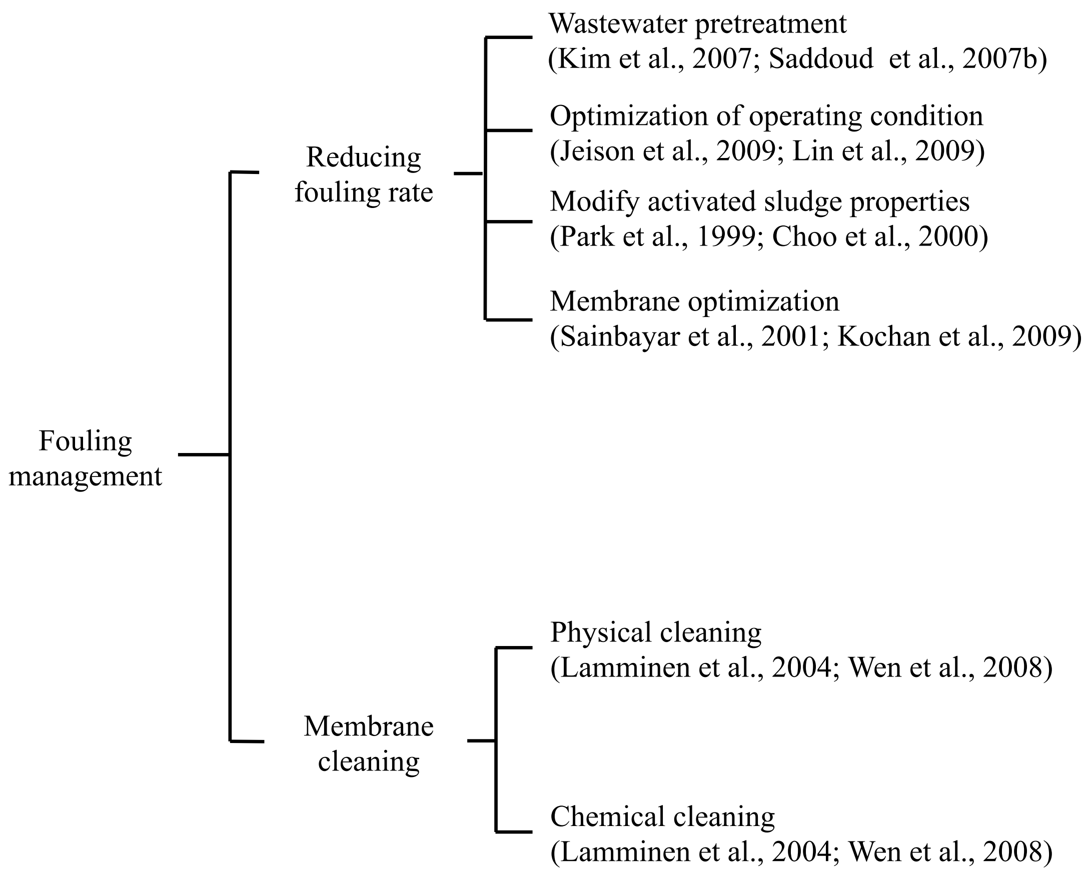


Figure 2.3 Strategies for membrane fouling mitigation

Since membrane fouling cannot be avoided and membrane cleaning should be adopted. However, frequent cleaning, particularly chemical cleaning, could reduce the lifespan of membrane, and therefore lead to costly membrane replacement. In this case, reducing the rate of fouling would elongate the length of time between cleanings and contribute to stable operation and economic efficiency. In particular, more attention should be paid attention to the following aspects when developing membrane fouling management protocol: avoidance of toxic shocks and pH shocks, maintenance of healthy biological system, careful selection of the applied SRT and HRT, good control of reactor temperatures at mesophilic condition (above 20 °C), and careful selection of membrane material.

2.3 Biogas production from AnMBRs

The slow growing nature of methanogenic organisms and microbial complexity in the systems have made the operation of biogas fermenters difficult. The success of efficient biogas production depends on the effective retention of methanogenic bacteria in the reactor through decoupling of SRT and HRT (Dereli et al., 2012). Research has mostly focused on retaining a high density of functioning anaerobic microorganisms, in order to achieve efficient biogas production. The most recent development in biogas production is the incorporation of anaerobic bioprocesses with membrane separation techniques in a membrane bioreactor (MBR), the purpose being to increase biomass concentration extensively in the bioreactor. In an AnMBR, high cell concentrations can be sustained under reasonably high hydraulic load and sufficient mixing due to completely decoupling HRT from SRT (Wang et al., 2012).

Moreover, due to the sufficient retention of active microorganisms, AnMBRs generally have high product concentration and productivity and relatively good toxic resistance, and simplify the separation of product and/or biomass by using micro-filtration or ultra-filtration, thus leading to an improved biogas production economy (Ylitervo et al., 2013). The reported biogas production was well documented with the methane yield up to 0.36 L CH_4/g $COD_{removed}$ (0.30 L CH_4 (STP)/g $COD_{removed}$, the volume of methane produced at 0° C Standard Temperature and 1 atm Standard Pressure) and the high methane content up to 90% (Liao et al., 2010). However, the optimization of biogas production from AnMBRs has not gained much attention due to the as yet under-developed nature of AnMBRs (Visvanathan and Abeynayaka, 2012). For extreme conditions, such as high salinity, thermophilic temperature, high OLR and presence of toxicity, membrane assisted anaerobic processes can be hampered and biogas productivity can be compromised.

Ylitervo et al. (2013) provided a general review of the MBR technology in ethanol and biogas processes and summarized the development of MBRs and the membrane technologies for these biofuels. Wang et al. (2012) reviewed the progress in biogas technology in China and briefly introduced AnMBRs as one of the emerging technologies. Mao et al. (2015) discussed advances in biogas production from anaerobic digestion in recent years and provided brief information on AnMBRs for biogas production. He et al. (2012) summarized the recent performance of AnMBRs for methane and hydrogen production. Minardi et al. (2015) reviewed membrane applications for biogas production and purification processes. The following sections provided a comprehensive overview of such advances in various AnMBRs, in view of both traditional and advanced reactor configurations for biogas production. Moreover, with the focus on inhibitors and operational dilemmas, a detailed assessment on the potential challenges that AnMBRs were facing was included.

2.3.1 Types of AnMBRs for biogas production

2.3.1.1 Conventional AnMBRs

- **CSTR-AnMBRs**

CSTR is by far the most frequently researched anaerobic process in AnMBR systems for biogas production due to the ease of construction and operation. In most cases, CSTRs are in cylindrical or rectangular shapes and employ mechanical turbines for mixing. Side-stream membranes are often used, resulting in high bioreactor liquid turnover rates and a well-mixed hydraulic flow regime. The potential for biological conversion from substrate to methane can be greatly increased due to the prevailing high shear stress and intensive mixing (Liao et al., 2006; Ozgun et al., 2013). Lab- and pilot-scale studies have been carried out with all three primary AnMBR configurations: external side-stream membrane (Bornare et al., 2014), submerged membrane (Lin et al., 2011), and submerged membrane with external membrane tank (Martinez-Sosa et al., 2012). In general, a CSTR coupled membrane system is able to achieve a promising methane yield up to theoretical value (Wei et al., 2014). Lin et al. (2011) found a methane yield rate of 0.26 L CH_4/g $COD_{removed}$ (0.23 L CH_4 (STP)/g $COD_{removed}$) and high content of methane up to 85% when operating a pilot-scale SAnMBR. The compact configuration of such module also allowed more convenient biogas collection. The research on fermentative H_2 production is also typically conducted in CSTR-AnMBRs. Having the largest amount of energy per mass unit than any other known substance (142 kJ/g), H_2 is an ideal energy carrier free from harmful emissions during utilization as it is combusted to form only water. As the hydrogen production stage occurs briefly prior to the methanogenic process, biohydrogen production can be realized by inhibiting the methanogenesis phase using various intervention means. These include the manipulation of hydrogen partial pressure, pH control, chemical inhibition, and promotion of ferric-reducing conditions (the addition of $FeSO_4$ solution) (Ylitervo et al., 2013).

Previously, most research has been carried out in an external cross-flow type, but the immersed type has become much more popular recently, due to requiring less energy consumption and less need for fouling mitigation (Jung et al., 2011). The carbon source conversion efficiencies in the anaerobic hydrogen producing membrane bioreactor are promising with two cases reaching 100% (Lee et al., 2009; Shen et al., 2009). $FeSO_4$ concentration, in particular, was seen as a crucial factor impacting on the dark fermentation pathway for H_2 production from AnMBRs (Lee et al., 2009). Hydrogen productivity from the AnMBRs ranged from 2.5 (Lee et al., 2008) to 66 L/L·d (Lee et al., 2007), and the hydrogen content of biogas could reach as much as 62.6% (Lee et al., 2009).

However, to ensure a well-mixed flow regime and sufficient mass transfer, rigorous mechanical mixing is required. Moreover, the disruption of particles as a result of sludge recirculation through the membrane feed pump can have a negatively impact on the orientation between acetogens and sensitive methanogens, thus limiting the essential hydrogen transport for acquiring a superior specific methanogenic activity (SMA) (Brockmann and Seyfried, 1997; Ozgun et al., 2013). The impaired syntrophism often leads to a higher concentration of VFA in the system and VFA inhibition is more severe particularly in the thermophilic system. Additionally, CSTR-AnMBR usually operates at a lower biomass concentration (e.g. 5 g/L MLSS) compared to other high rate anaerobic reactors due to fouling control issues, which results in a lower OLR applied to the system, limiting the biomethane potential from high loading wastewater.

- **UASB-AnMBRs**

The UASB concept was developed by Lettinga et al. in the 1970s for methane production. The secret of such a novel high rate reactor design lies in its ability to: firstly, retain a high concentration of biomass in the form of well settlable methanogenic sludge granules in a thick dense sludge bed at the bottom of longitudinal reactor; and secondly, capture produced biogas through a gas/liquid/solid (GLS) separator at the top. Many researchers have attempted to combine UASB with membrane to optimize the joint benefits such as enhanced methane production and less fouling problems (van Voorthuizen et al., 2008; Martin-Garcia et al., 2013). Xie et al. (2010) have investigated the feasibility of a submerged UASB-AnMBR system for recovering energy from kraft evaporator condensate at 36 °C to 38 °C for 9 months. The methane production rate of 0.35 ± 0.05 L CH_4/g $COD_{removed}$ (0.31 ± 0.05 L CH_4 (STP)/g $COD_{removed}$), which were very close to the theoretical yield of methane with 0.397 L CH_4/g $COD_{removed}$ at 37 °C (0.350 L CH_4 (STP)/g $COD_{removed}$), and the methane content in the biogas, reached 90% in this study. Lin et al. (2009) also reported a similar methane production rate and excellent fuel quality with 80 - 90% methane for both mesophilic and thermophilic SAnMBRs from kraft evaporator condensate treatment.

To assist with effective biomass retention, a hybrid UASB-AnMBR, in which fine fibers were placed at the top of the sludge zone as a biofilter, served to treat real domestic wastewater for biogas production at ambient temperature in Beijing, China (Wen et al., 1999). Due to the sufficient retention of biomass by the membrane (MLSS maintained as high as 21.5 g/L), this integrated G-AnMBR system achieved maximum biogas production of 0.42 L/L·d, and methane gas content of 66%. UASB reactors for methane fermentation from low strength wastewater at low and moderate temperatures are often encountered by a poor mixing regime, which undermines biogas productivity due to a decrease in soluble COD treatment efficiency. The granulation process for UASB using non-granular seeding is also very lengthy, requiring at the least a 3-month start-up period for stable biogas production. Direct Membrane intervention into UASB eliminated the hydraulic selection pressure for sound granules. This can negatively impact the granular sludge properties and hence the methane yield in the long-term (Ozgun et al., 2013).

- **Expanded granular sludge bed reactor (EGSB) AnMBRs**

EGSB reactors are tall reactors characterized by a higher ratio of height to width and internal or external effluent, and they can provide a very high mixing intensity and sufficient substrate-microbes contact induced by the high upflow force. However, to date, the studies on the feasibility of the EGSB combined membrane process are limited. This is probably due to the fact that manipulating sludge bed expansion in EGSB is relatively difficult due to the absence of solid carriers under high hydraulic upflow force. The only study available was Chu et al. (2005) who reported biogas production ranging from 0.28 to 0.58 L/L·d, and 63 - 72% methane at low HRT of 3.5 h from an EGSB reactor coupled with hollow fiber membrane filtration for energy recovery from domestic wastewater at 15 °C. However, granules fragmentation and sludge washout may occur due to the applied high upflow velocity (U_v >4 m/h), which may affect methane production. In addition, no granulation is expected in EGSB-MBR, which would alter the properties of granular biomass and affect the biogas production in the long-term.

- **AFBR-AnMBRs**

AFBR is regarded as an effective anaerobic process to be coupled with membrane filtration due to its good mass transfer characteristics and retention of high concentration of active microorganisms at short HRT lasting from a minutes to a few hours. Compared to other gas-sparged AnMBRs, membrane fouling was successfully controlled through the energy efficient scouring effect of fluidized GAC on the membrane surface, resulting in fouling mitigation having significantly reduced energy costs (Shin et al., 2014). A two-stage anaerobic fluidized bed system is often required to fully reclaim methane from wastewaters. Kim et al. (2011) proposed a SAF-MBR system, which consisted of an AFBR and an AFMBR for methane rebate from municipal wastewater primary-clarifier effluent. The methane production from this system was reported as 4.11 mol CH_4/m^3 (92.1 L CH_4 (STP)/m^3) with methane composition of 86% at HRT less than 5h. Using only 30% of the gaseous methane energy produced could satisfy total fluidization energy required for the system, meaning such a SAF-MBR is a promising AnMBR for bioenergy production. Similarly, Yoo et al. (2012) and Dutta et al. (2014) also worked on the SAF-MBRs and concluded that the

SAF-MBR system has excellent potential as a low-energy input, high-efficiency, and cost-effective system using methane energy. However, dissolved methane representing 63% of the total methane production continued to represent a big issue that needs to be solved if energy production is to increase (Yoo et al., 2012).

Methane production is significantly affected by the temperature, and dissolved methane, particularly in the winter period was a severe issue identified by researchers. Gao et al. (2014a) investigated an integrated anaerobic fluidized-bed membrane bioreactor (IAFMBR) system with simplified reactor operation and much smaller footprint compared with two-stage systems. In this study the methane content in biogas had typical value of (80 ± 2%) and nearly 50% of the influent COD was converted into methane, of which 25% of produced methane was lost in the liquid phase. As a result of the restrained organic degradation capacity of the AFBR-AnMBRs, VFA accumulation and inhibition was found at shorter HRT, which reduced the specific methane productivity. Moreover, Shin et al. (2014) reported that the specific acetoclastic methanogenic activity (SAMA) on the GAC was much lower than the enriched acetoclastic cultures, indicating low levels of such organisms in the GAC VSS. Therefore, future research on biogas production optimization should consider the facilitation of attached growth of syntrophic VFA-degrading acetogens and acetoclastic methanogens on GAC. Ye et al. (2017) integrated membrane aerators into an AFMBR to form an aeration membrane fluidized bed membrane bioreactor (AeMFMBR), and such an integrated system could achieve simultaneous removal of organic matter and ammonia without production of dissolved methane when treating domestic wastewater. Membrane fouling was well controlled by GAC particles fluidization and TMP was maintained below 3 kPa.

- **Jet flow anaerobic bioreactor (JFAB) AnMBRs**

A jet flow anaerobic reactor has certain advantages when coupling with membrane filtration to form an AnMBR for methane fermentation. The liquid circulation inside such a reactor by using an inner tube and a nozzle system offers an adequate homogenization and mass transfer (Ozgun et al., 2013). Applying an UF membrane coupled to the JFAB, Saddoud et al. (2007a) reported rich methane in biogas (70%) with the average methane yield being 0.27 L CH_4/g $COD_{removed}$ (0.24 L CH_4 (STP)/g

$COD_{removed}$) from domestic wastewater treatment. However, in another study, Saddoud et al. (2009) reported the inefficient methanization of such AnMBR with the average methane yield not exceeding even 0.1 L CH_4/g $COD_{removed}$ (0.088 L CH_4 (STP)/g $COD_{removed}$). This was due to the considerable fluctuations in the substrate composition and presence of toxic substances emanating from industrial effluents. Saddoud and Sayadi (2007), therefore proposed an innovative TPAD system coupling anaerobic fixed bed reactor for optimized acidogenesis and the AnMBR for optimized methanogenesis for biogas production from slaughterhouse wastewater. In this combined process, VFA inhibition was successfully overcome, and biogas conversion was significantly improved with an average value of 0.31 L CH_4/g $COD_{removed}$ (0.27 L CH_4 (STP)/g $COD_{removed}$). Table 2.2 summarizes the key features and advantages and challenges of conventional AnMBRs for biogas production.

Table 2.2 Key features, advantages and challenges of conventional AnMBRs

Conventional AnMBRs	Key features	Advantages	Challenges
CSTR-AnMBRs	• The first generation of high rate anaerobic reactor and most employed in AnMBR studies • A cylindrical or rectangular tank • Mechanical mixing	• Good substrate-sludge contact with slight mass transfer resistance • High liquid turnover rates and well-mixed flow regime • Enhanced biomethane potential due to prevailing high shear stress and intensive mixing	• Rapid acidification and VFA inhibition due to continuous mixing and high shear stress • Negatively impacted SMA • Lower organic loading leads to lower biomethane potential
UASB-AnMBRs	• A cylindrical or rectangular column • Biomass retention in the form of granules • Sufficient mixing provided by liquid upflow force and rising biogas bubbles	• Good wastewater-biomass contact • Superior quality of granular sludge for higher biogas production • Significantly higher organic/hydraulic loading rates compared to flocculent sludge bed reactor • Moderate tolerance to toxic compounds • No mechanical mixing device required • Reduced gas sparging demand	• Long start-up period • Dead space and poor mixing at psychrophilic conditions • Elimination of hydraulic selection pressure causes granules deterioration and disintegration, and unstable biogas production
EGSB-AnMBRs	• Tall column reactors with smaller footprint and effluent recirculation • High upflow velocity (> 4m/h), very high mixing intensity, and efficient biomass-substrate contact	• Improved mass transfer in a compact design • Resolve issues with UASB such as hydraulic short cuts, preferential flows, poor mixing regime (dead zones), and temperature constraints • Effective in generating biogas from soluble pollutants sources such as	• No granulation is expected, and this may affect the granule sludge properties and hence biogas production • Granules fragmentation and sludge washout due to the high upflow velocity

		domestic wastewater, and wastewater containing lipids and toxic/inhibitory compounds	
AFBR-AnMBRs	• Granular activated carbon as the medium for bacterial attachment and growth • Tall column reactors with a smaller foot print and effluent recirculation • Two stage submerged membrane configuration is most employed • A greater surface area per unit of reactor volume	• Higher organic and hydraulic loading and greater resistance to inhibitors for biogas production • Much lower capital cost due to reduced reactor volume. • Efficient in biogas production from pharmaceuticals and suspended particles of domestic wastewater	• Two stage systems are often required for effective biogas production • Dissolved methane remains as a big issue • VFA accumulation and inhibition at shorter HRT. • Low specific acetoclastic methanogenic activity (SAMA) on GAC
JFAB-AnMBRs	• An inner tube and a nozzle system for mixing • Jet flow module	• A sound homogenization inside the reactor • Sufficient mixing and compact design	• VFA inhibition causing reduction in biogas production • Ineffective in accommodating toxics and fluctuations in the feed

2.3.1.2 Modified AnMBRs

- **Anammox AnMBRs**

Anammox-AnMBR is a novel process combining energy recovery in the form of methane with effective nitrogen management. Compared to the conventional nitrification/denitrification process, anaerobic ammonium oxidization (Anammox) has many advantages such as high nitrogen removal, cost effectiveness and small footprint (Li et al., 2015a). The Anammox reaction allows microbial oxidation of ammonium to form nitrogen gas (N_2) under anoxic conditions using nitrite as the electron acceptor (Suneethi and Joseph, 2011). Due to the complete separation of HRT and SRT by membrane, effective domestication and cultivation of the slow growing anammox bacteria was guaranteed in an Anammox-AnMBR. Dai et al. (2015) investigated the simultaneous methane production and nitrogen removal from concentrated municipal wastewater by using a membrane-based process combining anaerobic digestion and nitritation-anammox under ambient temperature. The system achieved a stable methane yield of 0.223 L CH_4/g $COD_{removed}$ (0.206 L CH_4 (STP)/g $COD_{removed}$) while a total nitrogen (TN) removal efficiency of 81% was obtained in the sequential completely (CANON) MBR. This study concluded that the proposed process was a sustainable approach for biogas recovery and nitrogen removal. This research also revealed that further treatment is required to reclaim dissolved methane released into the environment as a powerful greenhouse gas (GHG), as well as to enhance the methane recovery efficiency.

Li et al. (2015a) investigated the sustainable operation of submerged Anammox AnMBR, and found biogas sparging could greatly reduce small flocs attaching to the membrane to form a cake layer, thereby alleviating membrane fouling. It should be noted that since nitrite and ammonia were converted to nitrogen gas by anaerobic ammonia-oxidizing bacteria (AnAOB), methane composition in the biogas would be lower than that of other types of AnMBRs as a result of the accumulation of produced N_2 in the biogas. The interaction between AnAOB and methane-producing bacteria is still unknown, requiring further studies to maximize the methane yield from the Anammox-AnMBR. Last but not least, the feed for the Anammox-AnMBR contains high levels of ammonia, which can inhibit the production of methane.

- **Anaerobic Dynamic Membrane bioreactors (AnDMBRs)**

Dynamic membrane (DM) technology is a new approach for resolving problems such as high cost of membrane modules, low membrane flux, and rapid membrane fouling encountered in conventional AnMBR processes. In an AnDMBR, the solid–liquid separation is mainly accomplished by the cake layer (e.g., dynamic membrane) formed on low cost supporting materials such as meshes and fabrics with macropores (Ersahin et al., 2014). Such a DM transforms one of the most critical disadvantages of AnMBRs, namely membrane fouling, into a competitive advantage. When the dynamic membrane is seriously fouled, the cake layer can be easily removed, cleaned and then replaced by a new deposited layer, thus significantly reducing the membrane cost (Ma et al., 2013). A new deposited layer after each membrane cleaning generally requires a short time to be established sufficiently so as to acts as an efficient solids filter (Ersahin et al., 2014). Alibardi et al. (2014) developed a bench scale anaerobic dynamic MBR using a large pore-sized mesh at 200 μm. They observed varying biogas production with maximum value of 1 L/d and methane content fluctuating from 50% to 79%, which was mainly due to the variability of COD removal and HRT during operation. Low crossflow velocity (CFV) (due to the use of larger pore size) contributed to the sustainable aspect of the AnDMBR by reducing high energy input and led to improved methanogenic activity by minimizing the shear stress on the biomass. Methane oversaturation in the effluent was reported in the study. Due to the higher methanogenic activities of the cake layer formed on the external dynamic membrane module, a great amount of biogas was produced by the membrane unit and subsequently released with the effluent stream.

Ersahin et al. (2014) applied a monofilament woven as support material for dynamic membrane formation in the AnDMBR for biogas production from synthetic concentrated wastewater. Average methane yields of 0.31 ± 0.02 L CH_4/g $COD_{removed}$ (0.27 ± 0.02 L CH_4 (STP)/g $COD_{removed}$) and 0.34 ± 0.04 L CH_4/g $COD_{removed}$ (0.30 ± 0.04 L CH_4 (STP)/g $COD_{removed}$) were reported at SRTs of 20 days and 40 days, respectively, which were very close to the maximum theoretical value. Methane was solubilized in the permeate. The studies also revealed that strong shear stress as a result of biogas sparging might create a physical interruption on the syntrophic relationship between anaerobes associated with methane forming, based on the fact that SMAs of the bulk sludge samples were lower than those of the seed sludge at both SRTs. Xie et al. (2014) achieved promising average methane yield at 0.34 L CH_4/g $COD_{removed}$ (0.30 L CH_4 (STP)/g $COD_{removed}$) and methane content of 70 - 90% using Dacron mesh (pore size = 40 μm) in the AnDMBR for the treatment of raw leachate, high heavy metal concentrations, and high total ammonium concentration above 3000 mg/L. Based on the archaeal taxonomic identification, aceticlastic methanogens were the dominant functional group that produced methane, while hydrogenotrophic methanogens were eliminated at the end of the experiment when ammonium inhibition was observed.

- **Anaerobic Membrane distillation bioreactors (AnMDBRs)**

Membrane distillation (MD) is a thermally driven separation process in which water vapor transfers across a thermal gradient through a hydrophobic, microporous membrane such as polypropylene (PP), polyvinylidene fluoride (PVDF) or polytetrafluoroethylene (PTFE) membranes to form water (Goh et al., 2015; Kim et al., 2015) The competitive advantages of anaerobic processes can be readily utilized when they are combined with the MD process, as the mesophilic or thermophilic operation for the methane fermentation can allow no or less heating requirement for the subsequent MD treatment (Kim et al., 2015). AnMDBRs usually require significantly reduced footprints and provide complete retention of incoming organics and microorganisms for maximum bioconversion from waste to energy in the form of biogas (Phattaranawik et al., 2008; Goh et al., 2015; Kim et al., 2015). The other highlight of the AnMDBR treatment is the complete removal of total phosphorus for the purpose of controlling eutrophication, which has been recognized as a significant environmental and ecological concern for decades (Kim et al., 2015).

In a typical AnMDBR system, biogas can be recovered from the system for gas sparging for mixing and fouling control purposes, and additional gas can be utilized for heating and energy use (Goh et al., 2015). However, post-treatment is required to recover ammonium nitrogen and methane dissolved in the permeate. Smith et al. (2012) reported methane loss in the liquid phase from the anaerobic MBR could be as much as 30% and 50% at 35 °C and 15 °C, respectively, due to the fact that the solubility of methane gas decreases in response to temperature increase. In an AnMDBR most of the methane is more likely to exist in the gas phase, thus allowing methane extraction and recovery to be much easier. Moreover, the dissolved methane is transported with the permeate via the slower gas diffusion process in the thermally-driven AnMDBR, while conversely, other AnMBRs are mostly pressure-driven, the methane gas would rapidly pass into the permeate across the porous MF/UF membrane via poiseuille flow. Therefore, dissolved methane in the permeate from the anaerobic MDBR will most likely be much less than those from the other AnMBRs (Goh et al., 2015). Xie et al. (2014) hybridized anaerobic moving bed biofilm reactor (AMBBR) with the MD process for the treatment of domestic wastewater. A small quantity of biogas with methane content at 58% - 72% was produced from the AMMBR while no other biogas data was available from the MD process. Further research regarding biogas production from the AnMDBR and the effects of MD process on the methane-producing species would be valuable so that the benefits of AnMDBR in the sense of bioenergy recovery can be fully explored. It would also be possible to identify possible challenges in biogas production from AnMDBRs.

- **Anaerobic osmotic membrane bioreactors (AnOMBRs)**

AnOMBRs constitute a novel integrated system combining AnMBRs with the forward osmosis (FO) process for effective retention of smaller sized contaminants and prolonging of their residence time in the reactor, thus leading to improved biodegradation efficiency and biogas yield in one integrated system (Gu et al., 2015). In an AnOMBR the FO membrane is usually used, in which water flows from a low-osmotic-pressure feed solution (FS) to a high-osmotic-pressure draw solution (DS) across a semi-permeable membrane. One of the greatest advantages of the FO process is that no energy input is required to drive the filtration process as compared to traditional energy-intensive pressure-driven separation processes such as MF/UF. Gu et al. (2015) evaluated the extent of energy recovery in the form of methane gas from an AnOMBR when treating low-strength wastewater at mesophilic temperature. A promising methane production of 0.25 - 0.3 L CH_4/g $COD_{removed}$ (0.22 - 0.27 L CH_4 (STP)/g $COD_{removed}$) was obtained although a loss of methane in the effluent and a high salinity environment (10 mM - 200 mM NaCl equivalent) was discovered in the system. Although the salt, alkalinity and ammonia accumulations in the reactor were reported to have no effects on the bioactivity and biogas production, a long-term examination of salt inhibition, pH stability and ammonia inhibition on the biogas production still requires a further assessment. Chen et al. (2014) reported an average value of 0.21 L CH_4/g $COD_{removed}$ (0.19 L CH_4 (STP)/g $COD_{removed}$) for the methane yield and a methane content of 65%-78%, demonstrating the feasibility of energy recovery from such a FO-AnMBR system. The methane yield represented only 58% of the maximal theoretical value due to the loss of methane dissolved in the permeate. Moreover, the lower methane yield compared to the theoretical value indicated that the methanogenic activity was partly inhibited under the high salinity environment. However, less obvious decrease in methane yield was observed in the last phase of one cycle. The authors attributed this observation to the fact that under high osmotic conditions, anaerobic biomass tended to consume substrate to produce compatible solutes and extracellular polysaccharides in order to survive. Therefore, a further investigation of salinity's effect on the microbial kinetics and methanogenic activity is required to optimize the biogas production rate. Other challenges associated with the FO-AnMBR were the disposal of inorganic-rich supernatant, and the membrane's low tolerance to high temperature solution and biological attachment to sustain stable biogas production in the long-term.

- **Anaerobic membrane sponge bioreactors (AnMSBRs)**

The low cost polyutrethane sponge has been considered as an ideal attached growth mobile media in many aerobic submerged MBR studies to improve overall system performance due to its high specific surface area and internal porosity, light weight and high stability to hydrolyse (Guo et al., 2010). Guo et al. (2008) indicated sponge addition could significantly enhance the treatability of a conventional submerged membrane bioreactor, resulting in 2-time increase in sustainable flux. Additionally, sponge addition into submerged MBR can effectively retain biomass and enhance the flocculation ability of sludge flocs, leading to better membrane fouling mitigation and better nutrient removal (Guo et al., 2009; Deng et al., 2015). Deng et al. (2015) also reported that sponge media could positively modify the sludge flocs, reduce SMP and EPS, and prevent cake layer formation and pore clogging, thereby alleviating membrane fouling.

For an AnSMBR, the medium for bacterial attachment and growth is low-cost polyurethane sponges. These sponges represent a viable mobile carrier in many MBR technologies due to their high porosity and endurance, which can immobilize microorganisms and remove organics and nutrients effectively. Their sound mechanical features in relation to membrane scouring are another advantage to counter-attack membrane fouling due to the continuous rubbing behavior of the moving media. Kim et al. (2014) investigated both single and two-stage sponge-submerged AnMBRs using an anaerobic rotary disk MBR (ARMBR) without the membrane cleaning and replacement. They found that disk rotation contributed to enhanced shear force and mass transfer of media, and led to the effective collision between the sponge and membrane surface, thus successfully alleviating fouling and enhancing the membrane permeability in the ARMBR. Apart from the disk rotation, sponges were utilized to maintain microbial growth in the mobile phase as well as effectively control membrane fouling. The reported methane production yield and methane composition in the single system were 12% and 13% higher than those of two-stage systems. Therefore it was suggested that the single ARMBR process was superior to the two-stage process due to higher energy production in the more simplified configuration of the single system. Further research on the effects of sponge size, density and shape, and shear stress of the disk rotation on the bioactivity and biogas production are very much appreciated.

- **Gas-lifting AnMBRs (Gl-AnMBRs)**

Gl-AnMBR is considered to be an advanced hybrid treatment process that combines anaerobic bioprocess with low-pressure membrane filtration. In both external cross-flow and immersed configurations of conventional AnMBRs, considerable energy input is needed for the gas scrubbing requirements of membrane. A Gl-AnMBR, instead, applies the airlift configuration by using headspace biogas for gas lift to maintain a reasonable membrane flux with minimal energy input, thus optimizing the overall energy footprint of AnMBRs. The utilization of biogas-assisted mixing also facilitates the methane stripping from the bulk liquid, avoiding super-saturation, and allowing a minimum amount of methane dissolved in the permeate (Gimenez et al., 2012). Prieto et al. (2013) developed a Gl-AnMBR and evaluated its ability to recover resource from sewage. This suspended-growth bioreactor coupled to a tubular PVDF UF membrane was able to produce 4.5 L/d (0.28 L/gVSS·d) biogas, which can be used for membrane scrubbing and energy recovery. Biogas injection was introduced at the bottom of this system where biogas was combined with the sludge to form a two-phase (liquid-gas) flow through the lumen of vertically-placed tubular membranes. The introduction of biogas bubbles into the membrane feed significantly reduced the membrane fouling because of the increased shear force and turbulence over the membrane surface. The ascending biogas bubbles also enhanced the sludge filterability by decreasing the feed density. Therefore, pumping cost for feed flow and permeate extraction was minimized, meaning there were less energy expenses. However, the biogas recirculation for scouring generated high shear force, which was reported to negatively impact on the SMA and subsequently compromise biogas production.

- **Vibrating AnMBRs (V-AnMBRs)**

Membrane fouling is typically controlled by the recirculation of biogas to create shear and turbulence on the membrane surface (Vrieze et al., 2014). However, the high cost of gas pumping, the difficulty of operating gas sparging in certain cases, and its shear stress on the anaerobic microbes remain as major concerns for the gas-sparged AnMBRs. V-AnMBRs, which utilize effective vibratory shear for enhancing the shear at the membrane surface, have attracted the interest of researchers in recent years. Kola et al. (Kola et al., 2014) introduced a transverse vibration as an innovative membrane fouling mitigation strategy into a membrane coupled UASB reactor. Based on the observations of significantly increased critical flux and more reversible fouling as compared with conventional fouling control, this study proved transverse hollow fiber membrane vibration provided alternative enhancement of mass transfer. This type of membrane vibration also created vortices in the wake of the vibrating surface, thus facilitating the permeate filtration where gas sparging was often unfavorable. By appropriately incorporating periodical backwash/relaxation with vibrational filtration, such a V-AnMBR would be a promising technology for biogas production. However, the effects of vibratory shear stress on the methane-producing microorganisms require further analysis for the optimal biogas production.

Vrieze et al. (2014) investigated a novel V-AnMBR using a magnetically induced vibration membrane filtration system as the solo shear enhancement device in anaerobic digestion for fouling mitigation. The biomethanation performance and membrane fouling of the V-AnMBR was compared with a conventional SAnMBR with biogas scouring (known as NV-AnMBR). Similar CH_4/CO_2 ratios (around 1.89) in the biogas were observed from both reactors only when treating diluted molasses wastewater but the V-AnMBR resulted in a noticeable increase in transmembrane pressure and failed to prevent the formation of a cake layer due to the absence of a mixing system. The authors also justified that V-AnMBR is still a promising technology and can be applicable if conventional mixing devices or other measures can be implemented to avoid cake layer build-up. VFA accumulation and a decline in methane production were reported when concentrated molasses were applied, which indicated the inhibitory effects of concentrated molasses on biomethanation.

- **Anaerobic bio-entrapped membrane bioreactors (AnBEMRs)**

In view of fouling being the major concern in AnMBRs, the anaerobic bio-entrapped membrane reactor (AnBEMR) has been developed as an alternative to the conventional AnMBRs, particularly those with high biomass concentrations. The competitive advantage of the entrapped biomass technique was the superior simultaneous removal of carbon and nitrogen within a simplified single throughput bioprocess. Its robust capacity to tackle complex organic compounds, and handle high dissolved organics loading at low suspended biomass concentration was observed in the aerobic bioprocesses (Ng et al., 2014). In addition, when combining the entrapped biomass technique with the membrane, membrane fouling can be greatly reduced since less soluble organics and suspended biomass were produced in the bio-entrapped system.

Ng et al. (2014) proposed a novel lab-scale anaerobic bio-entrapped membrane reactor (AnBEMR) packed with bio-ball carriers. In their study, both the traditional AnMBR and AnBEMR were tested for biogas production from pharmaceutical wastewater treatment. The authors found that the AnBEMR was able to produce around 15% more methane than the AnMBR (0.142 ± 0.034 L CH_4/g $COD_{removed}$ (0.130 ± 0.034 L CH_4 (STP)/g $COD_{removed}$) while that of the AnBEMR was 0.159 ± 0.035 mL CH_4/g $COD_{removed}$ (0.145 ± 0.035 L CH_4 (STP)/g $COD_{removed}$) after a 70 day start-up period. However, both systems encountered the inhibition of methane yield due to organic overloading, high salinity conditions and accumulation of toxic organics when increasing OLRs up to 34.0 ± 2.7 kg COD/m^3·d. Furthermore, the AnBEMR showed a longer membrane filtration operating period than the AnMBR due to the release of smaller concentrations of EPS and SMP, and lower suspended biomass concentration. Juntawang et al. (2017) also evaluated the treatment performances and fouling of

AnBEMR in comparision with traditional AnMBR treating domestic wastewater. The bulk organic removal efficiencies by two AnMBRs were found comparable but lower SS, bound EPS and SMP in the AnBEMR contributed to less fouling compared. Biogas production from AnBEMR and traditional AnMBR contained 18 - 22% and 21 – 25% of methane, respectively. The AnBEMR showed methane production of 0.23 ± 0.02 L/d and methane yield of 0.08 ± 0.01 L CH_4/g COD $_{removed}$, while S-AnMBR had methane production of 0.28 ± 0.03 L/d and methane yield of 0.10 ± 0.01 L CH_4/g COD $_{removed}$. Table 2.3 summarizes the key features and advantages and challenges of modified AnMBRs for biogas production.

Table 2.3 Key features, advantages and challenges of modified AnMBRs

Modified AnMBRs	**Key features**	**Advantages**	**Challenges**
Annamox-AnMBRs	• Completely autotrophic nitrogen removal • Anammox bacteria • Homogeneous distribution of substrates and biomass • Anammox bacteria in forms of flocs or granules	• Production of Anammox bacterial as flocs or granules with high growth rate • High nitrogen removal • Suitable for biogas production from wastewater containing a high ammonium concentration and low COD content • Overcome long start-up issue with the Anammox process	• Dissolved methane • AnAOB compete with methane-producing bacteria • Methane composition can be altered due to the production of nitrogen gas • Ammonia inhibition
AnDMBRs	• Dynamic membrane was formed on the supporting materials such as meshes and fabrics with macropores	• Much lower capital costs for membrane and its cleaning and replacement • Higher membrane flux • Reduced energy consumption and shear stress on the biomass by using low CFV • Cope well with large OLR, high heavy metal and high ammonium concentrations • Promising methane production due to the higher methanogenic activity	• Methane oversaturation in the permeate • Strong shear stress due to biogas sparging for fouling control can affect methanogenic activity • Biogas escape from the external membrane unit and effluent collection vessel • Ammonium inhibition on hydrogenotrophic methanogens
AnMDBRs	• Thermally-driven MD process using microporous hydrophobic membranes • The organic retention times	• Complete retentions of non-volatile organics • Lower operating pressures than conventional pressure-driven membrane	• High energy requirement for heating, and uneconomical for large-scale applications

	much greater than the hydraulic retention time • Thermophilic bacteria at about 50 °C • Stable fluxes can be sustained	processes • Particularly suitable for treating refractory organics which require a long residence time • Complete rejection of total phosphorus • Shorter start-up time • Less dissolved methane	• Post-treatment required to recover methane from the permeate
AnOMBRs	• High retention forward osmosis semi-permeable membrane • Draw solution (such as seawater) required • Separation driven by osmotic pressure difference • High rejection capacity	• Better organic removal and higher biomethane potential • Suitable for methane fermentation from dilute wastewater under mesophilic conditions • Retention of alkalinity and avoiding reactor acidification • Complete total phosphorus removal and partial ammonia and total nitrogen removal • Promising methane production	• Possible effects of alkalinity, salt and ammonia accumulation on the long term stable biogas production • The inorganic-rich supernatant disposal • Membrane's low endurance to biological attachment and high temperature solution for the long time operation
AnSMBRs	• Rotary disk-supporting media for membrane fouling control • Sponges for sustaining microbial growth and fouling control	• Enhanced membrane permeability • Low cost sponge media for mobile carrier • Successful membrane fouling control (scouring) without any membrane cleaning	• Limited data available to examine the biogas production from such AnSMBRs • Further studies required to determine the optimal sponge characteristics for the optimized biogas production

Gl-AnMBRs	• Two-phase flow in the membrane unit • Enhanced sludge retentate recirculation	• Lower cross-flow velocity • Biogas-assisted mixing can help with reducing methane super-saturation	• Helium gas required for the start-up • Continuous scouring can cause varying gas equilibriums, methane oversaturation and changing pH • High shear stress on the methanogens due to the gas recirculation
V-AnMBRs	• Membrane vibration system for fouling control instead of traditional gas-sparging means	• No mixing system required • Suitable for fouling control when biogas gas-sparging is not feasible • No biogas sparging shear stress on the biomass	• Inhibited methanogenic activity due to high salinity and toxic sulfide levels • Vibratory shear stress may affect the microbial activity for biogas production and requires further investigation
AnBEMRs	• Biomass entrapped in the bio-carriers/bio-balls	• Better organic removal and higher methane yield than conventional AnMBRs • High organic loading • Significantly reduced fouling • Suitable for claiming biomethane from complex wastewater (pharmaceuticals)	• Acidogenesis and methanogenesis inhibition at high salinity conditions and organics overloading • Dissolved methane causes the lower methane yield • Long start-up period

2.3.2 Inhibitors of biogas production

2.3.2.1 Ammonia

During methane fermentation, ammonia is generated by the biodegradation of the nitrogenous compounds mostly in the form of proteins (Chen et al., 2008). As ammonia concentration grows above 3500 mg/L, methane yield starts to suffer from decreasing below the theoretical value (Kanai et al., 2010). As ammonia concentrations climb up to the values of 4051 - 5734 mg NH_3-N L^{-1}, the methanogenic bacteria can lose 56.5% of its activity. Ammonia inhibition includes the increase of maintenance energy requirement, a change in the intracellular pH, and inhibition of a specific enzyme reaction. Free ammonia (FA) (NH_3) is more toxic than ionized ammonia (NH_4^+) because it is able to penetrate through the cell membrane, resulting in the disruption of cellular homeostasis, potassium deficiency and/or proton imbalance (Chen et al., 2008). A higher temperature and pH value can exacerbate the inhibition by releasing more FA (Meabe et al., 2013). Release of ammonia is a primary concern for a high rate or intensified process such as AnMBR treating high strength waste, where shorter HRT and higher OLR may cause substrate and inhibitory intermediates to accumulate in the reactor (Jensen et al., 2015).

Measures to minimize ammonia inhibition are demanded especially for thermophilic methane fermentation from AnMBRs. Kanai et al. (2010) proposed the Kubota Submerged Anaerobic Membrane Bioreactor, in which the membranes could retain methanogenic bacteria while ammonia could be filtered out with the permeate, allowing the efficient production of methane from Japanese garbage with high protein levels (TN concentration at 10,000 mg/L). The recovered energy from this process was well above the overall energy consumption, enabling such a SAnMBR to be a net energy producer. Meabe et al. (2013) suggested that the acclimatization of biomass due to the high SRT in the AnMBR system could affect the degree of ammonia inhibition, and no critical inhibition by ammonia was expected in the their mesophilic and thermophilic study.

Jensen et al. (2015) also reported a successful methanation process from anaerobic digestion of slaughterhouse wastewater containing high proteins using a pilot-scale

AnMBR. They found over 95% of COD in the wastewater feed was converted into biogas with 70% methane, and 78 - 90% of nitrogen was released to the permeate as ammonia, which meant ammonium inhibition was minor in this system. Landfill leachate also features high concentrations of organic contaminants and ammonia, which can be problematic when used for biogas production. Xie et al. (2014) have successfully applied an AnDMBR for biomethane production with average methane yield at 0.34 L CH_4/g $COD_{removed}$ (0.30 L CH_4 (STP)/g $COD_{removed}$) from high strength landfill leachate digestion. Although free ammonium nitrogen (FAN) concentration was completely inhibitory in this case, detrimental effects were not observed on the performance due to the microorganisms' adaption to high free ammonia concentrations. The authors also suggested an interesting finding that the acetate-consuming methanogens were less inhibited than hydrogen utilizing methanogens, despite high ammonium concentrations in the reactor (over 3000 mg/L NH_3-N at mesophilic conditions). This has proved to be controversial to other researchers (Koster and Lettinga, 1984; Chen et al., 2008) indicating that aceticlastic methanogens were more sensitive to ammonium inhibition compared with hydrogenotrophic methanogens.

2.3.2.2 Sulfide

Problems associated with the methane fermentation of sulphate-rich wastes are the toxicity of sulphide to anaerobic microorganisms and the competition for the influent COD from the sulphate reducing bacteria (SRB) (approx. 2g COD/g SO_4-$S_{removed}$), which suppresses methane productivity (Chen et al., 2008; Ferrer et al., 2015; Pretel et al., 2014). In particular, methane production from municipal wastewater can be challenging because it can be easily characterized by low COD/SO_4-S ratios. The fierce competition between methane producing bacteria (MPB) and SRB can negatively impact on the quantity and quality of the biogas produced. Although it is evident that the AnMBRs are more resistant to toxics due to the sufficient SRTs for methanogens, many studies have reported increased operational costs during the treatment of high sulphate containing wastewaters by the AnMBR, especially at psychrophilic conditions and lower SRTs. Both Ferrer et al. (2015) and Pretel et al. (2014) concluded that AnMBR systems represented more energy surplus potential, thus being a net energy producer when treating low-sulphate municipal wastewater in warm/hot climates. The cost savings of up to 28% (Ferrer et al., 2015) in treating low-sulphate can be achieved as compared to the scenario with sulphate-rich municipal wastewater. Liao et al. (2010) reported the complete inhibition of biological activity caused by feed toxic shock (high concentration of H_2S in feed) in a thermophilic SAnMBR with mesophilic sludge as the inoculum.

Thus, the pretreatment of the feed should be in place to remove toxic sulfur substances so that the biological activity of thermophiles can be maintained. Gimenez et al. (2011) also observed a low methane yield at 0.069 L CH_4/g $COD_{removed}$ (0.061 L CH_4 (STP)/g $COD_{removed}$) from a pilot-scale mesophilic SAnMBR treating wastewater with a low COD/SO_4-S ratio, and this was mainly attributed to the SRB competition for 90% of influent COD. The methane recovery efficiency from SAnMBRs was greatly influenced by sulphate content in urban wastewater, and higher biogas production would be expected if high COD/SO_4-S or no sulphate were present in the substrate (Gimenez et al., 2012). The effective countermeasure to sulfide toxicity includes the dilution of the wastewater, and the implementation of sulfide removal techniques such as physico-chemical measures (stripping), chemical reaction (coagulation, oxidation, precipitation), and biological conversions (micro-aerobic sparging and partial oxidation to sulfur) (Chen et al., 2008). Acclimatization of MPB to free H_2S to increase the tolerance of aceticlastic and hydrogenotrophic MPB to sulfide can also be a possible solution. Nevertheless, above-mentioned sulfide toxicity control techniques require further research to obtain valuable data from AnMBRs, in order to validate their applicability and effectiveness in sulfide control.

On the other hand, methanogenic activities are not inhibited if the ratio of COD/sulphate in substrate is higher than 10, and low concentrations of sulphate and sulphide are also necessary for effective biogas production (Rinzema et al., 1988). Li et al. (Speece, 1983) compared the performance of two AnMBRs with and without the addition of sulphate for the anaerobic co-digestion of coffee grounds, milk and waste activated sludge. They concluded that sulphate addition (at a COD/SO_4^{2-} ratio of 200:1 to 350:1) wielded positive effects on propionate degradation and methane fermentation in a thermophilic AnMBR at higher OLRs. Without the addition of sulphate, the thermophilic AnMBR system at higher OLRs entered a "sub-health state" as a consequence of propionate acid accumulation.

2.3.2.3 Salinity

The presence of high salt concentrations is common in many saline wastes from fish and seafood processing, chemical, petroleum and pharmaceutical industries. High salinity can cause bacterial cells to dehydrate due to the osmotic pressure. With its toxic effects on non-adapted biomass mainly attributed to cations, high salinity is regarded as one of the most important factors influencing methane fermentation processes (Chen et al., 2008). Enzyme inhibition, cell activity decline and plasmolysis are the typical manifestations of salt stress on anaerobic microbes (Dereli et al., 2012). Ng et al. (2014) investigated strong salinity conditions' inhibitory effect on methane yield from both conventional AnMBR and advanced AnBEMR when treating the pharmaceutical wastewater. They found microbial flora was negatively impacted (methane yield below 0.16 L CH_4/g $COD_{removed}$ (0.14 L CH_4 (STP)/g $COD_{removed}$) was reported) in a hypersaline scenario, which was due to the disrupted ordinary metabolic functions and degradation kinetics with high salt concentrations. Jeison et al. (2008) attributed the presence of very small sized and weak granules in both UASB and AnMBR systems to the high salinity of the wastewater, despite the fact that membrane enhanced retention of active halotolerant bacteria contributed to a superior sludge activity than UASB. They also revealed that the long-term continuous adaption periods resulted in better levels of sodium tolerance, with the observed 50% activity inhibitory concentration (IC_{50}) value for acetotrophic methanogenesis at approximately 25 g Na^+/L. In addition, high salinity as a consequence of salt accumulation in the reactors is regarded as a significant concern for the AnOMBR in terms of fouling and excess flux loss rather than inhibition or toxic effects on the biological processes (Chen et al., 2014; Gu et al., 2015).

2.3.2.4 Long chain fatty acids (LCFAs)

LCFAs, are potentially suitable substrates for biogas production. However, the toxicity of LCFAs is known to impair granule formation, sludge flotation and washout, suppress methanogenic activity, mass transfer limitations of substrate, nutrients and biogas in anaerobic granular sludge bed reactors when treating high strength-lipid wastewaters (Chen et al., 2008; Dereli et al., 2012; Dereli et al., 2014a; Dereli et al., 2015). Dereli et al. (2014a) assumed that the major drawbacks mentioned above could be addressed by membrane assisted biomass retention in AnMBRs. However, they found the AnMBR process still suffered from reversible LCFA inhibition at 50 days SRT and in turn process instability, which was mainly caused by LCFA adsorption, although the membrane guaranteed excellent biomass retention when treating lipid rich wastewaters. The authors suggested AnMBR operation at shorter SRTs was preferred due to the deliberate washout of adsorbed and free LCFA, thus reducing high concentration LCFA inhibition or transport limitation. Nevertheless, a major fraction of LCFA would not remain degraded, therefore lowering the biomethane potential. Dereli et al. (2015) concluded that sustainable methane fermentation from all LCFAs required only very low applied Lipid/Mass ratios. Furthermore, they observed LCFA inhibition at high SRTs in their lab-scale AnMBR system when treating wastewater with Fat, Oil, and Grease (FOG) concentration at 11.3 ± 0.5 g/L. The inhibitory effect accelerated biomass deflocculation and SMP release.

On the other hand the LCFA absorption on sludge flocs modified their hydrophobicity, resulting in less fouling propensity. Jensen et al. (2015) reported minor LCFA inhibition from an AnMBR when treating slaughterhouse wastewater with average FOG concentration of 1407 mg/L. Ramos et al. (2014) reported that the long-term sludge adaption to LCFAs was required for high rate methanogenesis from LCFAs in an UASB coupled membrane system. Acclimated sludge quickly reached maximum methane production from the digestion of substrate with high oil and grease (O&G) content (4.6 - 36 g O&G/L) at OLR of 17 kg COD/$m^3 \cdot d$, without any notable inhibitory effects. The advantages and disadvantages of AnMBRs to mitigate problems induced by inhibitors are summarized in Table 2.4.

Table 2.4 Advantages and disadvantages of AnMBRs for the mitigation of problems induced by inhibitors

Inhibitors	Inhibitory effects on biogas production	Advantages/Disadvantages of AnMBRs
Ammonia	• Disrupting intracellular pH • Increasing maintenance energy requirement • Inhibiting a specific enzyme reaction • Methanogenic activity loss	• Complete retention of biomass • Ammonia were filtered out with the permeate • Acclimatisation and adaption of biomass to free ammonia due to the applied high SRT in AnMBRs
Sulfide	• The toxicity of hydrogen sulphide to anaerobes • The reduction on quality and quantity of biogas production • H_2S can cause corrosion in boilers, engines, and pipes causing higher maintenance and replacement costs • Downstream oxygen demand required for oxidising H_2S	• Complete retention of slow-growing methanogens • Higher methane production potential at higher temperature and higher SRT • Sulfate content in the substrate significantly affected the overall operating cost. • Promising for treating low/non sulphate-loaded wastewater • Cost associated with membrane fouling
Salinity	• Reduced methanogenic activity • Biomass decay • Long adaptation time • Negative impact on granule stability and granule size.	• Enhanced retention of active halotolerant bacteria • Flux decline due to salt accumulation • Long term adaption leads to high tolerance
LCFAs	• Impairment of granulation • Sludge flotation, washout, and foam/scum accumulation • LCFA precipitation on sludge particles • Methanogenic inhibition due to mass transfer limitations	• No biomass washout • Lesser fouling due to increased sludge hydrophobicity • Inhibition due to floc deterioration and SMP release.

2.3.3 Influential factors on biogas production

2.3.3.1 Temperature

Temperature is a vital parameter that profoundly influences anaerobic processes. Attempts to produce biogas from AnMBRs have been made under all three different temperature ranges: psychrophilic (0 - 20 °C) (Chu et al., 2005; Martinez-Sosa et al., 2012), mesophilic (20 - 42 °C) (Martinez-Sosa et al., 2011; Wei et al., 2014) and thermophilic (42 - 75 °C) (Qiao et al., 2013). Thermophilic AnMBRs are known to have a rate-advantage over the others due to a faster reaction time and higher volumetric loading rate, thus demonstrating higher biogas productivity (Mao et al., 2015). Both Liao et al. (2010) and Lin et al. (2009) investigated thermophilic SAnMBRs for biogas production from kraft evaporator condensate treatment at 55 ± 1 °C, and proved that it was a feasible technology to produce a promising methane yield at average value of 0.35 L CH_4/g $COD_{removed}$ (0.29 L CH_4 (STP)/g $COD_{removed}$) with an excellent fuel quality close to 85–90% methane in the biogas. They attributed higher methane yield to the higher sludge digestion rate making a larger contribution under thermophilic temperatures. Qiao et al. (2013) also reported that producing methane from coffee grounds via the thermophilic co-digestion SAnMBR was feasible, and traceable hydrogen content of 100 - 200 ppm was found in the biogas. However, thermophilic AnMBRs are more sensitive to the presence of toxic compounds such as hydrogen sulfide (Liao et al., 2010) and inhibitory substances in the feed, and environmental changes. The thermophilic process's extreme temperature is also believed to cause severe ammonia toxicity, digestion inhibition and unstable fermentation processes (Angelidaki and Ahring, 1994; Weiland, 2010).

Furthermore, higher membrane flux can be retained using lower energy requirements at higher temperatures due to the reduced viscosity of the biomass suspension (Jeison and van Lier, 2006a; Dereli et al., 2012). Membrane permeability can be further enhanced by decreasing TMP due to a lower permeate viscosity at high temperature. However, Jeison and van Lier (2006b) observed long-term fluxes in the thermophilic SAnMBR were in fact 2 - 3 times lower than those attained under mesophilic conditions. Therefore, more studies regarding temperature's effects on the sustainable flux are required so that biogas production from AnMBRs is at its most efficient. Membrane fouling is another critical area of interest, which impedes the development of thermophilic AnMBRs for biogas production. Lin et al. (2009) compared thermophilic and mesophilic AnMBRs at different OLRs but under similar hydrodynamic conditions. A more compact, less porous cake layer with higher cake resistance was observed in the thermophilic AnMBR, which was mainly due to the higher concentration of fine particles and EPS release at higher temperatures. Furthermore, permeate from the mesophilic AnMBR had much better quality than the thermophilic one. More attention should therefore be paid to the sustainable fouling mitigation measures in order to ensure the economic feasibility of thermophilic AnMBRs for biogas production.

Psychrophilic AnMBRs has recently attracted significant attention particularly in terms of generating biogas from low strength wastewaters (Martinez-Sosa et al., 2012). It was found that both psychrophilic and mesophilic AnMBRs achieved comparable methane production rates (Trzcinski and stuckey, 2010; Martinez-Sosa et al., 2011), although the former corresponded to significant methane loss in the permeate (Martinez-Sosa et al., 2011; Smith et al., 2013) and a slightly higher fouling rate due to VFA accumulation (Martinez-Sosa et al., 2011; Martinez-Sosa et al., 2012) and protein-dominated EPS release (Gao et al., 2014b). Last but not least, Gao et al. (2011) reported temperature shocks led to a temporary increase in biogas generation rate, but shocks with larger magnitude at higher temperatures resulted in performance being significantly disrupted.

2.3.3.2 pH

Most AnMBR systems operate at near neutral pH since methane fermentation takes place within the pH 6.5 - 8.5 range with the optimal range from 7.0 to 8.0 (Weiland, 2010). Such a pH range was usually maintained through neutralization, which requires the excessive use of chemicals such as sodium carbonate/biocarbonate or calcium carbonate since some streams have extreme pH values, and hydrolysis and acidogenesis phases will decrease pH values. Extreme pH conditions during AnMBRs operation can not only upset biological performance and methane yield but also affect membrane permeability and lifespan (Lin et al., 2013). Gao et al. (2010b) investigated the effects of elevated pH shocks (pH 8.0, 9.1 and 10.0) on biogas production from a SAnMBR, and found that the pH 8.0 shock had a minor impact, yet pH 9.1 and 10.0 shocks did exert significantly negative impacts on the methane yield. This was mainly due to the ammonia toxicity and VFA accumulation at increased pH value. Serious membrane fouling resistance was reported, due to pH shock induced sludge flocs breakage and the accumulation of fine particles in the bulk sludge. In light of the difference in growth rates, and optimum pH for the growth of acidogens (5.5 - 6.5) and methanogens (6.5 - 8.2), many researchers have worked on phased AnMBRs, which separate acidogenesis and methanogenesis processes into the two-stage reactor configuration (Chaikasem et al., 2015). Optimizing each stage separately in its own reactor reduces VFA accumulation, facilitates process stability, and enhances the system's tolerance to greater loading rate and toxicity. These features will lead to higher methane production. Such a phased AnMBR has been successfully applied to high loading wastewaters treatment for maximum methane yield (Saddoud et al., 2007b; Saddoud and Sayadi, 2007; Chaikasem et al., 2015).

2.3.3.3 Hydraulic retention time (HRT)

HRT is a key parameter from an economic perspective as it has a significant impact on the capital cost, meaning shorter HRTs allow smaller biogas-producing AnMBRs (Stuckey, 2012). Many researchers have worked on the influence of HRT on biogas production from AnMBRs. Generally, HRTs can range from as low as 2 h (Kim et al., 2011) to as high as 30 d (Jeong et al., 2010b) depending on feed characteristics, system hydraulics and sludge properties. Ho and Sung (2009b) reported that methane recovery decreased by 13% from municipal wastewater as a result of the increased COD accumulation in the AnMBR when reducing HRT from 12 to 6 h. Therefore, AnMBR operation with relatively long HRTs may maximize methane recovery. Jeong et al. (2017) showed that stable methane yield were successfully achieved in an AnMBR at a slightly long hydraulic retention time (HRT). However, Yuzir et al. (2011) observed reduced methane productivity from AnMBRs with longer HRT due to less COD available as substrate for methane production. Significantly enhanced methane production was evident when high hydraulic shock load was applied (HRT of 1 d) and they attributed this high yield to enhanced levels of hydrogenotrophic methanogenesis rather than acetoclastic activity. Huang et al. (2011) also reported that a shorter HRT increased biogas production due to increased organic loading rate in a SAnMBR. However, too short an HRT was not recommended due to higher biomass concentrations and higher SMP that could worsen membrane fouling. Gao et al. (2014a) observed something different when they investigated the effects of decreasing HRT on biogas production from an integrated anaerobic fluidized-bed membrane bioreactor. They found that methane productivity increased when the HRT decreased from 8h to 6h, which was linked to the increased OLR. Meanwhile the productivity decreased as more VFAs accumulated with a much shorter HRT.

HRT was controlled as an independent parameter from upflow velocity in the studies using CSTR-AnMBRs as the main biological component. However, An et al. (2009) reported that in the UASB-AnMBRs without the recirculation, the impacts of HRT and upflow velocity could be assessed dependently because they are inversely correlated to each other. They reported that biogas yield almost doubled from 0.062 to 0.12 L CH_4/g $COD_{removed}$ (0.057 to 0.11 L CH_4 (STP)/g $COD_{removed}$), in which methane percentages also rose from 59.3 to 65.2% when HRT of a membrane coupled UASB reactor was gradually decreased from 10 h to 5.5 h. They attributed the enhanced gas production to the improved substrate distribution in the sludge bed and enhanced mass transfer between biomass and substrate at a higher upflow velocity.

Based on the studies conducted by the researchers, it could be concluded optimized HRT exists for each case depending on many factors such as sludge characteristics, reactor configurations and substrate types. Decreasing HRT below the optimal level would lead to VFA accumulation, reduced methane productivity and severe membrane fouling, while a HRT longer than the optimal value causes AnMBRs reactor volume to be used inadequately.

2.3.3.4 Solid retention time (SRT)

Unlike other types of anaerobic reactors, AnMBRs enable SRT to become completely independent from HRT, irrespective of the sludge properties. SRT values ranged from 20 d (Dereli et al., 2014a; Dereli et al., 2014b) to infinite days (Huang et al., 2011), although most researchers worked using SRT values higher than 160 d. As a thumb of rule, AnMBRs operating at longer SRTs produce greater quantities of biogas because any decrease in the SRT may reduce the extent of reactions needed for stable digestion. For example, Huang et al. (2011) reported that a longer SRT would enhance the dominance of methanogenesis and lead to more biogas generation. In their study, methane yield rates of 0.670 ± 0.203 L CH_4/d, 0.906 ± 0.357 L CH_4/d, 1.290 ± 0.267 L CH_4/d (0.610 ± 0.203 L $CH_{4\,(STP)}$/d, 0.825 ± 0.357 L $CH_{4\,(STP)}$/d, 1.175 ± 0.267 L $CH_{4\,(STP)}$/d) were reported at SRTs of 30, 60 and infinite days, respectively. Yeo and lee (2013) suggested that AnMBR operation under a long SRT could permit low dissolved methane concentration in AnMBR permeate, along with high methane recovery. They attributed 45% more methane production at higher SRT to supplemental methane formation originating from biomass electrons via endogenous decay.

It is obvious long SRTs are more favorable in AnMBRs' operation since it results in minimal sludge production and hence significantly reduces disposal cost. However, longer SRTs operation can also impact on methanogenic activity due to a decrease in viable biomass concentration (Dereli et al., 2012). The effects of long SRTs on membrane fouling, furthermore, require urgent attention. Prolonged SRTs can hinder sludge flocculation and reduce particle size, and increase the release of SMP (Huang et al., 2011). On the other hand, high sludge concentration at high SRT can result in a rapid cake formation and compaction, leading to excess flux decline (Dereli et al., 2012). Additionally, the accumulation of inorganic solids at high SRTs may also increase inorganic fouling, which in many studies was found to be serious (Kang et al., 2002; Dereli et al., 2015).

2.3.3.5 Organic loading rate (OLR)

AnMBR processes have the competitive advantage of accommodating fluctuations in the organic loading, and OLRs ranging from 0.23 (Saddoud et al., 2007a) to 33.7 kg COD/m^3·d (Qiao et al., 2013) have been applied in AnMBRs for biogas production. In general, OLR represents the quantity of volatile solids fed into a biogas digester per day under continuous operation (Weiland, 2010). When an increase in OLR occurs, therefore, the biogas yield is supposed to also increase to a certain extent. An et al.

(2009) reported that biogas yield from an AnMBR rose linearly with an increase in the organic loading. Wijekoon et al. (2011) also observed a continuous increase in biogas production rate from 5L/d to 35L/d with increasing loading rate from 5 to 12 kg COD/m^3·d in a two-stage thermophilic AnMBR. Bornare et al. (2014) reported an increase in the average biogas generation from 159 to 289 L/d but a decrease in the biogas yield from 0.48 to 0.42 L biogas/g $COD_{removed}$ when increasing OLR from 0.62 to 1.32 kg COD/m^3·d. They attributed this conflicting outcome to a better food-to-microorganisms (F/M) ratio (0.08kg COD/kg mixed liquor volatile suspended solids (MLVSS)·d) at a lower OLR. Dereli et al. (2012), however, stated that the effect of OLR should be assessed together with SRT and biomass activity as the system's OLR was not an independent parameter.

Applied temperature also exerts a profound influence on the applicable OLRs in AnMBRs. Thermophilic AnMBRs emerge as being more effective in coping with higher volumetric loading than AnMBRs operating in the mesophilic range (Skouteris et al., 2012). It should be also noted that as the organic loading rate increases, the risk of deteriorating biogas production due to VFA accumulation might occur due to the inhibition of microbial activity (Wijekoon et al., 2011). For example, Saddoud and Sayadi (2007) documented a process failure in their study, i.e. a drastic decrease in the methane yield at OLR of 16.3 kg COD/m^3·d due to VFA accumulation and methanogenic inhibition in a one-phase AnMBR. They subsequently suggested a two-stage AnMBR with the anaerobic filter as acidogenic reactor and jet flow AnMBR as methanogenic reactor at high OLR and achieved a significant improvement in biogas conversion in the staged AnMBR.

Serious fouling caused by the release of EPS/SMP and the accumulation of large fine particles, became another issue associated with high loading AnMBRs when using AnMBRs for biogas production (Liu et al., 2012a). An increase in the OLR could cause an increase in the production of specific EPS and macromolecules in the SMP/EPS fractions, worsening the flocculation ability of the mixed liquor in the AnMBR and promoting the fast cake formation. Moreover, the EPS tended to be more viscoelastic and hydrophobic at a higher OLR. Since the adhesion forces of the EPS-membrane and EPS-EPS were significantly enhanced as the OLR increased, cake fouling was significantly accelerated (Chen et al., 2017c). In the face of the comprehensive effects of OLR on methane production, Wei et al. (2014) proposed the concept of sustainable OLR to optimize energy recovery potential from typical municipal wastewater through mesophilic AnMBRs. They reported sustainable OLR of 6 kg COD/m^3·d could result in maximum methane yield up to a theoretical value of 0.382 L CH_4/g $COD_{removed}$ (0.318 L CH_4 (STP)/g $COD_{removed}$). Table 2.5 summarizes the operational factors affecting biogas production from AnMBRs and the recommendations for optimized biogas production.

Table 2.5 The effects of operational factors on biogas production from AnMBRs and possible suggestions for optimized biogas production

Factors	**The effects on biogas production process**	**Possible suggestions for optimized biogas production from AnMBRs**
Temperature	Thermophilic: • Faster reaction rates→higher-load bearing capacity→higher biogas productivity • Possible acidification→ inhibition of biogas production • Decreased stability and increased toxicity→poor methanogensis→higher net energy input and larger investments • Difficulty in anaerobic biomass immobilization→poor sludge settling characteristics→ reduced methanogenic activities→poor effluent quality • Less cooling required→improved process economics • Sludge decay with non-adapted mesophilic sludge→serious membrane fouling • Reduced sludge viscosity→A higher flux→process efficiency →Lower shear rates→ lower energy requirement • A lower permeate viscosity→increased membrane permeability by decreasing TMP • More compact cake layer→higher cake layer resistance→servere fouling issues→very low long-term flux→process inefficiency Mesophilic: • Better process stability, higher biomass richness, better permeate quality but possible low methane yields and poor biodegradability and nutrient imbalance Psychrophilic: • Enhanced methane solubility→loss of methane in effluent→lower methane recovery • TSS and soluble COD accumulation and a higher viscosity→increased filtration resistance→increased fouling and operational cost • Enhanced membrane removal and compensation for the decreased SMA and bulk sludge removal • Energy requirement for operating the system is lower • Reduced reaction and hydrolysis rates→reduced methanogenic activity Temperature changes: • Temperature decrease→decreases in the VFA production rate, the ammonia concentration, the substrate utilization rate and the metabolic rate of the microorganisms→increased start-up times→decreasing CH_4 and H_2 yields	• Two phase AnMBRs with thermophilic hydrolysis/acidogenesis and mesophilic methanogenesis • Avoidance of drastic temperature changes

	• Temperature increase→increase in pH, hydrolysis of organic particulates→increase in methane potential • Temperature increase→Free ammonia concentration→ methanogenic inhibition • Temperature fluctuation→stress on biomass→increase membrane fouling and operational cost	
pH	• Extremely low pH value→acidification and VFA accumulation→reduced methane yield • Extremely high pH value→increased ammonia toxicity and VFA inhibition→reduced methane yield • pH shocks→dispersion of sludge flocs→the accumulation of colloids, solutes or biopolymers in the bulk sludge suspension→deteriorated membrane performance and biogas production potential	• Two phase AnMBR with optimized conditions for both acidogenic and methanogenic reactor to bring biogas yield optimization • Minimize pH shock loading by neutralizing the feed with chemicals such as sodium biocarbonate
HRT	• Optimum HRT exists, which ensures the maximum methane productivity • HRT lower than the optimal value→VFA accumulation→ reduced methane yield→ servere fouling • HRT above the optimal value→insufficient utilization of biogas digester component→reduced methane production	• Avoid operation at too high or too low a HRT • Operate AnMBRs for maximum biogas production at optimal HRT
SRT	• Long SRT→enhance dominancy of methanogenesis→enhanced methane yield • Long SRT→reduced dissolved methane→higher methane recovery • Long SRT→ reduced sludge disposal and cost • Long SRT→reduced sludge particle size and release of SMP→membrane fouling • Long SRT→cake formation and consolidation→increased fouling cost • Long SRT→accumulation of inorganic solids→inorganic fouling	• Long SRT is generally recommended for AnMBRs operation • Additional care is required for fouling mitigation at long SRT
OLR	• Increased OLR→higher metabolic activity of methanogens→increase biogas yield and methane content in the biogas to certain extent • High OLR→VFA accumulation→irreversible acidification→risk of a deteriorated biogas yield • High OLR or organic shock loading→release of tight EPS/SMP and accumulation of fine particles→serious membrane fouling	• Operating AnMBR at sustainable OLR to maximize the methane yield. • Thermophilic systems and effluent recirculation can help relieve systems from the overloading issues

2.4 Granular bioreactors

2.4.1 Mechanisms of granulation

Granulation is an autoimmobilization process in which fluffy biosolids assemble and agglomerate as dense and compact granules under controlled operational conditions (Liu and Tay, 2004). Compared to the conventional bioflocs, granules have a regular and well-defined shape, strong structure, and good settling velocities. They enable high biomass retention and withstand high strength wastewater and shock loadings. This means that the formation of granules allows the decoupling of HRT and SRT, and therefore the efficient treatment of wastewater can be carried out at much higher OLRs with a significantly reduced reactor footprint. Granulation can be characterized into two groups: anaerobic granulation and aerobic granulation. These granules are particulate biofilms with a dense syntrophic microbial community and contain millions of organisms per gram of biomass (Liu et al., 2004). Each anaerobic granule is a functional unit comprising of all the different microorganisms necessary for the methanogenic degradation of organic matter. Each microorganism in the granule has its own role in degradation of organic matter in the wastewater. Figure 2.4 shows the proposed structure of anaerobic granule. Granule nucleus refers to inert matter or inorganic precipitates. A central core of acetoclastic methanogens is surrounded by a layer of hydrogen-producing acetogens and hydrogen consuming methanogens (Abbasi and Abbasi, 2012). There is also an outside layer of bacteria that hydrolyse and acidify complex organic matter. Methanogen-like organisms are also found in the exterior and consume any free hydrogen, and this can help avoid its diffusion to the second layer. When carbonhydrate gets in contact with the granule, this polymer gets decomposed as it goes through each layer, firstly decomposed to volatile fatty acids, acetic acid and then CH_4 (Figure 2.4). Anaerobic granulation technology has the following drawbacks impeding its application: (i) the requirement of extremely long start-up period, (ii) a relatively high operation temperature, (iii) unsuitability for low strength organic wastewater such as municipal wastewater, (iv) poor capability of removing nutrients, and (v) post-treatment required to meet discharge standards (Lim and Kim, 2014; Liu and Tay, 2004; Liu et al., 2004). Hence, aerobic granulation process was developed to overcome the problems associated with anaerobic granulation (Liu and Tay, 2004).

Nevertheless, aerobic granulation system strictly requires sequencing batch reactor (SBR) operation rather than steady-state continuous flow reactor operation. With long-term SBR operation, the aerobic granules gradually lose their stability under inappropriate operational conditions, hindering the practical application of aerobic granulation technology (Lee et al., 2012).

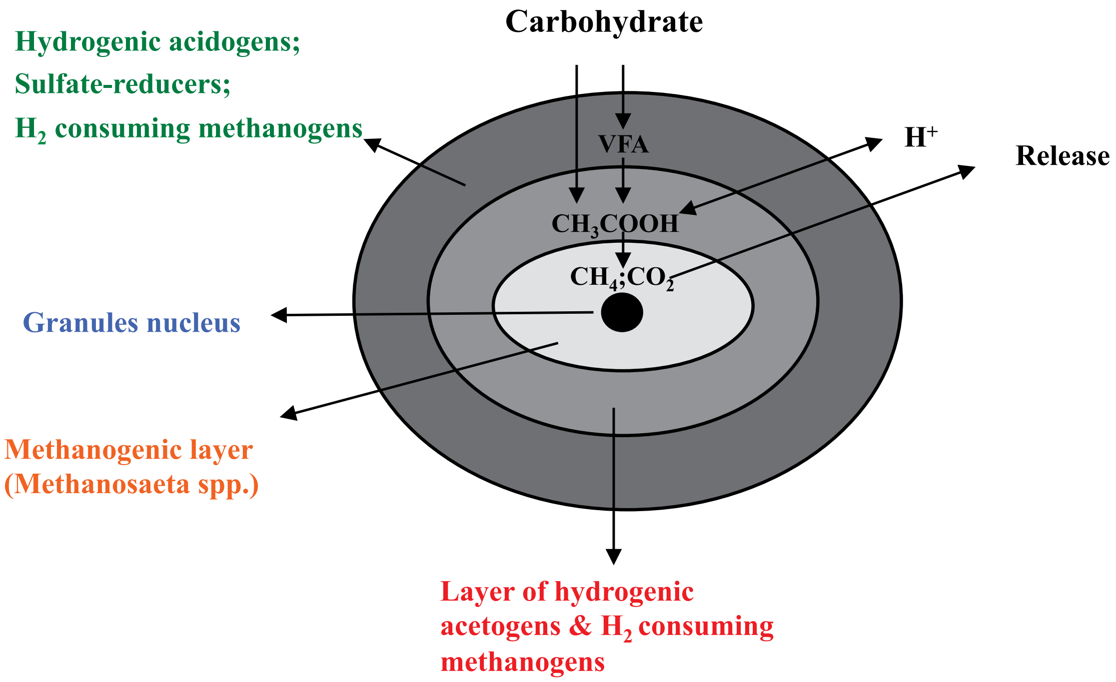


Figure 2.4 The proposed structure of a typical anaerobic granule

2.4.2 Factors affecting granulation

2.4.2.1 Innoculum

The use of anaerobic granular sludge as the seed enables quick start up of anaerobic granulation systems, and the greater the amounts of granules inoculated, the higher COD removal rates and greater loading rate that could be initiated as early as possible. Nevertheless, anaerobic granules, most often, are not readily available, and the expenses for purchasing and transporting the granular inoculum are extremely high. Consequently, studies on quick formation of anaerobic granules were heavily explored. Non-granular materials such as digested sludge, waste activated sludge and cow manure have been successfully adopted as inoculum to initiate UASB reactors and sludge granulation process. Anaerobic digested sludge is regarded as the most frequently used seed material for the production of anaerobic granules in the laboratory scale (She et al., 2006; Rajakumar et al., 2012; Zhang et al., 2011; Subramanyam and Mishra, 2013; Mitra and Gupta, 2014). The amount of inoculum for the granulation start-up was ranged from MLVSS of 7 to 32.8 g/L for these studies. In Hickey et al. (1991)'s detailed review, he summarized the findings by early researchers and found that the use of digested sludge with relatively high MLVSS ranging between 60 to 75 g/L was highly recommended to develop well settling granular sludge with minor washout. Inoculated digested sludge with MLVSS of 10 to 20 g/L in the reactor region was also suggested for the start-up.

Waste activated sludge (WAS) is also a readily available seed for an anaerobic granular sludge bioreactor. The growth of methanogens in the activated sludge is due to the existence of some anaerobic nuclei in the aerobic sludge flocs. Wu et al. (1987) reported that methanogenic bacteria of 10^8 g/SS were enumerated in the activated sludge from an aeration tank treating sludge and from a secondary sedimentation tank of an activated sludge plant treating textile dyeing wastewater. The characteristic of granulation process using WAS as the inoculum was similar to those using digested sludge. They also suggested activated sludge was an excellent alternative to the digested sewage sludge for several reasons. Firstly, it was not difficult to obtain a sufficient amount of the sludge from activated sludge plants. Secondly, the activated sludge also contained low amounts of sand and soil and was mostly composed of rich biomass, thus being ease of use and causing no issues with either dead space or scum layer coming from the seed material.

Nevertheless, fresh activated sludge is generally unstable and has a low density; therefore, it has to be incubated anaerobically and batch-fed for at least 10-20 days so as to be stabilized, to become denser, and to increase its methanogenic activity in advance of continuous feed. Wu et al. (1987) also suggested that the amount of aerobic inoculum should be kept at approximately 15 g/L. Ye et al. (2004), however, reported that granulation was not observed in reactors inoculated with acclimated inoculum from aerobic sludge. The authors attributed the reasons to much impurity and low methanogenic activity of this primary sludge. The sludge appeared flocculent and mainly consisted of cocci. On the contrary, the reactors inoculated with digested sludge efficiently developed black stable granules, which were predominantly composed of bacilli and filamentous bacteria. The higher methanogenic activity and high numbers of bacilli in the primary sludge might contribute to the successful granulation. The granulation took place readily with sludge containing bacilli and filamentous bacteria, which formed a matrix to which the other non-filamentous bacteria would then attach, resulting in formation of granular sludge.

In terms of aerobic granules, they are generally cultivated using conventional activated sludge (Lee et al., 2010), and crushed aerobic granules (Verawaty et al., 2012). Different microbial consortia showed different capabilities to agglomerate due to differences in their physico-chemical properties. The greater population of hydrophobic bacteria with lower surface charge density in the seeds could lead to faster aerobic granulation with excellent settleability (Lee et al., 2010). Other than surface properties, inoculum characteristics that profoundly impact the formation and properties of aerobic granlues also include macroscopic characteristics, settleability and microbial activity (Liu and Tay, 2004).

2.4.2.2 Substrate

So far, anaerobic granules have been developed using non-granular inoculum material for a rich variety of substrates, and it was found biogranulation was not strictly subjected to certain specific substrate. Characteristics of the feed exert a significant impact on the formation, composition, and structure of anaerobic granular sludge. Unlike aerobic bacteria, which grow fast, the effects of feed on aerobic granulation seem to be minor. As for the formation of anaerobic granules, high energy carbonhydrate feeding (such as glucose and sucrose) is favored to sustain the acidogens, enhance the formation of extracellular polymer substances, and maintain the availability of carbon source for methanogens (Liu and Tay, 2004). Lim and Kim (2014) argued that EPS was a significant factor affecting the formation and maintenance of anaerobic granules because EPS was required when forming granules, protecting anaerobes from the surroundings (Forster, 1992). Sufficient nitrogen and phosphorus are also necessary to maintain the healthy growth of various anaerobes. Last but not least, the addition of trace elements such as Ca, Fe, Ni, Co and Mg enhances granule formation, methanogenic activities and organic removal efficiency.

Literatures regarding the influential factors of trace nutrients were well documented (Liu and Tay, 2004, Abbasi and Abbasi, 2012). Vlyssides et al. (2008) found that the formation of the inorganic precipitate of ferrous sulphide constituted the inert nuclei around which the biomass was attached, and the formation of new granules was then initiated. They reported that the supplementation of iron in a UASB could achieve better COD removal (24% higher) and greater granule diameter (56%) than that of the control reactor (not receiving ferrous). Vlyssides et al. (2009) also suggested that the addition of ferrous iron greatly improved COD removal efficiency, increased granule diameter and settling velocity, and decreased the sludge bed porosity. They believed that the main responsible element for the enhanced sludge granulation was the formation of ferrous sulphide. Fermoso et al. (2008) reported the cobalt deprivation for methanogens in a methanol-fed anaerobic reactor could result in a decrease in SMA of granules, thus causing methanol accumulation, acidification and subsequent complete system failure. In face of the cobalt retention issues, Fermoso et al. (2010) suggested that Vitamin B12 was an excellent alternative cobalt source for methanol-fed UASB reactors. 3 times higher SMAs of the granular sludge in the vitamin B12 supplied reactor was reported, because of the improved the restoration of the methylotrophic methanogenesis activity being faster compared to the activity restoration upon a $CoCl_2$ pulse.

Granulation does not occur in all cases even if the above-mentioned conditions are satisfied. No granulation was found in a reactor fed with a VFA mixture containing high ammonium concentrations (1000 mg/L NH_4-N). Granular sludge did not develop using non-granular materials for treating cheese whey wastewater (Hickey et al., 1991; Hulshoff Pol et al., 1983). This was mainly due to the sensitivity of methanogens to the toxics such as heavy metals and chlorophenols. Free ammonia also exert an inhibitory effect on aerobic granulation (Yang et al., 2004a; Yang et al., 2004b), and aerobic granules only developed when its concentration was lower than 23.5 mg/L. Ye et al. (2004) also found biogranulation failed to occur using aerobic sludge as the seed to treat synthetic wastewater containing pentachlorophenol (PCP). It was also believed that high loading rates and excessive upflow velocity of wastewater containing toxic compounds could result in mortalities in granules during the granulation process in UASB reactors. The early studies on the biodegradation of toxic compound trichloroethylene (TCE) in UASB reactors have revealed the problematic granulation and sudden granular breakage and washout, due to the production of more toxic intermediate products such as dichloroethylene (DCE) and vinyl chloride (VC) (Sunil et al., 2007). Other researchers have found that with sufficient acclimatization period and step-wise feeding strategy, anaerobic granules could be developed under extreme conditions. Mitra and Gupta (2014) investigated the biodegradation of TCE in anaerobic hybrid reactor (AHR) with an influent TCE and COD concentrations of 50 and 2,000 mg/L, respectively. After the completion of the two phase acclimatization, compact granular sludge with diameter more than 2 mm was developed, yielding maximum 99.93 ± 0.13 and 97.81 ± 0.42 % of TCE and COD removals, respectively, at HRT of 24 h. Ye et al. (2004) successfully developed PCP degrading granules with an acclimated inoculum from anaerobic seed sludge. The compact granules had a maximum diameter of 2.5 and 2.2 mm, and were able to achieve over 98% COD removal rate and 99% of PCP removal rate.

2.4.2.3 Reactor configuration

So far, almost 100% of anaerobic and aerobic granular sludge only forms in column type air or liquid upflow reactors, and biogranulation is particularly, favored in the upflow reactors with a high ratio of reactor height to diameter (H/D). Unlike CSTR, which provides stochastic movement of an aggregate, upflow column configurations can facilitate a relatively homongenous circular flow along the reactor height, and therefore microbial aggregates are constantly subjected to a hydraulic attrition. The circular hydrodynamic flow can force those aggregates to form regular granules, which have a minimum surface free energy. The higher H/D can further allow a longer circular flowing trajectory and optimal interactive pattern between flow and microbial aggregates for granulation, thus creating a more effective hydraulic attrition to microbial aggregates. In contrast, with CSTR reactors, microbial aggregates are found to stochastically move with dispersed flow in all directions. Therefore, microbial aggregates are subject to varying localized hydrodynamic shear force, upflow trajectories and random collisions. Under such given circumstances, only flocs with irregular shape and size rather than regular granules occasionally form (Liu and Tay, 2002; Liu and Tay, 2004).

The predominantly applied bioreactor designs used to develop anaerobic granules are UASB reactors. The viability of other configurations in fostering anaerobic granules was rather limited and the reasons for this are not clear (Liu and Tay, 2004). Vanderhaegen et al. (1992) have challenged the traditional notion that upflow pattern was the necessity to the development of methanogentic granules. They successfully developed anaerobic granules in CSTRs, but granules disappeared within 3 weeks when reactors were incubated statically instead of being agitated. This finding reveals the importance of hydrodynamic shear force in maintaining the integrity of anaerobic granules. Therefore, anaerobic granulation may not rely on the types of configurations, but on the strategies of operation.

Anaerobic hybrid systems have gained researchers' interests, particularly in the recent years due to their capacity for better biomass retention, growth enhancement of methanogenic bacteria and non-clogging. The predominant hybrid design for the granule development is hybrid UASB reactors. Rajakumar et al. (2012) evaluated the treatment of poultry slaughterhouse wastewater in hybrid UASB packed with pleated poly vinyl chloride (PVC) rings. They found this advanced process enhanced the growth of anaerobic granules, and black matured granules with the size between 2.5 and 5 mm were observed at the end of 225 d operation. Similarly, Mitra and Gupta (2014) designed a pilot scale anaerobic hybrid reactor with filter media (300 mm height) above the reaction zone consisting of 25 mm diameters of PVC pipes. The flocculent sludge with size < 0.5 mm in diameter was converted to compact granules with 2 mm in diameter at the end of experiment. Zhang et al. (2009) investigated a rapid startup of a hybrid UASB-anaerobic fixing filter (UASB-AFF). The sludge interception by bio-filter contributed to minor sludge washout and the quick cultivation of granules with average size at 264 μm at the end of start-up period. Zhang et al. (2011) also reported an enhanced sludge granulation in a hybrid zero valence iron packed UASB (ZVI-UASB) reactor. The ZVI reaction was found to benefit the growth of methanogens by acid buffering and decrease of ORP. Fe^{2+} leaching from the ZVI could also advance the granule aggregation.

In addition, Feng et al. (2015) cultivated methanogenic granules in an anaerobic baffled reactor (ABR) toward the treatment of low-strength domestic wastewater. The main competitive advantage of ABRs over UASBs lied in its compartmentalized structure. This structural characteristic led to the separation of acidogenesis and methanogenesis longitudinally down the reactor, allowing the different bacterial groups to be developed under most favorable conditions (She et al., 2006). In the early 1996, a granular sludge blanket was successfully cultivated in an anaerobic sequencing batch reactor (ASBR) by Wirtz and Dague (1996). ASBR, as another piece of advanced design for the cultivation of anaerobic granules, was featured by the following characteristics: (1) no requirement of three-phase separator; (2) no requirement of a feed distribution system; (3) the absent of upflow hydraulic pattern; (4) discontinuous mode; (5) no short circuit and primary and secondary settles; (6) flexible control; (7) high efficiency for both COD removal and gas production, etc (Liu and Tay, 2004; Shao et al., 2008). Shao et al. (2008) employed a pilot scale ASBR with a floating cover for the treatment of brewery wastewater. The flocculent sludge was completely granulized after two months, and the properties of anaerobic granules in the ASBR differed from the ones in UASB with small size and greater density. The time required for granulation this study was significantly reduced compared to ten months operation in a non-fat dry milk fed ASBR (Sung and Dague, 1995). The authors attributed this to rich carbonhydrate in the wastewater and the reactor configuration, in particular, the employment of floating cover.

Angenent and Sung (2001) discovered a novel anaerobic migrating blanket reactor (AMBR) for wastewater treatment. It was a continuous fed, compartmentalized reactor that did not require an elaborate gas-solid separator and systems for feed distribution. The granules formed in the AMBR appeared darker in color, smaller and denser than granular sludge developed in a UASB reactor under similar operational conditions. The competitive advantages of such configuration over UASB were documented as a low biomass migration rate, less chance of short-circuiting, efficient removal of poorly biodegradable compounds and step feed operation mode for high strength wastewaters during loading shocks. Despite of these merits, AMBR was more complex in terms of the internal structure compared to UASB, and required multipoint mechanical mixing for the improvement of feed distribution and prevention of sludge clogging, which seemed less economically feasible (Liu and Tay, 2004). The EGSB reactor, as a family of the UASB reactor, has the definitive feature of high upflow velocity and the sludge granulation without carrier material. However, to date, the feasibility of the EGSB in the development of anaerobic granular sludge has not been demonstrated, and almost all EGSB studies were carried out with already available granular sludge. This was probably due to the fact that the maintenance of expanded sludge bed in EGSB was relatively difficult because of an absence of solid carriers during the preliminary biogranulation process.

As compared to anaerobic granulation, almost all the aerobic granulation phenomena have been only observed in SBR, while no successful example of aerobic granulation has been reported in continuous culture. The SBR operation consists of four steps: feed filling, aeration, settling and effluent decanting. The exchange ratio of liquid volumes and settling time in each cycle were the main screening factors for eliminating non-granular biomass from the reactor (Lee et al., 2010). A short cycle time (a short HRT) was generally preferred for accelerated aerobic granulation. Liu and Tay (2007) reported a decreased biomass growth rate of granules (from 0.266 to 0.031 d^{-1}) when increasing the cycle time from 1.5 to 8 h. Furthermore, the granules developed at 1.5 h cycle time were found the biggest in size whilst the granules cultivated at cycle time of 4 h were the most compact ones when comparing with those cultivated at other cycle times. Nevertheless, too short cycle time was not recommended for avoiding excessive washout of seed sludge.

2.4.2.4 Operational parameters

Most of studies for anaerobic granulation adopted the step-wise increase of OLR by decreasing HRT and increasing feed concentration. HRT values varied from 8 h to more than one day depending on the wastewater characteristics, whereas SRT values were not clearly specified in most cases, indicating that no sludge purging was undertaken in practice. No sludge waste was due to the slow growth rate of anaerobic microbes and naturally occurring sludge washout during the granulation process. OLR represents the degree of starvation of microorganisms in a biological system, and should be taken precautions for cultivating anaerobic granules. Most of the anaerobic granules were cultivated at the OLR ranging from 0.2 to 22 kg COD/m^3·d. A high OLR encourages fast microbial growth and rapid granulation while a low value could induce microbial starvation. Generally, the rule of thumb for the rapid granulation is to increase the OLR to achieve only 80% reduction of COD with close observation of sludge washout in the effluent. When applied OLR is too high, excessive biogas production can cause hydrodynamic turbulence, leading to washout of the seed sludge and small granules, and possible granules fragmentation. Although the step-wise decreased settling time is generally recommended to start up the aerobic granulation, directly using short settling time for cultivating aerobic granules is believed much less time-consuming (Liu and Tay, 2015). Employing combined strong hydraulic selection pressure (HSP) including short cycle time and short settling time with high OLR at 24 g COD/L·d, aerobic granules were cultivated only within 7 h (Zhang et al., 2013). This strategy was found the fastest and simplest granulation procedure ever reported till now. However, such a lab-scale strategy only lasted for 12 days, and hence the stability of the fast-formed granules in the long term needs to be further investigated. OLR also influences the size, settleability and activities of the granules, and a wide range of OLR (2.5 - 15 kg COD/m^3·d) has been successfully used to develop the aerobic granules (Peyong et al., 2012)

Most of the anaerobic granules were cultivated at 20 - 37 °C in the limited mesophilic range and at neutral pH values (Liu and Tay, 2004). It is because methanogens, as the core microbial element of granules, grow and function in an efficient manner in these ranges. Aerobic granules can be developed in SBRs at a dissolved oxygen (DO) level of 0.7 - 6 mg/L, room temperature of 20 - 25 °C and cycle time of 3 - 24 h (Liu and Tay, 2004; Lee et al., 2010). The role of reactor pH and temperature on aerobic granulation has not been thoroughly investigated and needs further study. Upflow velocity, which is correlated to the HRT for a specific granulation system, is the most important parameter contributing to the formation of granules. Aerobic granules could be developed only above a threshold shear force value with superficial upflow air velocity over 1.2 cm/s in a SBR (Liu and Tay, 2004). More regular, round, and compact aerobic granules were formed at higher hydrodynamic shear force due to the production of more EPS.

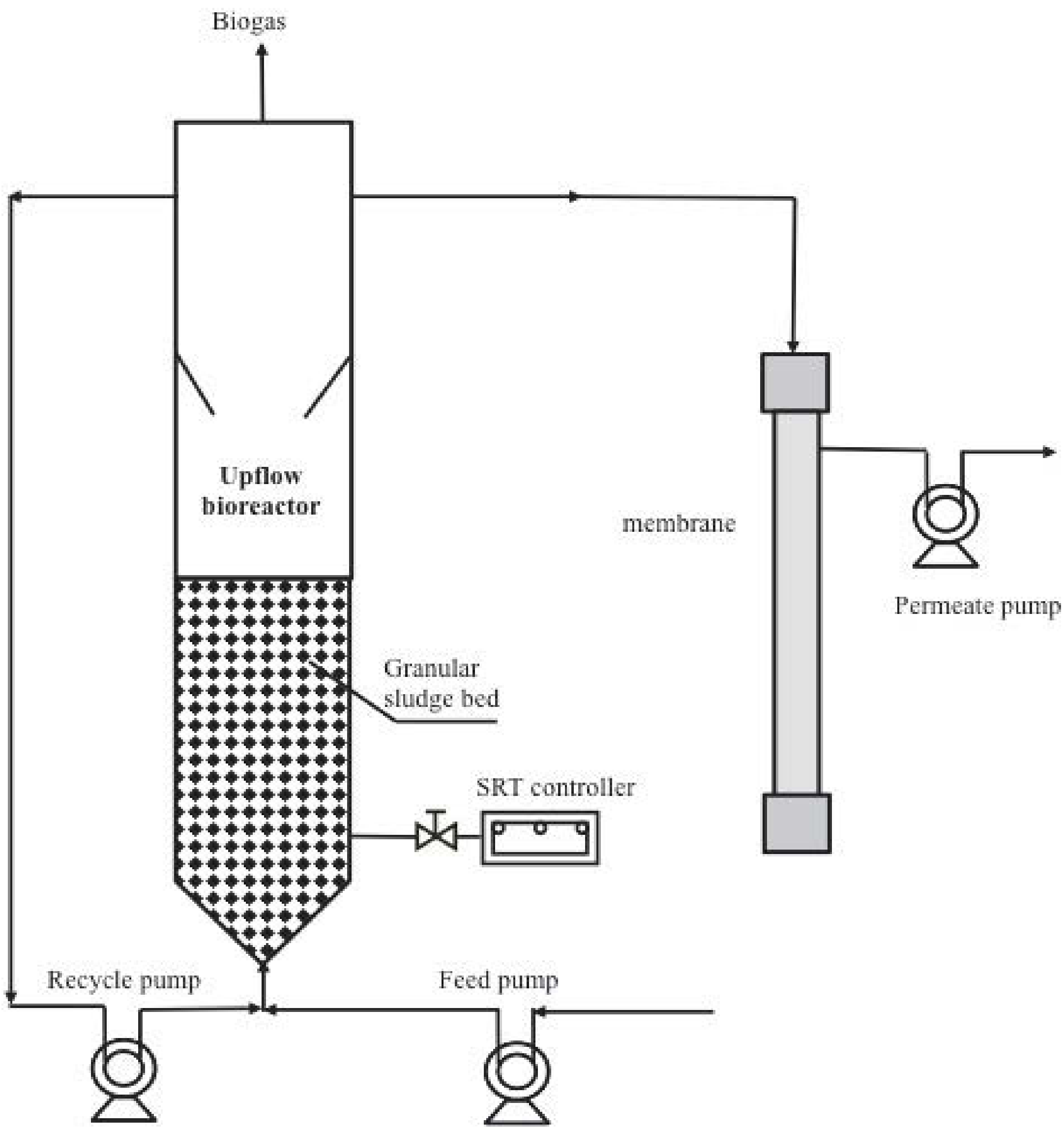
Anaerobic granulation can proceed successfully at relatively high liquid upflow velocity, and however, hardly occur under low hydrodynamic shear conditions (Alves et al., 2000; Liu and Tay, 2004). Most often, the effects of upflow liquid velocity on anaerobic granulation process are explained by the selection pressure theory (Hulshoff Pol et al., 1983). In this case, the selection pressure is the sum of the hydraulic loading rate and the gas loading rate (dependent on the sludge loading rate). Both factors are important in the continuous selection of sludge granules with different settling characteristics. A long HRT accompanied with a low upflow liquid velocity may allow dispersed bacterial growth and be less favorable for microbe granulation. Under conditions of high selection pressure, nongranulation competent bacteria will be washed out while heavier granular components can be retained in the reactor. These findings are found consistent with those reports for the aerobic granulation. Evidence shows that flocculent anaerobic sludge can be converted to a relatively active anaerobic granular sludge by enhancing agglomeration, manipulating only hydraulic stress, in a short time less than 8h (Noyola and Mereno, 1994). This finding provides a solid support to the hypothesis of Hulshoff Pol (1989) that the selection pressure of upflow velocity is the major cause of granulation, realising granulation at a threshold U_v of 10 m/h.

Zhang et al. (2009) reported a failure of anaerobic granulation due to a low mass transfer caused by low rates of hydraulic flow and biogas ascension. During wastewater treatment, the substrate was first transferred from the bulk liquid via the diffusion to the surface of the granule (i.e., external mass transfer), followed by successive intra-granule mass transfer and biochemical reaction within the granule. Accordingly, low external mass transfer due to low hydraulic load in the feed would limit the system performance, including microbial growth and biodegradation. The hydraulic recirculation by manipulating circulation ratio (defined as the ratio of circulation flow to feed flow, R), was quickly adopted after the startup fail, and enhanced the mass transfer between the granules and substrate, thus accelerating the growth of granules and the startup. However, excessive hydraulic circulation (at an R of 28) could result in the mutual collisions among sludge granules, resulting in disintegration of the granular sludge by a decrease in the diameter of the sludge granules from 0.25 mm to 0.15 mm. Zhang et al. (2011) conducted a further research on the hydraulic circulation (at an R of 15) operated around the ZVI bed, and they observed the apparent enhancement of the sludge granulation. It was concluded by them that the intensified effects of ZVI on the granulation, including promoted methanogenic growth due to acid buffering and decrease of ORP from ZVI reaction and Fe^{2+} leaching from the ZVI, advanced the granule aggregation.

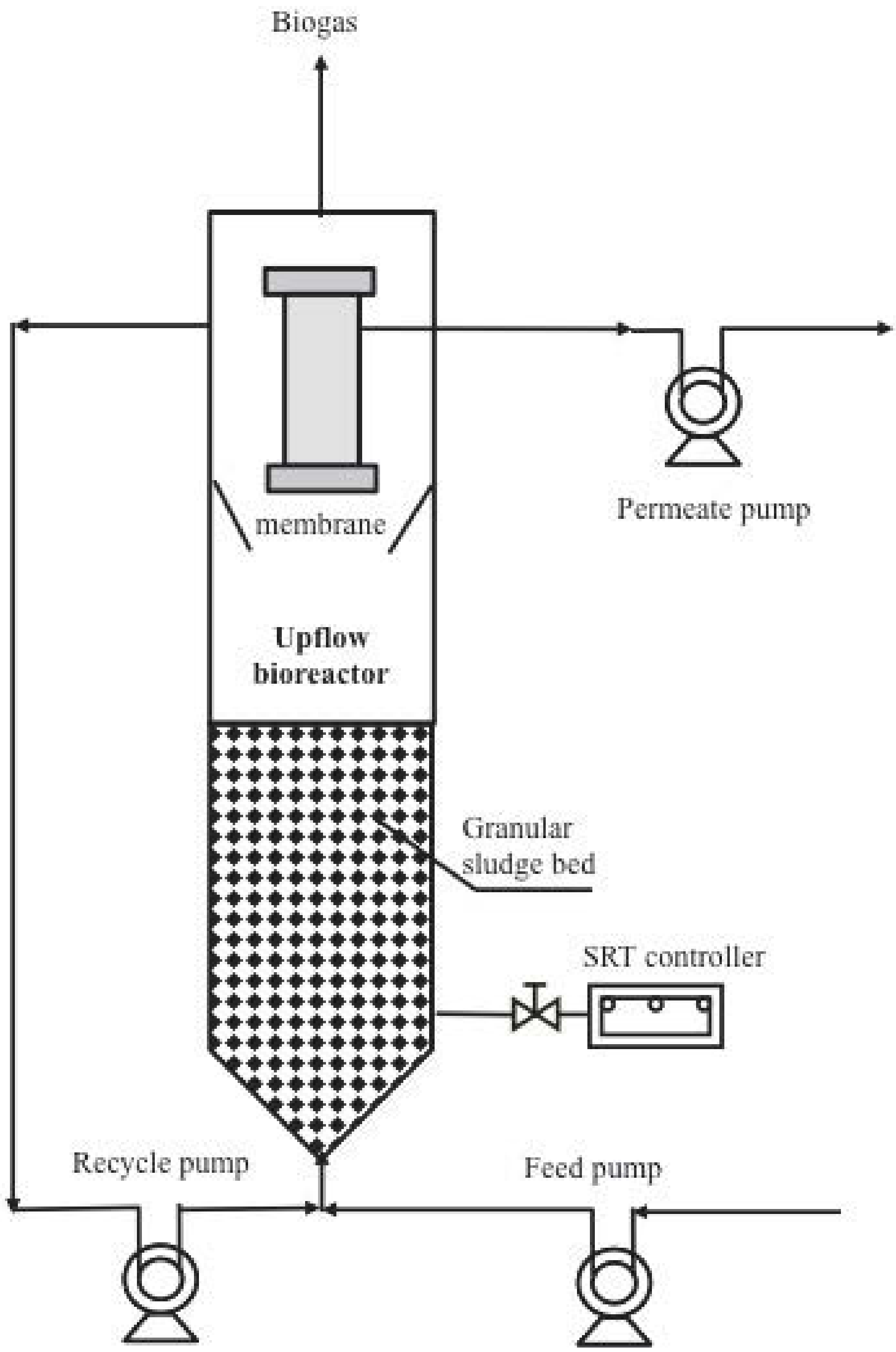
2.4.3 Recent development of granular membrane bioreactor

2.4.3.1 Granular anaerobic membrane bioreactor (G-AnMBR)

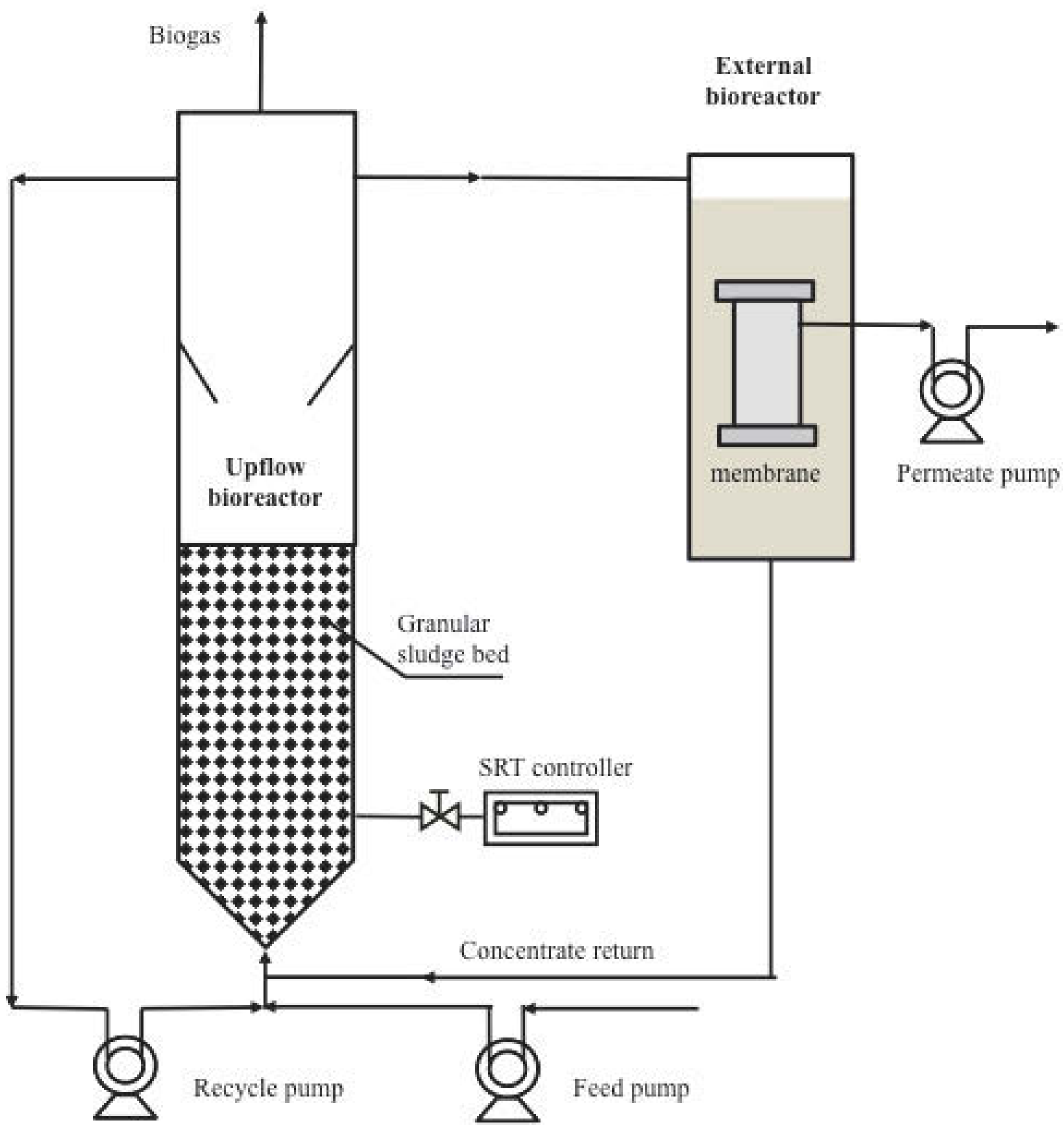
In the recent years, G-AnMBR has experienced increasing applications due to its superior performance such as high effluent quality, complete biomass retention, high biomass content, less sludge bulking problem, relatively low-rate sludge production, higher loading capacity, compact design and rapid start-up period. The G-AnMBR consists of an anaerobic granular growth bed reactor such as UASB and a membrane module. Membrane unit could be externally connected to the granule bed system as the side-stream mode (Figure 2.5a), submerged in the granular sludge bed reactor (Figure 2.5b) or immersed in a separated bioreactor (Figure 2.5c). Polymeric membranes such as PVDF and polyethersulfone (PES) were predominantly used in G-AnMBR mainly due to economic concerns. In regards to filtration, both MF and UF membranes are the most common ones, with membrane pores ranging from 0.4 µm (Diez et al., 2012), 0.2 µm (Lew et al., 2009) or 0.1 µm (Liu et al., 2013) in the MF region to values as low as 30 kDa (Gao et al., 2010a) in the UF region. Hollow fiber membrane configuration gained the most popularity as compared to flat sheet (plate or frame) and tubular modes in G-AnMBR studies.



(a) External crossflow G-AnMBR (membrane as solo polishing step)



(b) G-AnMBR with membrane place directly in the reactor (Membrane as part of G-AnMBR)



(c) G-AnMBR with the membrane immersed in a separated bioreactor (Membrane as part of G-AnMBR)

Figure 2.5 Schematic of G-AnMBR configurations

2.4.3.2 G-AnMBR versus suspended growth AnMBR (S-AnMBR)

G-AnMBR offers a promising alternative approach to the traditional suspended growth AnMBR (S-AnMBR). These conventional AnMBRs generally consist of CSTRs with either internal or external membrane separation devices, and they are operated based on suspended growth pattern. AnMBRs studies by far were predominantly S-AnMBRs

(Huang et al., 2011; Martinez-Sosa et al., 2011; Martinez-Sosa et al., 2012), because of the ease of use and construction. However, critical obstacles including membrane fouling, low flux and high operational costs still exist and limit the wider application of S-AnMBR. Traditional AnMBR is usually operated at a lower biomass concentration compared to HRARs mainly due to fouling issues, corresponding to a lower OLR (<10 kg COD/m^3d) (Lin et al., 2013). To maintain a well-mixed flow regime and sufficient mass transfer, rigorous mechanic mixing is required, which is energy intensive. In addition, the membrane unit is directly exposed to the bulk sludge, and the high suspended solid concentration subjected to MF/UF membrane filtration worsen cake deposition and compaction in all CSTR configurations (Liao et al., 2006; Ozgun et al., 2013). More rapid and dense cake layer build-up results in heavy membrane fouling and low flux, as under sufficient mixing, effluent solids concentration of CSTRs remains the same as the bulk solids concentration. To resolve this issue, frequent physical and chemical cleaning, interval operation, and likely sub-critical flux operation to sustain the flux should be in place. Moreover, a dramatic decrease in the sludge floc size owing to sludge recirculation through the membrane feed pump can result in severe membrane fouling (Ozgun et al., 2013), particularly in the side-stream membrane configurations. The high shear stress may also impact the biological activities of anaerobic microbes due to negatively impacted juxtapositioning of acetogens and methanogens, restricting the essential hydrogen transport for acquiring a superior SMA. Table 2.6 represents a comparision of conventional aerobic treatment, anaerobic treatment, S-AnMBR and G-AnMBR.

Table 2.6 Comparison of conventional aerobic treatment, anaerobic treatment, S-AnMBR and G-AnMBR

Feature	Conventional aerobic treatment	Conventional anaerobic treatment	S-AnMBR	G-AnMBR
Organic removal efficiency	High	High	High	High
Effluent quality	High	Moderate to poor	High	High
Organic loading rate	Moderate	Moderate	High	High
Sludge production	High	Low	Low	Low
Footprint	High	High	Low	Low
Biomass Retention	Low to moderate	Low	High	Excellent
Nutrient requirement	High	Low	Low	Low
Alkalinity requirement	Low	High	High to moderate	High to moderate
Energy requirement	High	Moderate to low	Moderate to low	Low
Bioenergy recovery	No	Yes	Yes	Yes
Mode of treatment	Total	Essentially pretreatment	Total or pretreatment	Total or pretreatment
Mode of Operation	Continuous	Batch/continuous	Continous	Continuous
Start-up time	2-4 weeks	2-4 months	< 2 weeks	<1 weeks or 2-4 month
Types of Wastewaters	Low to moderate	High to moderate	High to low	High to low
Membrane fouling	-	-	High to moderate	Moderate to low
Mode of Operation	Continuous	Batch/continuous	Continuous	Continuous

It is apparent that as a hybrid system, G-AnMBR combines the advantages of granular technology and MBR technology, yielding maximum joint benefits. Firstly, biomass retention is achieved by the spontaneous formation of granular sludge, and the granule bed systems are characterized by TSS concentrations ranging between 20 and 40 g/L reactor volume; whereas significantly lower effluent TS concentration at 50 mg/L was possible (An et al., 2009) and this makes them feasible for high organic and hydraulic loadings. Since biomass is not directly exposed to a membrane module in this reactor design, less apparent dense cake layer formation and consolidation will occur in comparison with conventional S-AnMBR when coupling these reactors with a membrane module. Secondly, the natural occurring turbulence caused by the rising biogas bubbles and liquid upflow force, which buoy the granular sludge, provides sufficient substrate and microorganism contact so as mechanical mixing is no longer required and relevant operational cost would be greatly decreased (Chong et al., 2012).

In addition, less severe fouling is found in G-AnMBR configuration, thus allowing enhanced operation with reduced gas sparging demand and increased fluxes (Martin-Garcia et al., 2010). Martin-Garcia et al. (2013) confirmed the lower fouling potential in the G-AnMBR as compared to the C-AnMBR, due to the reduced solid and colloidal load (by a factor of 10 and 3) to the membrane. Furthermore, the critical flux test also revealed the G-AnMBR required much lower gas sparging intensity, resulting in lower energy demand for fouling control. The filtration performance of three MBRs (i.e. C-AnMBR, G-AnMBR and conventional aerobic MBR) for domestic wastewater treatment was also investigated (Martin-Garcia et al., 2011). Comparing to the C-AnMBR, it was found that the G-AnMBR was characterized with 50% lower MLSS concentration and SMP, contributing to lower fouling rate than that of the C-AnMBR. Mathioudakis et al. (2012) reported that the net specific operational energy demand of a G-AnMBR based flowsheet treating 10,000 m^3/d domestic sewage was around 0.14 kWh/m^3 whereas the values for immerse AnMBR configurations could be as high as 3.57 kWh/m^3. The potential of the proposed G-AnMBR offers high treatment efficiency with significantly reduced energy demand as compared to traditional sewage treatment. For example, for the typical specific net energy demand of a typical activated sludge plant utilizing anaerobic sludge digestion can be more than 0.6 kWh/m^3 (Martin et al., 2011). Most importantly, the granule structure offers ideal conditions for syntrophic associations such as those between H_2-accumulating acetogenic bacteria and H_2-consuming methanogens, allowing high COD removal efficiencies even under presence of toxicity or hydraulic loading events (Ozgun et al., 2013). The compactness of the granules also renders a more compact reactor design, resulting in a much smaller footprint to apply this technology. Apart from the above-mentioned benefits, the capital cost of UASB reactors can also be reduced when coupling a membrane unit and it is by eliminating the necessity for a GLS separator in a UASB (Liao et al., 2006). However, the high sludge carry over to the effluent can occur with increased biogas production when operating in the absence of GLS. Last but not least, the granulation process and start-up period can be greatly reduced due to the membrane absolute barrier to provide the complete retention of methanogens.

2.4.3.3 G-AnMBR for municipal wastewater treatment

Recent studies on G-AnMBRs were predominantly applied to low strength municipal wastewater treatment rather than high strength organic industrial wastewaters such as brewery and alcohol-distillery wastewater. Municipal wastewater has long been categorized into complex wastewater due to its high fraction of particulate organic material, moderate biodegradability and its low strength. For this reason, domestic sewage treatment by anaerobic means is still challenging due to the kinetic limitations of anaerobic metabolism. Low substrate affinity of anaerobic biomass compared to aerobic bacteria and the rate-limiting step of hydrolysis of particulate matter into dissolved molecules particularly under low temperature (< 20 °C) conditions have made it hardly practical to achieve low effluent COD concentrations and to fulfill more stringent legislation for wastewater reclamation and reuse (Lin et al., 2013; Ozgun et al., 2013). G-AnMBRs have offered the most sustainable options of municipal wastewater treatment and reuse essential in all the land-scarce, water-short and energy-poor countries. Table 2.7 presents a number of studies that have used G-AnMBRs for treating domestic wastewater under various operating conditions. G-AnMBRs have typically shown high COD/TOC removal efficiencies with 77% - 97% and this high efficiency could be sustained even at psychrophilic temperature and high hydraulic loading rate.

Table 2.7 Summary of G-AnMBR performances for municipal wastewater treatment

Type of wastewater	Innoculum	Scale	Volume (L)	Reactor configuration	Characteristics of membrane	Operating condition	Removal efficiency (%)	Reference
Domestic wastewater (COD=100-2600 mg/L)	Digested sludge	L	17.7	Hybrid upflow anaerobic bioreactor+ submerged membrane	PE Hollow fibre UF pore size: 0.03 μm Surface area: 0.3 m^2	OLR= 0.5-12.5 kg $COD/m^3·d$, HRT=6 and 4 h Ambient temperature, SRT=150 d, Flux= 5 LMH, MLSS=16-22.5 g/L	COD= 97%	Wen et al. (1999)
Synthetic wastewater (COD=383-849 mg/L)	Granular sludge	L	4.7	EGSB with submerged membrane	PE Hollow fibre MF pore size: 0.1 μm Surface area: 0.1 m^2	Uv^b= 2-8 m/h, OLR= 1.6-4.5 kg $COD/m^3·d$, HRT=3.5, 4.6 and 5.7 h, Temp=25, 20 and 15 °C	COD= 85-96%	Chu et al. (2005)
Raw municipal wastewater (COD=58-348 mg/L)	Digested sludge	P	34	UASB+ external membrane	Polyacrlonitrile Module 1: ID/OD of 1.2/2.1 mm, 0.2m^2 Module 2: ID/OD of 1.9/2.9 mm, 0.2 m^2 Module 3: ID/OD of 3.0/3.9 mm, 0.2 m^2	OLR= 0.3-0.9 kg $COD/m^3·d$, Temp= 27-30 °C, HRT= 5.5-10 h, MLSS=12-32 g/L, SRT=∞	COD= 77-81%	An et al. (2009)
Pre-settled domestic	Granular	L	180	Upflow anaerobic	PVDF Hollow	OLR= 1.08, 2.16, 4.32 kg	COD> 88%	Lew et al.

wastewater (COD=540 mg/L)	sludge			bioreactor+ External membrane	fibre MF Pore size: 0.20 µm Surface area: 4 m^2	COD/m^3·d, Temp= 25 °C, SRT=∞, HRT= 12, 6 and 4.5 h, Flux=3.75, 7.5, 11.25 LMH, MLSS=14-80 g/L		(2009)
Synthetic wastewater (COD=500 mg/L)	Granular sludge	L	10	UASB+ External membrane	PVDF and PEI Flat sheet UF, 100 kDa MWCO and 30 kDa MWCO Surface area: 0.052 m^2	OLR= 5 kg COD/m^3·d, Temp= 30 °C, HRT= 24 h, MLSS=12-32 g/L, SRT= 50 d	COD= 96%	Gao et al. (2010a)
Real municipal wastewter (COD=646 mg/L)	-	L	3.5	UASB+ external membrane	Tubular UF, 40 kDa MWCO Surface area: 81 cm^2	Temp= 20-25 °C, HRT= 3 h, SRT= 60, 100 d	COD= 86-87%	Herrera-Robledo et al. (2010)
Raw sewage (COD=445±138 mg/L)	-	L	849	UASB+ external membrane	PVDF Tubular UF, 100 kDa MWCO Surface area: 5.10 m^2	Uv=0.64 m/h, Temp=22±3 °C, Flux=6LMH, HRT=6 h, SRT=180 d	COD= 93%	Herrera-Robledo et al. (2011)
Synthetic wastewater (COD=350±10 mg/L)	Granular sludge	L	12.5	UASB+ external membrane	PVDF Tubular UF, 100 kDa MWCO	UV^b= 0.122, 0.061 and 0.041 m/h, Temp= 27-30 °C, HRT= 4, 8 and 12 h, pH= 7, Flux=5 LMH	COD= 81-89%	Salazar-Pelaez et al. (2011a)

Real domestic wastewater (COD=285-2,088 mg/L)	Granular sludge	P	700	UASB+ External membrane	PVDF Tubular UF, 100 kDa MWCO Surface area: 0.84 m^2	SRT=150 d, Ambient temperature, HRT= 4, 8, 12 h	COD= 86-96%	Salazar-Pelaez et al. (2011b)
Settled primary wastewater (COD=338±74 mg/L)	Granular sludge	P	125	Upflow anaerobic tank+external membrane	PVDF Hollow fibre UF pore size: 0.08 μm Surface area: 0.93 m^2	Uv=0.7-1 m/h, Temp=8 – 22 °C, Flux=6LMH, HRT=16h	COD= 84-86%	Martin-Garcia et al. (2013)
Synthetic wastewater (COD=500±10 mg/L)	Granular sludge	L	10	UASB with submerged membrane	PVDF Hollow fibre MF pore size: 0.1μm Surface area: 1 m^2	UV^b= 2.5m/h, OLR= 1 and 6 kg $COD/m^3 \cdot d$, Temp= 27-30 °C, HRT= 2 and 12 h, Flux=5 LMH, MLSS= 1.6 and 10.1 g/L	COD= 97%	Liu et al. (2012a) Liu et al. (2013)
Synthetic wastewater (530±40mg/L)	Flocculent anaerobic sludge	L	7	UASB+ external MBR	PES Tubular UF pore size: 30nm Surface area: 0.0038 m^2	UV^b= 0.6 m/h, OLR= 2 kg $COD/m^3 \cdot d$, Temp= 25 °C, HRT= 6 h, Flux= 12.3LMH	COD= 92%	Ozgun et al. (2015)

1) Membrane as solo polishing step

A pilot scale UASB reactor followed by a post-treatment external UF membrane operating at different HRTs (12 - 4 h) was able to achieve permeate free of TSS, and with COD concentration less than 120 mg/L for the treatment of real domestic wastewater with a high variability in its characteristics (Salazar-Pelaez et al., 2011b). In spite of high variations in COD, TS, volatile total solids (VTS) and TSS concentrations by sediments washout from pipelines in the rainy seasons, the combined system consistently produced permeate fulfilling the Mexican standards established for wastewater reclamation in public services at all times. Similarly, such a system of lab scale yielded a slightly better total COD removal (89 and 82% respectively at HRT of 8 and 12h) than that of a UASB reactor alone at the steady state in treating synthetic wastewater with average COD concentration at 350 mg/L at ambient temperature (Salazar-Pelaez et al., 2011a). However, as suggested by the authors, the transition to lower HRT (12 to 4 h) deteriorated the AnMBR performance by inducing a higher production of SMP and EPS and particle release in UASB effluent, thus eventually increased COD and solids concentrations in both, UASB effluent and permeate (total COD removal 81% vs 89% in the case of Salazar-Pelaez et al. (2011a)). The authors attributed declined removal efficiency to the following reasons. Firstly, the operation of UASB reactor at a low HRT consequently resulted in higher upflow velocity and OLR, therefore increasing shear forces inside the reactor and biogas production. Both facts caused solids washout as well as particle disaggregation and stress in microorganisms and promoted biopolymer release in its effluent (Wang et al., 2009). Secondly, the lower removal efficiency was mainly due to the limited contact time between microorganisms and substrate for the physical and biological processes at low HRTs. The last but not least, the system was incapable of retaining UASB sludge, and therefore had the decreased filtration capacity of the sludge bed at higher upflow velocities (Leitão et al., 2005). The enhanced release of biopolymeric substances in the effluent at lowest HRT also worsened the fouling propensity in the UASB effluent by increasing the fouling rate and the specific cake resistance and decreasing particle sizes. Therefore, both studies recommended to avoid operating AnMBR (UASB + external UF arrangement) at HRTs lower that 4 hours in order to control SMP and EPS fouling potential, and maintain better COD removal performance.

On the other hand, An et al. (2009) showed that with gradually decreasing HRT from 10 to 5.5 h, the application of membrane filtration as a polishing unit of UASB effluent could achieve better TOC removal efficiency from 80% to 85%, and much higher biogas yield from 61.8 to 120.7 mL CH_4/g $COD_{removed}$ in treating raw municipal wastewater at ambient temperature in Singapore. Such a behavior at lower HRT was attributed to the increased upflow velocity, which improved water distribution and provided more even contact between substrate and microorganism, and higher sludge loading rate in a low-strength wastewater treatment system, which favored the anaerobic microorganisms activity. Their investigation also indicated that periodic 20 seconds backwash every 10 mins suction presented the best result to reduce fouling and membrane cleaning frequency and prolonged membrane longevity.

Herrera-Robledo et al. (2010) showed that with HRT (3h) more than three times lower than those in full-scale UASB applications for municipal wastewater treatment, the operation of UASB + MBR process at ambient temperature in southern Mexico City was still feasible. They compared the performance of this combined system operated at two different SRTs (100 and 60 d) at local temperatures of 20 - 25 °C, and found there was no difference on COD removal efficiency (TCOD and SCOD removal efficiencies of over 85% and 73% were achieved in the parallel systems, compared to 50% in the UASB reactors alone, suggesting that such a hybrid system operated at relatively low HRT and SRT is viable at the Mexican climate conditions. Their investigation also indicated that longer SRTs operation during long-term operation of 500 h resulted in more repetitive sudden TMP and flux changes which might be explained by a fouling lay collapse and compression hypothesis for cross-flow membrane ultrafiltration, suggesting a stronger fouling layer structure. Furthermore, Herrera-Robledo et al. (2011) demonstrated the efficacy of a high rate UASB-MBR for raw sewage treatment at HRT and SRT of 6 h and 180 d in producing an effluent free of suspended solids, pathogens (fecal coliforms) and parasite ova, with 93% COD removal and 73% phosphorous reduction (sorption by the biofouling or even chemical precipitation through biomineralization), that met official Mexican regulation for direct urban water reclamation. Based on fouling analysis, SMP with size lower than membrane pores (89 mg/L) tended to absorb on membrane surface or inside the pores, and were considered significant for fouling development. It was also suggested that a mild cleaning procedure using chlorine (NaClO at 300 mg/L, for 30 min) accomplished a limited removal of fouling mass per unit area (13%), and the biofouling remnants was partly resulted from biologically-induced mineralization materials that were synthesized (massive EPS secretion from colonizing cells) in response to cleaning procedure, and may be the basis of irreversible membrane fouling.

The studies mentioned above have used the membrane unit as a solo polishing step after the UASB reactor. In such a configuration, the concentrate streams were not recycled back to the bioreactors, and therefore the hydraulics and biogranulation in the UASB reactors remained undisturbed from the membrane incorporation, resulting in continuous selection of stable granules with good settling properties for efficient anaerobic digestion. Nevertheless, Ozgun et al. (2015) elucidated that the addition of membrane as an absolute barrier could cause a detrimental effect on the suspended solids (SS) accumulation in the membrane tank situated after UASB reactors, contributing to an intensifying increase in the SS loading on the membrane unit and subsequent high tendency to foul.

2) Membrane as part of G-AnMBR

A great number of studies have suggested employing membranes as an element of a G-AnMBR system to provide nearly absolute biomass retention and allow for operation at high SRTs via concentrate flow recycle to the granular reactor, thus leading to the enhanced reactor performance subject to psychrophilic methanogenesis, high loading events, loading shocks and climate temperature fluctuations (Chu et al., 2005; Liu et al., 2012a; Liu et al., 2013). Membranes in these systems are either located as an external side-stream (Lew et al., 2009; Martin-Garcia et al., 2013; Ozgun et al., 2015) in which the concentrate is recycled back to the granular bioreactor or submerged at the top of the reactor (Wen et al., 1999; Chu et al., 2005; Liu et al., 2013). In such a setup, the membrane is not simply regarded as a physical barrier, but also facilitates a general cultural adaptation to the prevailing loading conditions of the reactor environment.

Liu et al. (2012a) utilized a granule based AnMBR system in which a MF hollow fiber membrane was immersed in the expanded section to evaluate the impact of F/M ratio on the system performance and fouling treating low strength wastewater at 27 - 30 °C.

They reported impressive reclamation with the TOC removal efficiency of more than 96% in the high load AnMBR. However, they observed more severe fouling in the high load G-AnMBR as compared to the low load system, with cake resistance responsible for over 98% of the total fouling in both systems. This higher cake resistance was attributed to higher amounts of SMP and higher tightly-bound to loosely-bound EPS ratio in the cake layer whereas the greater amount of fine particles in the high loading system was also responsible for more serious fouling. The authors pointed out that membrane filtration deteriorated sludge bioflocculation, which in turn exacerbated membrane fouling, and a lower F/M ratio and strategies for cake layer elimination were preferred for the long term sustainable operation of AnMBRs.

Thereafter, Liu et al. (2013) used the same system to evaluate specific enhancement of the UASB performance by incorporating submerged membrane to the overloaded reactor at mesophilic temperatures, and obtained similar TOC removal of 97% at HRT of 2 h and specific organic loading rate (SOLR) of 3.8 kg COD/kg MLSS/d. Linear increased TOC removal from 55% to 91% by bulk sludge mainly accounted for the enhanced performance, implying that submerged membrane addition overcame the upper SOLR limit in the UASB reactor treating low strength wastewater and significantly enhanced biological activity of the suspended sludge when encountering the high loading events. One of the major advantages of this system was the membrane retention of biomass with sufficiently high SMA and an especially beneficial and diverse microbial community structure predominated by Methanothrix-like bamboo shaped rods when the loading rate is too high for sufficient sludge retention and when the high quality effluent is required for reclamation. The author also suggested that the enhancement by the membrane incorporation to the UASB was featured by two phenomenal behaviors: (1) the comparatively higher removal by the membrane at the early transition stage when the process was adapting to the introduced membrane, and (2) dramatic increase in the bulk removal during the adaption stage as well as gradual decrease in the membrane removal to the steady state.

On the other hand, Ozgun et al. (2015) reported that the introduction of membrane to a UASB reactor significantly affected the system in both biological and physical perspectives. Membrane incorporation induced the deterioration of sludge settleability and more frequent sludge washout, due to the decreasing of PSD caused by an accumulation of fine particles and decreasing EPS, thus causing a resultant increase in COD and TSS and SMP concentrations in the UASB effluent. The authors justified that, despite the SMA and stability of the UASB sludge deteriorated after membrane incorporation, the increase in microbial community index in both richness and evenness was found, and hence the enhancement in overall system performance was observed in terms of higher COD removal efficiency of 92% (72% COD reduction before membrane addition) and more methane production at HRT of 6 h, due to the complete retention of all particulate and colloidal matter and biomass inside the reactor by the membrane. In addition, the stable TMP was observed at 85 mbar in average during AnMBR operation, indicating no severe membrane fouling propensity was encountered.

Lew et al. (2009) on the other hand, suggested that at temperature of 25 °C, using an HRT and OLR ranging from 4.5 to 12 h, and 1.08 to 4.32 kg COD/m^3/d, an innovative external G-AnMBR, in which the traditional cross-flow external membrane unit was replaced by a microfiltration, hollow fiber, dead end external unit placed below the bioreactor effluent exit, would provide the most energy efficient measure of municipal wastewater treatment than other AnMBR configurations, due to the enough transmembrane pressure provided by the height difference between the bioreactor and the membrane which allowed no pump used for recirculation or transmembrane pressure enhancement, promoting energy savings. Furthermore, as suggested by the authors, intermittent backwash was adopted for fouling amelioration instead of gas bubbling scouring most often found in other studies (Lin et al., 2009; Huang et al., 2011; Martin-Garcia et al., 2013), and the best backwash frequency for energy savings and fouling mitigation of 30 - 60 min. EPS was found as the fouling agent during slow linear increase of fouling rate, according to the observation of the sulfate and aliphatic accumulation on the membrane.

In order to widely apply G-AnMBR technology, anaerobic reclamation of low strength municipal sewage should be maintained under ambient temperature (7 - 20 °C) due to the excessive energy cost for heating (Martin et al., 2011). However, problems still remain in terms of psychophilic methane fermentation. For instance, the anaerobic process requires a long start-up period, its performance may be unstable, and the kind of wastewater that can be treated is limited due to the activity of methanogen (Ikuo et al., 2010). Lettinga et al. (2001) suggested that AnMBR operation at psychrophilic temperature is technically feasible, although SRT must be maintained twice those commonly applied in mesophilic operation, causing SRTs of 120 - 160 d. Nevertheless, in light of the limitation of anaerobic metabolism below 20 °C, only partial solid hydrolysis and incomplete digestion of volatile fatty acids into methane are achieved, resulting in the increase in colloidal and soluble solids content in anaerobic effluents and on membrane fouling propensity (Kashyap et al., 2003).

Martin-Garcia et al. (2013) compared the treatment efficiency and membrane performance of a granular and suspended growth anaerobic membrane bioreactor (G-AnMBR and S-AnMBR respectively) for 250 days treating settled sewage under UK weather conditions, and concluded the impact of configuration was negligible with COD and BOD removal of 80 - 95% and >90%. As temperature dropped from 20 °C to 10 °C, COD removal efficiency experienced noticeable reduction from 97% to 78%, due to the production of non-biodegradable organics rather than the accumulation of VFAs at lower temperatures. This study also confirmed the lower fouling potential in the G-AnMBR as compared to the S-AnMBR, due to the reduced solid and colloidal load (by a factor of 10 and 3) to the membrane which was allowed by the enhanced interception of solids in the granule bed of the G-AnMBR as a result of mixed liquid recycle from the membrane tank to the bioreactor at a low upflow velocity. Similarly, van Voorthuizen et al. (2008) reported the much severer fouling in an anaerobic MBR as compared to an UASB coupled to membrane unit due to the accumulation of higher amount of colloidal matter. They suggested the reduced colloidal matter was mainly due to G-AnMBR biodegradation of dissolved organics taken place predominately within the granules, and colloidal particles arising from the influent solids (Lant and Hartley, 2007) physically adsorbed and retained in the granule bed protecting the membrane from their influence on fouling. Therefore, the granular system would require lower gas sparging intensity and lower energy requirements for fouling control (van Voorthuizen et al., 2008). Moreover, energy efficiency could be further enhanced especially when backwashing was implemented within the granular AnMBR (Martin-Garcia et al., 2013).

Singh et al. (2006) reported that the dead space of the UASB reactor was 10% - 11% and it was dependent upon the operating temperature, which meant the smaller volume of mixing zone the more by-pass flow could occur in the reactor when low temperature was applied. Therefore, UASB reactors treating municipal wastewater at low and moderate temperatures are sometimes characterized by a poor mixing regime, which causes a decrease in soluble COD treatment efficiency. To resolve this issue, tall reactors with a higher ratio of height to width and external or internal effluent recirculation, so-called EGSB reactors, are increasingly applied to provide a very high mixing intensity and efficient wastewater-biomass contact induced by the high upflow velocity. In the study of Chu et al. (2005), a U shape hollow fiber membrane submerged in the upper part of EGSB, was utilized for treating domestic wastewater during 7-month period in the range 11 - 25 °C, and at HRT of 3.5 to 5.7 h. At temperatures above 15 °C, the combined system had the capability of removing 85 - 96% of total COD and 83 - 94% of TOC despite HRT variations. However, at 11 °C, increasing HRT from 3.5 to 5.7 h contributed to the enhanced COD removal from 76% to 81%, which indicated the significance of HRT for low strength wastewater treatment at lower temperature. Upflow velocity, as the other important parameter governing hydraulic mixing, was found significant in achieving better effluent removal efficiency, and a higher membrane permeability due to a rinsing effect on the membrane at the low temperature. Nevertheless, the granule segregation was also induced by the high upward velocity, which was found from granule size distribution and SMA test along the sludge bed. This study showed that psychrophilic sewage treatment using EGSB reactor coupled with membrane technology is feasible, due to the intensified mass transfer between substrate and microbes and viable retention of granule sludge comprised predominantly of filamentous Methanothrix-like species.

Although many studies have proved the competitive advantages of such G-AnMBRs configurations in municipal wastewater treatment, hydraulic selection pressure required for granulation is minimized by the membrane barrier in these cases, through the avoidance of the washout of flocculent sludge with poor immobilization characteristics (Ozgun et al., 2013), thus resulting in a sludge bed with poor settling properties in granule reactors in the long term. Furthermore, the high fraction of partly degradable particulate matter in domestic wastewater, which can be entrapped and gradually accumulated in the granular sludge bed, may further impede the applicability of granular reactors in a G-AnMBR system configuration in the long-term operation.

2.4.3.4 Aerobic granular-membrane bioreactor (AG-MBR)

Compared to G-AnMBR studies, the research on AG-MBR was found relatively limited. Li et al. (2005) proposed a membrane bioreactor system, namely membrane bioreactor with aerobic granular sludge (MGSBR), for municipal wastewater treatment and membrane fouling control. The results showed excellent COD and nitrogen removal efficiencies and the membrane permeability of MGSBR was 50% higher than that of a conventional floc based membrane bioreactor (MFSBR), indicating significantly reduced fouling propensity. However, changes in characteristics of aerobic granules (smaller average diameter and poorer settleability) were also reported in MGSBR, which was mainly attributed to the overgrowth of filamentous bacteria (Li et al., 2007a). Further research on suppressing the overgrowth of filamentous bacteria is greatly needed in operating MGSBR system. Tay et al. (2007) investigated a novel AG-MBR and found such a system had much better filtration performance compared with conventional membrane bioreactor, which was due to much better filtration characteristics of AG-MBR mixed liquor as a result of the low compressibility of granular biomass. Moreove, Yu et al. (2009) suggested that EPS including proteins and polysaccharides, were largely bound with cells as a part of granules, hence leaving low amount of free EPS in supernatant causing membrane fouling. Wang et al. (2010) reported that application of aerobic granule could improve sludge filtering properties and control membrane fouling in the AG-MBR, due to better morphological structure and lower EPS content. Although aerobic granules could effectively reduce membrane fouling, severe pore blockage was still found in AG-AnMBR and foulants were mainly proteins and polysaccharides (Jun et al., 2007). Apart from the traditional AG-MBRs, the researchers have also started to explore novel AG-MBR such as continuous-flow bioreactor with aerobic granular sludge and self-forming dynamic membrane (CGSFDMBR) (Liu et al., 2012b), aerobic granule-mesh filter MBR (AG-MMBR) system (Li et al., 2012) and a continuous flow membrane bioreactor (CFMBR) (Sajjad et al., 2016) for low cost and efficient wastewater treatment.

The critical review of the current studies has revealed that there is a great need in developing a novel G-AnMBR for domestic wastewater treatment. Particularly, pollutants removal efficiencies, renewable energy generation, and fouling control would be the there main areas that need to be considered in developing such G-AnMBR.

CHAPTER 3
EXPERIMENTAL INVESTIGATION

3.1 Introduction

This chapter gives detailed information on materials used and procedure followed for a series of experimental investigations conducted to achieve the objectives of this research outlined in Chapter 1. In this study, four different types of systems were used, namely:

- Preliminary G-AnMBR system
- Submerged and external G-AnMBR systems
- Sponge assisted-granular anaerobic membrane bioreactor system (SA-GAnMBR)
- Sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor (SAAMB-AnGMBR)

The experimental setups, operation conditions, analytical methods used are included separately in this Chapter.

3.2 Materials

3.2.1 Synthetic wastewater

Synthetic wastewater simulating domestic wastewater just after primary treatment was used in the experiments to avoid any fluctuation in the feed concentration and provide a sufficient source of biodegradable organic pollutants. The synthetic wastewater was comprised of organics and macronutrients such as glucose, ammonium sulphate, potassium dihydrogen phosphate, and trace nutrients (Table 3.1). A concentrated stock solution was prepared for 5 days use and stored in a refrigerator at 4 ± 0.5 °C. The synthetic wastewater was produced by diluting the stock solution with tap water and was fed continuously and evenly to the treatment system. The synthetic wastewater (e.g. C/N/P = 100/5/1) contained dissolved organic carbon (DOC) of 100.0 - 120.0 mg/L, COD of 330.0 - 370.0 mg/L, ammonia nitrogen (NH_4-N) of 12.5 - 15.6 mg/L, nitrite nitrogen (NO_2-N) of 0 - 0.02 mg/L, nitrate nitrogen (NO_3-N) of 0.2 - 0.8 mg/L, and orthophosphate (PO_4-P) of 3.1 - 3.6 mg/L. Ammonia nitrogen might vary depending on

C/N ratios required for each experiment, and were specified in Chapter 4 - 7. $NaHCO_3$ or NaOH was utilized to adjust pH to 7.

Table 3.1 Characteristics of the synthetic wastewater (displayed in the case of C: N: P = 100: 5: 1)

Compounds	Chemical formula	Molecular weight (g/mol)	Concentration (mg/L)
Organics and nutrients			
Glucose	$C_6H_{12}O_6$	180.0	280.00
Ammonium sulphate	$(NH_4)_2SO_4$	132.1	72.00
Potassium phosphate	KH_2PO_4	136.1	13.20
Trace nutrients			
Calcium chloride	$CaCl_2 \cdot 2H_2O$	147.0	20.00
Manganese chloride	$MnCl_2 \cdot 7H_2O$	197.9	0.20
Zinc sulphate	$ZnSO_4 \cdot 7H_2O$	287.5	0.44
Ferric chloride anhydrous	$FeCl_3$	162.2	13.50
Cupric sulphate	$CuSO_4 \cdot 5H_2O$	249.7	0.39
Cobalt chloride	$CoCl_2 \cdot 6H_2O$	237.9	0.42
Sodium molybdate dehydrate	$Na_2MoO_4 \cdot 2H_2O$	242.0	1.26
Magnesium sulphate	$MgSO_4 \cdot 7H_2O$	246.5	5.00
Nickel sulphate	$NiSO_4$	154.8	0.50
Yeast extract	-	-	30.00
COD	-	-	300
C: N: P			100: 5: 1

3.2.2 Membrane

U-shape hollow fibre membranes manufactured from Tianjin Polytechnic University were used for experiments. Each membrane was comprised of 50 fibre threads with a length of 400 mm, and was made of PVDF material configured with a pore size of 0.22µm and a filtration area of 0.06 m^2. Fouled hollow fibre membrane was taken out for physical (sponge scrubbing) and chemical cleaning using the following three steps: 6 h in 0.4% sodium hydroxide, 6 h in 0.5% citric acid, and 6 h in 0.8% sodium hypochlorite.

Figure 3.1 U-shape hollow fiber PVDF membrane module

3.2.3 Sponge

Porous polyester-urethane sponge cubes (dimensions: 2 mm × 2 mm × 2 mm), namely S28 - 30/90 R (density of 28 - 30 kg/m^3 with 90 cells per 25 mm, Joyce Foam Products) were used for experiments.

Figure 3.2 Sponge carrier used in the studies

3.3 Experimental set-up and operation protocol

3.3.1 Preliminary experiments

In the preliminary experimental stage, a laboratory-scale G-AnMBR was studied for domestic wastewater treatment at 20.0 ± 0.5 ˚C in a temperature-controlled room (Figure 3.3).

Figure 3.3 Experimental setup for preliminary G-AnMBR

For the G-AnMBR, the hollow fiber membrane was immersed in the subsequent membrane tank located after UAGB. Membrane tank was fed with the UAGB effluent and a suction pump was operated with an intermittent suction cycle of 8 min on and 2 min off to acquire permeate from the membrane module. The purpose of the on/off cycle was to relax membrane unit and prevent the membrane fouling. Membrane flux was controlled by adjusting the pump speed and two calibrations were made each day. The membrane fouling was indicated by an increase in TMP, which was recorded by a pressure transmitter. When TMP reached 30kPa, the membrane module was taken out from the reactor and cleaned chemically. With regards to the reactor seed, the available acclimatized aerobic sludge was adopted as the inoculum. Aerobic activated sludge with MLVSS of 10 g was put into a glass carboy of 400 mL, and incubated at laboratory temperature for 14 days. Initially, a certain amount of synthetic wastewater containing 0.5 g COD was added into the carboy each day and the same amount of supernatant was removed. The amount of wastewater added was increased gradually, and finally contained 2 g COD each day. Subsequently, the sludge was transferred to the reactor. Before the continuous feed operation, the reactor has been fed intermittently for 14 days in advance of continuous feed. The main operational parameters were summarized in Table 3.2.

Table 3.2 Operation conditions for the preliminary research on G-AnMBR

Operational phase	A	B	C
Operational time (d)	1 - 30	30 - 60	60 - 90
Upflow velocity (m/h)	0.525	0.700	1.050
Inflow (mL/h)	257	343	514
HRT (h)	16	12	8
Flux ($L/m^2 \cdot h$)	4.3	5.7	8.6
OLR (kg $COD/m^3 \cdot d$)	0.46	0.61	0.92

3.3.2 G-AnMBR system (submerged and external configuration)

Two G-AnMBRs with equal working volume of 4 L, namely EG-AnMBR and SG-AnMBR were operated in parallel at 20.0 ± 0.5 °C in the Environmental Engineering lab at the University of Technology Sydney (Figure 3.4). Both G-AnMBRs were fed with identical inoculated anaerobic sludge with similar initial sludge concentration (21.48 ± 0.98 g/L for the EG-AnMBR, 21.41 ± 1.12 g/L for the SG-AnMBR) at the beginning of the experiments. For the EG-AnMBR, the hollow fiber membrane was immersed in the subsequent membrane tank located after UAGB. Membrane tank was fed with the UAGB effluent and a suction pump was operated with an intermittent suction cycle of 8 min on and 2 min off to acquire permeate from the membrane module. While in the SG-AnMBR, an identical membrane module was directly immersed into the mixed liquor at the settling zone of the UAGB. Both systems were operated at a constant filtration rate of 7 $L/m^2 \cdot h$, HRT of 12 h, and upflow velocity of

0.7 m/h. The membrane fouling was indicated by the development of the normalized TMP, which was recorded by a pressure transmitter. When TMP reached 30 kPa, G-AnMBR operation was terminated.

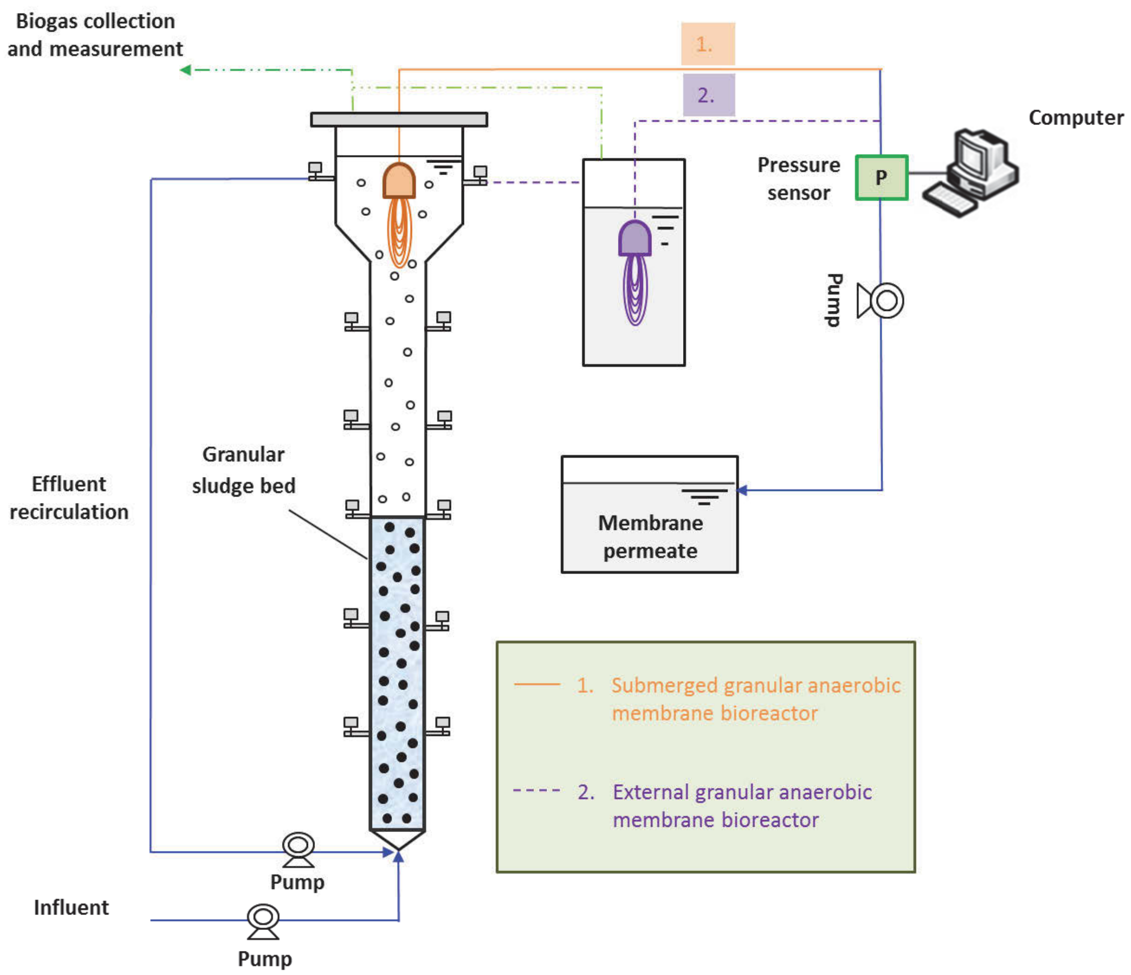


Figure 3.4 The experimental setup of SG-AnMBR and EG-AnMBR

3.3.3 Sponge assisted-granular anaerobic membrane bioreactor (SA-GAnMBR)

A CG-AnMBR and a SG-AnMBR with the same effective working volume (3 L) were operated in parallel at 20.0 ± 0.5 °C in a temperature controlled room (Figure 3.5). The anaerobic sludge (MLSS = 22.34 ± 0.41 g/L, MLVSS = 17.41 ± 0.38 g/L, SVI = 98.5 mL/g, Mean particle size = 58 µm, Temperature = 21 °C and pH = 7.5) was acclimatized to synthetic wastewater for 30 days until a stable treatment performance was reached. The two reactors were fed with identical acclimatized anaerobic sludge with MLSS of 20.50 ± 1.53 g/L in the reaction zone. A PVDF hollow fiber membrane was immersed in the mixed liquor at the settling zone of each reactor. A vacuum driven peristaltic pump was employed to feed influent into UAGB. The other suction pump was operated with an intermittent suction cycle of 8 min on and 2 min off to acquire permeate from the membrane module. The purpose of the on/off cycle was to relax membrane unit and prevent the membrane fouling. Porous polyester-urethane sponge cubes were added into the UAGB of the SG-AnMBR together with the inoculated sludge, and sponge volume fraction was 10% working volume. The CG-AnMBR and SG-AnMBR were operated at a constant filtration rate of 5.3 L/m^2·h with HRT of 12 h till membrane was fouled. Upflow velocity of 3.2 m/h was maintained using internal recirculation. The membrane fouling was indicated by development of the normalized TMP, which was recorded by a pressure transmitter. When TMP reached 30 kPa, operation was terminated.

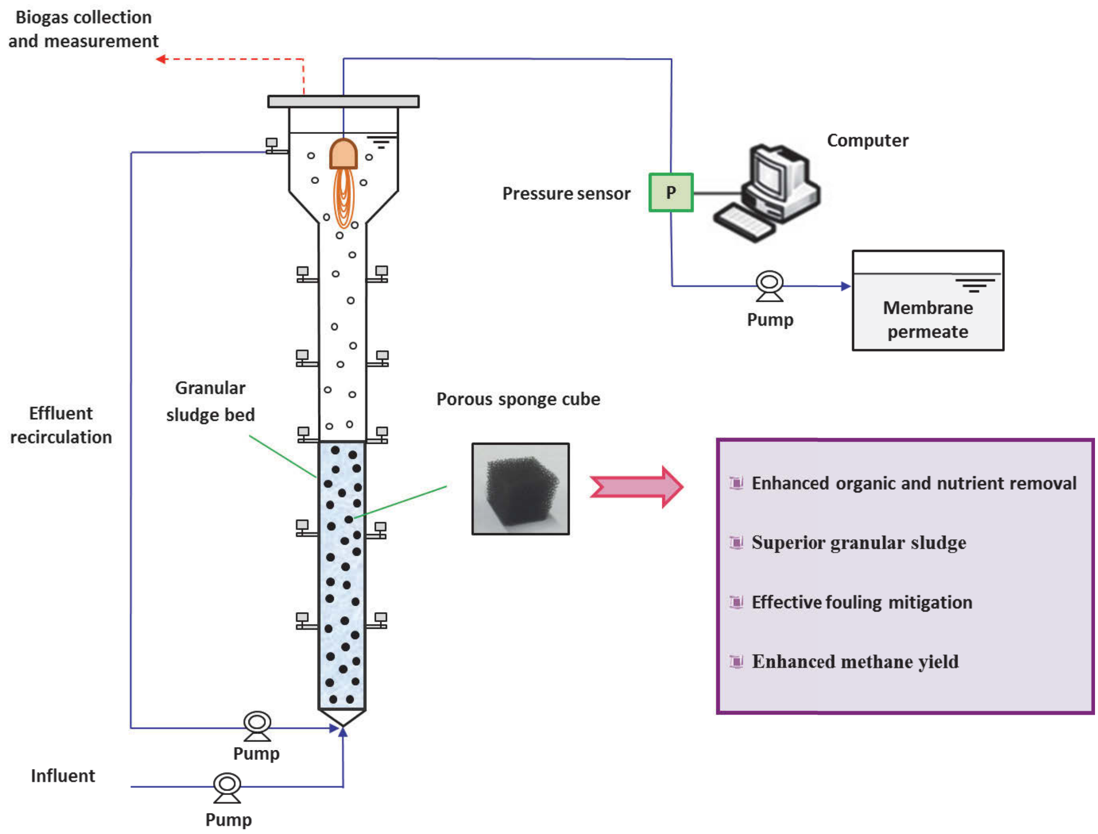


Figure 3.5 The experimental setup of SA-GAnMBR

3.3.4 Sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor (SAAMB-AnGMBR)

The hybrid SAAMB-AnGMBR, consisting of a sponge-assisted aerobic moving bed reactor (SAAMBR) and a submerged sponge-assisted anaerobic granular membrane bioreactor (SS-AnGMBR), was continuously operated for 282 days in a temperature-controlled room (20.0 ± 0.5 ºC). Each of the SAAMBR and the SS-AnGMBR had effective working volume of 3 L, and sponge fraction was 20% of working volume. At the bottom of the SAAMBR, fine bubble diffuser was set to supply air in order to provide complete liquid-solid mixing and moderate sponge up/down motion, and maintain dissolved oxygen (DO) concentration of 3.5 - 4.8 mg/L. Prior to continuous operation, the SAAMBR with fresh sponge was acclimatized to synthetic wastewater for 30 days at HRT of 12 h until the system reached relatively stable treatment performance. The attached growth on the sponge also reached steady state at 1.020 ± 0.041 g MLVSS/g sponge. The sponges and anaerobic granular sludge were acclimatised to synthetic wastewater for 30 days until a stable treatment performance was reached. The SS-AnGMBR was seeded with anaerobic granular sludge with initial MLSS concentration of 20.12 ± 1.21 g/L, and biomass grown on sponge cubes after acclimatization was 1.78 ± 0.093 g MLVSS/g sponge. A polyvinylidence (PVDF) hollow fiber membrane module with a pore size of 0.22 μm and surface area of 0.06 m^2 was immersed in the settling zone of the SS-AnGMBR.

The SS-AnGMBR was continuously fed with synthetic wastewater at a flow rate of 4.17 mL/min while wastewater from the SS-AnGMBR was continually transferred into the SAAMBR at the same flow rate. The SAAMBR effluent was recirculated back to the SS-AnGMBR through a nitrogen gas sparged buffer tank. The permeate pump in the SS-AnGMBR was operated in an intermittent mode with relaxation (8 min on and 2 min off) to acquire permeate from the membrane module with a constant filtration flux of 5.21 LMH. Both SAAMBR and SS-AnGMBR had HRT of 12 h, and upflow velocity in the SS-AnGMBR was maintained at 3.2 m/h using internal recirculation. The membrane fouling propensity was indicated by normalized trans-membrane pressure (TMP), which was recorded by a pressure transmitter. Operation was terminated when

TMP exceeded 30 kPa, and fouled membrane was taken out for ex situ cleaning (Deng et al., 2016a).

The entire study period was divided in 2 phases according to the research objectives. In phase 1, the hybrid SAAMB-AnGMBR was fed with wastewater having C/N/P ratio = 100/5/1 (0 - 75 day), 100/6/1 (76 - 126 day), 100/8/1 (126 - 151 day) and 100/10/1 (151 - 166 day), respectively, with the aim to investigate the impact of C/N ratios on the performance of the hybrid system. In phase 2, after overloaded nitrogen events, the hybrid system was operated with C/N/P ratios of 100/6/1 (167 - 210 day) and then 100/5/1 (211 - 282 days) to investigate the extent of system recovery after the overloading nitrogen event.

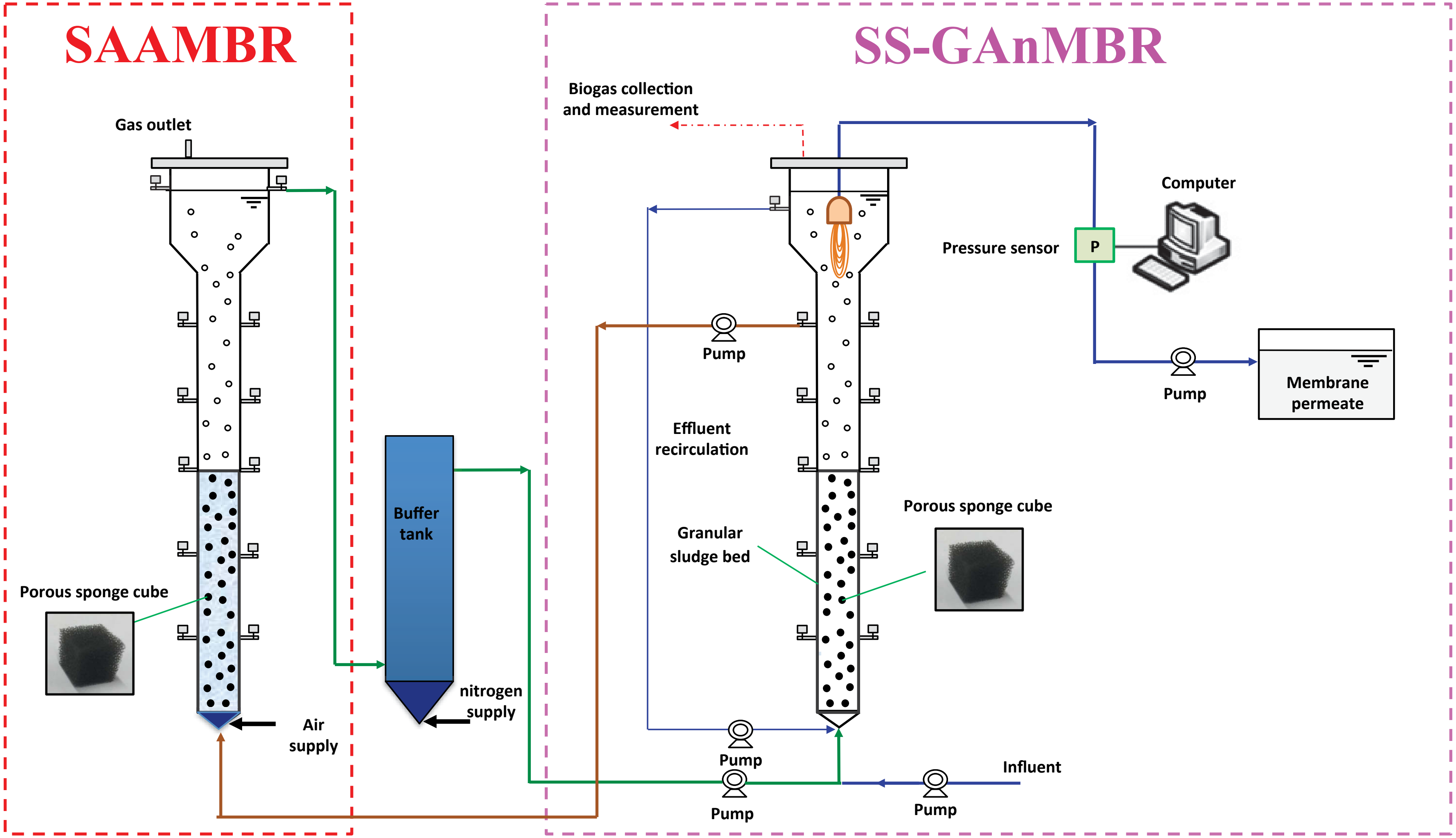


Figure 3.6 The experimental setup of SAAMB-AnGMBR

3.4 Analytical methods

For the experimental investigation, all the liquid, gas and sludge samples were tested in triplicate, with an average value and standard deviation for discussion.

3.4.1 Organics, nutrients, pH, ORP and DO

Influent and effluent (membrane permeate) were taken from the outlets of feed pump and permeate pump everyday for analysis. Both influent and effluent were required to undergo filtration by a 0.45μm filter to remove fine suspended material and any biomass, and stored in the refrigerator before being analyzed. Total organic carbon (TOC) and dissolved organic carbon (DOC) of the influent and effluent were measured using a DOC/TOC analyzer (Analytikjena Multi N/C 2000). The analysis of COD was carried out according to Standard Methods (APHA, 1999). NH_4-N, NO_2-N, NO_3-N and PO_4-P were measured by spectrophotometric method using Spectroquant Cell Test (NOVA 60, Merck). The pH and temperature of the reactor were measured everyday using pH meter (Hach Company, model no. HQ40d). Oxidation and reduction potential (ORP) meter (Vernier Labquest) and DO meter (Horiba Ltd. Japan, model no. OM-51E) were used to monitor ORP and DO everyday. The analytical instruments used were shown in Figure 3.7.

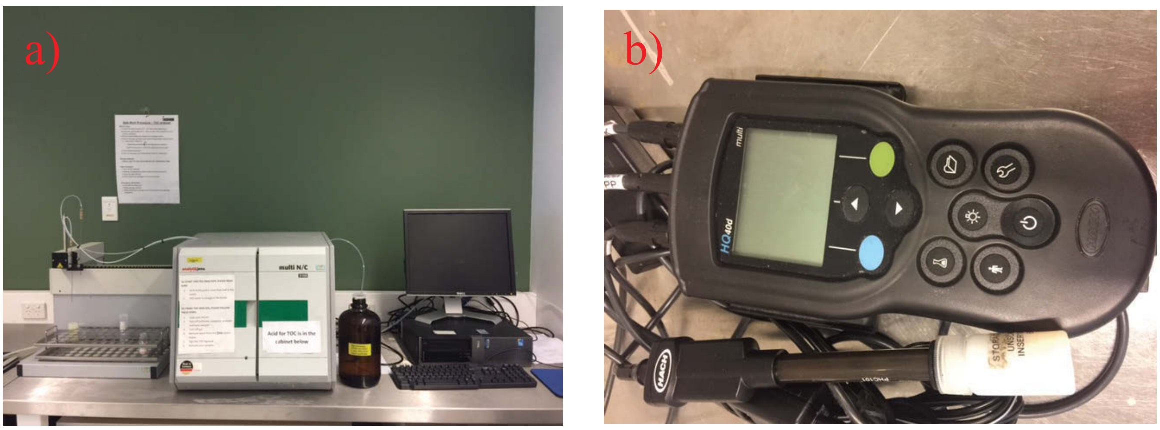


Figure 3.7 The analytical instruments used in the experimental investigation: a) DOC analyser; b) pH meter; c) ORP meter; d) DO meter

3.4.2 Sludge properties

3.4.2.1 Sludge volume index (SVI), settling velocity and zeta-potential

Physical characteristics of granular sludge such as SVI, settling velocity and Zeta-potential were analysed every three days. The determination of the SVI was carried out based on Standard Methods (APHA, 1999). The settling velocity was determined by monitoring the fall of randomly selected granules. This was accomplished by gently releasing the granules at the top of a 2 L vertical glass cylinder with 80 mm internal diameter and 560 mm height, filled with the treated reactor effluent. The zeta potential (surface charge) of activated sludge in this study was determined using Zetasizer Nano ZS (Malvern Instruments, UK) (Figure 3.8)

Figure 3.8 The instrument for Zeta-potential measurement

3.4.2.2 Particle size distribution (PSD) and microscopic observation

PSD of granule sludge was determined by using the laser particle size analysis system Mastersizer Series 2000 (Malvern Instruments Ltd. UK) with a detection range of 0.02–2000.0 mm. The scattered light was detected by means of a detector that converted the signal to a size distribution based on volume. Each sample was measured three times with a standard deviation of 0.1 - 4.5%. Mean diameter was used to represent the size of the sludge particles. D (0.1) (i.e. 10% of the volume distribution was below this value) was used to describe the colloidal and fine particle fractions. The sludge granules were examined by Olympus System Microscope Model BX41 (Olympus, Japan) and the images were captured and analyzed using Image-Pro Plus software.

3.4.2.3 Biomass concentration and growth rate

The granular sludge was collected at 3 sampling port at different heights of the UAGB (Port 1: 20 cm, Port 2: 40 cm and Port 3: 60 cm height from the bottom) twice a week. MLSS and MLVSS measurements were conducted based on the Standard Methods (APHA, 1999). A well-mixed sludge sample was first gone through filtration using 1.2 μm glass microfiber filter paper (47 mm DIA, Filtech). The retained sludge residue on the filter was dried in an oven at 105 °C for 2 h. The increment in weight of the filter paper indicated MLSS in the sample. Subsequently, the filter paper was ignited in a furnace at 550 °C for 20 min. The weight reduced during ignition represents MLVSS in the sample. The biomass growth rate is calculated based on following Equation 1:

$$\frac{\Delta \mathrm{MLSS}}{\Delta \mathrm{t}} = \frac{\mathrm{MLSS_B} - \mathrm{MLSS_A}}{B-A} \tag{1}$$

Where $\frac{\Delta \mathrm{MLSS}}{\Delta \mathrm{t}}$ indicates biomass growth rate (g/d); $MLSS_B$ is MLSS concentration of granular sludge sample when operation is terminated; $MLSS_A$ is MLSS concentration of granular sludge sample when operation is started, B - A is the number of days that the system is operated.

For the attached-growth biosolids on the sponge, three sponge cubes were taken out from the reactors, and were further hand-squeezed and thoroughly washed-off using with Mill-Q water. The attached growth rate was calculated according to Equation 2:

$$\frac{\Delta \mathrm{MLSS}}{\mathrm{Sw}} = \frac{\mathrm{MLSS_{B1}} - \mathrm{MLSS_{A1}}}{Sw} \quad (2)$$

Where $\frac{\Delta \mathrm{MLSS}}{Sw}$ indicates attached biomass growth rate (g/g sponge); $\mathrm{MLSS_B}$ is MLSS concentration of biosolids attached on the sponge when operation is terminated; $\mathrm{MLSS_A}$ is MLSS concentration of sponge-attached biosolids when operation is initiated, Sw is the weight of sponges obtained for analysis.

3.4.3 Nitrification and denitrification rate

Batch tests were conducted according to methods proposed by Gong et al. (2012), to evaluate the specific nitrification rate (SNR) and specific denitrification rate (SDR). The attached-growth biomass onto sponge carriers was determined by the method suggested by Nguyen et al. (2011). DO concentration was kept around 5 mg/L by aeration for determining specific nitrification rate (SNR) while the value was maintained below 2 mg/L for determining specific denitrification rate (SDR). Allylthiourea (ATU) of 1 mg/L was added as the nitrification inhibitor into the synthetic substrate to eliminate the influence of nitrifiers for the determination of SDR. SNR and SDR were calculated by the decreasing slope of NH_4-N and NO_3-N concentrations with time and divided by the initial MLSS concentration.

3.4.4 Volatile fatty acid (VFA) and biogas

VFA, namely acetate acid, propionic acid, butyric acid, isobutyric acid, iso-valeric acid and n-valeric acid were extracted using methyl-tert-butyl ether (MTBE) for liquid-liquid extraction according to the methods reported by Banel and Zygmunt (2012) (Figure 3.9).

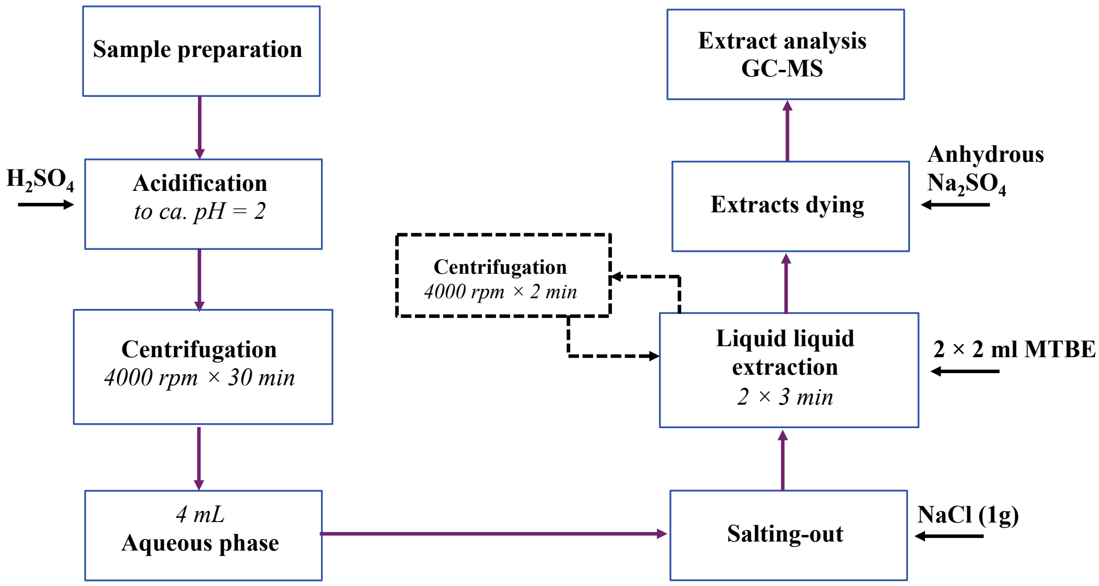


Figure 3.9 The schematics of the procedure for VFA extraction

Six VFAs were pretreated using 0.45 µm syringer filter, and were further quantified by gas chromatogram mass spectrometry method (GCMS TQ8040, Shimadzu, Japan) using an open tubular analytical column (VF-WAXms, Agilent, US). An injection port equipped with a 1 mm internal diameter (ID) liner operated in splitless mode (after 1 min, split ratio was 1:10) was maintained at a temperature of 230 ˚C. Temperature program started at 50 ˚C and was held for 5 min before ramping to 250 ˚C at 10 ˚C/min and was then held for 10 min. Helium was a carrier gas operated at a flow rate of 2.05 mL/min. Electron impact ion source was set at 230 ˚C while the injection port and transfer line temperatures were held at 230 ˚C. Mass spectrometer (MS) was operated in a selected ion monitoring (SIM) mode and in a full scan mode (m/z 15-550). Ions for detection of individual VFA in SIM mode were selected using the mass spectra of standards generated in SCAN mode. Biogas production was collected using a biogas sample bag and determined using a liquor displacement device. Biogas composition including CH_4, CO_2, H_2 and N_2 was determined using potable biogas analyser (Biogas 5000, Geotech, UK).

3.4.5 Membrane fouling determination and fouling rate

The experiments were terminated when TMP reached 30 kPa. Fouling rate indicated the development of membrane fouling, and its value was calculated based on Equation 3:

$$\frac{\Delta \mathrm{TMP}}{\Delta \mathrm{t}} = \frac{\mathrm{TMP_B} - \mathrm{TMP_A}}{B-A} \quad (3)$$

Where $\frac{\Delta \mathrm{TMP}}{\Delta \mathrm{t}}$ indicates membrane fouling rate (kPa/d); TMP_B is the transmembrane pressure value (kPa) when operation is terminated; TMP_A is the transmembrane pressure value (kPa) when operation is started, B - A is the number of days that the system is operated (d).

3.4.6 Soluble microbial products (SMP) and extracellular polymeric substances (EPS) extraction and measurement

The extraction and analysis of EPS and SMP in the granular sludge, cake layer and mixed liquor from the G-AnMBR were based on the methods provided by Deng et al. (2014) (Figure 3.10). The extracted samples were further analysed for protein (EPS_P and SMP_P) and polysaccharide (EPS_C and SMP_C) concentrations, using modified Lowry method (Sigma, Australia) and Anthrone-sulfuric acid method, respectively.

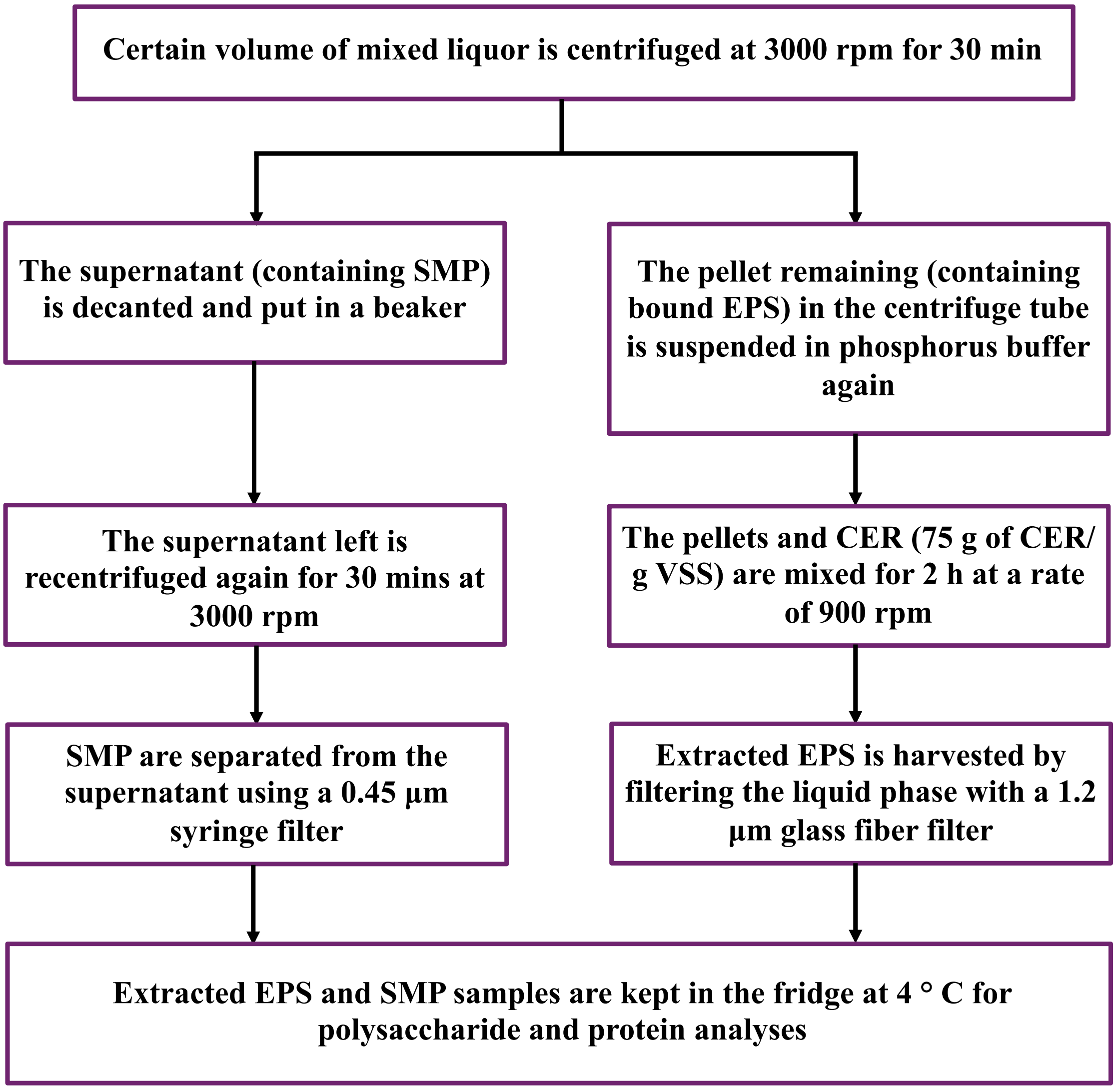


Figure 3.10 The procedure for SMP and EPS extraction (Modified from Deng (2015))

3.4.7 Fouling resistance determination

Based on the resistance-in-series model, fouling resistance of the G-AnMBR was determined after G-AnMBR experiments by using measurement protocol proposed by Deng et al. (2015) and applying Equation 4 and 5 (Choo and Lee, 1996):

$$J = \frac{\Delta P}{\mu R_T} \tag{4}$$

$$R_T = R_M + R_C + R_P \tag{5}$$

Where J is the permeation flux (m^{-3} m^{-2}/ h^{-1}); ΔP is the transmembrane pressure (Pa); μ is the dynamic viscosity of the permeate (Pa s); R_T is total resistance (m^{-1}); R_M is the intrinsic membrane resistance (m^{-1}); R_C is the cake layer resistance (m^{-1}); and R_P is the pore blocking resistance (m^{-1}). The linear relationship of flux∗viscosity and TMP can be established by plotting the TMP curve against membrane flux∗viscosity, and the gradient indicates the corresponding membrane resistance.

3.4.8 Foulant characterization

3.4.8.1 Liquid chromatography-organic carbon detection (LC-OCD)

Foulant attached on the surface of membrane was extracted with 2 L of 0.4% NaOH solution using a horizontal shaker for 3 h. The extract was filtered through 1.2 μm glass fibre filter and filtrate was then diluted to ensure the DOC level was less than 5 mg L^{-1} before being analyzed. Size exclusion LC-OCD, a TSK HW 50-(S) column and a 0.028 mol/L phosphate buffer (Figure 3.11) was used to analyse the hydrophilic and hydrophobic fractions of the membrane foulant. The hydrophilic fraction was further quantified as biopolymers, humic substances, building blocks, low molecular weight (LMW) acids and neutrals.

Figure 3.11 LC-OCD analysis instrument at University of New South Wales

3.4.8.2 Excitation emission matrices (EEMs)

Three-dimensional Fluorescence measurements (excitation emission matrix, (EEM)) of foulant were obtained using a Varian Cary Eclipse Fluorescence Spectrophotometer, USA, according to methods suggested by Hong et al. (2012).

CHAPTER 4

PRELIMINARY RESEARCH ON GRANULAR ANAEROBIC MEMBRANE BIOREACTOR (G-ANMBR)

4.1 Introduction

Anaerobic granular bioreactor (AnGBR) technology has been widely adopted in treating industrial and municipal wastewater since 1980 due to its competitive advantage in renewable energy production (Lettinga et al., 1984). However, AnGBR process has to face the following challenges to realize worldwide applications: (1) the requirement of extremely long start-up period; (2) a relatively high operation temperature required; (3) unsuitability for low strength organic wastewater such as municipal wastewater; (4) post treatment required to meet discharge standards (Liu et al., 2004; Liu and Tay, 2004; Lim and Kim, 2014). Recent research efforts have been mainly directed to the discovery of specific high rate AnGSB bioprocess that could resolve the above-mentioned demerits. Theoretically, the sufficient inoculation of seed granular sludge in operating AnGSB has the advantage of achieving high organics removal within an accelerated startup period. Some researchers also used additives such as natural polymers, cationic polymers and hybrid polymers to enhance particle agglomeration, in order to shorten startup time and enhance granulation (Show et al., 2004; Wang et al., 2004; Jeong et al., 2005; Tiwari et al., 2005; Cao et al., 2010). Other researchers devoted themselves into the development of hybrid AnGBR for the enhancement of anaerobic granulation. Zhang et al. (2009) showed that rapid startup could be successfully accomplished by utilizing a hybrid UASB - AFF reactor with internal hydraulic circulation and external sludge circulation. Jung et al. (2013) also reported high rate circulation could accelerate the formation of hydrogen-producing granules in a UASB.

Other researchers have been working on modifying the reactor design and developing hybrid anaerobic systems for treating various types of wastewater. Ikuo et al. (2010) reported that an EGSB bioreactor, a modified UASB configuration, had the capability of treating low strength wastewaters at low temperatures. Li et al. (2007b) studied the performance of an integrated EGSB-Zeolite bed filtration (EGSB-ZBF) hybrid system for the removal of carbon and nutrients from low strength wastewater at 35 °C for 7 months and the combined system could effectively reduce COD by 71.58%, and completely remove ammonia and phosphate. Anaerobic ammonium oxidation

(ANAMMOX) - EGSB combined system was reported as a novel nitrogen removal process that achieved the nitrogen removal efficiency up to 94.68% (Chen et al., 2011).

As advanced membrane-based separations are well suited to water recycling and reuse, membrane coupled AnGBR technology (G-AnMBR) is now experiencing a rapid growth as a tertiary treatment process for treating municipal wastewater as compared to suspended growth anaerobic membrane bioreactors (S-AnMBRs) (Herrera-Robledo et al., 2010; Herrera-Robledo et al., 2011; Liu et al., 2013). In this study, G-AnMBR was studied for the efficient granules development using aerobic sludge as inoculum. The effects of HRT on the granulation and G-AnMBR performance (organic and nutrient removal, sludge properties and fouling propensity) were also studied.

4.2 Modification of G-AnMBR configuration

Initially, a G-AnMBR was started up using an upflow anaerobic reactor with a 4 L working volume (Dimension: 13cm×13cm×45cm), in which a hollow fibre membrane was submerged. A vacuum driven peristaltic pump was employed to feed influent into the anaerobic reactor. The suction pump was operated with an intermittent suction cycle of 8 min on and 2 min off to acquire permeate from the membrane module. HRT was maintained at 16 h and SRT was infinite since no sludge discharge was implemented. The characteristics of the synthetic wastewater were shown in Table 3.1 (Chapter 3), and C/N/P ratio was 100/5/1. The membrane properties were depicted in Chapter 3 (Section 3.2.2). The highest COD removal efficiency observed in this trial was 69.2% on Day 35 and the average value was at 55.2 ± 14.3% (Table 4.1).

Organic removal was significantly lower than the values gained by other researchers. COD and TOC reduction over 90% by AnMBRs were reported by many researchers (Hu and Stuckey, 2006; Huang et al., 2011; Liu et al., 2013). As for the nutrient removal, TN removal was at 8.2 ± 3.7%, while PO_4^{3-}-P reduction was at 7.8 ± 4.5% (Table 4.1). Nutrient removal was not expected in the G-AnMBR system because the removal of TN and PO_4^{3-}-P required the anoxic or aerobic zone.

Table 4.1 Removal efficiencies of the G-AnMBR trail

Parameters	TOC	COD	NH_4-N	TN	PO_4-P
Influent concentration (mg/L)	100-120	330-370	12.5-15.6	12.7-16.6	3.1-3.6
Effluent Concentration (mg/L)	335-584	113-219	10.5-14.8	10.1-16.2	2.8-3.5
Removal efficiencies (%)	58.3 ± 17.7	55.2 ± 14.3	10.3 ± 5.8	8.2 ± 3.7	7.8 ± 4.5

The reactor design was regarded as the main cause leading to the unexpected low performance. In this experiment, a retangular shape reactor with a fairly large cross-sectional area was used for granulation process and G-AnMBR operation. Such a large cross-sectional area could not allow upflow wastewater to be evenly distributed across the area, resulting in poor mixing regime and low upflow hydraulic force. Upflow velocity was regarded as an important parameter to represent the upflow hydraulic force and the level of mixing regime. The calculated upflow velocity in this case was 0.0015 m/h, which was greatly below the reported values of 2.5 m/h (Liu et al., 2012a; Liu et al., 2013). By reducing the cross-sectional area, the upflow velocity can be increased since upflow velocity equals influent flowrate divided by cross-sectional area.

The reported dead space of the UASB reactor was 10% - 11% and it was dependent upon the operating temperature (Singh et al., 2006). The volume of mixing zone became small and the by-pass flow in the reactor increased when temperature was low. The anaerobic sludge was found deposited at the bottom of the reactor, which meant sludge in this case was not suspended in the reaction region. This implied that only limited space in the reactor was used to degrade substrate. The low rates of hydraulic flow at HRT of 16h and biogas ascension at a low organic loading rate of 0.6 kg COD/m^3·d further deteriorated the performance (Zhang et al., 2009). Specifically, the low biogas production during startup prevented effective lifting of sludge, resulting in a low mass transfer and slow startup. A long HRT accompanied with a low upflow liquid velocity might allow dispersed bacterial growth and be less favorable for granulation (Liu and Tay, 2004). Under conditions of such low selection pressure, the growth took place predominantly as dispersed biomass, which gave rise to the formation of a bulking type of sludge (Figure 4.1) and granule sludge was not found during observations.

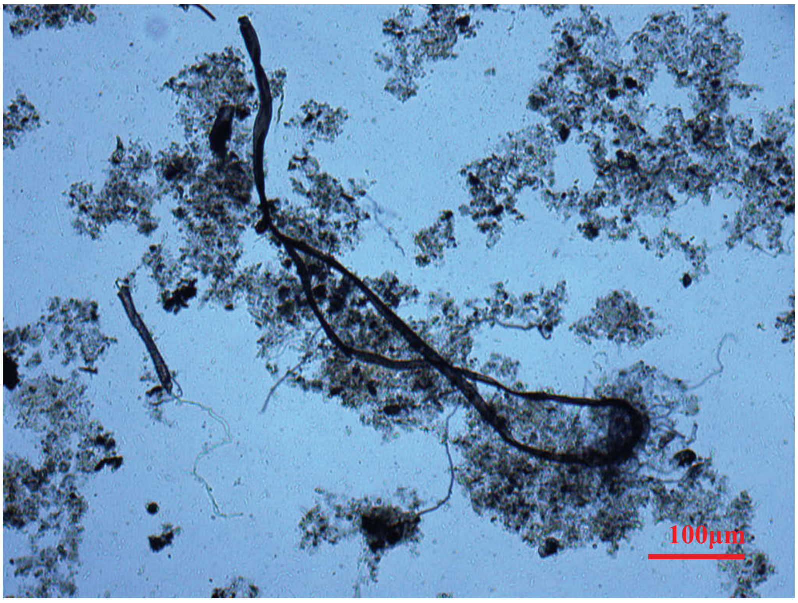


Figure 4.1 Microscopic observation of reactor sludge on Day 50

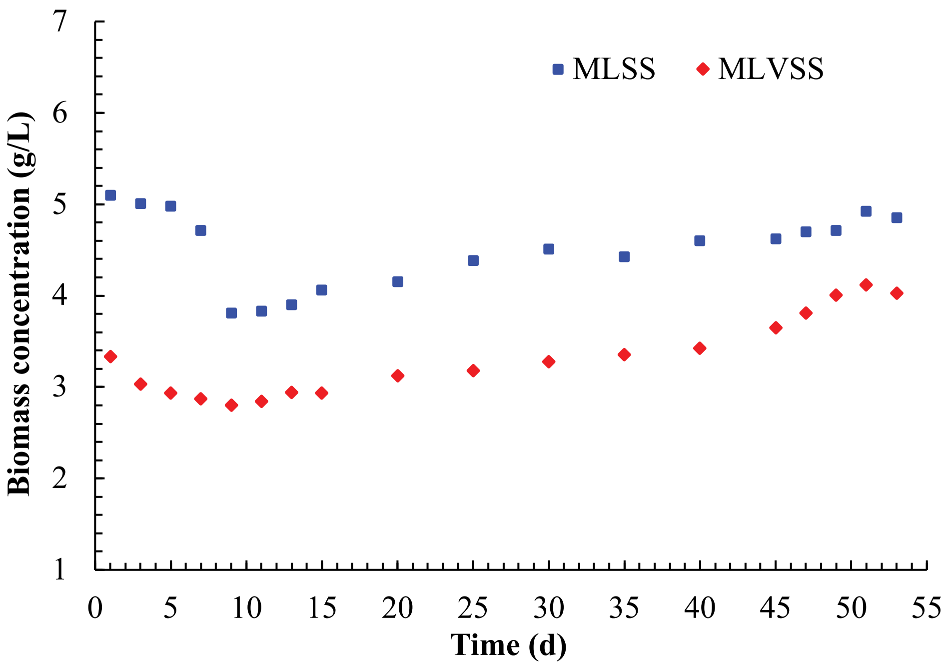


Figure 4.2 Biomass concentrations over the operation time

The initial inocula of sludge (MLSS) was 5 g/L in the reactor. After startup, the biomass concentrations in G-AnMBR experienced a rapid decrease. This was because the original sludge innoculum probably contained a significant amount of insoluble material such as fine solids, and cell debris or sand (Hu and Stuckey, 2006). When the MLSS concentrations decreased over time to a steady state, MLVSS values approached the MLSS values. The initial ratio of MLVSS to MLSS in G-AnMBR was 0.64, whereas the final ratio was 0.85, with a MLVSS of approximately 4.5 g /L (Figure 4.2). This implied that most of the inactive material probably passed through the membrane, or attached to the membrane surface during the run. In this study, no sludge purge was undertaken except the sludge withdrawn for analysis every 5 days. Even though such a long SRT was operated, the sludge concentration did not increase and it was because of the relatively short operation time and the low loading rate, which exclusively restricted the multiplication of microorganisms.

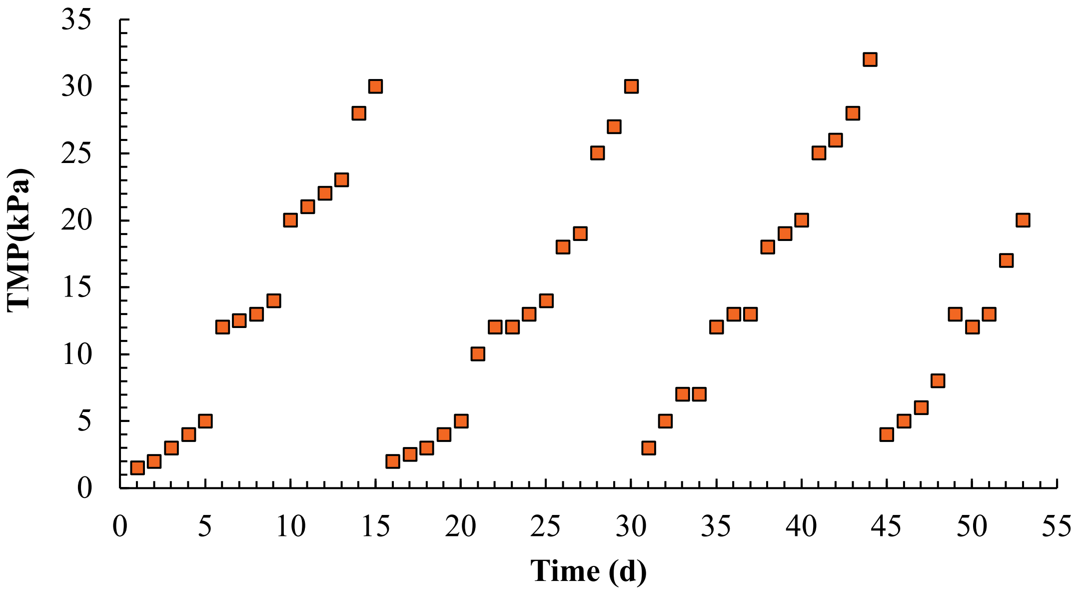


Figure 4.3 TMP profile over the operation time

TMP, as the indication of fouling propensity, was monitored continuously throughout the G-AnMBR operation. TMP value increased very quickly (Figure 4.3) as a result of serious fouling, and fouling rate was found at 2.04 kPa/d. Fouled membranes were taken out for physical and chemical cleaning for four times. The dense cake formation was observed on the surface of the membrane, and this was mainly due to the sludge bulking and floating issues. As mentioned above, a loosely intertwined structure of filaments, with very poor settling characteristics was obtained in the sludge zone. Submerged membrane was directly exposed to the bulking sludge and therefore was fouled very quickly. Moreover, through the attachment of gas bubbles to these loosely intertwined filaments, the sludge had a tendency to float to the membrane surface and cause sludge deposition on the membrane surface.

Therefore, the reactor design required further modification according to following comments, in order to achieve better treatment performance.

(1) Column with smaller cross sectional area (i.e. small diameter) is required to provide sufficient uniform upflow velocity for mixing and granular growth.

(2) Distinguished sludge zone and sedimentation zone are required to better retain sludge at the sludge zone, and produce low effluent TSS loading on membranes so as to reduce membrane fouling.

(3) Adequate expansion zone (higher height to diameter ratio) is required to avoid sludge washout to the sedimentation zone.

(4) Upflow velocity should be considered as the key parameter acting as the selection pressure for granule formation.

(5) More attention should be paid to the granule formation, and analysis of parameters such as granular bed height, granule diameters and microscopic images of granules were highly desired.

The reactor in the previous study was modified to the lantern shape reactor, with the main reaction region (sludge zone) as the small column and a settling zone at the top sedimentation tank (Section 3.3.1, Chapter 3). Therefore, the dead space of the reactor could be minimized and poor sludge distribution could be overcome when the wastewater was fed into the system from the bottom. The granules were expanded and the hydraulic mixing was intensified, giving granules a greater chance to contact with wastewater. Suspended solids were settled at the large sedimentation tank, resulting in effluent with low TSS before entering the membrane unit.

Therefore, membrane fouling was much less expected in this case since membrane was only challenged by sludge supernatant. The granulation is time consuming. The reasons for the slow granulation are slow growth of methanogens and excessive sludge washout. For the modified reactors, when the uplow velocity was imposed in the reactor, dispersed sludge was separated from the mature granules at the reaction region and temporarily stored at the top settling zone due to the absolute biomass retention by membrane. These filaments were periodically washed back to the reaction region due to the turbulence, and were further engaged in forming new granules.

4.3 Effect of HRT on organic and nutrient removal

The proposed system was tested against three different operation conditions at HRT of 16, 12 and 8 h. The detailed information regarding experimental set-up and procedure were shown in Chapter 3 (Section 3.3.1). At the startup operational stage (HRT = 16 h), the upward liquid velocity was maintained at 0.52 m/h at the reaction zone. This low level of hydraulic stress allowed flocculant sludge to bond and form the anaerobic microbial aggregates. The increased upflow velocity at HRT of 12 h acted as a selection pressure for biomass by washing out light and dispersed particles, while retaining heavier biomass at the bottom of the reactor to form the granular sludge bed. By the Day 55, the anaerobic granular sludge bed was well developed. Figure 4.4 showed that the G-AnMBR had the highest organic removal (92.89% of TOC reduction and 92.73% of COD reduction) at HRT of 12 h on Day 55.

For the longer HRT of 16 h, the highest COD and TOC removal was below 85%. At HRT of 16 h, upflow velocity was maintained at a low level of 0.52 m/h. Under such a long HRT accompanied with a low upflow liquid velocity, it allowed dispersed bacterial growth and was less favourable for the growth of methane-producing granules. Therefore, the treatment efficiency remained below the optimal value achieved at HRT of 12 h. Anaerobic sludge was also found deposited at the bottom of the reactor at HRT of 16 h, and hence not all the sludge was suspended in the reaction region degradating the substrates. This was because the insufficient hydraulic flow and the biogas ascension could not provide efficient biomass distribution in the sludge zone (Zhang et al. 2009). Specifically, the low biogas production during startup impeded the effective lifting of sludge, which resulted in low mass transfer and low level of treatment.

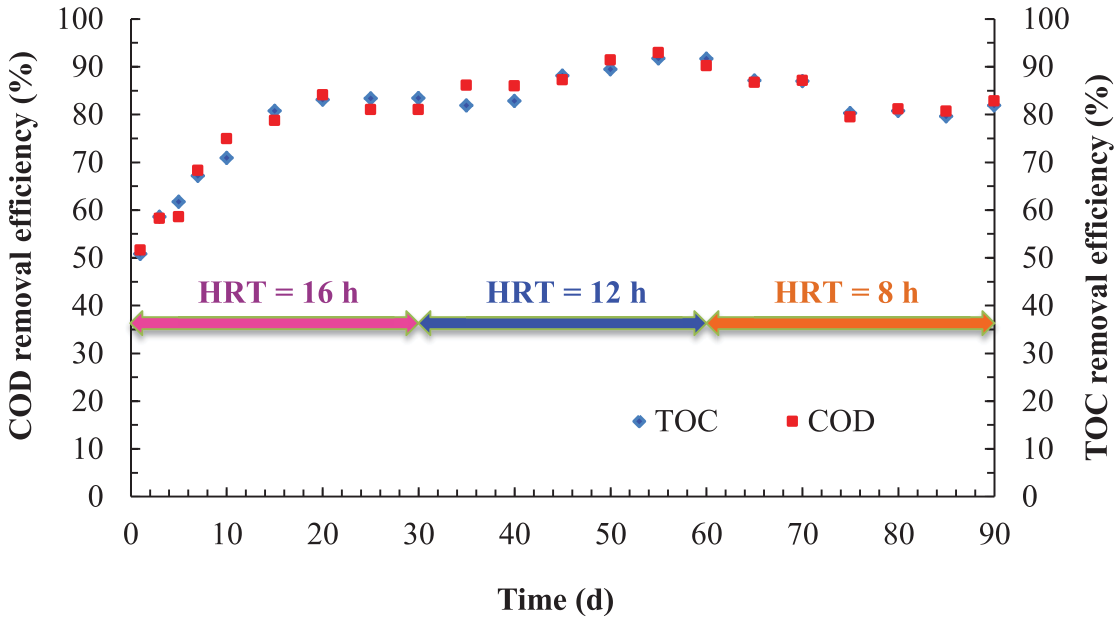


Figure 4.4 Organic removal efficiency of the modified G-AnMBR

When the reactor was operated at HRT of 8h, the upflow velocity (shear force) and biogas production were enhanced but organic removal efficiency was reduced to 84.23 ± 4.23% (Figure 4.4). This deteriorated removal efficiency was mainly ascribed to the granular sludge disintegration and the washout of granular sludge. With the growth of granules, more biogas that produced (due to the increased organic loading as a result of decreasing HRT) inside granules was more difficult to be released. Together with excessive external shear stress caused by upflowing liquid and rising gas bubbles, the fracture of large granules might occur (Yan and Tay, 1997). It was found that the mean diameter of granules was decreased from the highest value at 251 μm on Day 55 to 224 μm on day 90.

Zhang et al. (2009) reported that excessive hydraulic pressure could lead to the mutual collisions among sludge granules, resulting in the fragmentation of the granular sludge and a sharp decrease in the effectiveness of the reactor performance. They explained this using the structural model of granulation. Methanothrix acted as the core of the sludge granules, while acetate producers and fermentative bacteria formed the exterior layer around the core. Disintegration of the sludge might lead to the exposure of Methanothrix to an acidic environment, which might cause a sharp decrease in the organic removal. There are other explanations for the increase of COD and TOC in the effluent at HRT of 8 h. According to Salazar-Pelaez et al. 2011(b) and Wang et al. (2009), the higher shear stress forces inside of the reactor when it was operated at low HRTs, could cause stress conditions for microorganisms. Hence, more biopolymeric substances were released in its effluent and resulted in COD increase in the effluent. Leitão et al. (2005) also reported that when operating under low HRTs, the limited contact time between microorganisms and substrate, and the decreased filtration capacity of the sludge bed occurred at higher upflow velocities. Therefore, a part of influent COD was allowed to leave the reactor without proper treatment.

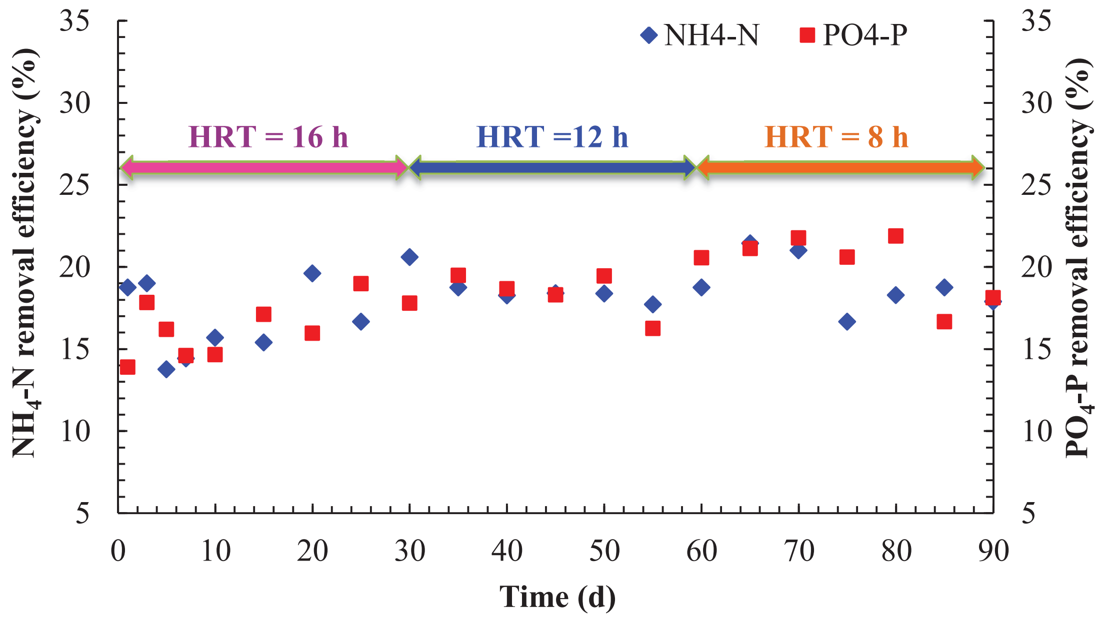


Figure 4.5 Nutrient removal efficiencies of the modified G-AnMBR

In terms of nutrients, Figure 4.5 showed that ammonium and phosphate removal efficiencies fluctuated around 18.0 ± 2.1% and 18.1 ± 3.2%. The low NH_4-N and PO_4-P elimination were attributed to the fact that nutrient removal was not much expected in anaerobic treatment. The results also revealed that HRT had neglegible impact on the nutrient removal of G-AnMBR.

4.4 Effect of HRT on sludge properties and fouling

Initially, floc sludge formed and a considerable number of flocs were washout to the sedimentation tank. Periodically, bulking sludge in the sedimentation tank was washed back to the main sludge zone due to hydrodynamic turbulence. On Day 7, granules-like flocs appeared at the bottom of the reactor (Figure 4.6a). Based on the theory of 'microbial nuclei growth', methanothrix bacteria formed very small aggregates at the initial stage, owing to the turbulence generated by the biogas production, or the attachment to finely dispersed matter. Once these precursors were formed, the growth of the precursor particles to form granules was inevitable. This was due to the growth of the individual bacteria and the entrapment of non-attached bacteria to the precursors. These spherical granules mainly consisted of loosely intertwined filamentous bacteria attached to an inert particle (filamentous granules).

By Day 55, the sludge zone was full of granules (Figure 4.6b) with mean diameter at 255 μm at HRT of 12h (Figure 4.7). The rapid sludge granulation might be attributed to the wastewater composition (biodegradable high-energy carbohydrate feeding) and reactor configuration. It was believed that more compact rod-type granules were formed from these filamentous granules at a high SRT, due to the increase in the density of the bacterial growth. According to Rajakumar et al (2012), lowering HRT could result in increasing upflow velocity and enhanced hydraulic shear force. The increased selection pressure, therefore, enhanced the formation of both short rod type (Methanobacterium formicum) and dense long fibers loosely intertwined filamentous bacteria granules of Methanothrix sp. (Methanosaeta concilii). Very small particles of these thin filamentous functioned as primary nuclei on which Methnothrix bacteria was attached, initiating the granulation process.

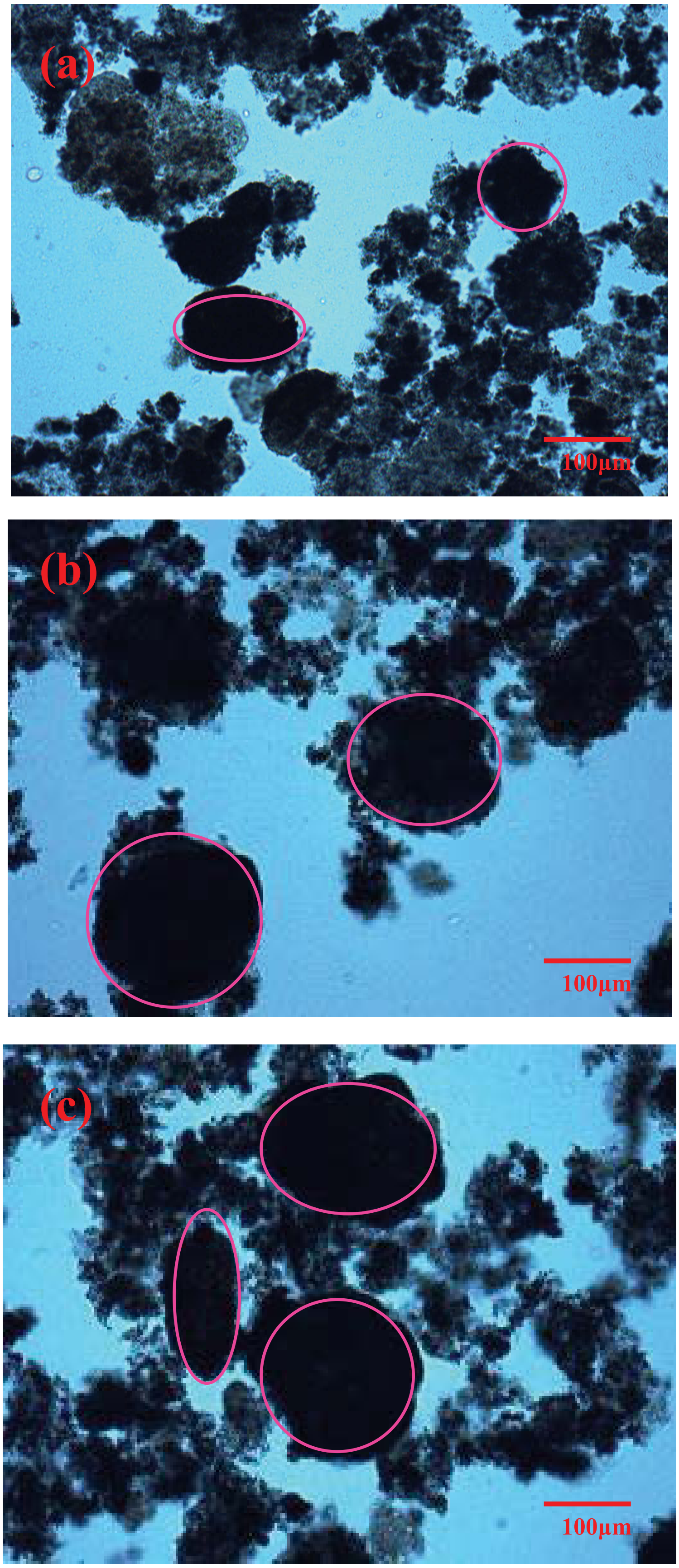


Figure 4.6 Microscopic observations of anaerobic granules on Day 7 (a), Day 55 (b), and Day 85 (c)

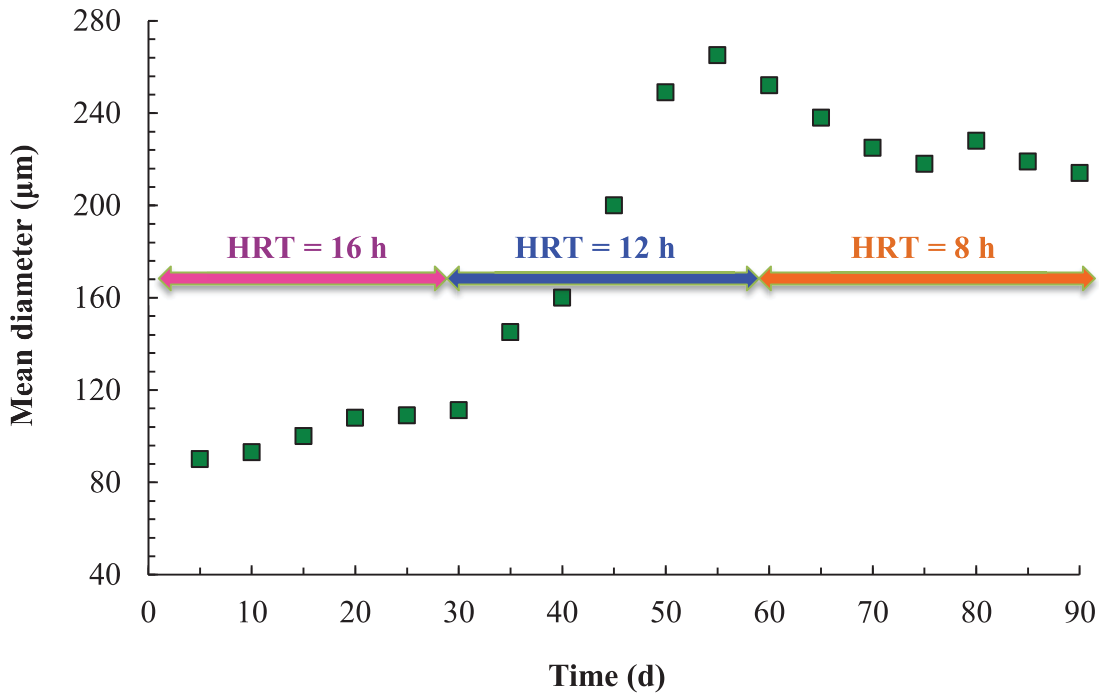


Figure 4.7 Granule growth course of the modified G-AnMBR

When HRT was reduced to 8h, the system was not able to sustain the healthy growth of granules and loosely structured granules were presented (Figure 4.6c). This could be seen from the reduced diameter of granules to 224 µm (Figure 4.7) and massive bed expansion from Day 65 to Day 75 (Figure 4.8). Kalyuzhnyi et al. (1996) suggested that when the system was operated at low OLRs, the granules mainly consisted of filaments of Methanothrix cells during the treatment of sucrose based synthetic wastewater. These loosely structured filamentous granules could be destructed more easily under high liquid and gas upflow force, thus leading to the granule disintegration and sludge washout.

Figure 4.8 showed the MLSS and MLVSS growth profile. The biomass growth course corresponded to the trend of the granular diameter growth (Figure 4.7), with slow growth at HRT of 16 h, rapid incline at HRT of 12 h, and final drop after HRT of 8h was imposed. Maximum granules diameter was observed on Day 55 with mean diameter of 255 um. This value was lower than the other reported (2 - 5mm), and this might be attributed to low organic loading, seed sludge characteristics, low strength wastewater characteristic and relatively shorter granulation period.

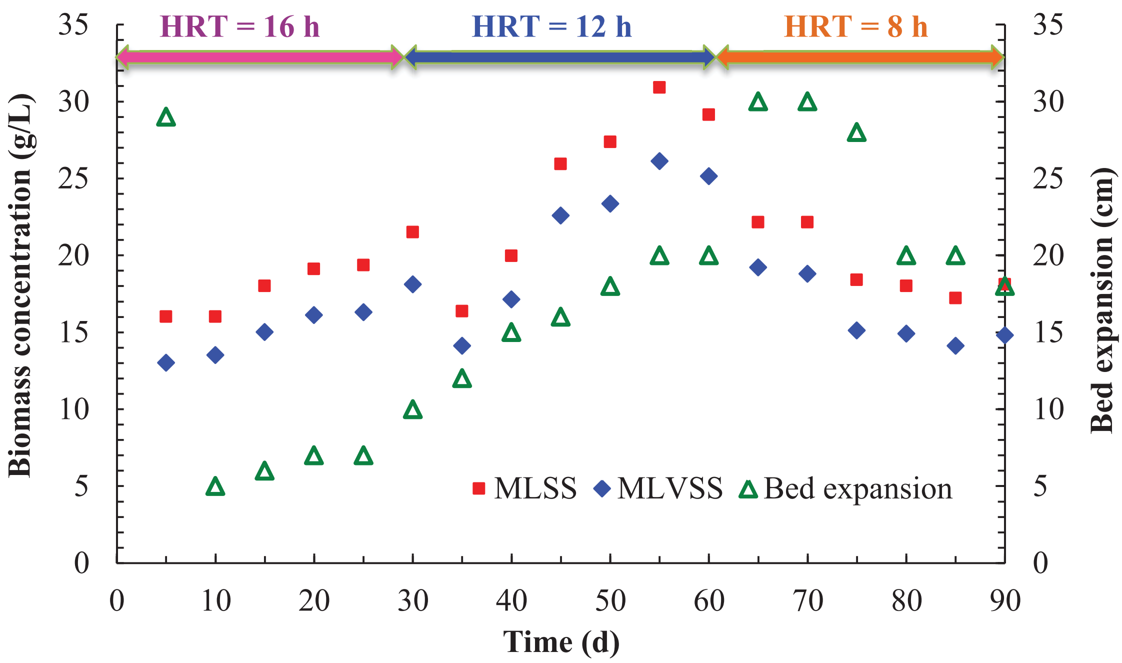


Figure 4.8 MLSS, MLVSS and bed expansion of the modified G-AnMBR

4.5 Conclusions

Flocculent aerobic sludge could be successfully granulated using low strength synthetic wastewater used as substrate. HRT exerted a profound influence on the performance of G-AnMBR including organic removal, granular sludge properties and fouling propensity. The mean diameter of granules increased to the maximum value of 255µm and the sludge was well distributed in the reacting region when HRT was reduced from 16 h to 12 h. Highest organic removal efficiencies were reached at more than 92% at HRT of 12 h. Good performance of the reactor occurred due to the good hydraulic conditions, rapid granulation, and the low loss of sludge. However, further decrease of 12 h to 8 h did not lead to increased granule size and COD removal, and actually caused a sharp decrease in the effectiveness of the granules and reactor performance. The size of the granules found in this study were much lower than some research studies and this was attributed to the seeded sludge characteristics (aerobic sludge with average floc diameter of 30 µm), low strength wastewater characteristics, low OLR (microorganisms may be starved of nutrients at low OLRs) and relatively short granulation period.

CHAPTER 5

IMPACT OF REACTOR CONFIGURATION ON THE PERFORMANCE OF A G-ANMBR

5.1 Introduction

Membrane fouling has remained as one of the most challenging issues impeding the progress of AnMBRs (Sanguanpak et al., 2015; Huang et al., 2012; Saleem et al., 2016), especially with high biomass concentration in widely used conventional AnMBRs (C-AnMBRs). In view of this concern, many researchers have devoted their efforts into developing various AnMBR configurations such as V-AnMBRs (Kola et al., 2014), Gl-AnMBRs (Gimenez et al., 2012), AnBEMRs (Ng et al., 2014), AnDMBRs (Saleem et al., 2016) and AnMSBRs (Kim et al., 2014) for sustainable fouling mitigation strategies (Section 2.3.1.2, Chapter 2). G-AnMBR, a hybrid anaerobic biotechnology that incorporates the granular technology with membrane-based separation, has offered a promising approach to the C-AnMBR in terms of fouling mitigation (Chen et al., 2016a) (Section 2.4.3.2, Chapter 2).

The predominated configuration of current G-AnMBR operation for municipal wastewater treatment was found as the external G-AnMBR (EG-AnMBR) where membrane filtration was applied as a polishing stage for UAGB effluent (Section 2.4.3.3, Chapter 2). The main advantages included undisturbed hydraulics in the UAGB, and the ease of operation and membrane cleaning. Nevertheless, Ozgun et al. (2015) elucidated that the EG-AnMBR might be encountered with the progressive increase in the SS loading on the membrane unit. Very few researchers employed submerged membrane in the SG-AnMBR to provide nearly absolute biomass retention and allow for operation at nearly infinite SRTs (Chu et al., 2005). Membrane, in this case, not only acted as a physical barrier for active biomass retention, but also promoted a general cultural adaptation to the prevailing organic loading conditions in the SG-AnMBR (Liu et al., 2013). On the other hand, Liu et al. (2012a) pointed out membrane filtration could exacerbate sludge bioflocculation in the SG-AnMBR and induced greater cake resistance, resulting in more serious fouling. To date, no references have been found to compare the two mainstream G-AnMBRs for the treatment of municipal wastewater.

The objective of this chapter is, therefore, to determine which type of G-AnMBR configurations is favourable for municipal wastewater treatment. To this aim, a direct comparison of external and submerged membrane operation in G-AnMBR (namely EG-AnMBR and SG-AnMBR) was conducted. The comprehensive evaluation of the two G-AnMBRs included the investigation of treatment efficiencies (organics and nutrient removal), granules properties (e.g. particle size, settling velocity and EPS), membrane fouling behaviour (TMP, potential foulants, fouling resistance analysis), and renewable energy recovery (methane yield).

A major part of Chapter 5 has been published in the following paper:

Chen, C., Guo, W.S., Ngo, H.H., Chang, S.W., Nguyen, D.D., Nguyen, P.D., Bui, X.T., Wu, Y., 2017a. Impact of reactor configurations on the performance of a granular anaerobic membrane bioreactor for municipal wastewater treatment. International Biodeterioration and Biodegradation 121, 131-138.

5.2 Effect on organic and nutrient removal

The experimental set-up and operation procedure were shown in Chapter 3 (Section 3.3.2). The characteristics of municipal wastewater could be found in Chapter 3 (Section 3.2.1) and COD: N: P ratio was maintained at 100: 2: 1. The membrane specification was depicted in Chapter 3 (Section 3.2.2).

Organic removal efficiency over 90% was achieved in both G-AnMBRs (Table 5.1). The EG-AnMBR removed 92.6 ± 2.3% of DOC and 91.9 ± 1.5% of COD while 91.8 ± 1.9% of DOC and 91.3 ± 2.1% of COD were eliminated by the SG-AnMBR. The relatively high organic removal efficiencies could be attributed to the complete retention of all particulate and colloidal matter, and biomass by membrane. The influent COD also contained the majority of readily biodegradable COD using glucose as the sole carbon source, which contributed to the high organic removal (Martinez-Sosa et al., 2011). The EG-AnMBRs exhibited NH_4-N reduction at 24.2 ± 6.3% and PO_4^{3-}-P elimination at 12.2 ± 4.3% while the SG-AnMBR had similar NH_4-N (23.1 ± 5.8%) and PO_4-P (11.3 ± 5.1%) removal. The low ammonia and phosphate removal efficiencies were due to the fact that nutrient removal was not expected in the anaerobic treatment. The results showed that the membrane addition methods had negligible impact on the organic and nutrient removal of G-AnMBRs.

Table 5.1 Removal efficiencies of DOC, COD, NH_4-N and PO_4-P in EG-AnMBR and SG-AnMBR during the operation period

G-AnMBR	DOC (%)	COD (%)	NH_4-N (%)	PO_4-P (%)
EG-AnMBR	92.6 ± 2.3	91.9 ± 1.5	24.2 ± 6.3	12.2 ± 4.3
SG-AnMBR	91.8 ± 1.9	91.3 ± 2.1	23.1 ± 5.8	11.3 ± 5.1

5.3 Effect on granular sludge properties

5.3.1 Granular sludge

The performance of UAGB relies on its capability to form a dense granular sludge bed in the reaction region for efficient physical entrapment and biological degradation of particulate and dissolved organic substances (Seghezzo et al., 1998). The granular sludge MLSS and MLVSS concentrations in the EG-AnMBR gradually increased during operation, reaching 25.23 g/L and 21.78 g/L, respectively at the end of operation on day 75 (Table 5.2). While for the SG-AnMBR, MLSS and MLVSS concentrations were relatively stable at 22.18 g/Land 17.92 g/L, respectively when operation was terminated on day 35. The biomass growth rate (ΔMLSS/Δt) was found at 0.050 g/d in the EG-AnMBR, which was higher than that of the SG-AnMBR (0.022 g/d). In the case of the EG-AnMBR, hydraulics in the UAGB reactor was not influenced by the external membrane incorporation in the subsequent membrane tank. This configuration allowed the selective washout of the flocculent sludge with poor settling ability, which could lead to an increased growth of retained sludge agglomerates and granules. In this case, biodegradation of organics occurred mainly within the granules, promoting the growth of granules rather than dispersed growth of free swimming bacteria (Martin-Garcia et al., 2013). In contrast, as for the SG-AnMBR configuration, the membrane direct addition into the UAGB eliminated the selection pressure on the granules due to a nearly complete retention of small and colloidal flocs, thus resulting in the accumulation of fine sludge flocs and suspended particles inside the UAGB (Ozgun et al., 2015). In this case, the growth of dispersed sludge with poor immobilization properties could predominately take place, resulting in the bulking type of sludge formed in the SG-AnMBR and undermining granular growth.

Table 5.2 Comparision of sludge characteristics of inoculated sludge, granular sludge from G-AnMBRs when operation was terminated

Sludge properties	Seed sludge (EG-AnMBR)	Seed sludge (SG-AnMBR)	Granular sludge (EG-AnMBR)	Granular sludge (SG-AnMBR)
MLSS (g/L)	21.48 ± 0.98	21.41 ± 1.12	25.23 ± 1.19	22.18 ± 0.78
MLVSS (g/L)	17.58 ± 1.25	17.42 ± 1.34	21.78 ± 1.08	17.92 ± 0.98
SVI (mL/g)	38.8 ± 3.1	37.5 ± 2.9	24.5 ± 1.9	72.5 ± 7.9
Settling velocity (m/h)	17.5 - 25.4	18.4 - 25.9	14.1 - 28.5	12.1 - 17.2
Zeta-potential (mV)	-17.5 ± 1.3	-17.9 ± 2.1	-13.1 ± 1.8	-19.1 ± 1.7

The settling ability of the sludge determines the level of biomass retention and physical removal of particulate organics in the UAGB, and influences the fouling propensity when coupling UAGB with membrane separation. Granular sludge from the EG-AnMBR had a better settling ability, which could be seen from the decreased SVI and increased settling velocity values compared to the seed (Table 5. 2). At the end of the EG-AnMBR operation, SVI was found at 24.5 mL/g while settling velocities of granular sludge were around 14.1 - 28.5 m/h. In contrast, the SG-AnMBR contained granular sludge with higher SVI of 72.5 mL/g and lower settling velocity of 12.1 - 17.2 m/h than those of seed sludge and the EG-AnMBR, which revealed the settleability of granular sludge was deteriorated. Zeta potential value of the granular sludge was measured in the EG-AnMBR (-13.1 mV), which was found higher than that in the SG-AnMBR (-19.1 mV). Higher zeta potential suggested that the negative charge on the surface of the flocs could be neutralized to form larger granular sludge with better settling properties (Deng et al., 2014).

5.3.2 Granules

Bhunia and Ghangrekar (2007) have defined that the required minimum granule size was 340 μm with specific gravity of 1.035. Chu and Huang (2005) have suggested granule size distribution ranging from 0.25 mm to 3.75 mm when treating sucrose- and phenol-containing synthetic wastewaters. The reported granule size in Zhang et al. (2011) was 125 - 830 μm while Pevere et al. (2006) recognized bioparticles with the size of 100 μm or even less as anaerobic granules. In this chapter, granular sludge with the size larger than 100 μm was defined as granules since low strength synthetic municipal wastewater was adopted as the feed for the G-AnMBRs. As can be seen from Table 5.3, seed sludge for both G-AnMBRs showed very high similarity in PSD, since the two systems were inoculated with sludge from the same source.

Table 5.3 PSD of seed sludge and granular sludge from G-AnMBRs (%)

PSD	Seed sludge (EG-AnMBR)	Seed sludge (SG-AnMBR)	Granular sludge (EG-AnMBR)	Granular sludge (SG-AnMBR)
< 50 μm	14.3	12.5	5.4	25.9
50 - 100 μm	29.1	28.3	13.2	33.2
100 - 300 μm	39.2	42.3	53.8	36.5
300 - 500 μm	9.4	8.9	14.2	4.2
500 - 1000 μm	7.2	7.3	10.5	3.4
> 1000 μm	0.8	0.7	2.9	0.8

However, significant differences were observed for the granular sludge PSD obtained from two systems at the end of the operation. One-way shift to fine flocs was found in the SG-AnMBR, which could be due to the membrane's complete retention of fine sludge particles (<100 μm) including small particles such as colloidal flocs and macromolecules of SMP, and non-settling particles in the SG-AnMBR. On the other hand, abundant larger-sized granules were developed in the EG-AnMBR. The fraction of granules (>100 μm) was found around 41.9% of the total granular sludge in the SG-AnMBR, which was about half of the value obtained from the EG-AnMBR (81.4%).

EPS serves as an essential element for the formation of anaerobic granules, and is vital for integrating cells into large aggregates and granules and preserving the sound structure of granules. Hence, the lower percentage of granules in the SG-AnMBR also indicated that granules segregation occurred in the granular sludge bed. The SG-AnMBR contained significantly lower total EPS concentration at 6.1 ± 1.2 mg/g MLVSS with both protein and polysaccharides amounts of EPS decreased by 83.3 ± 5.2

% and 76.3 ± 4.2 %, as compared to those of the seed sludge (total EPS, EPS_P, EPS_C at 23.5 ± 3.3, 17.2 ± 2.3, 5.3 ± 1.2 mg/g MLVSS, respectively). The dramatic decrease in EPS amount might suggest looser, scattered and weaker structures of sludge granules in the SG-AnMBR, meaning possible granule fragmentation and decrease in PSD, and SMP increase in mixed liquor (Ozgun et al., 2015). On the other hand, the EPS contents were around 40.5± 3.0 mg/g MLVSS in the EG-AnMBR, in which EPS_P and EPS_C were 31.8 ± 2.1 and 7.5 ± 1.2 mg/g MLVSS, respectively. The higher contents of EPS increased the electrostatic interactions between the microbial biomass and the polymeric material and therefore, rapidly increased the granular sludge particle size and promoted the formation of granules. The total SMP contents (38.5 ± 2.3 mg/g MLVSS) of the SG-AnMBR were found 5.6 times higher than those of the EG-AnMBR (5.8 ± 1.2 mg/g MLVSS). Much lower SMP values of the EG-AnMBR confirmed that the dominant fraction of the proteins and polysaccharides existed as the part of the anaerobic granules as their lower amounts detected in the soluble form as SMP.

5.4 Effect on membrane fouling behaviour

5.4.1 TMP and fouling rate

Figure 5.1 demonstrated the TMP profiles in both EG-AnMBR and SG-AnMBR. Obvious difference was observed in TMP increase from two distinctive G-AnMBRs. For the EG-AnMBR, the gradual increase in TMP was found over the operation period. The increase in TMP was found up to 30 kPa on day 75, and the fouling rate was approximately 0.4 kPa/d. On the contrary, the SG-AnMBR initially presented a gradual increase in TMP from 2.0 kPa to 11.2 kPa until day 24. A sudden TMP jump to 16 kPa was observed on day 25, and TMP reached 32.0 kPa after 35 days. The averaged fouling rate for the SG-AnMBR was around 0.9 kPa/d, which was 2.25 times to the corresponding value obtained from the EG-AnMBR. The results indicated that the SG-AnMBR experienced much faster fouling propensity than that of the EG-AnMBR.

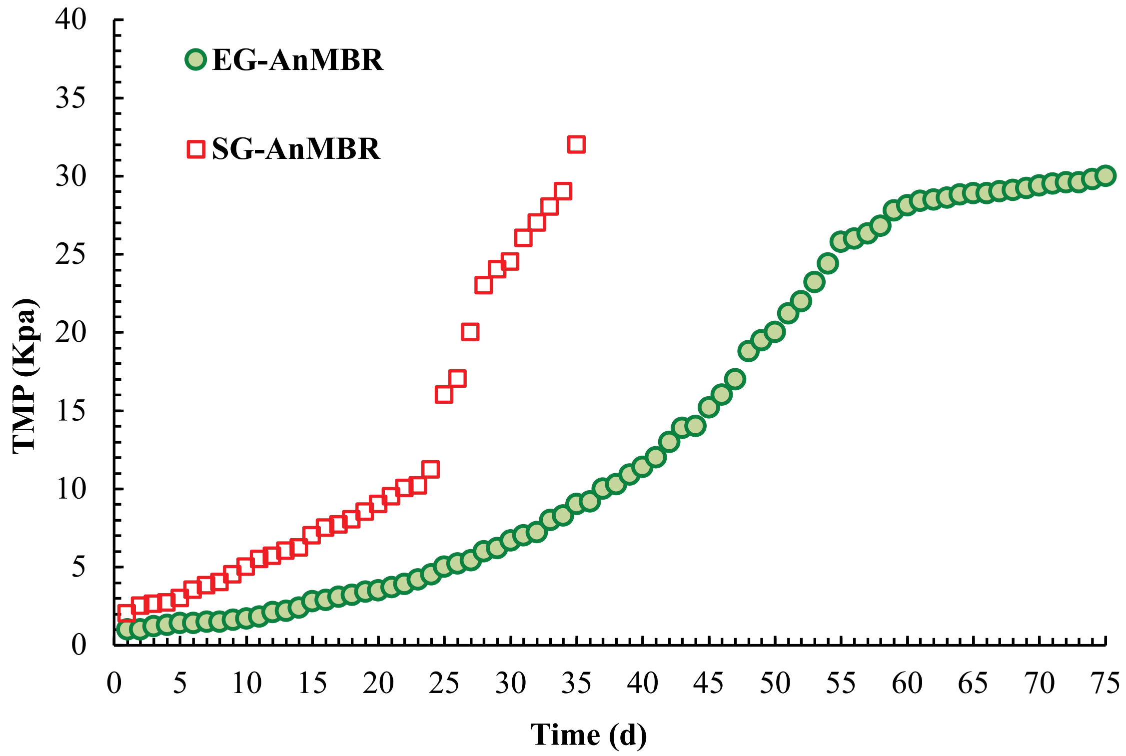


Figure 5.1 TMP values of G-AnMBRs over the experimental period

5.4.2 SMP and EPS in the mixed liquor

EPS clog the membrane pores, accelerating the formation of a strongly attached fouling layer on the membrane surface due to their multiple functions including cell aggregation, formation of a protective barrier and retention of water and adhesion to surfaces (Salazar-Pelaez et al., 2011a; Sun et al., 2016). On the other hand, accumulation of SMP in the pores and/or on the membrane surface causes pores blockage and a gel layer formation, limiting membrane permeability (Zuthi et al., 2013; Deng et al., 2014). In this chapter, SMP and EPS in the mixed liquor of SG-AnMBR (settling zone) and EG-AnMBR (membrane tank) were monitored, in order to explain the relationship between the properties of mixed liquor and membrane fouling since membranes were directly immersed in the mixed liquor. Figure 5.2 and Figure 5.3 demonstrated the EPS and SMP concentrations of the mixed liquor and their polysaccharides (SMP_C, EPS_C) and protein contents (SMP_P, EPS_P) from both G-AnMBRs at different designated TMP values.

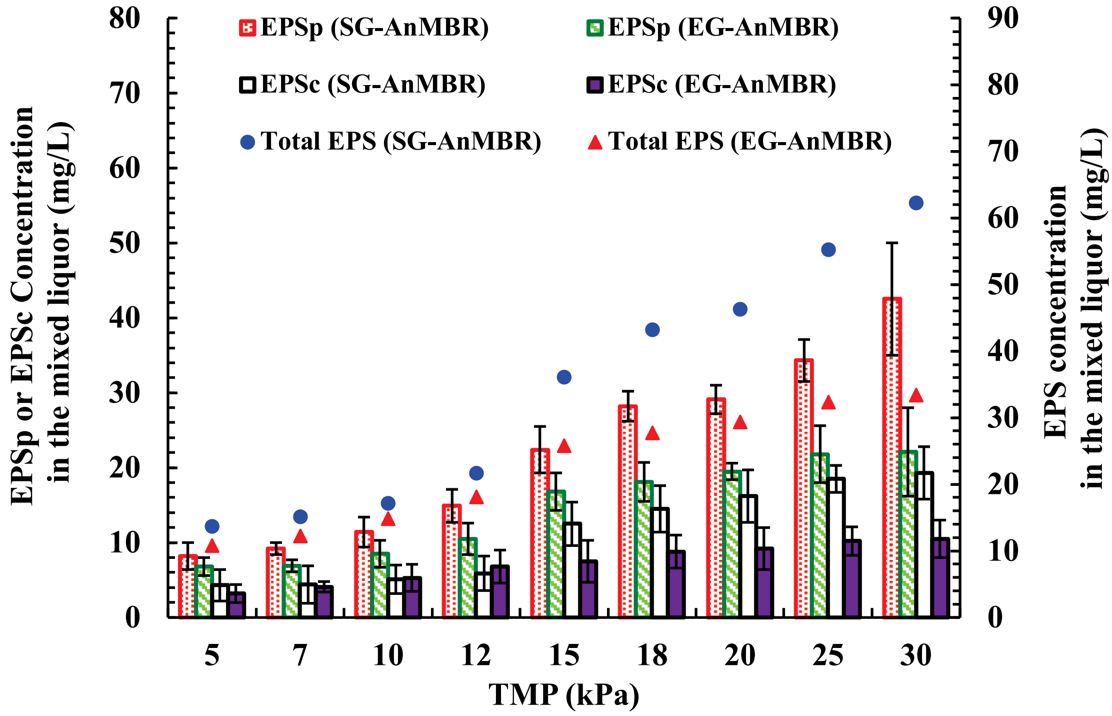


Figure 5.2 Variations of EPS (including EPS_P and EPS_C) concentrations in the mixed liquor of SG-AnMBR settling zone and EG-AnMBR membrane tank

It was obvious that EPS concentrations of both systems increased over the operation period. Prior to the sudden TMP jump (15 kPa), EPS (total EPS, EPS_P and EPS_C) were at low values and presented minor difference in both G-AnMBRs. At TMP of 15 kPa, noticeable differences of EPS levels between the SG-AnMBR and EG-AnMBR were observed. A notable increase in EPS was found in the SG-AnMBR (36.1 mg/L), while EPS_P and EPS_C rose to 22.4 mg/L and 12.5 mg/L. In contrast, the EG-AnMBR possessed lower EPS at 25.8 mg/L, with EPS_P and EPS_C at 16.8 and 7.5 mg/L. At the time when TMP reached 30 kPa, total EPS, EPS_P and EPS_C in the SG-AnMBR climbed up to the highest values of 62.2, 42.5, and 19.3 mg/L, respectively, which were approximately 1.8 times of EPS values from the EG-AnMBR (33.4, 22.1, and 10.5 mg/L, respectively). Higher concentrations of EPS in the mixed liquor of the SG-AnMBR were possibly ascribed to higher MLSS concentration in the settling zone of UAGB. The averaged MLSS in the SG-AnMBR was observed at 138.5 ± 8.89 mg/L, which was approximately three times higher than those of the EG-AnMBR (45.2 ± 7.18 mg/L). The MLSS concentration peaked at 180.2 ± 9.12 mg/L in the SG-AnMBR while the EG-AnMBR had MLSS of 50.2 ± 6.18 mg/L when TMP reached 30kPa. Much higher MLSS in the SG-AnMBR was mainly attributed to membrane's complete retention of dispersed sludge with poor immobilization properties under infinite SRT. The deterioration of the sludge settling capacity also encouraged inefficient solid entrapment of the sludge bed and hence promoted the accumulation of small and colloidal flocs in the mixed liquor, and the accumulation of small particles in the lumen (Ozgun et al., 2015).

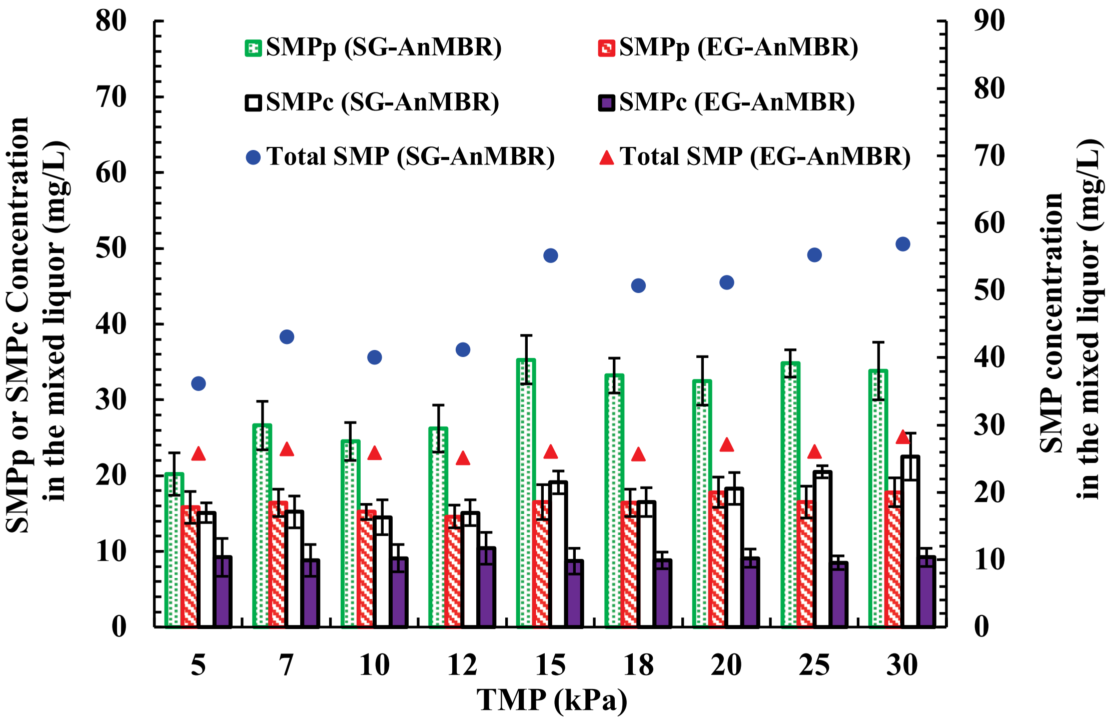


Figure 5.3 Variations of SMP (including SMP_P and SMP_C) concentrations in the mixed liquor of SG-AnMBR settling zone and EG-AnMBR membrane tank

With respect to SMP production in both G-AnMBRs (Figure 5.3), the EG-AnMBR showed very stable SMP concentration at 26.7 ± 1.1 mg/L while no obvious variation was observed in SMP_P (17.0 ± 0.7 mg/L) and SMP_C (8.9 ± 0.3 mg/L). The SG-AnMBR showed wide fluctuations of SMP concentrations between 36.2 and 56.9 mg/L, and SMP levels of the SG-AnMBR were substantially larger than those of the EG-AnMBR at all designated TMPs. The results revealed that both SMP and EPS in the SG-AnMBR were responsible for faster fouling period with rapid TMP development (Miyoshi et al., 2012; Prado et al., 2017).

5.4.3 Fouling resistance

G-AnMBR operation was terminated after TMP reached 30 kPa, and membranes were taken out for physical and chemical cleaning. The fouling resistance was calculated based on the resistance-in-series model and the results were shown in Table 5.4. R_T was found higher in the SG-AnMBR (20.7×10^{13} m^{-1}) as compared to that of the EG-AnMBR (13.4×10^{13} m^{-1}). R_P only contributed to a small portion of the total resistance. R_P of the SG-AnMBR (1.5×10^{13} m^{-1}) accounted for 7.1% of R_T, while the EG-AnMBR had R_P at 0.7×10^{13} m^{-1}, corresponding to 5.4% of R_T. As for both G-AnMBRs, R_C was responsible for over 92% of total resistance. Liu et al. (2012a) also reported that total filtration resistance was mainly governed by cake filtration resistance (over 98% of R_T). The results suggested the significance of minimizing the cake formation in G-AnMBRs, in order to increase membrane filtration efficiency. Cake layer resistance of the SG-AnMBR was found 50% higher than that of the EG-AnMBR. Lin et al. (2010) reported D (0.1), meaning the colloidal and fine particle fractions, had a significant negative effect on the cake formation rate. The D (0.1) of the SG-AnMBR was 69.1 μm, which was approximately 2 times to that of the EG-AnMBR (35.2 μm), and could be partly responsible for the higher cake fouling resistance in the SG-AnMBR.

Table 5.4 Fouling resistance distribution of G-AnMBRs at the end of the operation

Reactors		EG-AnMBR	SG-AnMBR
Fouling resistance distribution	R_M ($\times10^{11}$ m^{-1})	8.9 ± 1.2 (0.7%)	9.1 ± 1.8 (0.4%)
	R_P ($\times10^{13}$ m^{-1})	0.7 ± 0.2 (5.4%)	1.5 ± 0.4 (7.1%)
	R_C ($\times10^{13}$ m^{-1})	12.6 ± 2.5 (93.9%)	19.1 ± 2.8 (92.5%)
	R_T ($\times10^{13}$ m^{-1})	13.4 ± 4.1 (100%)	20.7± 5.1 (100%)

[a] R_T = total fouling resistance, [b] R_C = cake layer resistance, [c] R_P = pore blocking resistance, [d] R_M = clean membrane resistance

5.4.4 SMP and EPS content in the cake layer

Since the cake layer contributed to the dominant fraction of the total fouling resistance, the compositions of EPS and SMP of the cake layer on the membrane surface were further analysed (Table 5.5). Higher concentration in EPS_P was found in the SG-AnMBR (11.7 mg/g cake layer) than EG-AnMBR (8.5 mg/g cake layer), while minor difference in EPS_C was observed in both G-AnMBRs. EPS_P excreted from the anaerobic microbes exert an impact on the cake resistance via filling the void spaces between the particles in the cake leading to a severe reduction of permeate flux (Chu et al., 2005). Instead of EPS_P, structure collapse was responsible for cake filtration resistance (Liu et al., 2012a). Factors that promoted the compactness and dewatering of the membrane cake layer were also responsible for cake resistance in a long-term operation. With regards to SMP concentration of the cake layer, the SG-AnMBR exhibited higher SMP_P and SMP_C concentrations of 18.6 and 6.5 mg/g cake layer, respectively as compared with the lower values for the EG-AnMBR (10.2 and 5.1 mg/g cake layer). The accumulation of SMP_P could change the surface charge and morphology of the membrane, promoting the adhesion and growth of sludge flocs as cake layer (Liu et al., 2012a; Deng et al., 2014). Furthermore, the build-up of SMP_C could result in irreversible fouling by forming a thin gel layer on the membrane surface (Deng et al., 2015). Results implied that EPS_P, SMP_P and SMP_C were the main contributors to the higher R_C in the SG-AnMBR. Under higher TMP, higher drag force from the permeate pump enhanced formation of cake layer by exacerbating the accumulation of EPS_P and SMP on the surface of the membrane. Greater amount of sludge cake on membrane surface could promote cell lysis or endogenous decay at the bottom cake layer, which in turn led to more EPS_P and SMP released (Deng et al., 2014).

Table 5.5 Cake layer analysis of G-AnMBRs at the end of the operation

Reactors		EG-AnMBR	SG-AnMBR
	EPS_P [a]	8.5 ± 1.5	11.7 ± 2.2
Cake layer compositions	EPS_C [b]	3.7 ± 1.3	3.8 ± 0.9
(mg/g cake layer)	SMP_P [c]	10.2 ± 1.9	18.6 ± 1.2
	SMP_C [d]	5.1 ± 0.8	6.5 ± 1.4

[a] EPS_P = protein concentration of extracellular polymeric substances, [b] EPS_C = polysaccharides concentration of extracellular polymeric substances, [c] SMP_P = protein concentration of soluble microbial products, [d] SMP_C = polysaccharides concentration of soluble microbial products

5.4.5 Nature of foulant organics

The fractionation of organic matter in the foulant by LC-OCD analysis provides vital information for the understanding of fundamentals of the membrane fouling. LC-OCD divides the total foulant organics into two categories: hydrophobic and hydrophilic. The hydrophilic fraction, which is deemed to be the major foulant, is further subdivided into several groups including biopolymers, humic substances, building blocks, LMW neutrals and acids. According to Figure 5.4, the biopolymers showed the greatest difference between two G-AnMBRs, with 14.6 mg/L in the SG-AnMBR and 6.9 mg/L in the EG-AnMBR. In fact, Yue et al. (2015) and Johir et al. (2012) both recognized biopolymers as the major foulants responsible for membrane fouling in the aerobic MBR and AnMBR operations. Higher concentration of biopolymers might suggest hydrophilic layers build-up on the membrane surface in the SG-AnMBR (Hong et al. 2012). LC-OCD also indicated the SG-AnMBR possessed higher amount of building blocks and LMW neutrals and acids (6.3 mg/L, 9.7 mg/L, respectively) than those in the EG-AnMBR (4.6 mg/L, 5.9 mg/L, respectively). Building blocks and LMW neutrals and acids were the influential factors affecting fouling as their assemblage could promote biopolymers formation on the membrane surface, exacerbating fouling propensity (Aryal et al., 2009). Last but not least, minor difference in the concentration of humic substances was observed between two G-AnMBRs, indicating the impact of humic substances on the fouling could be neglected in this chapter.

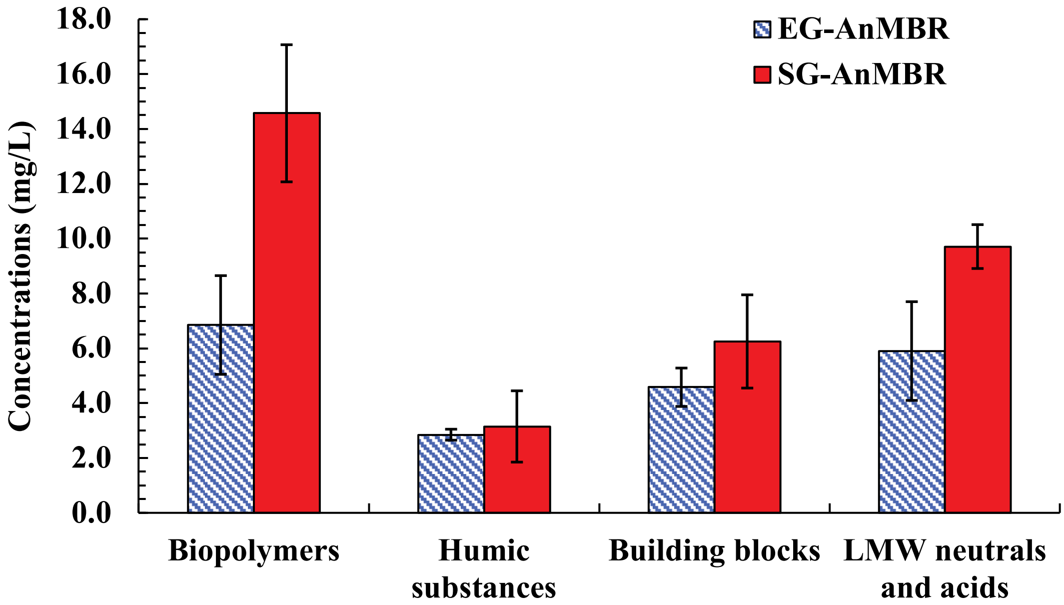


Figure 5.4 Nature of foulant organics in the G-AnMBRs by LC-OCD analysis

5.5 Effect on VFA accumulation and biogas production

VFA is regarded as the most vital process performance indicator for G-AnMBR operation since it not only serves as the essential intermediary compounds in the metabolic pathway of methane fermentation but also profoundly affects pH dynamics in the system (Ji et al., 2015). If presented in high concentration, VFA accumulation in G-AnMBRs can result in significant pH drop and microbial stress, particularly for methane-producing bacteria, ultimately resulting in reactor acidification and even failure (Chen et al., 2008; Chen et al., 2016b). The VFA of the mixed liquor, including acetic acid (C_2), propionic acid (C_3), iso-butyric acid (i-C_4), butyric acid (C_4), iso-valeric acid (i-C_5) and valeric acid (C_5) were monitored in this chapter. According to Table 5.6, C_2 accounted for the dominant fraction of VFA (97.0%) in the mixed liquor of the EG-AnMBR, with concentration as low as 5.2 ± 1.1 mg/L. Only very low amount of C_3 (0.2 ± 0.3 mg/L) was found randomly while C_4 - C_5 was not detected. This revealed that the EG-AnMBR was rarely encountered with VFA accumulation and reactor acidification over the operation time.

Table 5.6 VFA concentrations of the mixed liquor from the EG-AnMBR and SG-AnMBR

VFA	EG-AnMBR (mg/L)	SG-AnMBR (mg/L)
C_2 [a]	5.2 ± 1.1(97.0%) [h]	16.6 ± 2.5(65.8%)
C_3 [b]	0.2 ± 0.3(3.0%)	1.3 ± 0.9 (5%)
i-C_4 [c]	0	1.7 ± 1.3 (6.9%)
C_4 [d]	0	1.5 ± 1.1(5.8%)
i-C_5 [e]	0	1.7 ± 3.0 (6.6%)
C_5 [f]	0	2.5 ± 0.9 (10.0%)
V_T [g]	5.4 ± 1.1(100%)	25.3 ± 4.9 (100%)

[a] C_2=acetic acid, [b] C_3=propionic acid, [c] i-C_4=iso-butyric acid, [d] C_4=butyric acid, [e] i-C_5=iso-valeric acid, [f] C_5=valeric acid, [g] V_T=total volatile fatty acids, [h] = percentage of total volatile fatty acids (V_T)

On the other hand, the SG-AnMBR possessed much higher concentration of VFA at 25.3 ± 4.9 mg/L, which was 4 times more than the corresponding value in the CG-AnMBR. C_2 accounted for the major fraction (65.8%) in the VFA of the mixed liquor. C_3 - C_5 was also detected although their concentrations were lower than that of acetic acid. The higher concentration of VFA could be related to VFA release from the granule fragmentation, since the elimination of hydraulic selection pressure undermined the integrity of sound granules in the submerged configuration. A low VFA concentration (below 5 mg/L as acetic acid) was found from both G-AnMBRs permeate, suggesting G-AnMBR system was capable of eliminating most of the biodegradable organic material in the wastewater.

Biogas produced from the G-AnMBRs is a competitive advantage against the aerobic MBRs. Biogas commonly refers to a mixture of gases produced by the biological breakdown of organic matter in the absence of oxygen, of which methane, hydrogen and carbon monoxide can be combusted or oxidized with oxygen. The energy output/input can reach up to 28.8 MJ/MJ under favorable conditions, contributing to a very efficient use of the valuable biomass (Deublein and Steinhauser, 2008). Similar biogas production was observed at around 650 mL/d and 666 mL/d from the EG-AnMBR and SG-AnMBR. Biogas production was not profoundly affected, even though SG-AnMBR was encountered with VFA accumulation to some extent. Stable methane and carbon dioxide composition in the biogas were around 69.7 and 26.9% in the EG-AnMBR, while the SG-AnMBR had 68.5 and 26.0%, respectively. Tracable H_2 with 3 - 10 ppm was detected in the biogas from both G-AnMBRs.

The EG-AnMBR achieved methane yield of 160.6 ± 5.6 mL $CH_{4\,(STP)}$/g COD $_{removed}$ (volume of methane produced per g $COD_{removed}$ at 0 °C Standard Temperature and 1 atm Pressure), which was almost the same to the value from the SG-AnMBR (161.3 ± 4.6 mL $CH_{4\,(STP)}$/g COD $_{removed}$). Methane yield from two G-AnMBRs represented only about 50% of the optimal theoretical value at 20 °C (317.6 ± 4.6 mL $CH_{4\,(STP)}$/g COD $_{removed}$), and nearly half of the removed COD was lost. Galib et al. (2016) revealed that methane loss in the aqueous phase was significant and could be up to 58% of the total methane produced from AnMBRs. While Sanchez-Ramirez et al. (2015) also suggested a significant dissolved methane concentration, accounting for 20 - 40% of methane in the produced biogas, could be lost in the AnMBR effluent. Furthermore, when using AnMBR for the treatment of municipal wastewater under low temperatures (i.e. 15 - 20 ºC), the release of this dissolved methane into atmosphere could be a more critical issue as greater amount of methane could be dissolved in the permeate under the lower temperature and emitted to the atmosphere (Pretel et al., 2016). Hence, quantifying and recovering dissolved methane in the permeate would be of great significance in the future research since greenhouse gas potential of dissolved methane is significant and may impact the environmental sustainability of G-AnMBRs.

5.6 Conclusions

In this chapter, external and submerged G-AnMBRs were operated in parallel treating municipal wastewater, in order to investigate differences with respect to overall performance of two different configurations of G-AnMBRs. The specific conclusions were drawn as follows:

1) In terms of the organic removal and methane yield, negligible differences were noticed in both G-AnMBR systems, although VFA accumulation exhibited in the SG-AnMBR.
2) Deteriorated granular sludge properties were found in the SG-AnMBR with granules fragmentation, reduced granules EPS content and settleability.
3) The SG-AnMBR demonstrated higher fouling propensity, which could be attributed to higher concentrations of SMP and EPS in mixed liquor, much higher cake layer resistance, and more deposition of EPS_P and SMP in the cake layer.
4) The characterization of foulant organics demonstrated that biopolymers were the major foulants followed by LMW neutrals and acids and building blocks.
5) The results proved that the EG-AnMBR was a more promising configuration for municipal wastewater treatment due to the better granular sludge quality and prolonged operation time.

CHAPTER 6
EVALUATION OF THE PERFORAMANCE OF A SA-GANMBR

6.1 Introduction

In the past decades, AnMBRs have been found particularly attractive for wastewater treatment because it can not only achieve total biomass retention, high effluent quality, small footprint and low sludge production, but also significantly contribute to renewable bioenergy generation for the substitution of fossil fuel in power and heat production (Mnif et al., 2012; Galib et al., 2016; Pretel et al., 2016). In particular, G-AnMBR, a hybrid anaerobic bioreactor incorporating granular technology with membrane-based separation, offers a promising approach compared to C-AnMBR, predominantly in the form of continuous stirred tank reactor configuration. The competitive advantages of G-AnMBR include no requirement for mechanical mixing, significantly low energy demand and much more compact reactor design (Chen et al., 2016a).

However, recent research has shown that the integration of membrane into the granular systems could affect the hydraulics of granular sludge bed by eliminating the washout of fine floc sludge, thereby negatively impacting granular sludge properties (Ozgun et al., 2013; Ozgun et al., 2015). The accumulation of fine and colloidal flocs in sludge supernatant may also contribute to membrane fouling. In addition, at high liquid and biogas upflow velocity, vigorous up and down movements of granules may break granules, resulting in granules fragmentation due to the high shear force (Abbasi and Abbasi, 2012; Chen et al., 2012; Wu et al., 2015). It is essential to seek for strategies to maintain the quality of granules for long-term operation of submerged G-AnMBR, since the integrity of the anaerobic granules determines the efficiency and stability of anaerobic biological treatment and guarantees the sludge supernatant quality for controlling fouling propensity.

The use of low cost polyutrethane sponge for the enhancement of aerobic submerged MBR was found particularly successful in recent years (Guo et al., 2009; Deng et al., 2015). As far as we know, the effects of sponge, as the inert material, on the enhancement of granular sludge characteristics, and membrane fouling mitigation in the G-AnMBR have yet to be investigated for domestic wastewater treatment. Thus, this chapter aimed to evaluate the overall performance of a SA-GAnMBR and a CG-AnMBR. Granule properties (e.g. PSD, SMP, EPS and SVI), fouling propensity (e.g.

TMP, SMP and EPS of the mixed liquor and cake layer, and foulants) and biogas production were also assessed.

A major part of Chapter 6 has been published in the following paper:

Chen, C., Guo, W.S., Ngo, H.H., Liu, Y., Du, B, Wei, Q., Wei, D., Nguyen, D.D., Chang, S.W., 2017b. Evaluation of a sponge assisted-granular anaerobic membrane bioreactor (SG-AnMBR) for municipal wastewater treatment. Renewable Energy 111, 620-627.

6.2 Impact of sponge addition on treatment performance

The experimental set-up and operation protocol were depicted in Chapter 3 (Section 3.3.3). The characteristics of municipal wastewater were shown in Chapter 3 (Section 3.2.1), and COD: N: P ratio was maintained at 100: 2: 1. The membrane properties and sponge specification could be found in Chapter 3 (Section 3.2.2 and Section 3.2.3).

Both AnMBRs achieved organics removal efficiency higher than 90% (Figure 6.1). More specifically, the SA-GAnMBR demonstrated slightly higher removals of DOC (92.4 ± 2.2%) and COD (93.7 ± 1.7%) when compared to those of the CG-AnMBR (90.1 ± 0.9% and 90.8 ±1.4 %, respectively). The high organics removal efficiencies could be attributed to the influent COD contained the majority of readily biodegradable COD using glucose as the sole carbon source. The complete retention of all particulate and colloidal matters by membrane also contributed to the high organics removal (Martinez-Sosa et al., 2011). In general, TN and PO_4^{3-}-P removal in the CG-AnMBR was low, which was found to be 15.0 ± 4.1% and 17.6 ± 6.2%, respectively (Figure 6.1). However, higher removal efficiencies were observed in the SA-GAnMBR (31.7 ± 6.8% for TN removal and 36.2 ± 7.9% for PO_4^{3-}-P removal), which is in line with the findings in Nguyen et al. (2011). The results revealed that the addition of sponge could not only enhance the removal of organic matter but also encourage nutrient removal in the G-AnMBR.

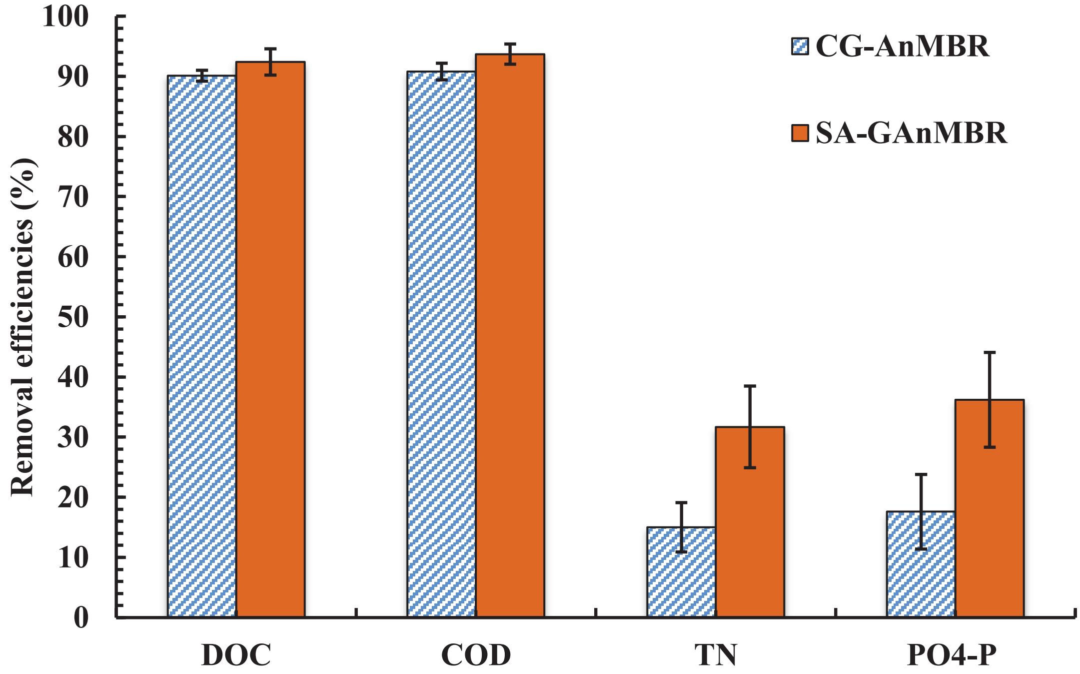


Figure 6.1 DOC, COD, TN and PO_4-P removals in the CG-AnMBR and SA-GAnMBR

6.3 Impact of sponge addition on granular sludge properties

6.3.1 Granular sludge

The successful implementation of anaerobic granular bioreactor technology relies on its capacity to retain a dense granular sludge bed for efficient physical entrapment and biodegradation of particulate and dissolved organic substances (Seghezzo et al., 1998). The CG-AnMBR and SA-GAnMBR have been operated for 25 and 55 days, respectively till TMP reached up to 30 kPa. As can be seen from Table 6.1, at the end of experimental period, MLSS concentrations of granular sludge increased to 23.82 g/L and 21.30 g/L in the SA-GAnMBR and the CG-AnMBR, corresponding to the growth rate (ΔMLSS/Δt) of 0.060 g/L·d and 0.032 g/L·d, respectively. The higher biomass growth rate in the SA-GAnMBR indicated that the sponge addition encouraged the growth of retained sludge agglomerates in the granular sludge bed. Furthermore, the SA-GAnMBR also presented higher MLVSS concentration with 19.10 g/L than that of the CG-AnMBR (16.59 g/L). The biomass attached to the sponge was found at 1.28 ± 0.41 g/g sponge. In addition, the granular sludge from the SA-GAnMBR also presented superior settling properties. At the end of the operation, granular sludge from the SA-

GAnMBR had SVI of 20.1 mL/g with settling velocity varying from 17.5 to 32.5 m/h (Table 6.1).

Table 6.1 Summary of sludge characteristics of seed sludge and granular sludge in G-AnMBRs

Sludge properties	Seed sludge	Granular sludge (CG-AnMBR)	Granular Sludge (SA-GAnMBR)
MLSS (g/L)	20.50 ± 1.53	21.30 ± 0.91	23.82 ± 1.83
MLVSS (g/L)	16.21 ± 1.85	16.59 ± 1.28	19.10 ± 1.11
Zeta-potential (mV)	-15.5 ± 3.5	-21.1 ± 2.5	-13.8 ± 1.8
SVI (mL/g)	38.8 ± 4.8	58.5 ± 5.1	20.1 ± 4.2
Settling velocity (m/h)	15.51 - 25.42	14.1-18.4	17.5 - 32.5

Compared to the settling properties of the seed sludge, reduced SVI and increased settling velocities indicated that the settling properties of granular sludge were enhanced in the SA-GAnMBR. On the other hand, granular sludge in the CG-AnMBR exhibited higher SVI of 58.5 mL/g and lower settling velocity of 14.1-18.4 m/h than those of the seed sludge, suggesting the sludge settleability was deteriorated. Zeta potential of the granular sludge in the SA-GAnMBR (-13.8 mV) was found higher than those of the CG-AnMBR (-21.1 mV) and the seed sludge (-15.5 mV). With increased zeta potential, the negative charge of the flocs could be neutralized and form large sludge aggregates with better settling characteristics (Deng et al., 2014; Ozgun et al., 2015). Since the development of well settling granular sludge requires selective washout of flocculent sludge with poor immobilization properties, the complete retention of small and colloidal flocs in a G-AnMBR by membrane barrier eliminated the hydraulic selection pressure required for granular sludge with good settling capacities. In this case, the growth of dispersed sludge would predominately take place, resulting in the bulking type of sludge formed in the CG-AnMBR with poor settling properties (Deng et al., 2014). However, sponge addition could somehow improve granular sludge properties of the SA-GAnMBR, and further alleviate the deterioration of granular sludge settling properties.

6.3.2 Granules

Generally, it has been reported the formation of sludge aggregates on or over 500 μm could be considered as granules (Bhunia and Ghangrekar, 2007). However, a few studies have regarded sludge particles with 160 μm or less as granules (Chou et al., 2005; Pevere et al., 2006; Zhang et al., 2011). Abbasi and Abbasi (2012) suggested that granules size could range from 100 μm to 5 mm while Zhang et al. (2009) reported average granule size increased from 111 μm to 264 μm from a hybrid anaerobic granular system with internal hydraulic circulation. Thus, in this chapter, bioparticles over 100 μm was considered as granules since synthetic domestic wastewater with low organic loading rate of 0.53 - 0.59 kg COD/m^3·d was used as the feed and relatively short operation time was adopted. As compared to the seed sludge, one-way shift to fine particles was observed in the CG-AnMBR while bigger size granules tended to form in the SA-GAnMBR (Figure 6.2).

Based on the PSD of the granular sludge, the SA-GAnMBR presented granules with increased diameter, compared to those of the CG-AnMBR. Fig. 1 shows that the percentage of granules (>100 μm) was approximately 84% of the total granular sludge in the SA-GAnMBR, which was almost two times to the corresponding value obtained from the CG-AnMBR (42.5%). As membrane functioned as an absolute barrier in the CG-AnMBR, fine sludge particles (<100 μm), such as colloidal flocs, macromolecules of SMP and non-settling particles, could not be effectively discharged and rather accumulated in the CG-AnMBR, presenting lower percentage of granules. In contrast, sponge addition could assist granular growth by immobilizing fine particles on or inside the sponge pores, contributing to larger fraction of granules.

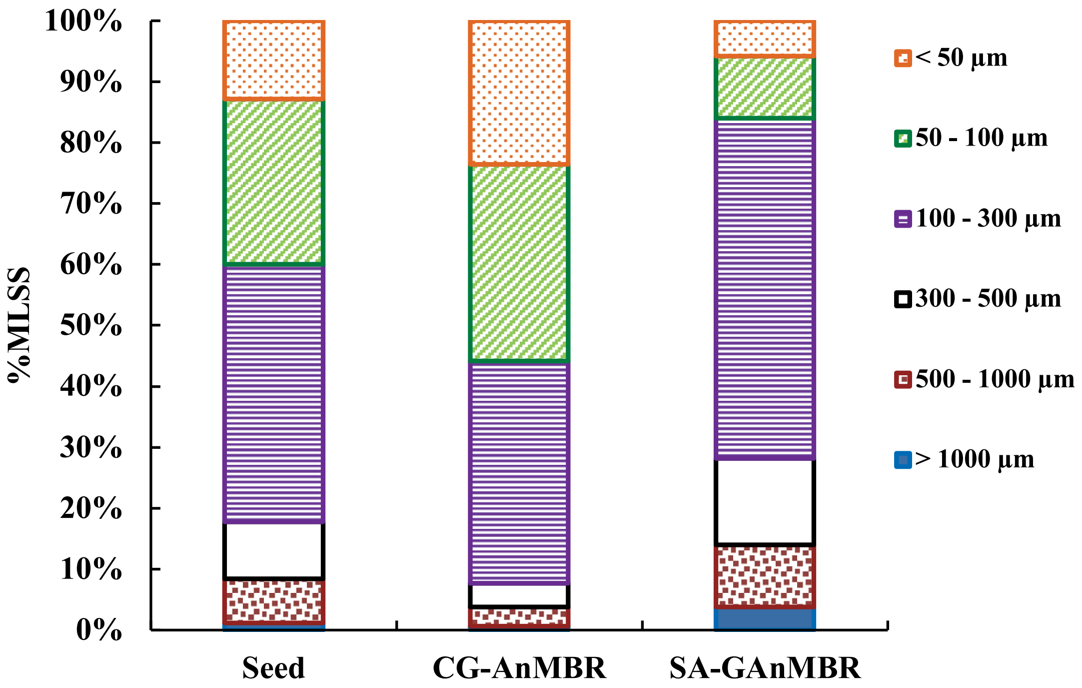


Figure 6.2 Particle size distribution of seed sludge, and granular sludge for both G-AnMBRs

Apart from the complete retention of fine sludge particles, granules breakage could be another explanation for the lower amount of granules in the CG-AnMBR. Normally, EPS in the sludge plays a vital role in the synthesis of anaerobic granules, and is crucial for integrating cells into granules and maintaining intact structure of the granules. At the end of experiment, both protein and polysaccharides amounts of EPS decreased by 81.1% and 77.1% in the CG-AnMBR, as compared to the seed sludge EPS (EPS_p and EPS_c: 20.2 and 6.9 mg/g VSS), respectively. Therefore, the significant decrease in EPS amount might indicate scattered, looser and weaker structures of granules (Figure 6.3), meaning granule fragmentation and decrease in particle size, as well as SMP increase in the mixed liquor (Ozgun et al., 2015). On the contrary, the stable EPS production in the SA-GAnMBR was observed with the average values of 28.8 and 8.6 mg/g VSS for EPS_P and EPS_C, respectively. Therefore, the higher contents of EPS promoted granule growth in the SA-GAnMBR. Additionally, the amount of SMP from the CG-AnMBR sludge granules (Protein: 25.1 mg/g VSS, polysaccharide: 8.2 mg/g VSS) were found approximately 7 times higher than those from the SA-GAnMBR (3.2 mg/g VSS, and 1.1 mg/g VSS). Much lower SMP values of the SA-GAnMBR confirmed the majority of proteins and polysaccharides existed as the part of the anaerobic granules. As a result, the sponge addition had profound impacts on the EPS production of the anaerobic granules, as well as the granules abundance, structure and stability.

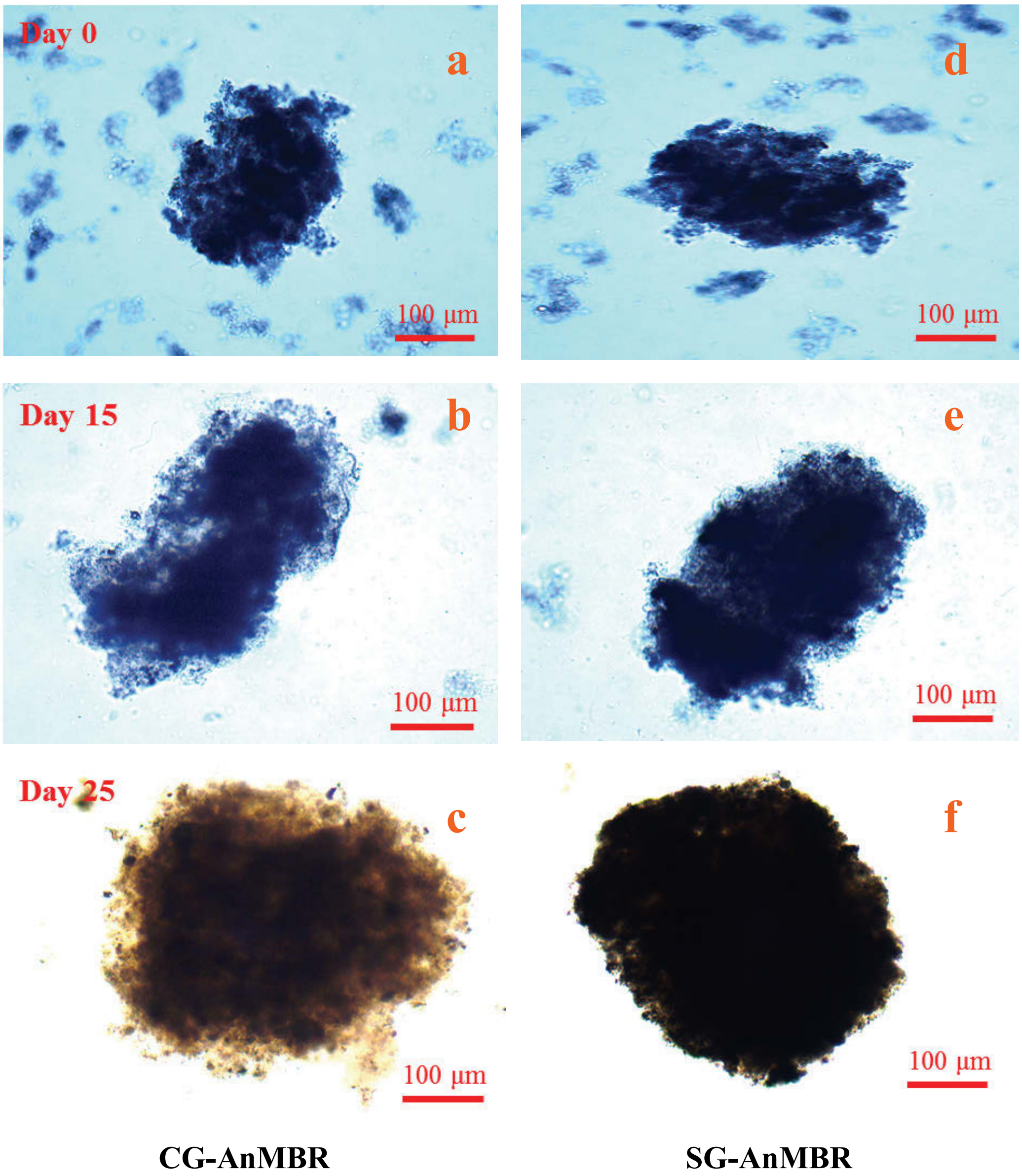


Figure 6.3 Microscopic observations of seed sludge (day 0) and sludge granules from the CG-AnMBR (a, b, c) and SA-GAnMBR (d, e, f) on day 15, 25.

6.4 Effects of sponge addition on membrane fouling behaviour

6.4.1 TMP and fouling rate

Membrane fouling profiles indicated by TMP development in two G-AnMBRs were shown in Figure 6.4. Both systems showed significant differences in TMP profiles. As for the CG-AnMBR, the increase in TMP with time was characterized by a gradual rise at 0.3 kPa/d from day 1 to day 15, and then a rapid increase at 2.4 kPa/d till membrane was severely fouled on day 25. On the other hand, TMP in the SA-GAnMBR was maintained well below 6 kPa within the first 25 days of operation and reached 30 kPa on day 55, indicating a relatively lower fouling rate of 0.5 kPa/d compared to the averaged 1.2 kPa/d for the CG-AnMBR. The results revealed that the sponge addition could greatly reduce fouling rate and improve the filtration performance of the G-AnMBR.

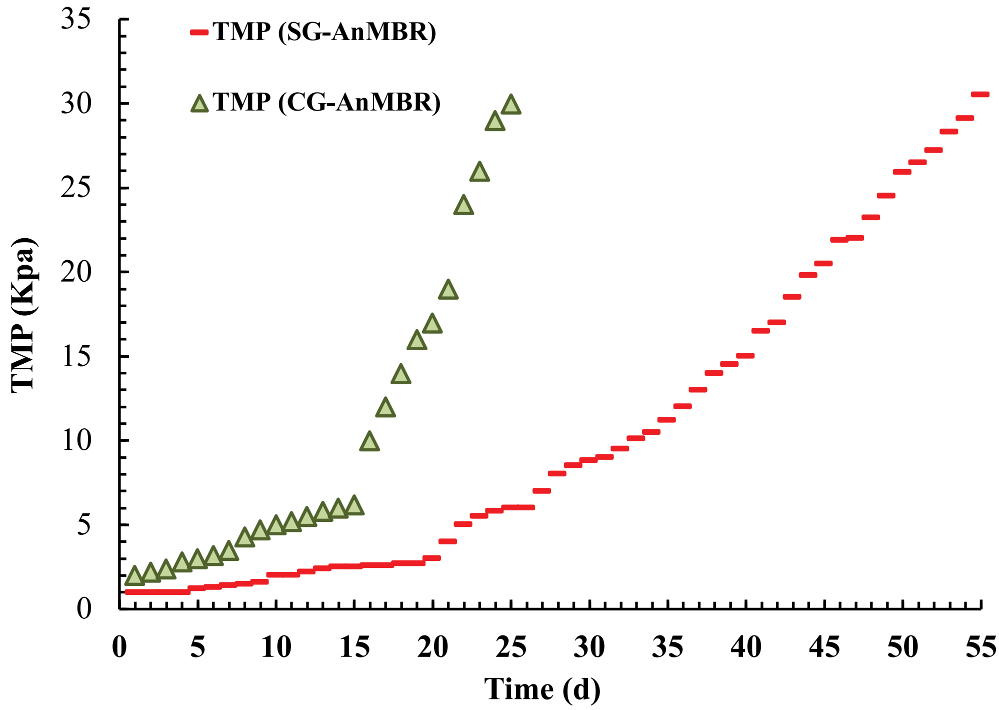


Figure 6.4 TMP profile of the CG-AnMBR and the SA-GAnMBR over the experimental period

6.4.2 SMP and EPS in the mixed liquor

Membrane fouling was often attributed to the accumulation of organics in or on the membrane in the form of EPS and SMP (Guo et al., 2012). Studies have reported that EPS clogged the membrane pores, promoting the formation of a strongly attached fouling layer on the membrane surface while SMP could be absorbed onto the membrane surface, thereby blocking its pores and forming a gel layer acting as a barrier for permeate flux during filtration (Deng et al., 2015; Xiong et al., 2016). Since the membrane was submerged in the mixed liquor of the G-AnMBR settling zone, SMP and EPS of the mixed liquor in both G-AnMBRs were analysed in order to explain the relationship between the mixed liquor properties and membrane fouling. As shown in Figure 6.5, averaged SMP concentration in the CG-AnMBR was 47.3 ± 7.6 mg/L, which was almost three times higher than the value obtained in the SA-GAnMBR (15.9 ± 3.5 mg/L). The significantly higher SMP amount in the CG-AnMBR was due to the release of biopolymeric susbstances to the mixed liquor as a result of granule and floc breakage and cell lysis (Kunacheva et al., 2017). This observation was further supported with particle size analysis, and EPS analysis of the granular sludge in Section 3.2. The bound EPS in the sludge could also be dissolved/ hydrolyzed into small fractions by bacterial hydrolysis (Guo et al., 2012). Their subsequent dissolution into the water phase could result in more SMP release from microbial aggregates into the mixed liquor (Ozgun et al., 2015).

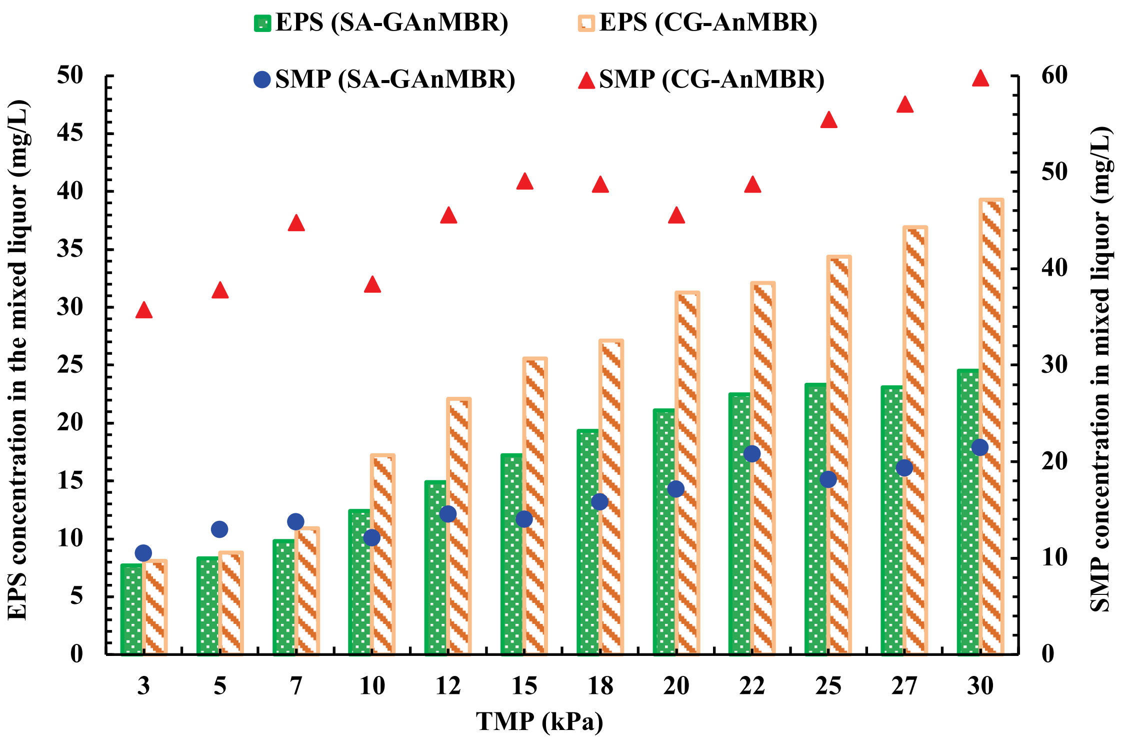


Figure 6.5 Variations of EPS and SMP concentrations in the settling zone of G-AnMBR at designated TMPs

EPS concentrations of both systems remained increasing (Figure 6.5) with the MLSS build-up in the mixed liquor. The MLSS concentrations in both G-AnMBRs increased gradually throughout the experimental period. At the end of experiment, the MLSS concentration in the CG-AnMBR reached up to 770.2 mg/L, which was nearly 3 times higher than that of the SA-GAnMBR (260.2 mg/L). The build-up of MLSS in the mixed liquor was mainly due to the membrane's complete retention of small and colloidal flocs that would be otherwise selectively washed out from the system. The EPS concentration averaged at 17.0 ± 6.2 mg/L (SA-GAnMBR) and 24.5 ± 11.0 mg/L (CG-AnMBR), and peaked at 24.5 mg/L (SA-GAnMBR) and 39.3 (CG-AnMBR) when TMP reached 30 kPa. In the SA-GAnMBR, sponge addition could help to limit the suspended growth (Deng et al., 2014), thus significantly reducing SMP and EPS concentrations in the mixed liquor by the means of adsorption onto the sponge and biodegradation by the attached biomass of the sponge. In addition, well-balanced granular and attached growth provided a sound environment for granules growth in the SA-GAnMBR. Thus, the biodegradation of organics occurred mainly within the granules and attached biomass of the sponge, limiting the dispersed growth of light flocs. Colloidal particles coming from the influent solids could therefore be physically adsorbed and retained in the thick and dense granule bed, preventing their impact on the fouling (Martin-Garcia et al., 2013).

6.4.3 Fouling resistance

The fouling resistance was calculated according to the resistance-in-series model and the results were shown in Table 6.2. The R_T of SA-GAnMBR and CG-AnMBR were 9.7×10^{13} m^{-1} and 19.7×10^{13} m^{-1}, respectively, indicating sponge addition into SA-GAnMBR reduced the R_T by 50.7%, compared to the CG-AnMBR. Higher R_P was also found for the CG-AnMBR compared to the SA-GAnMBR, corresponding to 9.5×10^{12} m^{-1} and 4.6×10^{12} m^{-1}, respectively. R_C of the CG-AnMBR (18.7×10^{13} m^{-1}) accounted for 94.9% of R_T, whereas the SA-GAnMBR had much lower R_C at 9.2×10^{13} m^{-1}, corresponding to 94.8% of R_T. The resistance caused by R_C presented dominant proportion of total resistance for both systems. Hence, minimizing the cake formation is of great importance to lower the fouling propensity of the G-AnMBR. Contrarily, pore clogging, due to particles or colloids with equal or smaller size than the membrane pores, contributed to small portion of fouling resistance. The results were consistent with the findings of Liu et al. (2012a) in which sludge cake formation was the main mechanism of membrane fouling in the G-AnMBR. Jeison and van Lier (2007) and Jeison et al. (2008) also reported that TMP and flux were mainly governed by cake formation. The higher cake layer resistance in the CG-AnMBR could be ascribed to higher MLSS concentration in the mixed liquor where membrane was immersed. Assisted by sponge, the SA-GAnMBR demonstrated the efficient solids entrapment of the dense granular sludge bed and contained much reduced MLSS of the mixed liquor. Lin et al. (2010) identified that the cake formation rate was significantly affected by colloidal and fine particle size D (0.1) of PSD. D (0.1) of the CG-AnMBR was 30.1 μm, which was much smaller than those of the SA-GAnMBR (62.5 μm). Considering the denser structure and reduced back transport velocity of the fine flocs, Liu et al. (2012a) suggested that the greater amount of fine particles in the CG-AnMBR were more likely to deposit on the surface of membrane, which in turn facilitated a cake layer denser than that with larger particles. Therefore, the results proved the sponge addition could greatly alleviate membrane fouling mainly by reducing the cake layer formation and pore clogging.

Table 6.2 Fouling resistance for both G-AnMBRs

		CG-AnMBR	SA-GAnMBR
Fouling resistance (m^{-1})	R_T [a]	19.7×10^{13}	9.7×10^{13}
	R_C [b]	18.7×10^{13}	9.2×10^{13}
	R_P [c]	9.5×10^{12}	4.6×10^{12}
	R_M [d]	5.7×10^{11}	5.1×10^{11}

[a] R_T = total fouling resistance, [b] R_C = cake layer resistance, [c] R_P = pore blocking resistance, [d] R_M = clean membrane resistance

6.4.4 SMP and EPS content in the cake layer

The compositions of bound EPS and SMP of the cake layer from both reactors were also analysed and compared. As shown in Figure 6.6, sponge addition could efficiently reduce EPS_P and SMP production in the cake layer of the SA-GAnMBR. Higher concentration of EPS_P (12.1 mg/g cake layer) was found in the CG-AnMBR than that in the SA-GAnMBR (10.7 mg/g cake layer), while minor difference could be observed on EPS_C of the cake layer from both G-AnMBRs. The CG-AnMBR demonstrated higher concentrations of SMP_P and SMP_C in the cake layer (8.2 and 4.1 mg/g cake layer, respectively) compared to the SA-GAnMBR (5.6 and 2.5 mg/g cake layer, respectively). These results implied EPS_P, SMP (including SMPp and SMPc) on the surface of the membrane were responsible for the higher R_C in the CG-AnMBR. At relatively high TMP, more EPS_P, SMP_P, and SMP_C could be deposited onto the membrane surface due to the high drag force from the permeate pump. Furthermore, the endogenous decay or cell lysis at the bottom layer could result in the release of more EPS_P and SMP due to more sludge cake accumulated on the membrane surface (Deng et al., 2015).

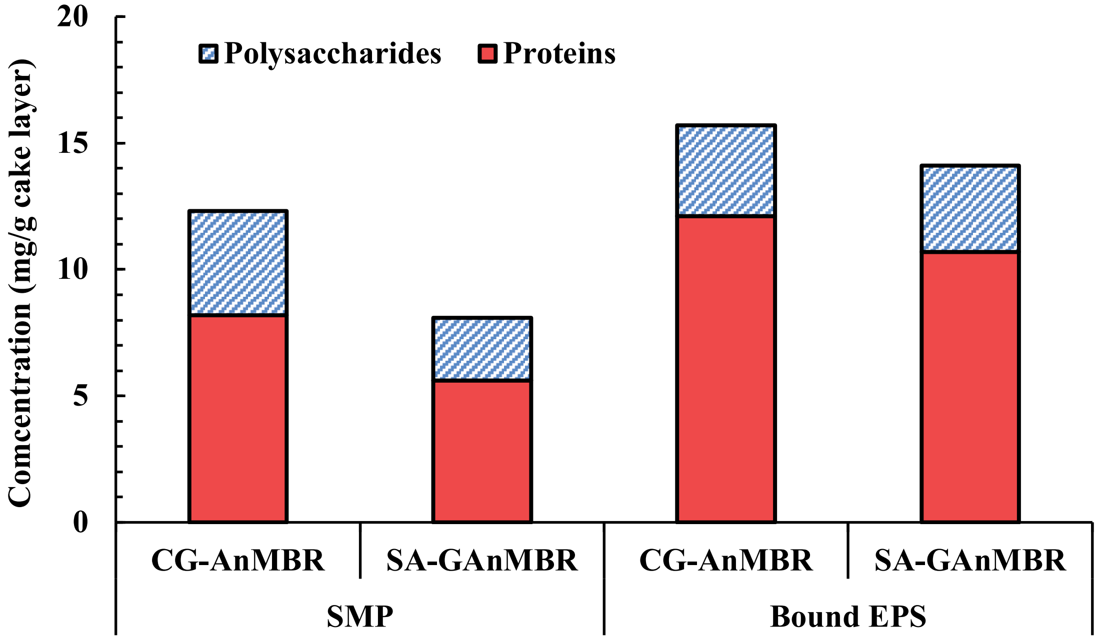


Figure 6.6 SMP and EPS content in the cake layer of G-AnMBRs

6.4.5 Characterization of foulant organics

LC-OCD provides important information regarding the fraction of organic matter in foulants by dividing the total organics into hydrophobic and hydrophilic groups. The hydrophilic fraction can be further subdivided into biopolymers, humic substances, building blocks, LMW neutrals and acids. As can be seen from Table 6.3, hydrophilic organics mainly contributed to membrane fouling, in which biopolymers were regarded one of the major foulants (Johir et al., 2012). The value of biopolymers for the CG-AnMBR was found twice higher (34.6%) as compared to that for the SA-GAnMBR (17.1%). The higher biopolymers concentrations in the CG-AnMBR indicated more hydrophilic layers built up on the membrane surface (Hong et al., 2012). Furthermore, bridging between inorganic compounds and deposited biopolymers could encourage the formation of more compact and dense fouling layer, leading to sever fouling (An et al., 2009). Greater amount of building blocks (17.0% vs. 13.9%) and LMW neutrals and acids (35.1% vs. 31.2%) were also found in the CG-AnMBR compared to the SA-GAnMBR. Aryal et al. (2009) reported that building blocks and LMW neutrals and acids were vital factors causing fouling and enhancing the formation of biopolymers on the surface of the membrane possibly through their assemblage. Nevertheless, the CG-AnMBR exhibited lower humic substances (10.5%) than the SA-GAnMBR (31.3%). Since the building blocks were the breakup of humic substances, lower fraction of humic substances might be related to the higher amount of building blocks in foulants of the CG-AnMBR (Hong et al., 2012).

Table 6.3 Organic fractions of membrane foulants based on LC-OCD analysis

Operating conditions		DOC [a] dissolved	HOC [b] Hydrophobic	CDOC [c] Hydrophilic	Biopolymers	Humic substances	Building blocks	LMW neutrals and acids
Description	G-AnMBRs	ppb-C (% DOC)	ppb-C (% DOC)		ppb-C (% DOC)			
Foulant	SA-GAnMBR	5360 (100%)	508 (9.5%)	4852 (90.5%)	918 (17.1%)	1675 (31.3%)	743 (13.9%)	1516 (31.2%)
Foulant	CG-AnMBR	5373 (100%)	152 (2.8%)	5221 (97.2%)	1857 (34.6%)	565 (10.5%)	915 (17.0%)	1884 (35.1%)

[a] DOC = dissolved organic carbon, [b] HOC = hydrophobic organic carbon , [c] CDOC = chromatographic dissolved organic carbon

6.5 Effects of sponge addition on VFA accumulation and biogas production

VFA serves as the most important process indicator for biogas production from G-AnMBRs not only because it can significantly influence pH value of the reactor but also due to the fact that it is the vital intermediary substrate for the methane generation (Gulhane et al., 2017). Approximately 75% of methane yield comes from decarboxylation of acetic acid (main component of VFA) and the rest 25% is from CO_2 and H_2 (McCarty and Smith, 1986). If existing in high concentrations, VFA can also cause significant pH drop and pose enormous stress on sensitive methane-producing bacteria, thus ultimately resulting in G-AnMBRs reactor acidification and low biogas production (Jeihanipour et al., 2013; Chen et al., 2016b; Peng et al., 2016; Roberts et al., 2016). In this chapter, seven types of VFAs including acetic (C_2), propionic (C_3), iso-butyric (i-C_4), n-butyric (n-C_4), iso-valeric acid (i-C_5), n-valeric (C_5) and caproic acid (C_6) were monitored. The SA-GAnMBR exhibited much lower level of acetic acid with the average value of 3.5 ± 0.8 mg/L, while other acids were at undetectable level (Table 6.4). The results revealed that there was no VFA accumulation in the SA-GAnMBR, and reactor acidification was rarely encountered over the operation time. Therefore, the sponge could help to maintain a well-functioning granular sludge bed and efficient VFA degradation.

In contrast, the CG-AnMBR demonstrated much higher VFA concentrations with an average value of 20.2 ± 2.7 mg/L (5.8 times higher than that of the CG-AnMBR). VFA accumulation was mainly attributed to the existence of acetic acid (67.4 ± 7.7%) in the mixed liquor. C_3, i-C_4, n-C_4, i-C_5 and n-C_5 were also detected in the CG-AnMBR. The accumulation of intermediate products VFA might be related to the VFA release as a result of granule disintegration or deteriorated methanogenic process. The stability of methanogensis process is the key to the efficient biogas production. Since methanogens are very sensitive to environmental factors (oxidation/reduction potential (ORP), pH, etc), any variations in the operating conditions may cause inhibition for biogas production. Average pH values were found at 7.3 ± 0.3 and 6.9 ± 0.2 in the SA-GAnMBR and CG-AnMBR, respectively, even though pH was not controlled. In the CG-AnMBR, the higher VFA concentrations were accompanied by lower values of pH

(Zhang et al., 2016). The changes of the ORP were also recorded. The ORP value in the SA-GAnMBR was -318.4 ± 8.9 mV, which was 58.9 ± 8.9 mV lower than that in the CG-AnMBR on average. Lower ORP favoured the survival and growth of methanogens, therefore enhancing the transformation of VFAs into CH_4 (Zhang et al., 2011).

Table 6.4 VFAs concentrations in the CG-AnMBR and the SA-GAnMBR

VFA	CG-AnMBR		SA-GAnMBR	
	Concentration (mg/L)	Fraction of VFA (%)	Concentration (mg/L)	Fraction of VFA (%)
C_2 [a]	13.6 ± 2.6	67.4 ± 7.7	3.5 ± 0.8	100
C_3 [b]	1.4 ± 0.9	6.9 ± 4.8	0	0
i-C_4 [c]	1.0 ± 0.6	5.2 ± 3.5	0	0
n-C_4 [d]	0.9 ± 0.7	4.6 ± 3.7	0	0
i-C_5 [e]	1.1 ± 0.8	5.6 ± 4.4	0	0
n-C_5 [f]	2.2 ± 2.2	10.3 ± 9.3	0	0
C_6 [g]	0	0	0	0
V_T [h]	20.2 ± 2.7	100	3.5 ± 0.8	100

[a] C_2=acetic acid, [b] C_3=propionic acid, [c] i-C_4=iso-butyric acid, [d] C_4=butyric acid, [e] i-C_5=iso-valeric acid, [f] C_5=valeric acid, [g] C_6= caproic acid, [h] V_T=total volatile fatty acids

According to Table 6.5, the SA-GAnMBR produced more biogas (486 ± 12 mL/d) than the CG-AnMBR (456 ± 9 mL/d) with similar methane and carbon dioxide composition in the biogas (69.8 and 26.5%, 67.5 and 28.1%, respectively). Very small amount of H_2 with 5 - 12 ppm was also detected in the biogas from both reactors. The CG-AnMBR achieved methane yield at 133.3 ± 5.3 mL $CH_{4\ (STP)}$/g $COD_{removed}$, volume of methane produced at and 0 °C Standard Temperature and 1 atm Pressure), while the SA-GAnMBR had higher methane yield of 156.3 ± 5.8 mL $CH_{4\ (STP)}$/g $COD_{removed}$. The methane yield from the SA-GAnMBR represented around 50% of the optimal theoretical value of 318 mL $CH_{4\ (STP)}$/g $COD_{removed}$. As it was reported that methane loss in the liquid phase from the anaerobic MBR could be as much as 30% and 50% at 35 °C and 15 °C, respectively (Smith et al., 2012), nearly half of degraded COD might convert to dissolved methane and lost. Considering the economic and environmental impacts, methane leakages have to be paid much attention to and minimized (Chynoweth et al., 2001; Karakurt et al., 2012). The development of feasible and effective recovery process for dissolved methane is highly desired for the optimization of bioenergy recovery and minimization of greenhouse gas emissions to the atmosphere. The available recovery processes include biological oxidation of dissolved methane using down-flow hanging sponge reactor (Hatamoto et al., 2011), removal of residual dissolved methane using degassing membrane (Moreno et al., 2016) and post-treatment aeration to strip of AnMBR effluent (McCarty et al., 2011).

Table 6.5 Biogas yield from the CG-AnMBR and the SA-GAnMBR

Parameter	SA-GAnMBR	CG-AnMBR
Biogas volume (mL/d)	486 ± 12	456 ± 9 mL/d
Methane yield (mL CH_4/g COD $_{removed}$)	156.3 ± 5.8 at STP [a]	133.3 ± 5.3 at STP
Methane (%)	69.8 ± 4.2	67.5 ±4.8
Carbon dioxide (%)	26.5 ± 4.8	28.1 ± 4.5
Hydrogen (ppm)	9.2 ± 2.8	8.1 ± 3.1

[a] STP = volume of methane produced at and 0 °C Standard Temperature and 1 atm Pressure

6.6 Conclusions

In this chapter, the overall performance of SA-GAnMBR was comprehensively evaluated. The specific conclusions were drawn as follows:

1) The sponge addition into G-AnMBR could not only improve organics and nutrient removal, but also retain superior granular sludge properties and enhance methane yield.
2) The SA-GAnMBR exhibited prolonged operation time due to effective fouling mitigation. Assisted by sponge, the SA-GAnMBR showed lower SMP and EPS levels in settling zone mixed liquor, less EPS_P and SMP production in the cake layer as well as much lower cake layer and pore clogging resistance compared to those of the CG-AnMBR.
3) Fouling resistance analysis revealed that sponge addition could reduce the R_T by 50.7% via decreasing both cake layer and pore logging resistance.

4) LC-OCD analysis confirmed that lower biopolymers, LMW neutrals and acids and building blocks were presented in the SA-GAnMBR foulant.

CHAPTER 7

Evaluation of the performance of hybrid SAAMB-AnGMBR under different influent C/N ratios

7.1 Introduction

G-AnMBRs offer a promising opportunity to transform conventional municipal wastewater plants into net producers of renewable energy with significantly reduced sludge handling costs and energy demand while occupying a small footprint (Chen et al., 2017b). Owing to the competitive advantages of granular biomass, G-AnMBRs have gained particular interest for fouling mitigation since membrane fouling has remained as one of the most critical challenges, hindering the progress of C-AnMBRs, predominantly in the form of continuous stirred tank reactor configuration (Chen et al., 2017a). Martin-Garcia et al. (2011) reported that the G-AnMBR had much slower fouling than the C-AnMBR because the G-AnMBR sludge had a lower MLSS and 50% less of SMP than those of the C-AnMBR. More fouling reduction in G-AnMBR due to the significantly reduced solid and colloidal loading (by a factor of 10 and 3, respectively) on the membrane was also reported in another study of Martin-Garcia et al. (2013).

Recent research has found that the incorporation of membrane into the granular systems could negatively affect the integrity of anaerobic granules and lead to severe membrane fouling, thus exacerbating the long-term performance of G-AnMBRs (Ozgun et al., 2015; Chen et al., 2017b). The low-cost polyutrethane sponge, an ideal attached growth mobile carrier, has been successfully applied in many aerobic membrane bioreactors (AMBRs) studies to enhance the overall performance of AMBRs due to its high internal porosity and specific surface area, high stability to hydrolyse and light weight (Guo et al., 2010). Chen et al. (2017b) worked on a sponge-assisted G-AnMBR, and indicated that sponge addition into G-AnMBR could enhance organic and nutrient removal, and maintain superior granular quality. Additionally, sponge media could not only positively affect the concentration and properties of microbial products (e.g. SMP and EPS) in granular sludge, cake layer as well as settling zone mixed liquor, but also reduce fouling resistance by 50.7%, thereby alleviating membrane fouling.

Although studies have proved that the sponge addition could improve nutrient removal (Nguyen et al., 2011), nutrient removal efficiencies were still considered quite low in the SG-AnMBR (Chen et al., 2017b), limiting its universal appeal for municipal wastewater treatment (Smith et al., 2012). C/N ratio is one of the most influential parameters affecting nutrient removal process as it affects the population and biodiversity of functional microorganisms (Lin et al., 2016). Moreover, membrane fouling can be significantly influenced by C/N ratio because C/N ratio profoundly affects the physiological property of microorganisms and chemical composition of biomass, and influences the concentrations of EPS and SMP and their protein and polysaccharides contents (Hao et al., 2016).

In this chapter, a new hybrid SAAMB-AnGMBR was developed to overcome the two major issues (i.e. fouling and low nutrient removal) impeding the progress of G-AnMBRs. Based on the literature, it is the first development of the hybrid configuration for enhancing nutrient removal and fouling control of G-AnMBR during municipal wastewater treatment. The main aim of this chapter was to evaluate the effects of C/N ratio on the performance of such a hybrid system in terms of pollutants removal (particularly for nutrient removal) and membrane fouling. The system recovery after the overloaded nitrogen event was also evaluated in the chapter.

A major part of Chapter 7 will be submitted to Bioresource Technology:

Chen, C., Guo, W.S., Ngo, H.H., Chang, S.W., Nguyen, D.D., Zhang, J., Liang, S, Guo, J.B., Zhang, X.B., 2017. Effects of C/N ratio on the performance of a hybrid sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor for municipal wastewater treatment. Bioresource Technology.

7.2 Effect of C/N ratio on organic and nutrient removal

The experimental set-up, operation protocol and wastewater characteristics were depicted in Chapter 3 (Section 3.3.3). The membrane properties and sponge specification could be found in Chapter 3 (Section 3.2.2 and Section 3.2.3).

The hybrid SAAMB-AnGMBR system showed sound organic removal even though influent C/N ratio varied. COD and DOC removal efficiencies were kept over 94.8%

and 94.5% regardless of C/N ratios (Table 7.1). The results suggested that decreasing C/N ratio from 100/5 to 100/10 did not adversely influence the organic removal since complete retention of all particulate COD and macromolecular COD components was achieved by the membrane and most of the readily biodegradable COD was removed by aerobic and anaerobic biodegradation (Martinez-Sosa et al., 2011).

Table 7.1 Treatment performance of the hybrid SAAMB-AnGMBR under various C/N ratios

Removal efficiency (%)	C/N = 100/5	C/N = 100/6	C/N = 100/8	C/N = 100/10
COD	95.5 ± 3.5%	95.2 ± 2.4%	95.0 ± 1.8%	94.8 ± 2.0%
DOC	95.3 ± 3.1%	95.1 ± 3.2%	94.8 ± 1.7%	94.5 ± 2.2%
NH_4-N	93.9 ± 4.5%	93.4 ± 2.0%	93.2 ± 2.3%	92.5 ± 1.6%
TN	91.3 ± 4.7%	84.4 ± 3.1%	68.9 ± 3.8%	44.1 ± 5.8%
PO_4-P	93.3± 5.5 %	88.6 ± 5.6%	63.5 ± 5.7%	39.3 ± 7.2%

Similarly, stable NH_4-N removal (Table 7.1) over 92.5% was observed in spite of varying C/N ratios in influent, because the population of autotrophic nitrifying bacteria was not impacted by the change of C/N ratios (Lin et al., 2016). The removal efficiencies were higher than those reported by Khan et al. (2013), who obtained NH_4-N removal efficiencies of 89.3 - 90.5% at C/N ratios of 20 - 10. The higher NH_4-N removal in this study was mainly ascribed to sufficient aeration for nitrifying process and the enhanced population of nitrifying bacteria retained by sponge cubes (Deng et al., 2016a). The attached biomass on sponge carriers in the SAAMBR showed minor change, with 1.07 ± 0.033, 1.05 ± 0.047, 1.01 ± 0.077 and 1.03 ± 0.095 g MLVSS/g sponge at varying C/N ratios from 100/5 to 100/10, respectively. Furthermore, with the decreased C/N ratio, SNR of attached biomass was enhanced (Table 7.2), which contributed to stable NH_4-N removal.

When C/N ratio was maintained at 100/6 or higher, more than 84% of TN removal was achieved (Table 7.1). Due to effective simultaneous nitrification and denitrification (SND) process in the hybrid system, effluent NO_3-N concentrations were maintained at low levels with 0.39 ± 0.15 and 1.63 ± 0.54 mg/L at C/N ratios of 100/5 and 100/6. Table 7.2 showed the SDR values for each type of functioning biomass in the SAAMB-

AnGMBR system. Anaerobic granular biomass exhibited the highest SDR, hence contributing the most to the denitrification process. SDR of aerobic sponge-attached biomass were found comparable to that of anaerobic sponge-attached biomass, although DO in the aerobic compartment was maintained at 3.5 - 4.8 mg/L. The main reason was that nitrification occurred on sponge surfaces while declining DO levels along the sponge inward depth favored the formation of anoxic/anaerobic zones, contributing to efficient denitrification process (Khan et al., 2011; Deng et al., 2016b).

Table 7.2 Specific nitrification rates and denitrification rates (SNR and SDR) of biomass under different C/N ratios

	SNR (g NH_4-N/g MLSS· d)	SDR (g NO_3-N/g MLSS· d)
C/N = 100/5	0.40 ± 0.23 [a]	0.44 ± 0.11 [b]
		0.25 ± 0.02 [c]
		0.22 ± 0.05 [d]
C/N = 100/6	0.45 ± 0.24 [a]	0.40 ± 0.10 [b]
		0.23 ± 0.09 [c]
		0.21 ± 0.03 [d]
C/N = 100/8	0.49 ± 0.16 [a]	0.31 ± 0.16 [b]
		0.22 ± 0.13 [c]
		0.18 ± 0.17 [d]
C/N = 100/10	0.52 ± 0.21 [a]	0.25 ± 0.13 [b]
		0.21 ± 0.12 [c]
		0.19 ± 0.09 [d]

[a] = SNR of aerobic attached biomass on sponge cube; [b] = SDR of anaerobic granular biomass; [c] = SDR of anaerobic attached biomass ; [d] = SDR of aerobic attached biomass on sponge cube

However, lower C/N ratios of 100/8 and 100/10 showed significant reduction on TN removal efficiencies to 68.9% and 44.1%, respectively. Biological nitrogen removal is achieved by nitrification (an autotrophic bioprocess), followed by denitrification (a heterotrophic bioprocess) (Kim et al., 2016). Since the NH_4-N removal efficiencies were kept more than 92.5% at all designated C/N ratios, the low TN reduction was mainly attributed to the poor denitrification when insufficient carbon was present for heterotrophic denitrifier for cell growth and nitrate reduction (Yang et al., 2012). Insufficient carbon source restrained the denitrification process, caused relatively higher NO_3-N concentrations in the effluent (5.33 ± 1.08 and 12.46 ± 1.19 mg/L for C/N ratios of 100/8 and 100/10, respectively) and broke the balance between nitrification and denitrification processes (Lin et al., 2016).

Low carbon source not only negatively affected the abundance of heterotrophic population but also significantly influenced vital parameters of denitrification process such as SDR. SDR of granular biomass was significantly reduced when decreasing C/N ratio to 100/8 and 100/10 (Table 7.2). This finding might be related to the deteriorated granule quality (details discussed in Section 3.2.3) as a result of decreasing C/N ratio. However, the attached growth in the SS-AnGMBR kept relatively stable at 1.74 - 1.84 g MLVSS/g sponge over the entire operation period. The impact of C/N ratio on the SDR of aerobic and anaerobic attached biomass could be neglected since their values did not vary significantly. Nitrite production, known to strongly inhibit denitrification process (Yang et al., 2012), was found not the reason for the low TN removal under low C/N ratio since very low nitrite concentrations (<1.52 mg/L) were detected in both SAAMBR and SS-AnGMBR.

Monclús et al. (2010) suggested that phosphate could be occupied by phosphate accumulating microorganisms (PAOs) in the oxic/anoxic zones or consumed for biomass growth during the operation. Since the SND process effectively reduced NO_3-N content at higher C/N ratios of 100/5 and 100/6, low amount of NO_3-N in the anoxic zones of the biofilm contributed to effective PO_4-P release, hence promoting 93.3% and 88.6% of PO_4-P removal, respectively (Deng et al., 2016b). However, when further reducing C/N ratio to 100/8 and 100/10, PO_4-P removal declined significantly to 63.5% and 39.3%, as higher concentration of NO_3-N could inhibit phosphorus release and result in sharp reduction of PO_4-P removal. Lower C/N ratio may also cause excessive growth of heterotrophic bacteria, hence limiting the growth of PAOs and slow growing denitrifiers (Lin et al., 2016; Khan et al., 2013).

VFAs were deemed to be the most preferred carbon source for denitrifier and PAOs (Singhania et al., 2013). It is interesting to observe that more VFAs were accumulated with increased nitrogen content in the feed, and total VFAs concentration for each C/N ratio was 2.77 ± 0.56 (100/5), 5.72 ± 1.12 (100/6), 9.38 ± 1.02 (100/8) and 11.51 ± 1.28 (100/8) mg/L. Acetic acid was found the most sensitive to the change of C/N ratio, whose concentration at C/N ratio of 100/10 (8.92 ± 0.95) was nearly 4 times higher than that at C/N ratio of 100/5 (1.82 ± 0.35 mg/L). Higher levels of residual VFAs, especially for acetic acid at lower C/N ratio, might indicate the inhibition of denitrification and phosphate removal processes. In addition, decreasing methane yields were found at 137.40 ± 3.83, 126.58 ± 3.07, 115.25 ± 2.29 and 102.12 ± 1.92 mL CH_4 $_{(STP)}$/g COD $_{removed}$ (STP: volume of methane produced at and 0 °C Standard Temperature and 1 atm Pressure) at C/N ratios of 100/5, 100/6, 100/8 and 100/10, respectively.

7.3 Effect of C/N ratio on membrane fouling behavior

7.3.1 TMP profile

Figure 7.1 showed that the TMP of the hybrid system increased from 0.90 to 30.80 kPa in 75 days (C/N ratio = 100/5); 1.20 to 30.80 kPa (100/6) in 50 days; 1.10 to 30.20 kPa in 25 days (100/8); 1.20 to 30.90 kPa in 15 days (100/10), respectively.

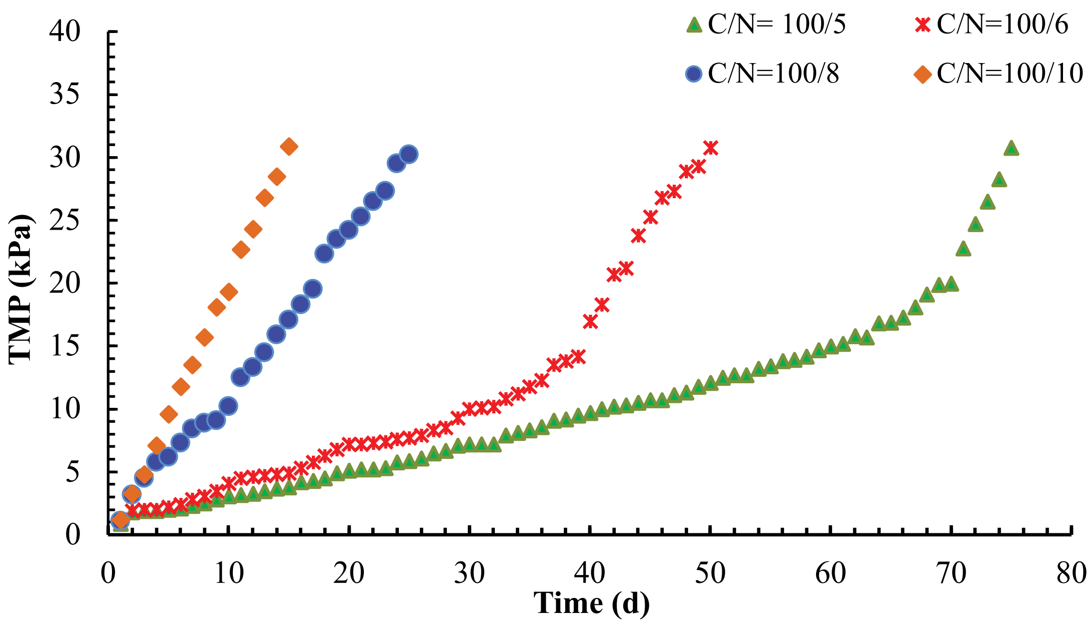


Figure 7.1 TMP profiles for the hybrid SAAMB-AnGMBR at different C/N ratios

Accordingly, the SAAMB-AnGMBR had the fastest fouling rate of 1.98 kPa/d at C/N ratio of 100/10, which was almost 4 times higher than the corresponding value (0.40 kPa/d) obtained at C/N ratio = 100/5. While for C/N ratios of 100/6 and 100/8, the fouling rates were 0.59 and 1.16 kPa/d, respectively. These results implied that lowering influent C/N ratio could deteriorate membrane filterability and shorten the operation period before membrane cleaning. The cause is discussed in Section 7.3.2.

7.3.2 EPS and SMP in the mixed liquor

In the SS-AnGMBR, the membrane module was only challenged by the mixed liquor in the settling zone. Hence, the EPS and SMP concentrations of the mixed liquor and their polysaccharide (EPS_C, SMP_C) and protein contents (EPS_P, SMP_P) were analysed in order to explain the relationship between the mixed liquor properties and fouling propensity, and the results were shown in Table 7.3. EPS at C/N ratio of 100/10 (48.41 mg/L) were found more than 4.27, 2.04, 1.29 times to the corresponding values obtained at C/N ratios of 100/5, 100/6, 100/8 (11.33, 23.66, 37.40 mg/L), respectively. At the lowest C/N ratio (100/10), the SS-AnGMBR contained the highest EPS content, which was mainly responsible for the highest fouling rate. Studies have reported that

EPS were biopolymers attached on flocs or cells surface, and were known to significantly influence membrane fouling (Hao et al., 2016), because EPS were closely associated with cake layer formation. Any variation in EPS concentration and composition would alter the interactions between floc surface and membrane surface or between fouling layer surface and floc surface, thus affecting cake layer formation rate and structure (Hao et al., 2016). Lower EPS_c/EPS_p ratio in sludge flocs was reported to increase floc hydrophobicity, thus deteriorating membrane fouling by their deposition on the membrane (Deng et al., 2016c). In this study, EPS_C/EPS_P ratio remained stable, mainly fluctuating from 0.46 to 0.61, suggesting that influent C/N ratio exerted a limited impact on EPS composition. Hence, EPS composition was not an influential factor affecting fouling.

SMP, known as biopolymers released from microorganisms into solution, has the strongest relationship with fouling rate (Zhang et al., 2015). In this study, despite the increase in N concentration in the feed, SMP showed the relatively low concentrations (5.58 - 8.08 mg/L) at all four C/N ratios. This might be due to the fact that sponge addition into hybrid system could significantly reduce the mixed liquor SMP values (Deng et al., 2014). The highest SMP value was observed at C/N ratio of 100/5 (8.08 mg/L), followed by those at C/N ratios of 100/6 (7.53 mg/L), 100/8 (6.85 mg/L) and 100/10 (5.58 mg/L). The higher SMP levels at higher C/N ratios in the hybrid system might be due to the SMP accumulation with evolution of time in the mixed liquor since prolonged operational time was recorded at higher C/N ratios (i.e. operation time were 75, 50, 25, 15 days for C/N ratios of 100/5, 100/6, 100/8, 100/10, respectively). In this case, the total SMP concentration was not responsible for faster fouling at lower C/N ratios.

Nevertheless, an increase in SMP_C/SMP_P was positively correlated with faster fouling rate at lower C/N ratio. The averaged values of SMP_C/SMP_P increased from 0.54 to 0.79, 1.14, and 1.44 when lowering C/N ratios from 100/5 to 100/6, 100/8 and 100/10, respectively, and it suggested that higher amount of hydrophilic SMP_C than SMP_P were presented in the mixed liquor at lower C/N ratios. SMP_C were more susceptible to membrane fouling since they could penetrate into the cake layer and membrane pore and lodge insides, thus inducing severe pore clogging and gel layer formation, and causing significant membrane filterability loss (Deng et al., 2016a).

Table 7.3 The concentration and properties of EPS and SMP in the mixed liquor of the SS-AnGMBR settling zone and anaerobic granular sludge

		C/N = 100/5	C/N = 100/6	C/N = 100/8	C/N = 100/10
Mixed liquor	EPS (mg/L)	11.33 ± 4.41	23.66 ± 6.04	37.40 ± 6.89	48.41 ± 7.13
	EPS_C/EPS_P	0.53 ± 0.03	0.46 ± 0.04	0.61 ± 0.04	0.54 ± 0.08
	SMP (mg/L)	8.08 ± 1.49	7.53 ± 1.78	6.85 ± 1.47	5.58 ± 1.07
	SMP_C/SMP_P	0.54 ± 0.04	0.79 ± 0.04	1.14 ± 0.28	1.44 ± 0.31
Granular sludge	EPS_P (mg/g VSS)	27.03 ± 5.13	13.30 ± 3.75	7.57 ± 2.12	4.92 ± 1.08
	EPS_C (mg/g VSS)	9.86 ± 2.36	5.15 ± 1.63	2.99 ± 1.02	2.18 ± 0.93
	SMP_P (mg/g VSS)	4.68 ± 1.23	9.58 ± 2.58	13.02 ± 2.88	14.21 ± 3.23
	SMP_C (mg/g VSS)	2.13 ± 0.85	1.95 ± 0.91	2.08 ± 0.95	2.12 ± 1.02

7.3.3 EPS and SMP in the granular sludge

The SMP and EPS of the mixed liquor were originated from the SMP and EPS of the granular sludge in the SS-GAnMBR. Hence, the analysis of the granule sludge properties could help understand the changes of mixed liquor SMP and EPS as a result of the impact of C/N ratio. According to Table 7.3, both EPS_P and EPS_C concentrations of the granular sludge were noticeably reduced when influent N was increased. The lowest EPS concentration (EPS_P and EPS_C of 4.92 and 2.18 mg/L, respectively) was observed at C/N ratio of 100/10. Since EPS played an vital role on integrating cells into granules, the dramatic decrease in EPS content at lower C/N ratio might indicate weaker, scattered and looser structures of flocs and granules, contributing to granules breakage (reduced sludge particle size) and to the increase in EPS of the mixed liquor in settling zone (Chen et al., 2017b).

Table 7.4 Summary of sludge properties of seed sludge and granular sludge in the SS-AnGMBR at different C/N ratios

	SVI (mL/g)	Settling velocity (m/h)	Median particle size (μm)	Zeta-potential (mV)
Seed	34.8 ± 2.3	15.8 - 24.8	259 ± 22	-16.8 ± 3.9
C/N = 100/5	21.3 ± 5.8	17.1 - 32.4	355 ± 18	-14.2 ± 2.3
C/N = 100/6	34.2 ± 6.1	15.6 - 27.2	308 ± 28	-15.9 ± 2.4
C/N = 100/8	45.5 ± 4.1	12.4 - 21.8	223 ± 21	-17.4 ± 3.0
C/N = 100/10	53.5 ± 8.3	10.2 - 17.4	165 ± 19	-20.3 ± 4.2

The granular growth rates (ΔMLSS/Δt) were found at 0.052, 0.045, 0.038 and 0.035 g/L·d, at C/N ratios of 100/5, 100/6, 100/8 and 100/10, respectively, suggesting that lower C/N ratio discouraged the growth of retained sludge agglomerates in the granular sludge bed. PSD analysis confirmed that smaller anaerobic granular sludge particles existed when operating at lower C/N ratios. Granular sludge with the biggest median particle size of 355 μm was observed at C/N ratio of 100/5, while lower values of 308 μm, 223 μm and 165 μm were found at C/N ratios of 100/6, 100/8 and 100/10, respectively. As a result, the disintegration of granules decreased settling capacity of the granular sludge, which could be seen from decreased settling velocity and zeta-potential and increased SVI (Table 7.4).

Additionally, the deterioration of the granular sludge settling capacity also promoted inefficient solid entrapment of the granular sludge bed. Hence, this encouraged the accumulation of fine sludge particles such as non-settling particles and small colloidal substances in the mixed liquor of the settling zone due to the membrane complete retention of particulate and colloidal matter and biomass. The elevated EPS levels in the mixed liquor (Section 7.3.2) at lower C/N ratios were mainly attributed to the higher MLSS concentrations in the mixed liquor of SS-AnGMBR settling zone as a result of granule breakage. At the end of experiment, the MLSS concentration in the SS-AnGMBR reached up to 293.8 mg/L at C/N ratio of 100/10, which was nearly 1.5, 2.7 and 3.9 times higher than those at C/N ratios of 100/8, 100/6 and 100/5, respectively. Moreover, the highest SMP level in the granular sludge (SMP_P and SMP_C of 14.21 and 2.12 mg/g VSS) was observed at C/N ratio of 100/10 (Table 7.3), which confirmed that the major fraction of the proteins and polysaccharides existed in the soluble form rather than being the part of the anaerobic granules in the form of EPS. These results highlighted that severe fouling at lower C/N ratios was not only associated with larger amounts of EPS, higher SMP_C/SMP_P ratio of mixed liquor but also due to the deteriorated granule quality.

7.3.4 Fouling resistance and cake layer analysis

Membrane fouling resistances were obtained for each of C/N regimes when cleaning of membrane was performed (Figure 7.2). With increasing N dose in the feed, R_T increased from 2.61×10^{12} to 3.65×10^{12} m^{-1} at C/N ratios of 100/5 and 100/6, which further rose to 5.62×10^{12} and 6.56×10^{12} m^{-1} at C/N ratios of 100/8 and 100/10, respectively. R_C and R_P values followed the similar trends as shown in Figure 7.2 in which R_C (2.03×10^{12} to 5.19×10^{12} m^{-1}) increased with a greater magnitude than R_P (3.9×10^{11} to 1.18×10^{12} m^{-1}), while R_M (1.89×10^{11} to 1.92×10^{11} m^{-1}) remained stable for all C/N ratios. These results suggested that higher N dose in the feed deteriorated membrane permeability due to more cake formation and pore clogging. Although R_p contributed to declined permeability, the values of R_C/R_T (over 77% at all C/N ratios) indicated that cake formation was the predominant fouling mechanism of the hybrid system, contributing more significantly to membrane fouling. As discussed in Section 7.3.2, the mixed liquor containing SMP with higher SMP_C/SMP_P ratio and higher amounts of EPS was responsible for the elevated R_C and R_P at lower C/N ratios. More EPS resulted in cake formation whist SMP with higher SMP_C/SMP_P ratio could modify the surface properties of membrane and outer cake layer to promote the self-accelerating fouling phenomena (accelerate cake formation rate) (Hao et al., 2016).

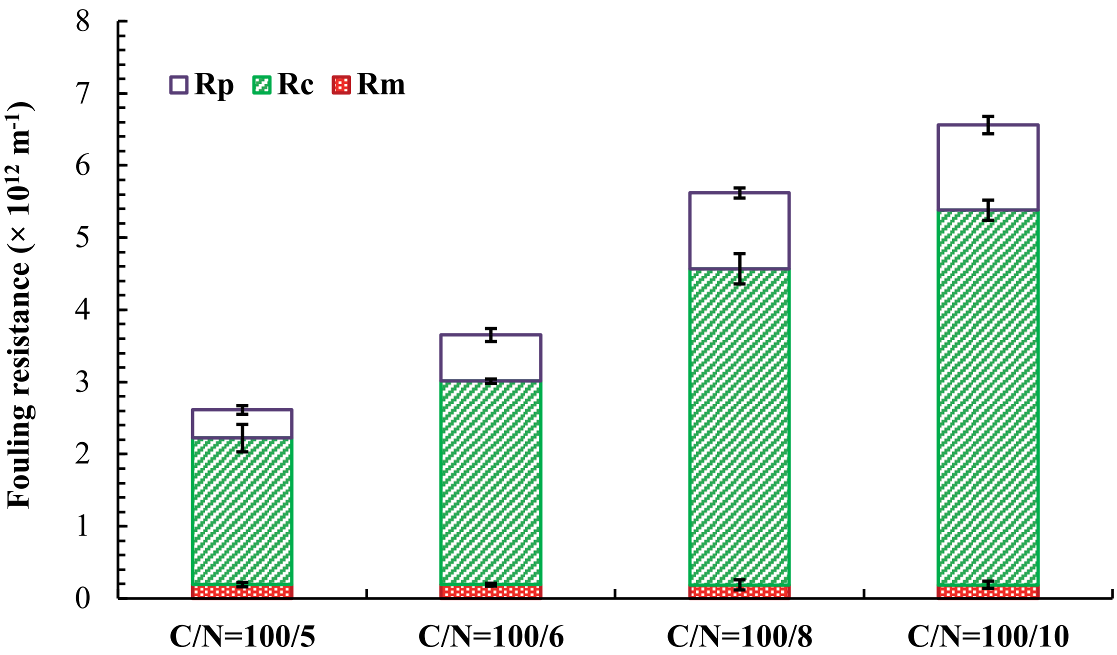


Figure 7.2 Fouling resistance distribution under different C/N ratios

Cake layer as the dominant mechanism of R_T was further investigated and characterized by the composition of EPS and SMP. As shown in Figure 7.3, the cake layer contained the lowest concentration of EPS_P and EPS_C (1.82 mg/g MLSS and 3.28 mg/g MLSS, respectively) at the C/N ratio of 100/5. The reduction of the C/N ratio to 100/6 induced a noticeable rise of EPS_P and EPS_C to 4.08 mg/g MLSS and 7.32 mg/g MLSS, respectively. Further increasing N dose to the C/N ratio of 100/8 facilitated a notable increase of EPS levels (EPS_P = 8.28 mg/g MLSS and EPS_C = 15.25 mg/g MLSS, respectively). The highest EPS_P and EPS_C levels were observed at the lowest C/N ratio of 100/10, which were 6.0 and 7.8 times to the corresponding values obtained at the C/N ratio of 100/5. The results revealed that higher nitrogen dose increased R_C by the accumulation of EPS within the sludge cake on the membrane surface. At lower C/N ratio, higher suction force due to faster TMP increment and increased drag force toward the membrane could enhance deposition of more EPS on membrane surface to form cake layer (Deng et al., 2016a). On the other hand, SMP extracted from the cake presented significantly low concentrations and slight variations. SMP_P amounts were maintained in the range of 0.47 - 0.52 mg/g MLSS while SMP_C ranged from 0.87 to 1.08 mg/g MLSS. It implied that SMP in the cake layer had minimal influence on the membrane fouling, which further confirmed that SMP were not the primary fouling factor for this study as discussed in Section 7.3.2.

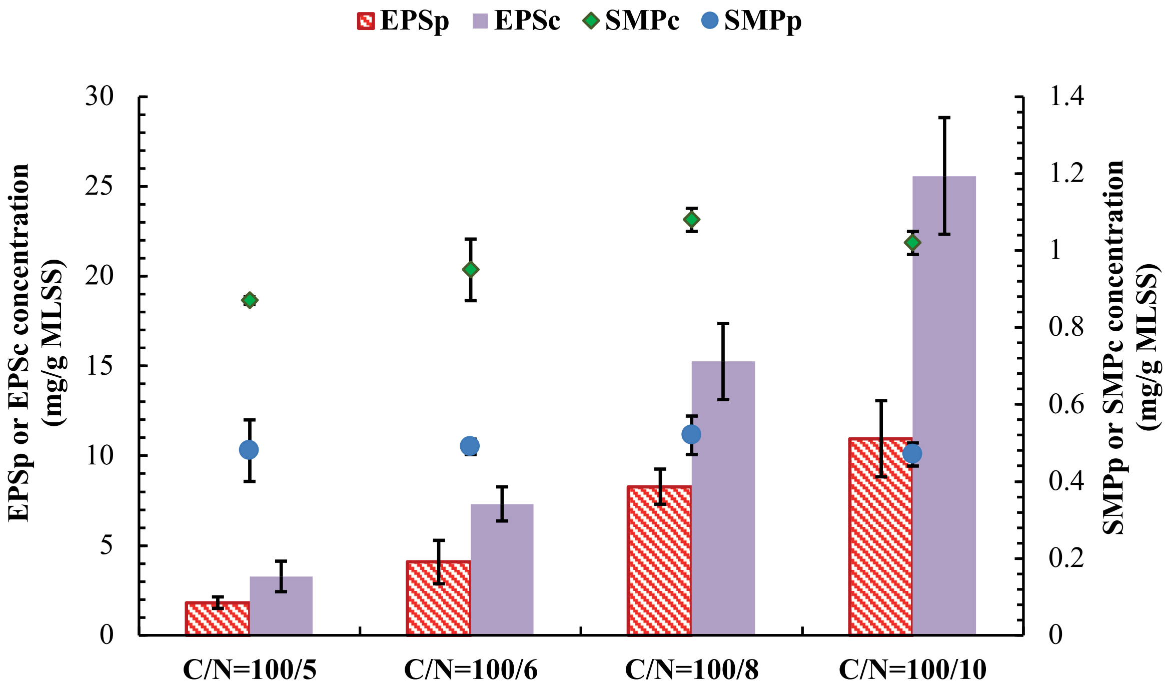


Figure 7.3 Compositions of EPS and SMP of cake layer in the hybrid system at different C/N ratios

7.3.5 LC-OCD and EEM analysis

LC-OCD provides critical information regarding the different components of the foulant organics by dividing the total organics into two groups: hydrophobic organics (HPO) and hydrophilic organics (HPI). The hydrophilic group can be further characterized into subgroups such as biopolymers, humic substances, building blocks, and lower molecular weight (LMW) neutrals and acids (Johir et al., 2012). Figure 7.4 showed that larger HPO occurred at lower C/N ratios, but they might not be the main foulants due to their much lower concentrations as compared to those of HPI. Within HPI, lower nitrogen dose favoured the accumulation of biopolymers in the foulant while humic acids were dominant at higher N dosage, contributing to the faster fouling rate. Building blocks, as the breakup of the humics, were found decreasing in response to the increase in influent nitrogen content. Though building blocks and LMW neutrals and acids were found in relatively lower concentrations, Aryal et al. (2009) suggested that they were the significant factors affecting fouling as their assemblage could promote biopolymers formation on the membrane surface, exacerbating fouling propensity.

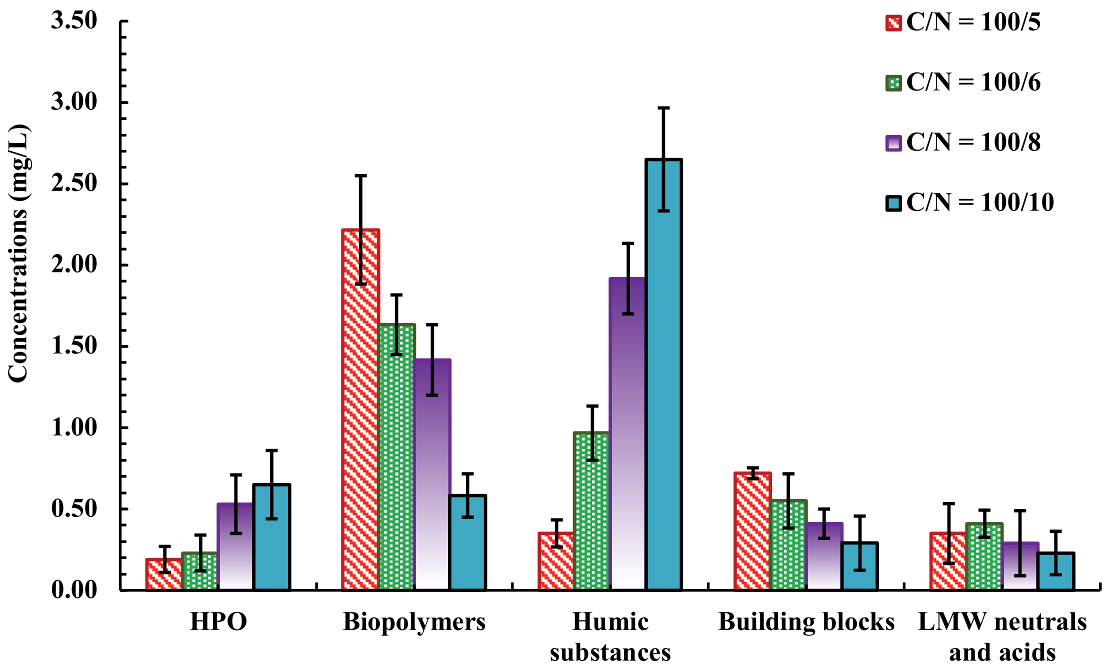


Figure 7.4 Nature of foulant organics at different C/N ratios

Figure 7.5 presented the spectra of membrane foulant organics obtained at different feed C/N ratios. The observation indicated that the hybrid system at the different C/N ratios yielded different organics structures and components in the membrane foulants. Peak A and peak B at Exitation/Emission (Ex/Em) of 270/300 and 220/320 nm under C/N ratio of 100/5 (Figure 7.5 (a)) were associated with biopolymers and their precursors (LMW small amino acid type organics) in the foulant. In comparison, the foulant at C/N ratio of 100/6 (Figure 7.5 (b)) demonstrated less intense spectra at the similar peak A and B locations, indicating the foulant had lower content of biopolymers along with their precursors. In contrast, Figure 7.5 (c) and Figure 7.5 (d) displayed humic and fulvic acid types peaks (Peak C and D) at Ex/Em of 310/390 and 230/390 nm, which suggested that foulant formed at C/N ratios of 100/8 and 100/10 contained higher content of humics but less biopolymers. The EEM results were found in agreement with the LC-OCD results in the sense that the higher N dosage at lower C/N ratio favoured the relative dominance of the humic-like substances in the extracted foulant, resulting in faster fouling. Similar results were reported by Hong et al. (2012) that humics were prevalent when the fastest fouling was observed.

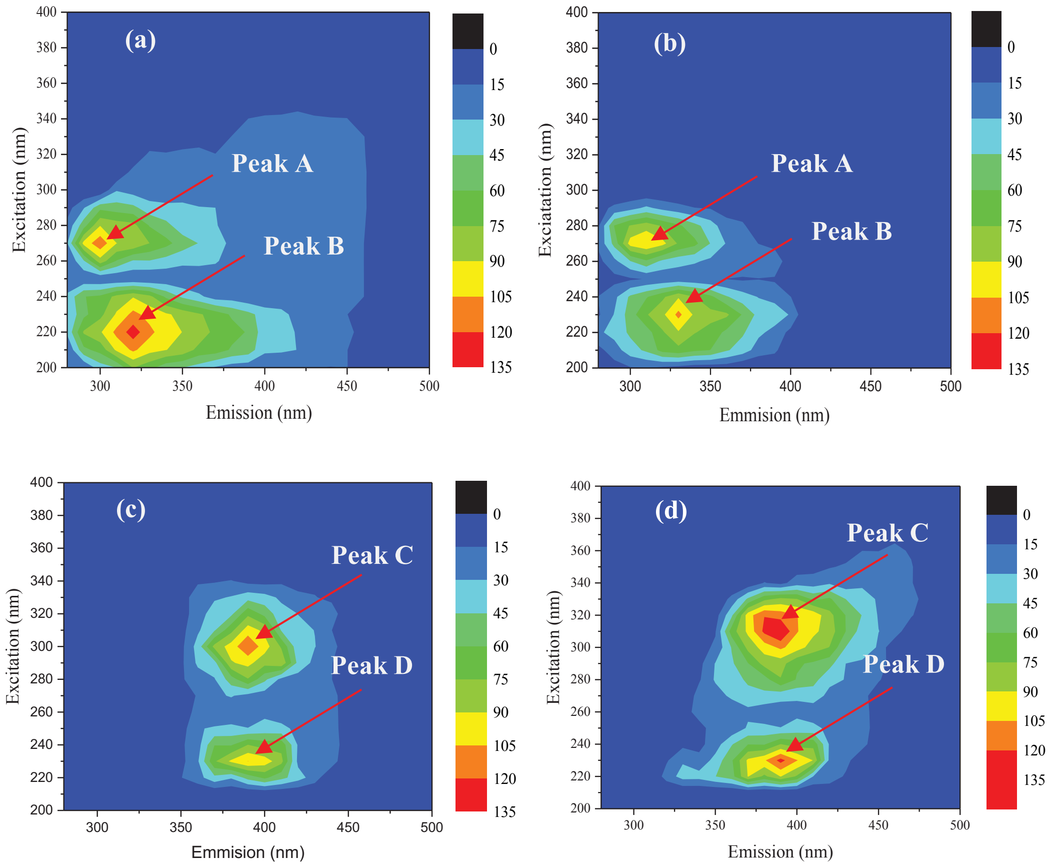


Figure 7.5 EEM fluorescence spectra of membrane foulants at different C/N ratios

7.4 System recovery from overloaded nitrogen events

As mentioned above, the performance of hybrid system including nutrient removal and membrane filtration was deteriorated in phase 1 as a result of the decrease in C/N ratios from 100/5 to 100/10. Therefore, the hybrid system was further investigated in phase 2 by gradually increasing C/N ratio back to 100/5 to observe the extent of system recovery after the overloaded nitrogen event.

7.4.1 Organic and nutrient removal

The system performed reasonably well in term of organic and NH_4-N removal in phase 2, with the comparable removal efficiencies of DOC and COD over 95.2% and NH_4-N over 93.5%. On the other hand, TN and PO_4-P removal efficiencies (82.3 ± 8.8% and 85.7 ± 6.7%, respectively) were found significantly improved after C/N ratio was increased from 100/10 to 100/6. When further increasing C/N ratio to 100/5, the hybrid system could achieve TN and PO_4-P removal efficiencies of 90.1 ± 6.8% and 92.1 ± 8.2%, respectively, indicating that the inhibitory effects on denitrification and phosphate removal processes imposed by overloaded nitrogen were not permanent. The system recovery in terms of nutrient removal was mainly attributed to efficient SND process and effective elimination of residual NO_3-N in the system (Deng et al., 2016b).

Table 7.5 System recovery in terms of organic and nutrient removal in phase 2

Removal efficiency (%)	COD	DOC	NH_4-N	TN	PO_4-P
C/N (100/6)	95.2 ± 7.3%	95.3 ± 7.7%	93.6 ± 5.7%	82.3 ± 8.8%	85.7 ± 6.7%
C/N (100/5)	95.4 ± 6.9%	95.6 ± 8.2%	93.8 ± 4.5%	90.1 ± 6.8%	92.1 ± 8.2%

7.4.2 Membrane fouling

Table 7.6 showed that the fouling rate was significantly reduced when increasing the C/N ratio from 100/10 to 100/6 and 100/5 in phase 2. The system showed much lower fouling rates of 0.69 and 0.47 kPa/d at the C/N ratios of 100/6 and 100/5, compared to that at the C/N ratio of 100/10 (1.98 kPa/d). The lower fouling propensity at lower feed nitrogen content was mainly due to much reduced pore clogging and cake layer formation on the membrane surface. As compared to fouling resistance at the C/N ratio of 100/10 (6.56×10^{12} m^{-1} of R_T), the hybrid system in phase 2 exhibited much lower R_T of 3.89×10^{12} m^{-1} and 2.80×10^{12} m^{-1} at the C/N ratios of 100/6 and 100/5, respectively. Accordingly, R_C were found at 2.97×10^{12} m^{-1} (76.3% of R_T) and 2.18×10^{12} m^{-1} (77.9% of R_T) while R_P were observed at 7.31×10^{11} m^{-1} (18.8% of R_T) and 4.32×10^{11} m^{-1} (15.4% of R_T) at the C/N ratios of 100/6 and 100/5, respectively, compared with those higher values obtained at the C/N ratio of 100/10 (5.19×10^{12} m^{-1} of R_C, 1.18×10^{12} m^{-1} of R_P). According to Table 7.6, EPS contents in the mixed liquor were maintained at 28.38 and 13.21 mg/L at the C/N ratios of 100/6 and 100/5, respectively, which were about only 59% and 27% and of the corresponding value obtained at the C/N ratio of 100/10 (48.41 mg/L).

Table 7.6 The summary of membrane fouling analysis results during system recovery after overloaded nitrogen event

		C/N = 100/6	C/N = 100/5
Fouling rate	R (kPa/d)	0.69	0.47
Fouling resistance (m^{-1})	R_M (m^{-1})	$(1.93 \pm 0.08) \times 10^{11}$	$(1.88 \pm 0.06) \times 10^{11}$
	R_C (m^{-1})	$(2.97 \pm 0.11) \times 10^{12}$	$(2.18 \pm 0.20) \times 10^{12}$
	R_P (m^{-1})	$(7.31 \pm 0.22) \times 10^{11}$	$(4.32 \pm 0.35) \times 10^{11}$
	R_T (m^{-1})	$(3.89 \pm 0.21) \times 10^{12}$	$(2.80 \pm 0.30) \times 10^{12}$
SMP and EPS in the mixed liquor	EPS (mg/L)	28.38 ± 8.34	13.21 ± 6.83
	EPS_C/EPS_P	0.48 ± 0.05	0.51 ± 0.04
	SMP (mg/L)	6.24 ± 1.85	8.38 ± 1.08
	SMP_C/SMP_P	0.88 ± 0.11	0.59 ± 0.05
SMP and EPS in the cake layer	EPS_P (mg/g MLSS)	6.12 ± 0.89	2.35 ± 0.51
	EPS_C (mg/g MLSS)	9.83 ± 1.05	4.58 ± 0.91
	SMP_P (mg/g MLSS)	0.75 ± 0.12	0.49 ± 0.16
	SMP_C (mg/g MLSS)	0.98 ± 0.07	0.92 ± 0.11
Major foulant organics	Biopolymer (mg/L)	1.72 ± 0.34	2.33 ± 0.73

The analysis of the cake layer revealed that the hybrid system in phase 2 contained less EPS_P (6.12 and 2.35 mg/g MLSS) and EPS_C (9.83 and 4.58 mg/g MLSS) at the C/N ratio of 100/6 and 100/5, respectively, compared to those at the C/N ratio of 100/10 in phase 1 (EPS_P, EPS_C = 10.95, 25.59 mg/g MLSS, respectively). Increasing C/N ratio helped reduce membrane fouling mainly through reducing EPS in the mixed liquor and cake layer, so as to alleviate cake layer fouling which was considered as the dominant mechanism of membrane fouling (Chen et al., 2017b; Hao et al., 2016). Moreover, compared with SMP_C/SMP_P (mixed liquor) of 1.44 at the C/N ratio of 100/10, lower SMP_C/SMP_P (0.88 and 0.59, respectively) under the C/N ratios of 100/6 and 100/5 also revealed less severe fouling in the hybrid system since less SMP_C/SMP_P led to lower R_P

and R_C (Deng et al. 2015). LC-OCD and EEM analysis of membrane foulant indicated that the major foulant organics were biopolymers. The results indicated that the impact of overloaded nitrogen on fast fouling propensity was also temporary.

7.5 Conclusions

In this chapter, the impact of C/N ratio on the performance of hybrid SAAMB-AnGMBR was comprehensively investigated. The specific conclusions were drawn as follows:

1) Reducing C/N ratio negatively affected TN and PO_4-P removal.
2) The SAAMB-AnGMBR exhibited faster fouling rate at higher C/N ratios due to worsened the granular quality, and exacerbated cake layer formation and pore blocking.
3) EPS was the influencing factor to membrane fouling at varying C/N ratios.
4) R_C and R_P increased at lower C/N ratio as a result of increased EPS and increased SMP_C/SMP_P ratio in the mixed liquor.
5) LC-OCD analysis confirmed that humics were the dominant organic foulant at lower C/N ratio when the fastest fouling occurred.
6) The system recovery could be achieved by increasing the C/N ratio from 100/10 to 100/5 through improving SND process and the properties of mixed liquor and cake layer.

CHAPTER 8
CONCLUSIONS AND RECOMMENDATIONS

8.1 Conclusions

This PhD thesis aimed at developing a specific integrated G-AnMBR for municipal wastewater treatment, and optimizing the G-AnMBR system by considering the effects of various factors such as HRT, membrane addition method, sponge incorporation and C/N ratio on the G-AnMBR performance in terms of treatment performance, renewable energy production, granular sludge quality and membrane fouling. The specific conclusions of the research were shown as follows:

(1) Preliminary research on G-AnMBR system

- After a failed startup, granules could be rapidly cultivated within 55 days using aerobic sludge as the inoculum in the modified G-AnMBR for municipal wastewater treatment.
- At optimal HRT of 12 h, COD and TOC removal efficiencies of more than 92% were achieved. Organic removal of 85% was found at HRTs of 16 h and 8 h due to low mass transfer (16 h) and granules disintegration (8 h). The biggest granules mean diameter was found at 255μm at HRT of 12 h.
- Either increasing HRT to 16 h or reducing HRT to 8 h decreased the granular size. At HRT of 12 h, the system retained the highest concentration of biomass as compared to HRT of 8 h and 16 h.

(2) External and submerged G-AnMBR

- Compared to the SG-AnMBR, the EG-AnMBR was found a better G-AnMBR configuration for municipal wastewater treatment due to less fouling propensity and superior granule quality. Both systems presented similar COD removal efficiencies and methane yield over 91%, and 160 mL $CH_{4\,(STP)}$/(g $COD_{removed}$) although VFA accumulation was found in the SG-AnMBR.
- Membrane direct incorporation into the SG-AnMBR significantly affected the concentration and properties of microbial products (e.g. SMP and EPS) in the cake layer, mixed liquor and granular sludge, as well as granular sludge size and settleability.
- The EG-AnMBR demonstrated less SMP and EPS in the mixed liquor and cake layer, which might reduce the cake layer resistance and lower the fouling rate.

- LC-OCD analysis of foulant revealed that biopolymers along with low molecular weight neutrals and acids and building blocks were responsible for higher fouling propensity in the SG-AnMBR.

(3) SA-GAnMBR system

- The SA-GAnMBR performed better than the CG-AnMBR in terms of treatment performance, granular sludge properties, membrane fouling and biogas production.
- The SA-GAnMBR showed better organics and nutrient removal, and enhanced methane yield at 156.3 ± 5.8 mL $CH_{4\ (STP)}$/g $COD_{removed}$. Granular sludge from the SA-GAnMBR had superior quality with better settleability, larger particle size, higher EPS content and more granule abundance.
- The SA-GAnMBR also exhibited slower fouling development with 50.7% lower total filtration resistance than those of the CG-AnMBR. Sponge addition effectively affected the concentration and properties of microbial products (e.g. SMP and EPS) in granular sludge, cake layer as well as settling zone mixed liquor, thus alleviating the fouling propensity.
- The LC-OCD analysis suggested that sponge addition reduced the concentrations of biopolymers, low molecular weight neutrals and acids, and building blocks of the foulants. GC-MS analysis confirmed the accumulation of volatile fatty acids, particularly acetic acid in the CG-AnMBR compared to those of the SA-GAnMBR.
- This work revealed that the SA-GAnMBR could be a promising solution for improving overall G-AnMBR performance and substantially mitigating membrane fouling.

(4) Hybrid SAAMB-AnGMBR

- The impact of C/N ratio on the performance of a new hybrid SAAMB-AnGMBR in municipal wastewater treatment was found significant. Decreased C/N ratio deteriorated TN and PO_4-P removal, worsened the granular quality, and exacerbated cake layer formation and pore blocking, thereby aggravating membrane fouling.

- R_C and R_P increased as a result of increased EPS and increased SMP_C/SMP_P ratio in the mixed liquor. This study also revealed humics were the dominant organic foulant at lower C/N ratio.
- Additionally, the system recovery could be achieved by increasing the C/N ratio from 100/10 to 100/5 through improving SND process and the properties of mixed liquor and cake layer.

8.2 Recommendations

This PhD thesis has comprehensively investigated the overall performance of different integrated G-AnMBR systems including SG-AnMBR, EG-AnMBR, SA-GAnMBR and SAAMB-AnGMBR. Nevertheless, further studies on these G-AnMBRs should be pursued in future works as summarized below:

1) Further research on microbiological analysis is needed to look into microbiological community structure and population in G-AnMBRs and DNA in the foulant;

2) Lab-scale experiments should be conducted for real domestic wastewater treatment for assessing the feasibility of G-AnMBRs in their practical application since the physical and chemical properties of real wastewater vary significantly over time;

3) Methane loss is a critical issue, especially at low temperatures, and hence quantifying, minimizing and recovering the dissolved methane in the permeate of G-AnMBR systems are of great necessity in the future study;

4) In the future research, the mechanism of enhanced granulation by sponge addition should be investigated to optimize the performance of sponge-assisted G-AnMBR systems.

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APPENDIX

LIST OF PUBLICATIONS

Peer Reviewed Journal articles:

Chen, C., Guo, W.S., Ngo, H.H., 2016a. Advances in granular growth anaerobic membrane bioreactor (G-AnMBR) for low strength wastewater treatment, Journal of Energy and Environmental Sustainability 1, 77 - 83.

Chen, C., Guo, W.S., Ngo, H.H., Lee, D.J., Tung, K.L., Jin, P.K., Wang, J., Wu. Y., 2016b. Challenges in biogas production from anaerobic membrane bioreactors. Renewable Energy 98, 120 - 134.

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Chen, C., Guo, W.S., Ngo, H.H., Liu, Y., Du, B, Wei, Q., Wei, D., Nguyen, D.D., Chang, S.W., 2017b. Evaluation of a sponge assisted-granular anaerobic membrane bioreactor (SG-AnMBR) for municipal wastewater treatment. Renewable Energy 111, 620 - 627.

Chen, C., Guo, W.S., Ngo, H.H., Chang, S.W., Nguyen, D.D., Zhang, J., Liang, S, Guo, J.B., Zhang, X.B., 2018. Effects of C/N ratio on the performance of a hybrid sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor for municipal wastewater treatment. Bioresource Technology 247, 340 - 346.

Book chapters:

Guo, W.S., Ngo, H.H., **Chen, C.**, Pandey, A., Tung, K.L., Lee, D.J., 2016. Anaerobic membrane bioreactors for future green bioprocess, in: Ngo, H.H., Guo, W.S., Surampalli, R.Y., Zhang, T.C. (Eds.), Green technologies for sustainable water management. American Society of Civil Engineers, USA, pp. 867 - 901.

CONTRIBUTIONS TO SCIENTIFIC FORUN

Chen, C., Guo, W.S., Ngo, H.H., Chang, S.W., Nguyen, D.D., Zhang, J., Liang, S, Guo, J.B., Zhang, X.B., 2017. Effects of C/N ratio on the performance of a hybrid sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor for municipal wastewater treatment (Oral presentation). 7th International Forum on Industrial Bioprocesses (IFIBiop 2017). Wuxi, China, 20 - 24 May, 2017.

AWARDS

2017 HDR Students Publication Award from Faculty of Engineering and Information Technology (FEIT), University of Technology Sydney (UTS) for publishing in high quality journals.

2016 HDR Students Publication Award from Faculty of Engineering and Information Technology (FEIT), University of Technology Sydney (UTS) for publishing in high quality journals.