FOULING BEHAVIOR AND NITROGEN REMOVAL IN THE AEROBIC GRANULATION MEMBRANE BIOREACTOR

by

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A dissertation submitted in partial fulfillment of the requirement for the degree of Doctor of Engineering

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Abstract

This study investigated the fouling behavior and nitrogen removal of two operational modes of granulation MBR, namely Batch Granulation MBR (BG-MBR) and Continuous Granulation MBR (CG-MBR). Additionally, the characteristics of sludge and supernatant were examined during the experimental course. Coupling of Sequencing Batch Airlift Reactor (SBAR) with MBR in a batch operation (*i.e.*, BG-MBR) showed better performance than the CG-MBR based on granule stability, simultaneous nitrification denitrification and fouling control. On the other hand, due to granule breakage after two weeks of operation, the CG-MBR system functioned similar to conventional MBR.

At the OLR of 2 kgCOD/m^3.d, granular sludge in the BG-MBR was maintained up to 18 gVSS/L while sludge flocs in CG-MBR was only up to 5 gVSS/L. Aerobic granules in the BG-MBR system were unstable under anaerobic conditions and with long retention of granules. By contrast, the anoxic/aerobic conditions in the reactor enhanced the retention, settling ability, denitrification and filterability of flocculent sludge.

The simultaneous nitrification denitrification was achieved in the BG-MBR system due to the spherical structure of the granule where aerobic and anoxic conditions exist in the surface and the core of the granule respectively. It was observed that the TN removal rate was 47% or 22 mgTN/L.h (1.76 mgTN/gVSS.h) at OLR of 2 kgCOD/m^3.d.

The fouling rate of the BG-MBR system was much lower than the CG-MBR or conventional MBR. Soluble microbial products were found to be the main cause for fouling where soluble polysaccharides and protein were deposited on membrane. More to the point it was noted that the polysaccharides were the dominant deposited substances. The specific deposition loading on membrane during membrane filtration was 11 mg/L.m^2 and 8 mg/L.m^2 for soluble polysaccharides and soluble protein respectively. High aeration rate and anoxic phase in the granulation reactor released different types of soluble microbial products which had influenced the filtration behavior of the system.

The release of soluble microbial products in the BG-MBR system depends on the HRT maintained in the MBR. Besides, the HRT of 2-5 h was found to be the suitable range for operating the MBR which can reduce the fouling, increase the supernatant quality and reduce the energy consumption. However, the granule breakage in the BG-MBR increased the fouling propensity due to release of soluble microbial products.

It was found that the filtration resistance of the SBAR effluent was higher when compared with the mixed liquor in anoxic/aerobic operational conditions. Furthermore, the resistance and irreversible resistance rates of SBAR effluent were increased at aeration rate of granulation process as high as 2.2 cm/s due to release of macromolecules (30-50 kDa) and small particles while the soluble microbial products were only released at lower aeration rate. Around 60% of the hydrophobic fraction was found at high aeration rate (2.2 cm/s) in the soluble fraction of SBAR effluent with low hydrophobic intensity. On the other hand, at the low aeration rate (0.6 cm/s) with anoxic growth, 20% of the hydrophobic fraction was noticed with high hydrophobic intensity.

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List of Abbreviations

AIT	Asian Institute of Technology
BAS	Biofilm Airlift Suspension Reactor
BG-MABR	Batch Granulation Membrane Airlift Bioreactor
BG-MBR	Batch Granulation Membrane Bioreactor
CASP	Conventional Activated Sludge Process
CG-MBR	Continuous Granulation Membrane Bioreactor
COD	Chemical Oxygen Demand
DO	Dissolved Oxygen
DRT	Dimensionless retention time
EEM	Emission Excitation Matrix
EPS	Extra-cellular Polymeric Substances
F/M	Food to microorganism ratio
HRT	Hydraulic Retention Time
INSA	Institut National des Sciences Appliquées
MBR	Membrane Bioreactor
MFI	Modified Fouling Index
MLSS/MLVSS	Mixed Liquor Suspended Solids/Mixed Liquor Volatile Suspended Solids
MWCO	Molecular Weight Cut-off
NTA	Nitrilotriacetic acid
OLR/NLR	Organic Loading Rate/Nitrogen Loading Rate
PHB	Poly-β-hydroxybutyric acid
PN	Protein
PS	Polysaccharides
PSD	Particle Size Distribution
RBC	Rotating Biological Contactor
SBAR	Sequencing Batch Airlift Reactor
SBBC	Sequencing Batch Bubble Column
SBC	Settled Biomass Concentration
SBR	Sequencing Batch Reactor
SEC	Size Exclusion Chromatography
SEM	Scanning Electron Microscope
SMP	Soluble Microbial Products
SND	Simultaneous Nitrification Denitrification
SOUR	Specific Oxygen Uptake Rate
SRT	Sludge Retention Time
SVI	Sludge Volume Index
TF	Trickling Filter
TMP	Trans-membrane Pressure
TN	Total Nitrogen
TOC/DOC	Total/Dissolved Organic Carbon
UASB	Upflow Anaerobic Sludge Blanket Reactor
USBR	Upflow Sequencing Batch Reactor
UVA_{254}/ SUVA	Ultraviolet Absorbance (at 254 nm)/ Specific Ultraviolet Absorbance
WW	Wastewater

Chapter 1

Introduction

1.1 Background of the study

The rapid industrialization and population growth has increased the water demand in day to day life. The useable water in the earth is inadequate to satisfy the human demand. In order to meet this demand and to slow down the rapid degradation of water environment, recently all the industries were enforced to employ appropriate treatment technologies by introducing stringent standards and regulations.

There are several treatment technologies available to treat industrial and domestic wastewater. However, the selection of appropriate treatment technologies depends on several factors such as the wastewater characteristics, end use, footprint left and investment cost. Among the existing technologies, a shift towards advanced biological or membrane treatment processes from the conventional biological treatment technologies are noticed in recent days.

Application of membrane technology has increased recently for its production of good quality treated water which can be reused and recycled for domestic, agriculture and industrial purposes. Currently, the membrane bioreactor (MBR) process is an emerging technology that has been successfully applied which operates based on combination of activated sludge and membrane unit. The activated sludge and membrane play role in removal of pollutant and liquid/solid separation respectively. However, the conventional MBR technologies still stay behind of limitations, namely inability to achieve simultaneous organic and nitrogen removal in a single reactor and fouling control. To overcome these issues, the combination of granular sludge and membrane process could be an attractive treatment process. The granular sludge has been recognized to possess the following properties such as excellent settling ability, compacted structure, microbial diversity, ability to sustain high organic/nitrogenous loading rate, removal of toxic substances, and adsorption of heavy metals. In addition it alleviates fouling due to less contact between sludge and membrane due to its large spherical structure.

1.2 Rationale

The combination of the granular sludge process with membrane filtration represents a hybrid treatment system which comprises advantages of both the processes. This technology is capable of achieving high organic and nitrogen removal efficiencies and fouling control. Further, aerobic granular sludge is preferable in batch operating reactors due to existence of periodical feast and famine period in every batch (Tay et al., 2001; Beun et al., 2002). Thus, to maintain the stability of granular sludge, the granule-based MBR systems should be operated in the batch and/or semi-continuous feeding mode. This study proposed to have two coupling modes between aerobic granulation process and membrane operation as follows:

- An external submerged MBR followed a sequencing batch airlift reactor (SBAR) to filter its effluent. A granular sludge reactor was not able to meet the effluent standards due to high suspended solids (SS) in the effluent. It was noticed that the SS concentration in the effluent of granulation reactor was in the range of 75-1200 mgSS/L

(Beun et al., 2002; Arrojo et al., 2004). Thus a coupling with membrane filtration is essential to eliminate SS from the effluent of the granulation reactor. Besides, the external submerged MBR was found to be more advantageous than internal submerged MBR in terms of maintenance, fouling elimination, foaming control, and elimination of short-circuiting. This system was named as the Batch Granulation MBR (BG-MBR) which was thought to alleviate fouling due to less substrate and biomass coming to MBR.

- A continuous granulation MBR (CG-MBR) could be operated stably with the continuous feeding and filtration. Moreover, a periodical removal of light biomass fraction was required to maintain the stability of granular sludge. This operating mode might satisfy the granule stability while combining with MBR. The system avoided the complication of batch operation such as using storage tank, transfer tank and high pump capacity.

1.3 Objectives of the study

This research includes the three following objectives:

(1) Study on organic removal and simultaneous nitrification and denitrification of aerobic granule and its stability in the SBAR;

(2) Characterization of fouling behavior of an external submerged MBR treating the SBAR effluent (BG-MBR);

(3) Study on the granule stability and fouling potential of the continuous granulation MBR (CG-MBR) at various organic loading rates.

1.4 Scopes of the study

For the BG-MBR: Synthetic wastewater with glucose (2 kg COD/m^3.d) and ammonia nitrogen (0.6-1 kgN/m^3.d) were used to cultivate shell granules. Characteristics of shell granule and effluent were investigated along with fouling behavior and nitrogen removal.

For the CG-MBR: Fouling behavior and nitrogen removal were investigated at organic loading rate (OLR) of 2, 4 and 8 kgCOD/m^3.d and fixed nitrogen loading rate (NLR) of 0.6 kgN/m^3.d.

Effect of aeration rates (0.6, 0.8, 2.2 cm/s) and anoxic/aerobic condition on sludge and effluent of SBAR in terms of resistance rate modification, resistance, molecular weight, hydrophobic interaction chromatography, and fouling ability of sludge fractions, namely suspended solids, colloids and solutes.

Sludge characteristics were investigated in terms of morphology, particle size distribution, settling velocity, SVI, biomass concentration, bound extracellular polymeric substances (EPS), microbial examination, granule to floc ratio, and capillary suction time (CST). Liquid characteristics were measured such as nitrogen species, dissolved organic carbon (DOC), soluble EPS, ultraviolet absorbance (UVA_{254}) and specific ultraviolet absorbance (SUVA). While the fouling behavior was quantified by trans-membrane pressure, resistances, specific cake resistance, modified fouling index (MFI) of sludge fractions, and deposition of soluble matters.

Chapter 2

Literature Review

2.1 Overview and application of aerobic biological treatment processes

Biological treatment is the series of microorganism-based processes for water, wastewater and sludge treatment. The objectives of the biological treatment of domestic wastewater are to oxidize dissolved and particulate biodegradable matters into acceptable end products, to convert suspended and nonsettleable colloidal solids into a biological floc or biofilm, to transform and remove nutrients, such as nitrogen and phosphorus, and in some cases, to remove specific inorganic constituents.

Oxidation and synthesis:

$$\text{COHSN (organics)} + O_2 + \text{nutrients} \xrightarrow{bacteria} CO_2 + NH_3 + C_5H_7NO_2 \text{ (new cells)} + \text{end products}$$

Endogenous respiration:

$$C_5H_7NO_2 + 5O_2 \xrightarrow{bacteria} 5CO_2 + 2\,H_2O + NH_3 + \text{energy}$$

Nitrification:

$$2NH_4^+ + 3\,O_2 \xrightarrow{nitrosomonas} 2NO_2^- + 4H^+ + 2\,H_2O$$

$$2NO_2^- + O_2 \xrightarrow{nitro-bacteria} 2NO_3^-$$

Biological treatment process was wildly used in most of treatment plants around the world. The biological processes include aerobic, anoxic and anaerobic operations and their combination. Among these, aerobic process has been recognized to be the most convenient wastewater treatment method because of its ease in operation and management. Moreover, aerobic treatment became popular because it could produce treated water quality at effluent standards without coupling with other processes. By contrast, anaerobic treatment processes were often combined with an aerobic operation for complete treatment. The development of aerobic unit processes was original starting from the conventional activated sludge process (CASP) in Figure 2.1 and then trickling filter, biofilter, submerged attached growth, activated sludge with fixed film packing, fluidized bed bioreactor and membrane bioreactor (MBR). Popular systems and their typical operating conditions are presented in Table 2.1 and Table 2.2.

The MBR is a derivative of the CASP in which the sedimentation tank is replaced by a membrane separation unit. In other words, activated sludge and treated water were separated by a microfiltration or ultrafiltration instead of a conventional gravitational settler. MBR became attractive due to less footprint consumption and high permeate quality which could meet the need of water reuse and recycling of the society. However the membrane fouling was still a constraint of the process which limits the widespread application of membrane process. Therefore a number of researches studying on membrane fouling behavior and its control have been conducted.

In recent years, aerobic granular sludge process was found to be very attractive compared to activated sludge operation due to the compactness, regularity, high

bioactivity, high biomass retention and excellent settling velocity of biogranules. One of the limitations of granular sludge process is that the suspended solids concentration in effluent is quite high, in the range of 75-250 mgVSS/L at OLR of 2.5 kgCOD/m^3.d (Beun et al., 2002) and 200-450 mgTSS/L at OLR of 6 kg COD/m^3.d (Arrojo et al., 2004). Based on the above reasons, the combination by using granular sludge instead of activated sludge in MBR would create a hybrid treatment system which includes the advantages of both processes. The advantages of this technology include simultaneous organic and nitrogen removal, fouling control and water reuse.

Due to water scarcity water recovery, reuse and recycling are becoming more and more popular, which require alternative treatment systems. The wastewater from industries contain high organic and nitrogenous substances, therefore, the conventional treatment processes would not be appropriate for the reuse purpose due to low effluent quality. Hence, the need for a new treatment technology which owns higher loading rate, high settling ability of sludge and high toleration with toxic substances is inevitable. Thus, to meet this requirement, biological process using aerobic granular system combined with the membrane technology could be an attractive alternative treatment process.

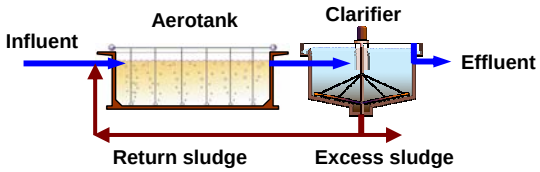


Figure 2.1 Typical configuration of the conventional activated sludge process

Table 2.1 Aerobic biological processes (adopted from Metcalf & Eddy, 2004)

Type	Common name	Use
Suspended growth		
	Activated sludge process (ASP)	Organic removal and nitrification
	Aerated lagoon	Organic removal and nitrification
	Aerobic digestion	Stabilization, organic removal and nitrification
Attached growth		
	Trickling filter (TF)	Organic removal and nitrification
	Rotating biological contactor (RBC)	Organic removal and nitrification
	Packed-bed reactor	Organic removal and nitrification
Hybrid suspended and attached growth processes	TF/ASP	Organic removal and nitrification
Lagoon process	Aerobic lagoon	Organic removal
	Maturation lagoon	Organic removal and nitrification
	Facultative lagoon	Organic removal

Note: TF: Trickling filter, ASP: Activated sludge process

Table 2.2 Typical design parameters for aerobic biological processes (adopted from Metcalf & Eddy, 1991 & 2004)

Process name	Reactor type	SRT (d)	F/M (kgBOD/kgVSS.d)	VLR (kgBOD/m^3.d)	MLSS (g/L)	HRT (h)	RAS (% of influent)	Additional information
High-rate aeration	Plug flow	0.5-2	1.5-2.0	1.2-2.4	0.2-1	1.5-3.0	100-150	
High purity oxygen	Plug flow	1-4	0.5-1.0	1.3-3.2	2-5	1-3	25-50	
Plug flow	Plug flow	3-15	0.2-0.4	0.3-0.7	1-3	4-8	25-75	
Step feed	Plug flow	3-15	0.2-0.4	0.7-1.0	1.5-4.0	3-5	25-75	
Complete mix	Complete mix	3-15	0.2-0.6	0.3-1.6	1.5-4.0	3-5	25-100	
Extended aeration	Plug flow	20-40	0.04-0.10	0.1-0.3	2-5	20-30	50-150	
Oxidation ditch	Plug flow	15-30	0.04-0.10	0.1-0.3	3-5	15-30	75-150	
SBC	batch	10-30	0.04-0.10	0.1-0.3	2-5	15-40	NA	
Trickling filter[(a)]	NA	NA	NA	0.07-3.20	NA		0-200 (recirculation)	Loading: 1-75 m^3/m^2.d packing materials: rock/plastic
RBC	NA	NA	NA	4-20 gBOD/m^2.d	NA	0.7-1.5	NA	Loading: 0.08-0.16 m^3/m^2.d
TF/SC	NA	0.2-2.0	NA	0.3-1.2	1-3	0.2-1.0	NA	Clarifier peak overflow rate 1.8-3.0 m/h
RF/AS	NA	2-7	NA	1.2-4.8	2.5-4.0	0.2-1.0	NA	Clarifier peak overflow rate 2.0-3.5 m/h
ABF	NA	0.5-2.0	NA	0.36-1.20	1.5-4.0	NA	NA	Clarifier peak overflow rate 1.8-3.0 m/h
BF/AS	NA	2-7	NA	1.2-4.8	1.5-4.0	2-4	NA	Clarifier peak overflow rate 2.0-3.5 m/h
Moving bed biofilm reactor (MBBR)	Complete mix	NA	NA	1.0-1.4	NA	3.5-4.5	NA	Clarifier overflow rate 0.5-0.8 m/h
Facultative lagoon	Partial mix	NA	NA	0.5-0.8 d^{-1}	0.05-0.2	4-10 d	NA	Sludge accumulates in lagoon
Aerobic flowthrough lagoon	Partial mix	3-6	NA	0.5-1.5 d^{-1}	0.1-0.4	3-6 d	NA	Sludge accumulates in external sedimentation
Aerobic with solids recycling lagoon	Complete mix	10-20	NA	2-10 d^{-1}	1.5-3.0	0.25-2 d	NA	Sludge recycled to process from sedimentation tank
Oxidation pond	Intermittent mix	NA	NA	16-180 kg/ha.d	0.04-0.26	4-40 d	NA	Algae, CO_2, bacteria Pond depth: 0.3-1.5 m
Submerged MBR	NA	5-20	0.1-0.4	1.2-3.2	5-20	4-6	NA	Vacuum pressure of 4-35 kPa

Note: (a) from low to high rate trickling filter (TF); SBC: Sequencing Batch Reactor; RBC: Rotating Biological Contactor; SC: Solid Contact; AS: Activated Sludge; RC: Roughing filter; ABF: Activated Biofilter; MBR: Membrane bioreactor

2.2 Granular sludge: aerobic granule vs anaerobic granule

The bio-granulation can be divided into aerobic and anaerobic granulation and it is formed through cell self-immobilization process of microorganisms. The anaerobic granulation has been extensively used in up flow anaerobic sludge blanket (UASB) by several industries to treat high strength wastewaters. But the anaerobic granulation system has some shortcomings such as long start up period, a relatively high operation temperature, and unsuitability for low strength wastewater and nutrient removal (Liu and Tay, 2004). In recent years, application of aerobic granular sludge system is becoming very attractive in wastewater treatment due to compactness, regularity, high bioactivity, high biomass retention, excellent settling velocity and simultaneous nitrification denitrification. Settling velocity is much greater than that of conventional activated sludge (much greater than 10 m/h compared to about 1 m/h)) (Tay et al., 2001; Beun et al., 2002, Linlin et al., 2005), sludge volume index (SVI) is up to 12 mL/g (de Kreuk et al., 2005). In addition, the aerobic granular sludge system can sustain an OLR as high as 9 kg COD/m^3.d (Tay et al., 2003) and 15 kg COD/m^3.d (Moy et al., 2002). Further, it can resist the organic shock loading up to 200% (Thanh, 2005), remove toxic substances and absorb heavy metals. The characteristics of aerobic and anaerobic granular sludge process are described in Table 2.3 and Table 2.4.

Table 2.3 Comparison of aerobic and anaerobic granular sludge process

Process	Anaerobic granular sludge	Aerobic granular sludge
Upflow velocity	+ 0.6-2.0 m/h	+ higher than 43 m/h (1.2 cm/s) (Tay et al., 2001)
Biomass concentration	+ 5-40 g/L (top reactor) & 50-100 g/L (bottom)	+ 5 – 15 g/L
Operating DO	+ Anaerobic (~ 0 mg/L)	+ Saturated DO concentration
Organic loading rate	+ Up to 50 kgCOD/m^3.d (Hulshoff Pol et al., 2004)	+ Up to 15-30 kgCOD/m^3.d (Moy et al., 2002; Liu et al., 2003; Thanh, 2005)
Sludge loading rate	+ 0.10-7.86 kgCOD/kgVSS.d (Singh et al., 1999)	NA
Substrate degradation	+ Not completely degrade the influent waste	+ Complete degradation to end products
Formation	+ Upflow anaerobic sludge blanket (UASB)	+ Types of batch reactors with high H/D ratio + Possible with continuous reactor (continuous upflow sludge blanket reactor) (Mishima and Nakamura, 1991; Tijhuis et al., 1994)
Effluent suspended solids	+ 30-150 mg/L	+ 80-1000 mg/L
Reactor start-up	+ about 3 months	+ about 1 month
Operating temperature	+ Preferable at mesophillic and thermophilic temperature range	+ stable at 8-15^{o}C (de Kreuk et al., 2005), 55^{o}C (Zitomer at al., 2007)
Wastewater strengh	+ High strength wastewater	+ Low to high strength
Nutrients (N,P) removal	+ Low	+ High
Simultaneous nitrification denitrification (complete N removal)	+ Impossible	+ High

Table 2.4 Characteristics of sludge types

Sludge	Activated sludge	Anaerobic granule	Aerobic granule	Reference
Size (mm)	~ 0.1	2-5 (or larger)	0.5-9	Thanh, 2005
Specific gravity (kg/L)		1.033-1.065	1.0069	Tay et al., 2002
Water content (%)		97.2	94.3	Linlin et al., 2005
Settling velocity (m/h)	< 10	72	22-60	Linlin et al., 2005
VSS/SS ratio	0.85	0.57	0.71	Linlin et al., 2005
Settled biomass conc. ($g/L_{granule}$)		-	12-60	Beun et al., 2002
Cell surface hydrophobicity (%)	< 50	-	> 75	Linlin et al., 2005
PS/PN	-	-	9	Tay et al., 2001b
Microbes	Aerobic	Anaerobic	Aerobic, facultative anaerobic & obligate anaerobic	Tay et al., 2006

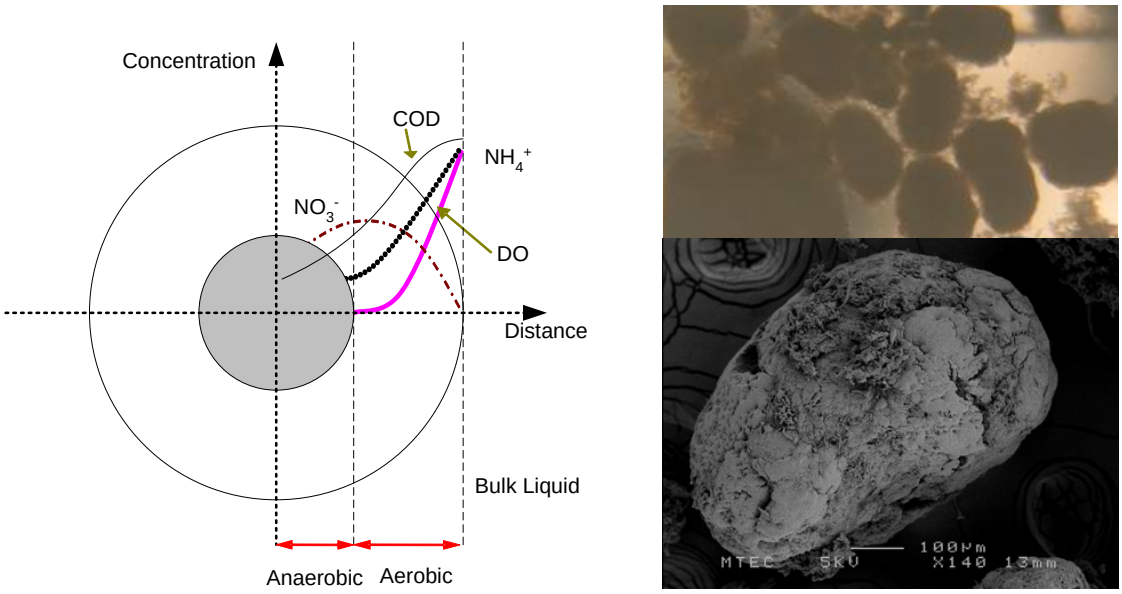


Figure 2.2 Substrate profile, microscopic and scanning electron image of aerobic granule (Thanh, 2005)

2.2.1 Mechanisms of aerobic granulation

The bio-granulation can be divided into aerobic and anaerobic granulation and it is formed through self-immobilization process of microorganisms. Cell immobilization technology has been used in environmental engineering field for several years and can be classified into three categories namely,

- Biofilm: Microorganisms are immobilized or attached onto a solid surface, such as activated carbon, basalts, plastics, polymers, ceramics, etc. (Liu and Tay, 2002).
- Microbial aggregates and granular sludge: Aerobic and anaerobic granules can be considered a self-immobilization community of bacteria.
- Entrapped microorganisms: Microorganisms may be entrapped in hydrophobic gels of photo-cross linked polymers or in other types of gels, such as polyacrylamide (Liu and Tay, 2002).

Similar to biofilm formation, granular sludge can be regarded as a form of cell immobilization. So far, it has been recognized that the formation of biofilm and microbial aggregates are multiple-step process, to which physico-chemical and biological forces make significant contributions (Beun et al., 1999; Tay et al., 2001b; 2004b & 2006). Based on previous studies, it is encouraged to propose that cell immobilization can be roughly described as a four-step process as follows:

Step 1: Physical movement to initiate bacterium-to-bacterium contact or bacterial attachment onto a solid surface. The forces involved in this step are hydrodynamic force, diffusion force, gravity force, thermodynamic force and cell mobility. Cell can move by means of flagella, cilia or pseudopods.

Step 2: Initial attractive forces to keep stable bacterium-bacterium and multicellular contacts, including (1) physical forces: van der Waals forces, opposite charge attraction, thermodynamic forces, hydrophobicity and cross-link by filaments. Among the physical forces, the increase of cell surface hydrophobicity promotes cell-to-cell interaction and further self-aggregates. In addition, filamentous microorganisms assist in building up a three-dimensional structure for the growth of attached microorganisms; (2) Chemical forces: hydrogen liaison, formation of ionic pairs/ionic triplet, interparticulate bridge and so on; (3) Biochemical forces: cellular surface dehydration and cellular membrane fusion.

Step 3: Microbial forces to make aggregated bacteria mature:

- Production of extracellular polymers such as exopolysaccharides;
- Growth of cellular cluster;
- Metabolic change and genetic competence induced by environment, which facilitate and further strengthen the cell-cell interaction and result in the high density of adhering cells.

Step 4: The outer shape and size of microbial aggregates are finally determined by the interactive strength between aggregate and shear force, microbial species, and substrate loading rate, etc. Shear force and selection pressure are the important factors influencing the formation of aerobic granule (Tay et al., 2001b; Liu and Tay, 2002; McSwain et al., 2004; Qin et al., 2004; Liu et al., 2005b).

2.2.2 Process of aerobic granulation

Aerobic granule could be cultivated from various kinds of seed sludge, namely conventional activated sludge (Morgeroth et al., 1997; Etterer and Wilderer, 2001; Beun et al., 2002; Jang et al., 2003; Tay et al., 2002; Arrojo et al., 2004; Schwarzenbeck et al., 2004&2005; Wang et al., 2006c), anaerobic granule (Linlin et al., 2005) and mixture of activated sludge and anaerobic granules (Thanh, 2005).

a. Aerobic granule formation from conventional aerobic activated sludge process

In general the granulation process of conventional activated sludge in SBR can be categorized into three phases, namely acclimation, granulation and maturation. Wang et al. (2004) reported that the granules were first initiated as mycelial pellets in the reactor and began to accelerate growth, the "granule initiated". The corresponding period from the start-up operation to the "granules initiated" was the sludge acclimation phase. Similarly, the initial granules could grow out fully and the biomass concentration was not changed,

the matured point. The granulation stage was considered from the 'granule initiated' to the granule matured point.

The activated sludge inoculated in the SBR was a mixture of filamentous sludge with brown colour and weak settling. Since the settling time was kept short, a washout of biomass took place. The sludge concentration in the reactor decreased from this wash out and more suspended solids were observed in the effluent because of bad settling ability. During this time, most of the sludge in the reactor became flocs. Over the next weeks, the floc-like sludge gradually changed to granular sludge. After 67 days operation, granular sludge began to appear whereas flocs still remained dominant in the reactor. The initial granular sludge formed in the SBR was smaller in size with fluffy edges. The small granules grew rapidly in the following weeks, while more floc-like sludge was washed out, resulting in the accumulation of the granules. Eleven weeks after inoculation, the sludge in the reactor was nearly completely granulized, and visually no suspended biomass was present. Due to the intensive mixing by aeration, the granular sludge became spherical with a smooth surface. The diameter of the granular sludge increased to 6–9 mm.

After the granules matured point, the granules were stable and dynamically balanced in the maturation phase. In this phase, the granular size might still be shifting mainly between 6-9 mm, but slowly and slightly. The matured granule color was white and somewhat transparent (Figure 2.4).

Jang et al. (2003) cultured aerobic granule from the initial seed sludge (size of 0.08-0.18 mm and SVI of 210-230 mL/g). After 50 days, granules formed with the size of 0.95-1.35 mm and SVI of 70-90 mL/g. The floc-like sludge changed gradually to granules over time. Granulation of the seed sludge could be achieved through accumulation by interparticle bridging under a condition of turbulent flow mixing. After 40 days of operation, the seed sludge in reactor was nearly totally granulated. First, the seed sludge was not in the form of large flocs, rather irregular and unstable filaments were dominant. The particles eventually started to join together to form biomass aggregates and the aerobic floc-like sludge form was accomplished within 10 days. Second, the aerobic floc-like sludge was heterogeneously mixed, with irregular and soft granules which started to appear in around 30 days. On day 40, the aerobic granular sludge formed. At that time most of granules had an uneven surface and soft texture. Finally, the irregular granules became stable and were smoother and round-shaped with a solid surface after 50 days.

Etterer and Wilderer (2001) found that by keeping the short settling time, biomass in the SBR was washed out during the start-up period. First, the filamentous granules appeared after two weeks whereas flocs still remained dominant. In the following weeks granules accumulated and four weeks after inoculation, biomass in reactor consisted of mainly aerobic granules. Due to the intensive mixing with hydrodynamic shear force by aeration, granules became spherical with smooth surface. Besides, it indicated that fungi and filamentous organisms in general were present in the overall structure of the aggregates.

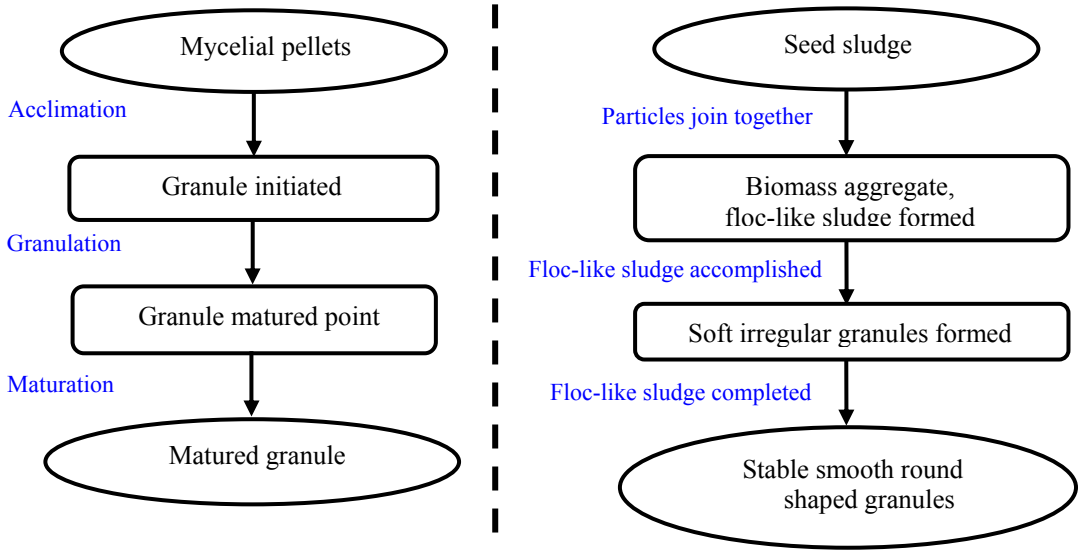


Figure 2.3 Formation process of aerobic granule (modified from Wang et al., 2004: left and Jang et al., 2003: right)

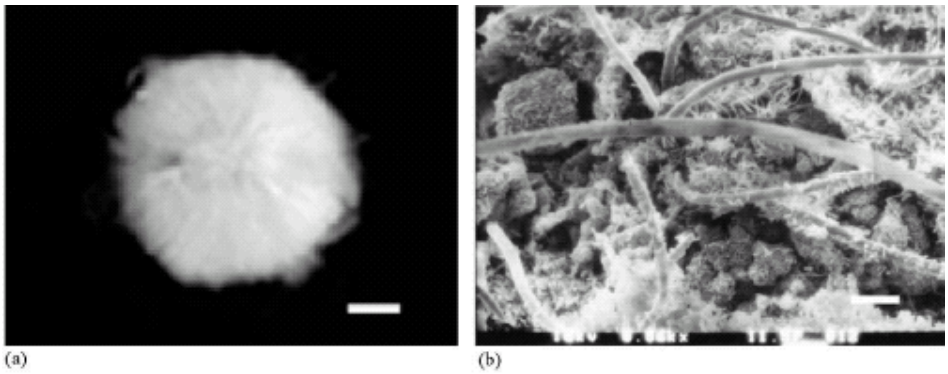


Figure 2.4 Microscopy images of mature granules after 120 days: (a) Microscope overview image, bar = 2 mm, (b) SEM of the granules surface, bar =11 µm (Wang et al., 2004)

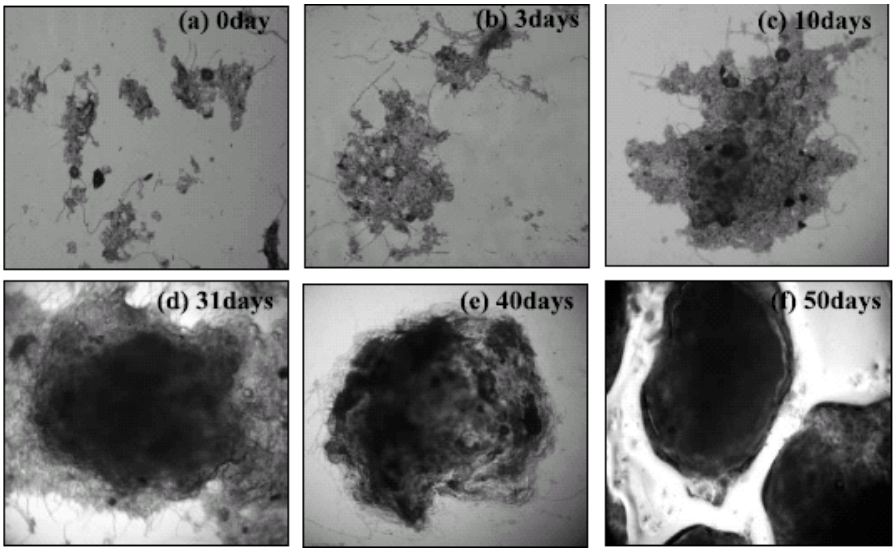


Figure 2.5 Granule development process: (a) 0 day, seed sludge; (b) 3 days; (c) 10 days; (d) 31 days, flocs-like; (e) 40 days and (f) 50 days, granule (Jang et al., 2003)

b. Aerobic granulation from anaerobic granule

Linlin et al. (2005) reported that aerobic granule could be cultivated from anaerobic granule. Firstly, the anaerobic granular sludge (regular shape, black colour and size of 1.1 mm) disintegrated under aerobic conditions after inoculation, forming irregular and small flocs, and highly filamentous granules. These granules were not stable at all and broke up into pieces after a few days. Subsequently, large part of the biomass was washed out, the remaining debris from the disintegrated granules recombined under aerobic conditions; and finally the granules grew up, resulting in the formation of aerobic granule (yellow color, size of 1.2 mm). The granules formed in this stage hardly contained any filament and consisted dominantly of bacteria. The author concluded that the disintegrated anaerobic sludge might play a role of nucleus for the granulation of aerobic sludge. Figure 2.6 shows the process of morphological change of granules in the reactor.

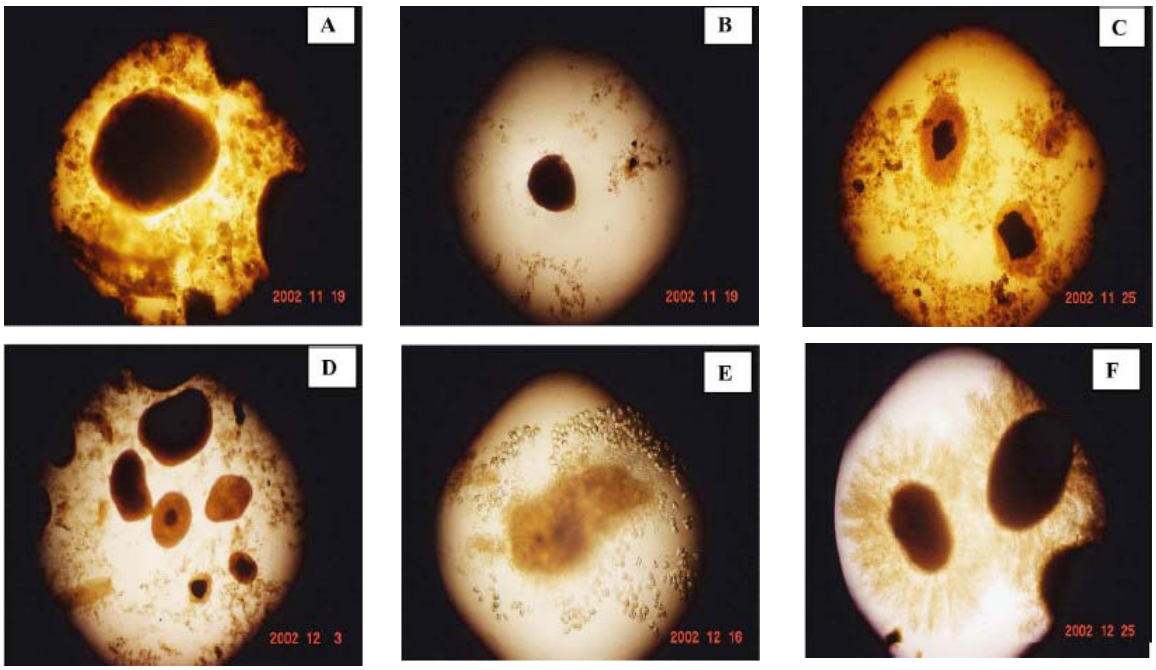


Figure 2.6 Morphological evolution of granulation process from anaerobic granule (40x). (A) Seed anaerobic granule; (B) after 1 week; (C) after 2 weeks; (D) after 3 weeks; (E) after 5 weeks and (F) after 5 weeks (Linlin et al., 2005)

2.2.3 Production of aerobic granules

a. Reactor configuration for aerobic granule formation

The aerobic granule can be cultivated by either batch or continuous system. However, from several researches, it was revealed that the batch operating reactor was preferable for granulation process due to the existence of periodic feast-famine stages and high gradient of substrate concentration (Beun et al., 2002). Granules can be cultivated in batch system such as SBAR (Sequencing Batch Airlift Reactor), SBR (Sequencing Batch Reactor), and SBBR (sequencing batch bubble column). A batch reactor for granulation process is also operated in sequencing cycles of feeding, aeration, settling and supernatant withdrawal which is similar to conventional batch reactor. To produce granules, besides the necessity of high hydrodynamic shear force by aeration, the time for settling and

supernatant withdrawal has to be short enough to maintain the selection pressure (washout of light biomass fraction).

Aerobic granule can also be formed by the continuous reactor such as BAS (Biofilm Airlift Suspension Reactor) (Tijhuis et al., 1994). However, short hydraulic retention time should be controlled together with the addition of support media. Thus there is no denitrification in this type of reactor. The granule size and density of continuous reactor were smaller compared to that of batch operating reactors. Therefore, batch operation is the better option for aerobic granule formation. The method comparison among different types of reactors to cultivate granule is presented in Table 2.5.

Table 2.5 Comparison of types of granulation reactor

Items	SBAR (Beun et al., 2000)	SBBC (Beun et al., 1999)	BAS (Tijhuis et al., 1994)
System	Discontinuous	Discontinuous	Continuous
External settler needed	No	No	No
Riser needed	Yes	Yes	Yes, plus 3 phase separator
Carrier needed	No	No	Yes
Selection variable	Settling time	Settling time	HRT
Detachment determined by	hydrodynamic conditions	hydrodynamic conditions	bare carrier conc.
Nitrification & denitrification	Possible	Possible	No denitrification
Settled biomass conc. ($g/L_{granule}$)	48	12	15
Granule diameter (mm)	1	2	0.35 ($d_{carrier}$ = 0.26 mm)

b. Substrate source

Table 2.6 describes that aerobic granules can grow on a wide variety of synthetic wastewaters (acetate, glucose, peptone, sucrose, alcohol, phenol and their mixture) and industrial wastewaters (molasses, abattoir, paper making, dairy, brewery, etc.). Figure 2.7 presents that glucose-fed biogranules possess a filamentous structure, while acetate-fed biogranule have a non-filamentous and compact bacterial structure.

These indicate that the granule formation is independent of the wastewater characteristics while the microbial structure and diversity of granule are closely related to the types of feeding wastewaters (Tay et al, 2001; Liu and Tay, 2002; Liu et al., 2003b).

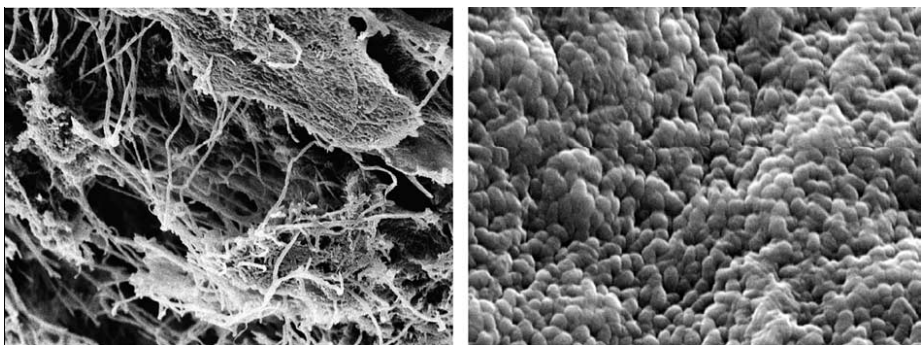


Figure 2.7 Microscopic scanning electron images of glucose-fed (left) and acetate-fed (right) aerobic granule (Liu and Tay, 2002)

Table 2.6 Characteristics of aerobic granule

Substrate source	Loading rate ($kg/m^3.d$)	Formation time	Granule diameter (mm)	SVI (mL/g)	Settling velocity (m/h)	Settled biomass conc. ($gSS/L_{granule}$)	MLVSS (g/L)	Reference
Acetate	5	-	0.35[a]		-	15-20	-	Tijhuis et al., 1994
Acetate	2.3	50 days	1		24	11.9	-	Beun et al., 1999
Acetate	2.5	> 63 days	2.5	-	> 10	60	7-10	Beun et al., 2002
Acetate	6.0	-	0.35	50	-	-	6	Tay et al., 2002
Acetate	-	3 weeks	0.35	50-140	-	-	-	Qin et al., 2004
Acetate	-	4 weeks	0.25-0.32	-	-	-	-	Yang et al., 2004
Acetate	6	21 days	0.33 -0.39	46-62	-	40-60	-	Tay et al., 2004b
Acetate	-	50 days	1.2	30-40	22-60	-	5 (SS)	Linlin et al., 2005
Acetate	1.2 - 1.6	48 days	1.2	12-15	-	-	-	De Kreuk et al., 2005
Acetate	6.0	-	0.85-3.67	31-88	-	-	-	Wang et al., 2006
Acetate	3.0	27 days	0.55-0.75	40	-	-	6	Liu & Tay, 2006
Acetate & peptone	8	15 days	0.2	30-40	-	-	-	Li et al., 2006b
Barley dust WW	3.4	4 weeks	2-4	30-40	-	-	-	Schwazenbeck et al., 2004
Brewery WW	3.5	41 days	2-7	32	91	-	8-11	Wang et al., 2007
Dairy WW	7	60 days	0.25-4	60	-	10-15	-	Arrojo et al., 2004
Ethanol	-	40 days	0.4-1.9	-	-	-	-	Yang et al., 2003
Glucose	4.8	67 days	6-9	40	32.7	-	7.8 (SS)	Wang et al., 2004
Glucose	5	50 days	1.2	< 65	-	45.2-45.7	-	Cai et al., 2004
Glucose	2.5 - 30	4 weeks	0.5-4	18-35	-	20-62	-	Thanh, 2005
Glucose & acetate	2.5	50 days	1.0-1.3	70-90	25.2-28.8	-	-	Jang et al. 2003
Glucose & acetate	1.76-2.84	51 days	0.35	83	-	-	9.5	Kim et al., 2008
Glucose & peptone	2.4	120 days	-	46-114	-	-	-	McSwain et al., 2004
Glucose, acetate & peptone	3.6	56 days	1.1-6.5	-	35	-	-	Etterer & Wilderer, 2001
Molasses	2.9	40 days	2.35	-	-	-	-	Morgenroth et al., 1997
Papermaking WW	8-11	19 days	0.5-3.5	75	-	-	3.89 (SS)	Hailei et al., 2006
Phenol	< 2.5	-	-	40-65	-		-	Jiang et al., 2004
Phenol	20 mg/L	3 weeks	0.53-0.67	19-25	-	-	-	Liu et al., 2005c
Slaughterhouse (beef) WW	2.6	4 days	1.7	22	51	62	8	Cassidy and Belia, 2005
Sucrose	-	68 days	0.5-1.2	23	-	-	-	Zheng et al., 2005b
Sucrose	6	30 days	1	44	130	-	6.43	Zheng et al., 2005
Tert butyl alcohol	0.6	90 days	0.32	57	-	-	4.54	Zhuang et al., 2005

(a) include carrier diameter of 0.26 mm; WW: Wastewater;

c. Seed sludge

To cultivate aerobic granule, seed sludge was often taken from conventional activated sludge processes such as aerotank and SBR (Beun et al., 1999; Tay et al., 2001; Jang et al., 2003; Arrojo et al., 2004; Wang et al., 2004; Qin et al., 2004; Schwarzenbeck et al., 2004; Thanh, 2005). Anaerobic granule could also succeed in forming aerobic granule (Linlin et al. al, 2005). Additionally, granular sludge could be produced by bioaugementation process between activated sludge and superior mixed flora (SMF) which consists of *Coriolus versicolor*, *Phanerochate chrysosporium* and *Azotobacter* sp (Hailei et al., 2006). Therefore, types of seed sludge do not play a role in cultivating biogranules as significantly as operating conditions.

d. Support media for aerobic granules

There are various support media available for microbial attachment such as basalt, sponge, sand, plastic bead, shells, etc. According to Tijhuis et al. (1994), the usage of basalt as the support media or carrier shows the good potential for biofilm development when compared to other media which is commonly found solidified lava (a type of igneous rock) mainly consisting of calcium rich feldspar and pyroxene. Thanh (2005) found that bivalve shell support could be a good support for aerobic granule cultivation. Shell support performed better than non-support granule in terms of faster settling ability (SVI of 14 mL/g and particle settling velocity of 63 m/h at OLR of 15 kgCOD/m^3.d), compactness and especially the ability to resist organic shock loading (200%). Moreover, at the initial stage of granulation, shell support could act as self-cleaning media to prevent attached biofilm growing on the reactor walls effectively. Biofilm growth on the reactor walls was observed to inhibit the granulation process (Morgenroth et al., 1997; Beun et al., 2002).

The supports used for aerobic granulation should have good settling ability, high porosity, circular structure and small size. Since the shell support and basalt possessed these conditions, it was suggested to be a good support for aerobic granulation process.

2.2.4 Factors affecting aerobic granulation

a. Hydrodynamic shear force

Hydrodynamic force by aeration in the reactor favors the formation, structure and metabolism of microbial community of the biogranules. Hydrodynamic shear force is created by superficial air velocity. At high shear force, more compacted, stable and denser granules could be formed. The shear force has significant influences on the structure, mass transfer and production of polysaccharides as well as on metabolic/genetic behavior of biofilm of aerobic and anaerobic granules.

Tay et al. (2001b) reported that at a low superficial air velocity of 0.008 m/s no granules were formed in the up flow sequencing batch reactor but only fluffy flocs were observed. On the contrary, when it was of high superficial air velocity of 0.025 m/s, regular shaped granules were successfully developed in the reactor (Figure 2.8). In addition, Beun et al. (1999) observed that at low superficial air velocity of 0.014-0.020 m/s there was no granule formation in SBR.

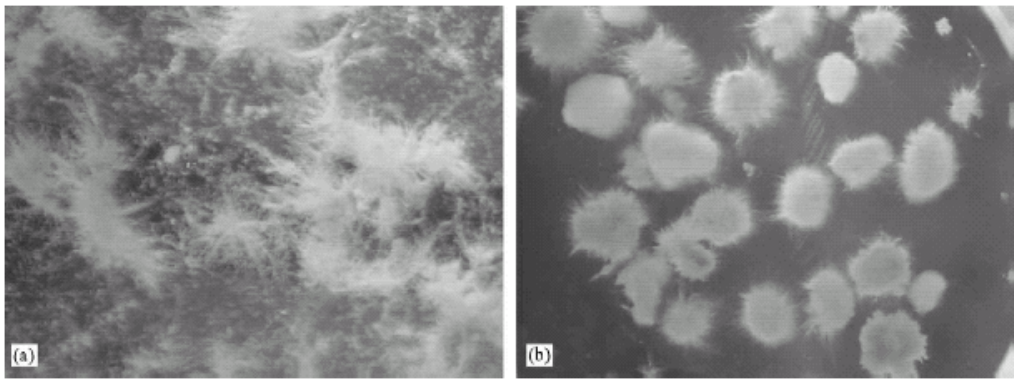


Figure 2.8 Bioflocs cultivated at a superficial air velocity of 0.008 m/s (a) and granules formed at a velocity of 0.025 m/s (b) (Tay et al., 2001b)

It was found that the specific gravity and polysaccharides of aerobic granules were increased while SVI and size of granule were decreased with the increase of hydrodynamic shear force (Figure 2.9 & 2.10). High specific gravity and low SVI of granular sludge ensured the good solid-liquid separation. The superficial air velocity had to be greater than 1.2 cm/s (43.2 m/h) to form aerobic granule in a reactor. This is one of the most important factors which influence the aerobic granulation process.

Hence, it can be concluded that the hydrodynamic shear force has significant effect on aerobic granulation and granule characteristics. It is one of the prerequisites for aerobic granule formation.

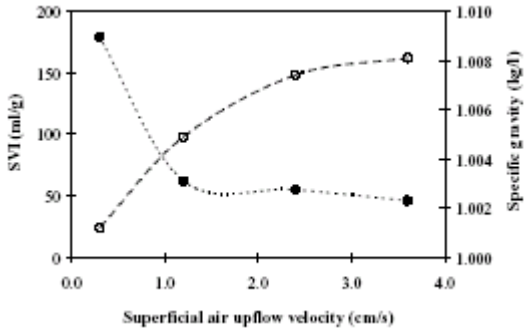


Figure 2.9 Effects of superficial air upflow velocity on the specific gravity and SVI of aerobic granules developed in USBR (•): SVI; (o): specific gravity (Tay et al., 2001b)

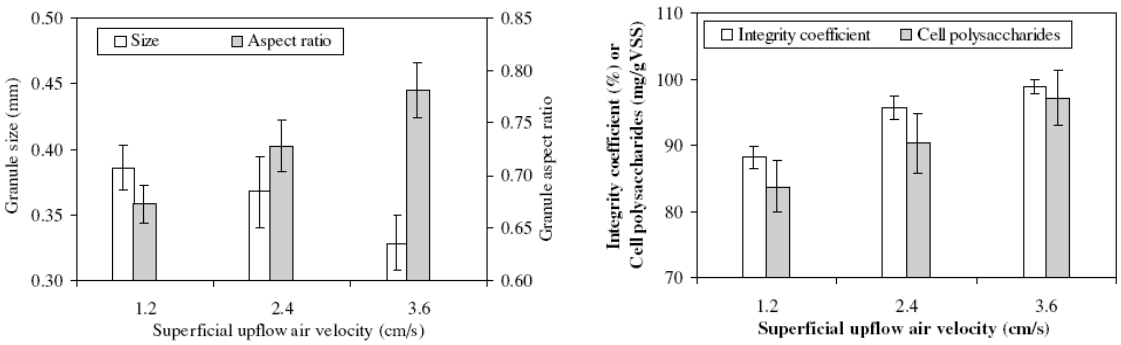


Figure 2.10 Effects of superficial air velocity on the granule size and polysaccharides (Tay et al., 2004b)

b. Settling and discharging time

Settling time controls particles which are maintained in reactor or washed out through supernatant of a batch granulation reactor. Flocs have settling velocity less than 10 m/h so a sludge particle is considered to be a granule when its settling velocity is higher than this value. In aerobic granulation process, the settling velocity of particles is selected first (about 10 m/h) to calculate the settling time (Beun et al., 2002).

Settling time (h) = [settling height (m) / selected settling velocity (m/h)]

Settling velocity or settling time affected the granule characteristics and ratio between granule and flocs in reactor. Fraction of aerobic granules in reactor and settling ability of granules were high when settling time was short (Qin et al., 2004). In aerobic granulation system, settling time has been commonly kept short to enhance granulation process (Tay et al., 2001b; Beun et al., 2002; McSwain et al., 2004; Thanh, 2005; Chen et al., 2008). Respectively, discharging time of a granulation reactor should be short together with the setting time. Liu et al. (2005b) established the relationship between settling time and discharge time for a granulation SBR (Figure 2.11).

Thus, settling and withdrawal time were also the triggering factors to maintain aerobic granules in a batch granulation reactor.

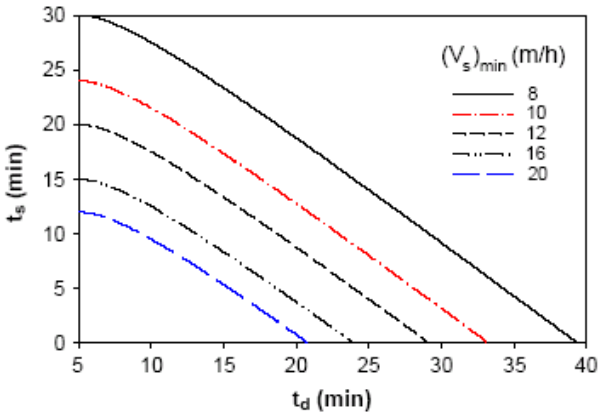


Figure 2.11 Relationship between settling time (t_s) and discharge time (t_d) (Liu et al., 2005b)

c. Volume exchange ratio

Volume exchange ratio or discharge depth which is the depth difference between influent and effluent point in the reactor is an important selection pressure. Wang et al. (2006d) reported that at the high volume exchange ratio the granulation process was faster than that of small volume exchange ratio. High ratio favored large size of granules with low SVI which led to high settling ability. Moreover, the excessive production of EPS and, subsequent calcium accumulation at high volume exchange ratio facilitated the formation and, further improved the settleability of granules.

d. Feeding strategy

The intermittent feeding of a system was found to be more advantageous than that of the continuous feeding due to the existence of substrate gradient or feast-famine period in the bulk liquid (Beun et al., 2002). It was confirmed that promoting a strong substrate gradient in the SBR resulted in good sludge settleability (SVI < 120mL/g) (Martins et al., 2003). Additionally, the intermittent feeding favored the formation of compact and dense aerobic granules. Under starvation, microorganisms became more hydrophobic which facilitated microbial adhesion and aggregation in the reactor (Liu and Tay, 2004). McSwain et al. (2004) reported that a high feast-famine ratio, or pulse feeding provided by dump fill (without aeration during feeding) in the SBR, was necessary for the formation of compact and dense granules. The intermittent feeding affected the selection and growth of floc-forming and filamentous organisms, which in turn influenced the structure of aerobic granules.

Therefore, intermittent feeding with the high substrate gradient is necessary to form compact and dense aerobic biogranules.

e. Organic loading rate (OLR)

Too high or too low OLR was found to be unfavorable for the formation of a compact sludge bed, and to maintain the stability of performance of the reactor. Moy et al. (2002) identified that the acetate fed system could create the compact spherical morphology of granules at OLR of 6 and 9 kg COD/m^3.d and the loose fluffy morphology dominated by the filamentous bacteria at low OLR. Tay et al. (2003) reported that the best aerobic granules were cultivated at 4 kg COD/ m^3.d with size of 5.4 mm, SVI of 50 mL/g and COD removal rate of 99%. While at OLR of 1 kg COD/m^3.d only the patchy flocs and at OLR of 8 kg COD/ m^3.d both granules and fluffy flocs were observed. Thanh (2005) described that aerobic granulation system with or without shell media could be operated at OLR from 2.5-30 kgCOD/m^3.d. Granules were stable at high loading and removal efficiency was always greater than 97 %. However at OLR greater than 15 kg COD/m^3.d the system often got clogging because granule stuck between tubes of reactor. Kim et al. (2008) found that the SVI of the granular sludge was increased with increasing OLR but the mean diameter of the granules was reduced. The operated OLR range depended on the hydraulic shear force. Higher shear force could sustain a higher OLR. Good reactor performance and well granule characteristics could maintain and operate under the aeration rate of 3.2 cm/s in a wide OLR range (6-15 kg COD/m^3.d). While under the aeration rate of 2.4 cm/s, the OLR was limited to 6–9 kg COD/m^3.d (Chen et al., 2008).

OLR influences on the formation time, structure and microbial diversity of aerobic granule. Li et al. (2008) claimed that granules formed at different OLRs had different morphology, structural properties and bacterial species. A higher loading rate resulted in faster formation of larger and loose granules, while a lower loading rate resulted in slower formation of smaller and denser granules. The reactor with the highest substrate loading rate had the lowest species diversity and vice versa.

This presents that granulation system could sustain a high OLR (2-15 kgCOD/m^3.d). The physico-chemical and microbial characteristics of aerobic granule depend on the operating OLR.

Table 2.7 The characteristics of aerobic granules/aggregates at superfical air velocity of 0.041 m/s (Tay et al., 2003)

OLR (kg COD/m^3.d)	8	4	1
SOUR (mg O_2/g VSS.h)	148	131	82
Mean diameter by number (mm)	8.8	5.4	4
Granule roundness	1.49	1.29	2.23
Specific gravity (kg/L)	1.024	1.034	1.011
SVI (mL/g)	65	50	138
COD removal efficiency (%)	0.79	0.99	0.95
VSS/SS ratio	0.91	0.87	0.88

f. Starvation conditions

The SBR system consists of four phases in a cycle of operation namely feeding, aeration, settling and supernatant discharge. During the aeration phase, the granules start to degrade the substrates, produce EPS and then starve due to the depletion of substrate. Under the starvation condition, the microorganisms become more hydrophobic which facilitates microbial adhesion and aggregation due to usage of EPS produced during the famine period (Liu and Tay, 2004). The starvation period which causes decrease in EPS is a prerequisite for aerobic granulation when no anaerobic microenvironment is initially available (Li et al., 2006b). During the starvation period, the microorganisms can produce stronger and denser granules. Shorter starvation time speeded up the granulation and granules formed with cycle time (short starvation time) of 1.5 h were unstable (Liu and Tay, 2008). By contrast, it has been noted by Wang et al. (2005) that the diameter, VSS ratio and strength of granular sludge under a long starvation period (cycle time of 12 h) was reduced compared to that under short starvation (short cycle time of 3 h).

It can be concluded that the reasonable starvation time is necessary to maintain the long-term stability of the aerobic granules.

g. Metal elements

According to Liu and Fang (2003) mineral cations tend to complex with EPS, affecting bioflocculation, settling and dewaterability of the sludge. Cations serve as a bridge between negatively charged EPS of neighbouring cells. The bridging could stabilize the floc network and thus improved sludge bioflocculation, settling and dewaterability. Especially, calcium ion was suggested either to stimulate granulation by neutralizing negative charges on bacterial surfaces, or to function as cationic bridges between cells.

Wang et al. (2004) found that most of the metal elements in the sludge changed significantly during the start-up operation because of the different chemical composition of the influents. Calcium and potassium amount were increased in matured aerobic granules. Therefore, calcium might play an important role in the cultivation of aerobic granular sludge which was similar to that for anaerobic granules. The change of granule color from brown to white was probably due to the change of the biomass composition, especially for decrease in the content of iron, magnesium, copper and cobalt in the sludge. Thanh (2005) noted that calcium in granules was found to be higher than that in seed sludge at any loading rates in the range of 2.5-15 kg COD/m^3.d. Qin et al. (2004) reported that calcium could accumulate in granule as high as 187.6 mg/g VSS. However, Liu et al. (2003c) reported that the ratio of cell calcium to carbon in aerobic granule at different C/N ratios was two-fold lower than that in the seed sludge. Further, Jiang et al. (2003) noted that the high amount of calcium in the feed

could accelerate the granulation process. The formation of granule took 16 days (100 mg/L Ca^{2+}) compared to 32 days (~0 mg/L Ca^{2+}). Further, Ca was found to accumulate in the center of granule and majority was in form of $CaCO_3$ (Ren et al., 2008).

Table 2.8 Metal elements in seed sludge and granule (mg/g) (Wang et al., 2004)

Type	**K**	Na	**Ca**	Mg	Fe	Cu	Mn	Co	Zn
Seed sludge	9.60	9.16	30.20	5.93	26.40	0.23	0.37	0.024	1.12
Matured granule	43.58	8.00	45.70	2.58	0.76	0.01	0.01	0.012	0.17

It can be concluded that calcium in the feed can accelerate granule formation process but it is not a triggering factor of the microbial granulation.

h. Extracellular Polymeric Substances (EPS) of aerobic granule

The EPS promotes the cohesion and adhesion of cells, and maintains structural integrity of the biofilm where the proportion of EPS produced can be between 50-90% of the total organic substances present in the system. Generally, the EPS includes bound EPS attached to cell wall and, soluble EPS suspended in bulk liquid. The EPS contains various organic substances like Polysaccharide (PS), Protein (PN), DNA, humic acid and uronic acid which are used during the starvation period of the microorganisms (Wingender et al., 1999).

The content of the PS was higher than the PN content present in the biofilm and aerobic granule (Liu and Tay, 2002; Yang et al., 2004). Recently many authors found that the content of PN is more than PS in aerobic granule. McSwain et al. (2005) found that PN were more dominant than PS and the PN content of granules was 50% more that of flocs. Adav and Lee (2008) reported that the PN/PS ratio was approximately 0.9 for sludge flocs and 3.4–6.2 for aerobic granules. The ratio of polysaccharides to proteins (PS/PN) depends on hydrodynamic shear force (Liu and Tay, 2002), volume exchange ratio (Wang et al., 2006d), intermittent feeding (Liu and Tay, 2004), starvation condition (Li et al., 2006b), and inhibitor such as ammonia (Yang et al., 2004).

In addition, the outer shell of aerobic granule was found to be more hydrophobic and with less EPS content compared to the inner core of aerobic granule. EPS content in the core part of granule was nearly 5 times higher than that in the shell part of granule (Wang et al., 2006b). In situ EPS staining of granules showed that cells and PS were localized to the outer edge of granules, whereas the center was comprised mostly of PN, dead and lyzed cells (Ivanov et al., 2004; McSwain et al., 2005).

The concentration of EPS of granular sludge is dependant on operating conditions. The role of EPS in granulation process enhancement was clearly recognized. However, the dominant role of PS and PN in granule formation is still unclear. The ratio of PS/PN is still not a signal of granulation process.

i. Cell surface hydrophobicity

Similar to EPS content, cell self-immobilization and attachment process mainly depend on the cell surface hydrophobicity in which hydrophobicity is induced by culture conditions (Liu et al., 2003). Cell hydrophobicity is often determined by the contact angle (CA) measurement of microbial adhesion to hydrocarbon in form of liquid or solid (Liu et al.,

2004b) or adherence percentage with hexadecane (Jin et al., 2003). The cell hydrophobicity is classified into three categories based on contact angle as follows:

+ CA > 90^o : hydrophobic surface
+ 50^o < CA < 60 : medium hydrophobic surface
+ CA < 40^o : hydrophilic surface

The cell hydrophobicity for the glucose-fed and the acetate fed aerobic granules were 68% and 73% respectively while for the suspended seed sludge it was only about 39%. Hence, the cell hydrophobicity of aerobic granules was about two times higher than that of the conventional activated sludge (Tay et al., 2003). Other studies found that the starvation and high shear stress conditions in the reactor could facilitate the cell surface hydrophobicity which could favor the microbial adhesion and the aggregation (Tay et al., 2001; Liu et al., 2004b). On the other hand, under condition of high free ammonia exposed, the nitrifying bacteria could not form granules due to the low surface hydrophobicity.

j. Suspended solids in the feed wastewater

Suspended particles in feed wastewater can be a factor for enhancing aerobic biogranulation because of its available surface area for cell attachment. Firstly, it helps to increase shear stress which is the main factor for granulation process (Tay et al., 2001b). Secondly, exopolysaccharides has a trend to be produced on the surface of any support media and exopolysaccharides is the bridging factor of cells (Wingender et al., 1999; Liu and Tay, 2002). For example, Arrojo et al. (2004) and Schwarzenbeck et al. (2004) successfully cultivated aerobic granules with the concentration of suspended particles of 1.2 g/L and 0.95 g/L in the feed. So support media (inorganic or organic) also play an important role in enhancing aerobic granule formation. Moreover, supports could act as self-cleaning media to prevent attached bio-film growing on the reactor walls at the start-up stage (Thanh, 2005).

l. Inhibition to aerobic granulation by free ammonia

The high concentration of free ammonia of the feed wastewater is an inhibitor to most of the microbial processes and granulation process as well. High free ammonia concentration inhibits nitrification, cell hydrophobicity, production of EPS and nitrifying activity. Particularly, it reduces the cell hydrophobicity and the EPS content which in turn affects the granulation process. Metabolic activity of the heterotrophic bacteria (SOUR) decreases with increase of free ammonia.

The cell hydrophobicity decreased from 70.6% to 40.6% with the increase of the free ammonia concentration from 2.5 mg/L to 39.6 mg/L. The PS/PN ratio decreased from 2.8 to 0.55 when free ammonia concentration increases from 2.5 to 39.6 mg/L. The PS/PN ratio in R4 and R5 was 0.62 and 0.58, which is comparable with that of seed sludge (0.55), no granule was observed in the reactors (Yang et al., 2004).

Table 2.9 Effects of free ammonia to aerobic granule (Modified from Yang et al., 2004)

Reactor	R1	R2	R3	R4	R5
N/C ratio	5/100	10/100	15/100	20/100	30/100
Free ammonia (mg N/L)	2.5	9.2	18.0	23.5	39.6
PS/PN ratio	2.80	1.90	1.00	0.62	0.58
Granular size (mm) after 4 weeks	0.51	0.32	0.25	-	-
Morphology	Smooth, regular, dense	Smooth, regular, dense	Less smooth than R1, R2	Flocs	Flocs

m. Failure of granular sludge system due to long sludge retention time

The global sludge retention time (SRT) is not suitable to describe granule retention time for granular sludge system because the sludge removed from the effluent consists of the detached debris, suspended solids rather than granules. Thus, actual SRT of granule is longer than the calculated SRT based on the definition (SRT = sludge in the reactor over sludge discharged daily). The sludge retention time of granulation reactor is long and may be equal to the operating duration since matured granules are formed till the end of operation. The long SRT favors slow growing bacteria and filamentous growth which is commonly phenomenon in aerobic granular sludge reactor (Moy et al., 2002; Tay et al., 2004b; Schwarzenbeck et al., 2005). When the SRT is longer than 10 days filamentous growth usually occurs. Further, Liu and Liu (2006) reported that the low- or moderate-levels of filamentous growth did not cause the operational problems and could even stabilize the granule structure. However, the overgrowth of filaments led to poor settling of granule, washout of filamentous granules, filamentous granules outcompeting the non-filamentous granules, increase in effluent suspended solids and eventually disintegration of granule. The end result of filamentous growth was a failure of the aerobic granular sludge SBR. Thus, the SRT should be carefully managed within the suitable range of floc-forming bacteria (normally about 10 days) to get success in operating granular sludge system.

2.2.5 Treatment performance of aerobic granular sludge systems

a. Organic and nitrogen removal

The specific structure of aerobic granule containing the aerobic-anoxic zone could enhance simultaneous nitrification denitrification due to incomplete oxygen diffusion through granule radius. In the view of microbiology heterotroph, nitrifying and denitrifying populations could co-exist in microbial matrix of granules. Oxygen could diffuse into the biofilm depth of 150–300 μm (Tijhuis et al., 1994).

Table 2.10 presents types of wastewater containing organic matter and/or nitrogen can be treated by aerobic granular sludge process in a single aerobic reactor. Organic and nitrogenous compounds are simultaneously removed. Granular sludge reactors can function well in a wide loading range of 1-15 kgCOD/m^3.d. Organic matter is promptly degradable with the removal efficiency greater than 95 % in a high granular sludge concentration (3-20 g/L of VSS). In the case of wastewater containing high influent solid content (up to 0.95 g/L), the total COD removal efficiency is 50%. It is a bit low because suspended solids partly pass through the effluent (Schwarzenbeck et al, 2004). Further, the granular sludge system can sustain a NLR of 0.1-1.5 kgN/m^3.d. Nitrogen can be removed even in the organic or inorganic form (synthetic or real wastewater). Simultaneous nitrification denitrification occurs in the system to produce nitrogen gas (Beun et al., 2002; Arrojo et al., 2004; McSwain et al., 2004; Cassidy and Belia, 2005; Schwarzenbeck et al., 2005; Mosquera-Corral et al., 2005; Qin and Liu, 2006). To enhance complete nitrogen removal, there are some options such as alternative anoxic/aerobic cycle (Arrojo et al., 2004; Cassidy and Belia, 2005); aerobic/anoxic cycle (Yang et al., 2003; Qin and Liu, 2006); a stage of low DO (Beun et al., 2002; Mosquera-Corral et al., 2005); addition of external carbon source; static fill (Etterer and Wilderer, 2001). Complete nitrogen removal is achieved by apply low DO concentration of 0.5 mg/L (Yang et al., 2003) or by adding external organic substrate (Qin and Liu, 2006). In case of absence of external carbon sources, pre-accumulated poly-β-hydroxybutyric acid (PHB) in microbial granules can be utilized for cell maintenance and denitrification. The potential role of PHB for

denitrification (reducing power) by microbial granules is limited, less than 28 mg NO_3-N/L has been found to be denitrified with internally accumulated PHB (Qin and Tay, 2005). However, it is impossible to obtain stable granular sludge at low oxygen concentration (40% saturation). Granules break up when operated long term under low aeration rate (Mosquera-Corral et al., 2005).

The P removal efficiency is high in aerobic granular sludge system, normally more than 70%. The efficiency can be improved to be more than 90% when an enhanced P removal process applied (alternative anaerobic/aerobic). The anaerobic condition can be created by static fill or low mixing. The P is released by the granules during the anaerobic fill period, and then rapidly taken up during the aerobic react. The P content in aerobic granule is in the range of 1.9-9.3%, depending on the ratio of P/COD of the influent (Mosquera-Corral et al., 2005; de Kreuk et al., 2005).

As a result, nitrogen and phosphorus removal can happen effectively in granule even aerobic condition exists in system and depends on granule structures, size, oxygen concentration, external carbon source, PHB and microbial population.

b. Recalcitrant removal

Aerobic granule is biodegradable not only substances such as glucose, acetate, peptone, etc. but also recalcitrant such as phenol (Jiang et al., 2004; Tay et al., 2004 & 2005), pyridine (Adav et al., 2007c), p-nitrophenol (PNP) (Yi et al., 2006), 2,4- dichlorophenol (Wang et al., 2007b), methyl t-butyl ether (MTBE) (Zhang et al., 2008) and nitrilotriacetic acid (NTA) and ferric–NTA complex (Nancharaiah et al., 2006).

Tay et al. (2004) found that the granules degraded phenol at a specific rate exceeding 1 g/gVSS.d at 500 mg/L of phenol or at a reduced rate of 0.53 g/gVSS.d at 1900 mg/L of phenol. Aerobic granule could treat wastewater containing phenol up to 2.4 kg/m^3.d (Tay et al., 2005). In addition, Adav et al. (2007) noted that aerobic granules could remove phenol at 1.18 g/gVSS.d. Furthermore, Yi et al. (2006) reported that the specific degradation rate of p-nitrophenol (PNP) increased with corresponding increase in PNP concentration up to 40.1 mg/L with a peak at 19.3 mg/gVSS.h, and declined with any further increase in PNP concentration as substrate. Granules acclimated and quickly stabilized one week after phenol was introduced. Granules exhibited good settling ability with good biomass retention and metabolic activity. No significant inhibitory effects from phenol toxicity were observed at the intermediate phenol loadings of 0.6 and 1.2 kg/m^3.d, except for a slight lag in the ability of the granules to degrade phenol during the initial cycles of loading of 2.4 kg/m^3.d. However, this buildup quickly dissipated as the granules adapted rapidly to the high phenol concentrations (Figure 2.12)

Aerobic granules can efficiently degrade pyridine over initial concentrations of 200–2500 mg/L (Adav et al., 2007c). The specific degradation rate of pyridine has been found to be 73.0 and 66.8 mg/gVSS.h at 250 and 500 mg/L of pyridine, respectively. Phenol granules can be applied for the removal of phenol in the presence of pyridine in industrial wastewater (Adav et al., in press).

Aerobic granules can eliminate recalcitrant metal chelating agents, namely free nitrilotriacetic acid (NTA) and Fe(III)–NTA. Most of the influent NTA in the reactor is removed during the aeration period. Pre-cultivated granules completely degrade 2mM of free

NTA and Fe–NTA in 14 and 40 h at the respective specific degradation rates of 0.7 and 0.37 mM/gSS.h.

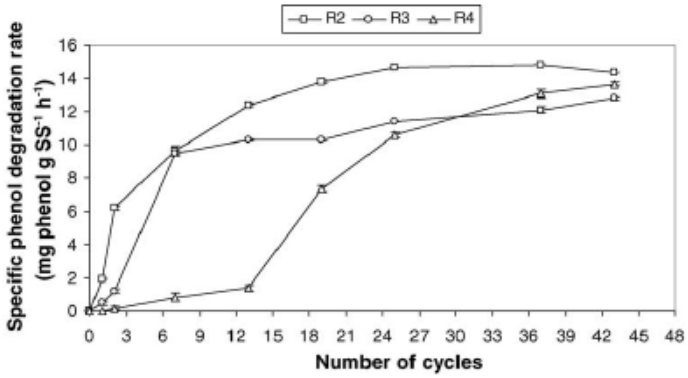


Figure 2.12 Specific phenol degradation rate at phenol loading rate of 0.6 (R2), 1.2 (R3) and 2.4 (R4) kg/m^3.d (Tay et al., 2005)

Efficient degradation of a recalcitrant synthetic chelating agent by aerobic biogranules suggests their potential application in situations where heavy metals are co-disposed with metal chelating agents (Nancharaiah et al., 2006).

c. Adsorption of heavy metals and dyes

Aerobic granule can be a good candidate of biosorbent for metals and dye removal from wastewater because of its strong points such as compactness, porous structure and excellent settling ability as compared to conventional bioflocs. Heavy metal ions (cadmium, copper, nickel, magnesium, zinc, etc.) and dyes (Rhodamine B, Malachite) were found to adsorb by aerobic granules effectively. Especially, the excellent settleability of aerobic granules could ensure a rapid solid liquid separation of the treated water, which in turn leads to a simple process design (Liu et al., 2003c).

The uptake capacity of the aerobic granules was found to be in the range of 43-566 mg Cd^{2+}/g granules. The maximum sorption capacity was 35 mg Ni^{2+}/g granule at pH 6. The Ni^{2+} biosorption and the zeta potential of aerobic granules was pH dependant (Xu et al., 2006). In addition, the large quantity of K^{+}, Mg^{2+} and Ca^{2+} released during the Ni^{2+} biosorption indicates that the ion-exchange mechanism was involved in the Ni^{2+} biosorption. However, it was observed that per 100 mg Ni^{2+} adsorbed by aerobic granules. There was simultaneously release 48.27 mg of Ca^{2+} and 8.25 mg of Mg^{2+} (Liu et al., 2003c).

It was reported that the maximum biosorption capacity of individual Cu^{2+} and Zn^{2+} by aerobic granules were closely related to the initial concentrations of Cu^{2+} and Zn^{2+} in the reactor and was 246.1 mg/g and 180 mg/g granule respectively (Xu et al., 2004). The metal affinity to aerobic granules was found to be in the order of $Cd^{2+} > Cu^{2+} > Ni^{2+}$ (Xu and Liu, 2008). The maximal adsorption capacity of the granules has been found to be 55.25 mg/g Co at pH 7 and 62.50 mg/g Zn at pH 5. Binary-metal addition induced competitive sorption among the metals with reduction of the maximal adsorption capacity by 1.2 mg/g and 6.0 mg/g for Co^{2+} and Zn^{2+}, respectively, compared to the single-metal sorption. The rate limiting step in this sorption process may be chemisorption involving valent forces between sorbent and sorbate. The initial biosorption rate was $Co^{2+} > Zn^{2+} > Co^{2+}$ (Co–Zn) > Zn^{2+} (Co–Zn) (Sun et al., 2008).

Additionally, the biosorption of Malachite Green (MG) was highly dependent on initial concentration, adsorbent dosage, pH and temperature. The biosorption capacity increased with the increase of pH in the range of 2–11 which fitted Langmuir, Freundlich and Redlich Peterson isotherms. The maximum biosorption capacity of MG onto aerobic granules was 56.8 mg/g (Sun et al., 2008b).

These above results indicate that aerobic granules have a high biosorption capacity for heavy metals and dyes removal from industrial wastewaters.

d. Severe temperature conditions

The biological treatment systems are operated day by day and usually face some trouble during the change of the seasons due to the decrease of the metabolic activities of microorganism at low temperature. De Kreuk et al. (2005) found that start-up temperature of granular SBR was at 20^{o}C, and then lowering to 15 or 8^{o}C, did not lead to any effect on granule stability and biomass could be retained well in the system. On the other hand, start-up at 8^{o}C resulted in irregular granules which caused severe biomass washout and instable operation. Due to the decreased activity in the outer layers of granules at lower temperature, the oxygen penetration depth could increase, which in turn resulted in a larger aerobic biomass volume, compensating the decreased activity of individual micro-organisms. Consequently, the denitrifying capacity of the granules decreased at the reduced temperatures, resulting in an overall poorer nitrogen removal capacity.

Thermophilic aerobic sludge systems have shown advantages over other operations including low waste biomass production, higher degradation rate and less aeration tank volume, elimination of cooling requirements for high temperature wastes, enhanced solubility and degradation of low-solubility substrate, and rapid inactivation of pathogens.

Zitomer et al. (2007) demonstrated that thermophilic aerobic biomass with good settling property (SVI values as low as 60 mL/g) was achieved in SBRs. The well-settling biomass contained granules and/or floc particles, with average granule diameter as high as 1.2–1.9 mm and granule resistance to disintegration was comparable to mesophilic aerobic granules. Two thermophilic bacteria were isolated from the thermophilic granules, namely *Anoxybacillus flavothermus and Pseudoxanthomonas taiwanensis*. High alkalinity and/or CO_2 in aerobic thermophilic systems could help the selection for a microbial population of granules.

Therefore, aerobic granular sludge systems can work well throughout the year in cold areas where temperature significantly changes in seasons when the reactor start-up is carried out in warm seasons (summer or spring). In addition, aerobic thermophilic granular sludge functions well for the hot wastewaters. The hot waste stream can be potentially treated via themorphilic granular sludge treatment with gravitational solid liquid separation without a pre-cooling step.

Table 2.10 Treatment performance of aerobic granular sludge system

Carbon/nitrogen sources	OLR (kg/m^3d)	NLR (kg/m^3d)	HRT (h)	SRT (d)	v (cm/s)	COD removal (%)	TN removal (%)	P removal (%)	N-NH_4 effluent (mg/L)	N-NO_2 effluent (mg/L)	N-NO_3 effluent (mg/L)	VSS (g/L)[a]	VSS eff (mg/L)[b]	Reference
Acetate/AC	1.2 - 1.6	69 mg/L	5.6-7.4	-	2.02	-	44-75	95-97	-	-	-	18-20	-	de Kreuk et al., 2005
Barley dust WW	3.4	-	12	-	0.5-0.7	50-80	-	-	-	-	-	6-7	-	Swazenbeck et al., 2004
Brewery WW	3.5	0.24	-	-	1.77	88.7	88.9	-	14.4	-	50	(10-11)	-	Wang et al., 2007
Dairy WW	7	0.7	-	-	-	80	70		20-30	-	15-18	(5-6)	50-800	Arrojo et al., 2004
Glucose, acetate/AS	2.5	0.12		15	0.42	95	97		< 1			4-5		Jang et al. 2003
Glucose, acetate/AS	1.76-2.84	0.10-0.16	-	-	-	93-98	47-99	-	1-29	0.1-13	2.3-10	(7.7-9.5)		Kim et al., 2008
Glucose & peptone	2.4	-	-	-	-	96	-	-	-	-	-	(3.2-9.0)	(170-290)	McSwain et al., 2004
Glucose, acetate, peptone/ AC	3.6	-	-	10		93	99	73	-	< 0.23	0.02	-	-	Etterer & Wilderer, 2001
Phenol	1.0-2.5	-	8	-	2.97	> 96	-	-	-	-	-	(5.5-8.5)	-	Jiang et al., 2004
Slaughter house WW (beef)	2.6			20	1.57	98	97	>98	2	0	26	8	42	Cassidy and Belia, 2005
Ethanol/AC	2.0	0.15-0.45	12	20	2.54	95-97	> 99	-	-	0.22-0.45 (external C added)		3.0-5.5	-	Qin and Liu, 2006
AS	-	0.13-1.50	7.6-120		0.85	-	> 90	-	< 10-40	< 400	< 500	-	-	Tsuneda et al., 2003
AS, nitrite (anamox)	-	400 mg/L each	24	-	-	-	-	-	< 100	< 30	-	1.25	0	Trigo et al., 2006
Ethanol/AC	1.5	0.08-0.45	7.7	-	0.5-2.4	> 95	100	-	-	-	-	9	-	Yang et al., 2003
Acetate/AC	1.6	0.2	5.8	25	2.52		8-45		0.13-0.70	0.04-3.90	0.13-22.0	5	< 120	Mosquera-Corral et al., 2005
Dairy effluent WW	4.5-5.9	0.12-0.49	16		1.1	90	80	67	-	12-85		2-7	-	Swazenbeck et al., 2005

Note: a,b values in bracket are MLSS and suspended solids; WW: wastewater; AC: Ammonium chloride; AS: Ammonium sulfate; v: superficial air velocity; NLR: nitrogen loading rate

2.3 Membrane Bioreactor

2.3.1 Development of membrane bioreactor

Municipal and industrial wastewaters are often treated biologically, such as by the activated sludge process, using microorganisms for degradation of organic pollutants. The conventional activated sludge process, which is one of the most popular wastewater treatment processes, has the major disadvantages, namely (1) Treated water quality is dependant on the settling properties of the biological particles. If the settling ability of the system is poor, it may result in presence of suspended solids in the effluent and a progressive washing out of the biomass from the bioreactor; (2) Hydraulic retention time of the bioreactor and secondary sedimentation tank is long. Thus, the volume of the tank has to be large which leads to large area requirement and high construction costs for the system; (3) Bulking and foaming due to filamentous growth; (4) Insufficient pathogens removal and potential of chlorinated by-products limit the reuse of treated effluent.

Nowadays, due to the water scarcity the water reuse and recycling are becoming more and more vital. As such, an advanced treatment is required with the following properties (Wisniewski, 2007):

- Disinfection without any oxidation step that induces carcinogen molecule formation;
- Compactness to optimize aesthetics, environmental impacts;
- Reliability notwithstanding the influent characteristic variation;
- Standards regarding sustainability (energy, chemicals and waste production).

An alternative treatment technology is the membrane bioreactor (MBR) which replaces two stages of the conventional activated sludge process, clarification and settlement, with a single integrated biotreatment and clarification step (Chang et al., 2002). The advantages of MBR are presented in Table 2.11.

2.3.2 MBR process and its potential application

The different membrane processes resulted from the various demands on the separation process. Subdivision of the different processes occurs according to:

- Driving force behind the filtration process (Vacuum, pressurized, gravity);
- Type of the inserted membrane (submerged/side-stream or dead-end/crossflow);
- Kind of the matters to be separated as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO).

In real, the suitable membrane types for biological wastewater treatment are MF, UF and, less commonly, NF. There has been MBR configurations described as Figure 2.13. Firstly, in the crossflow filtration MBR or side-stream MBR, the membrane can be either outer- or inner-skinned, and permeate is extracted by circulating the mixed liquor at high pressure along the membrane surface. Secondly, in a submerged MBR or deadend filtration MBR, the membrane should be outer-skinned which was developed by Yamamoto et al. (1989). This mode is currently used for most of MBR treatment plants around the world due to less energy consumption. In this mode, permeate is extracted by suction or, less commonly, by gravity forces. Lastly, the recent MBR configuration called semi-deadend MBR developed

by Polymem Company which is based on the combination of the two above configurations. Membrane module is placed externally to the bioreactor and operated in dead-end filtration mode with a recycling of the concentrate into the aeration tank, which generates a very small liquid velocity in the module. This system can be named an "external loop dead-end MBR" or "semi-deadend MBR". Its advantages are low energy consumption (comparable to submerged MBR), less contact with biomass, fouling control and ease in cleaning and maintenance (Bouchot et al., 2006).

Table 2.11 Advantages of MBR (adopted from Visvanathan et al., 2000)

Advantages	Details
High rate decomposition	- Treatment efficiency is improved by preventing leakage of undecomposed polymeric substances. If these polymers are biodegradable, they can be broken down with a reduction in the accumulation of substances within MBR; - Other dissolved organic substances with low molecular weights, which can be eliminated by membrane separation alone, can be broken down and gasified by various microorganisms or produced new bacteria cells.
Treated water quality	- MBR produces the effluent (BOD_5 < 5 mg/L); - MBR process is 15-20 fold higher in substrate conversion rate compared to CASP; - Solid/liquid separation is conducted by membrane filtration. Therefore, the final effluent does not contain suspended matters as CASP and enables the direct discharge into receiving sources and/or reuse of the effluent for cooling, toilet flushing, watering or process water.
Flexibility in operation	- SRT can be controlled completely independent so the system can be run at very long SRT providing favorable conditions for the growth of slow-growing microorganisms, which are able to degrade bio-refractory compounds and control fouling.
Compact plant	- Biomass concentration can be maintained up to 40 g/L in the reactor. Therefore, the system can stand for high OLR resulting in the reduced size of the bioreactor; - Secondary clarifier, filter, sludge thickener or post treatment are not required for MBR process.
Low sludge production	- Excess sludge from MBR is lower than CASP about one fifth fold (0.22-0.53 $kgSS/kgBOD_5$); - Low F/M ratio and longer SRT (50-100 d) causes low sludge production rate; - SRT increased causes reduction in filaments, increase in rotifiers and nematodes
Disinfection and odor control	- The removal of bacteria and viruses can be achieved without any chemical addition; - All the process equipment can also tightly close, no odor dispersion occurs.

Table 2.12 Key facets of two MBR configurations (Modified from Judd, 2004)

Sidestream MBR	Submerged MBR
Since early 1970s.	Most recent development (since 1990).
Membrane placed external to bioreactors.	Membrane placed in bioreactor.
Pump system with permeation rate determined by TMP and crossflow.	Permeate removed under hydrostatic head, with or without permeate suction, at rate partly determined by aeration.
Higher flux and hydraulic resistance.	Lower flux and hydraulic resistance.
Lower aeration and membrane area requirement.	Greater aeration and membrane area requirement.
Stabilised flux with periodic chemical cleaning.	Stabilised flux with periodic chemical cleaning (flat plate membrane configuration); Short backwash cycle, periodic chemical cleaning (hollow fibre configuration).
Greater overall energy demand (2-4 kWh/m^3).	Lower energy demand (0.2-0.8 kWh/m^3).
Greater process (hydrodynamic) control.	Reduced process (hydrodynamic) control.

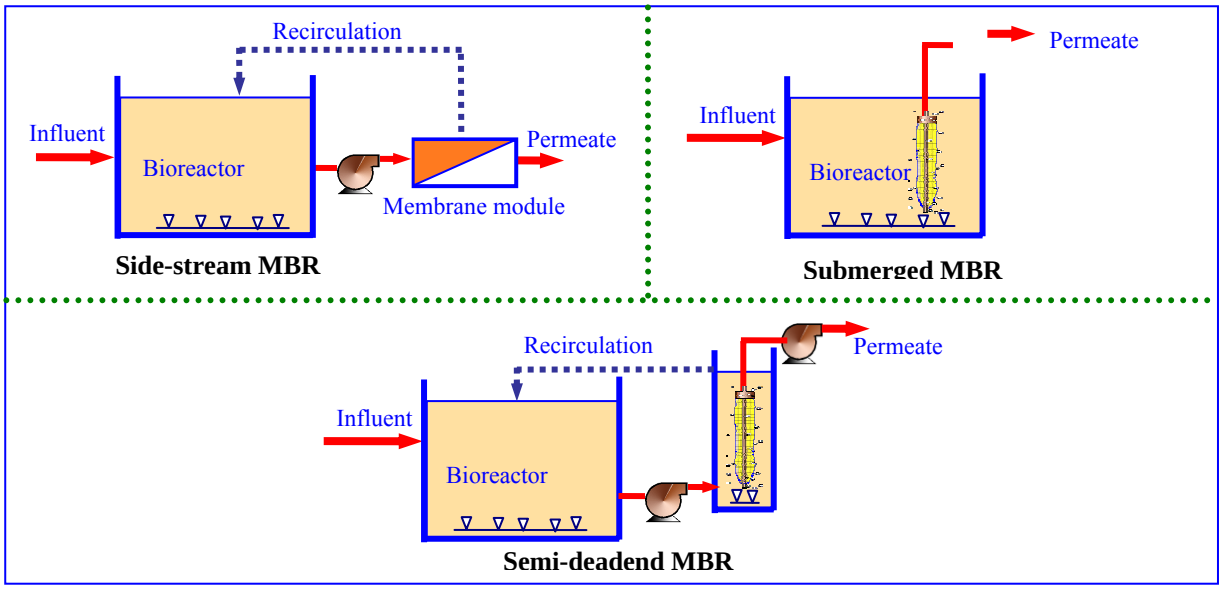


Figure 2.13 Configurations of MBRs

2.3.3 Membrane fouling

Membrane fouling results from interactions between membrane materials and the components of the biomass which includes substrate components, cells, debris, and microbial metabolites. As soon as the membrane surface comes into contact with the biological suspension, deposition of biosolids onto it takes place leading to flux decline (Chang et al., 2002). There are two main fouling mechanisms namely reversible fouling and irreversible fouling. Reversible fouling stands for the fouling caused by cake layer which is readily removable under a physical washing protocol. By contrast, irreversible fouling indicates the internal fouling caused by the adsorption and/or deposition of soluble matters into the membrane pores which is generally removed by chemical cleaning. Factors influencing on membrane fouling are summarized in Figure 2.14.

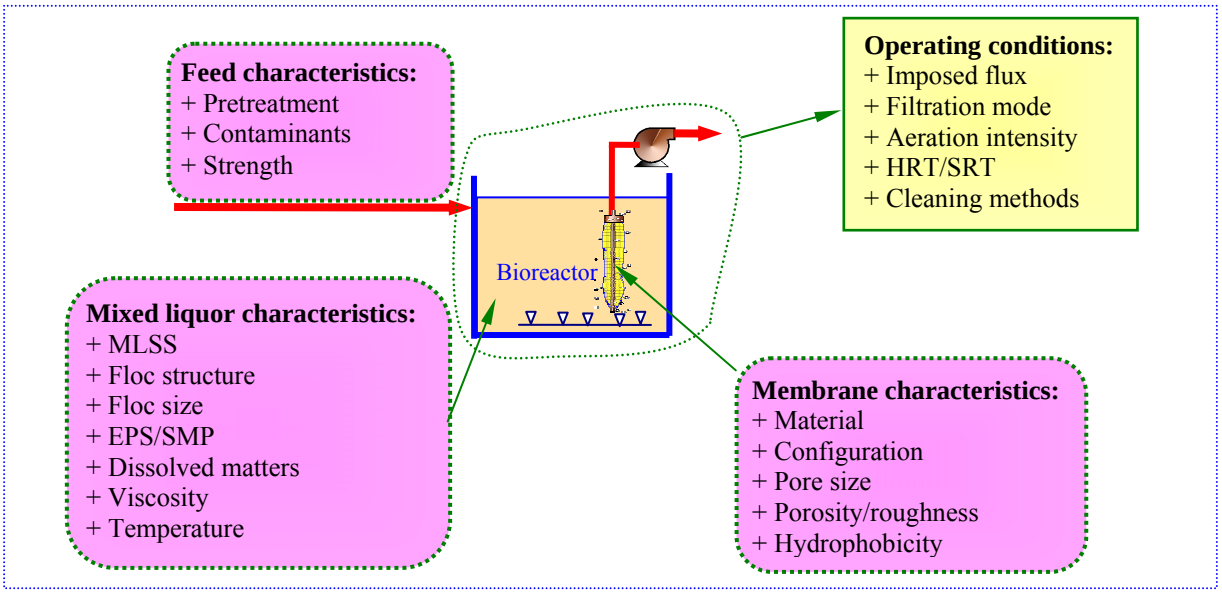


Figure 2.14 Factors contributing to membrane fouling in a membrane bioreactor

Effect of membrane fouling on the flux decline can be explained by the resistance-in-series model. In this model, the relationship between permeate flux and trans-membrane pressure (TMP) is described by the following equations:

$$J = \frac{\Delta P}{\mu . R_t}$$

Where,
J: Permeate flux ($m^3/m^2.s$)
ΔP: TMP (Pa)
μ: Viscosity of the permeate (Pa.s)
R_t: Total resistance (1/m)

$$R_t = R_m + R_c + R_f$$

Where,
R_m: Intrinsic membrane resistance
R_c: Cake layer resistance
R_f: Fouling resistance due to irreversible and pore plugging

Characteristics of the cake layer play an important role in membrane fouling. Effects of cake layer characteristics could be described by the Carman-Kozeny equation reported by Boerlage et al. (2002) as follows.

$$\alpha = \frac{180(1-\varepsilon))}{\rho_p . d_p^2 . \varepsilon^3}$$

Where,
P_h : Hydraulic permeability through the cake layer
d_p: Particle diameter
ε: Porosity of the cake layer
ρ_p: density of particles forming cake

From the above equation, it is obvious that greater the particle size and porosity, the lower the specific cake resistance (higher permeability). Many attempts have been conducted to improve the permeability of the cake layer by the addition of filter aids into MBR such as ferric chloride, alum coagulants, powder activated carbon (PAC) (Visvanathan et al., 2000; Le-Clech et al., 2006).

There are two operation modes namely, constant TMP operation and constant flux operation. In reality, the latter one is preferable for submerged type MBR. According to Le-Clech et al. (2006) the constant flux operation has three stages of membrane fouling namely conditioning fouling, steady fouling and TMP jump. Factors contributing to membrane fouling are shortly explained as follows:

a. Membrane characteristics

Membranes with small pores reject various ranges of materials which in turn result high cake resistance compared to membranes with large pores. However, due to

deposition of organic and inorganic material inside the membrane pores, the large pore membranes show poor long term performance. Further the rougher membranes are more prone to membrane fouling when compared to the smoother ones. Membrane configuration too plays an important role in fouling. The hollow fibre type has high possibility for fouling compared to the tubular type which leads to frequent washing and cleaning of membrane. In addition, the membrane fouling is severe in hydrophobic than the hydrophilic membranes (Le-Clech et al., 2006).

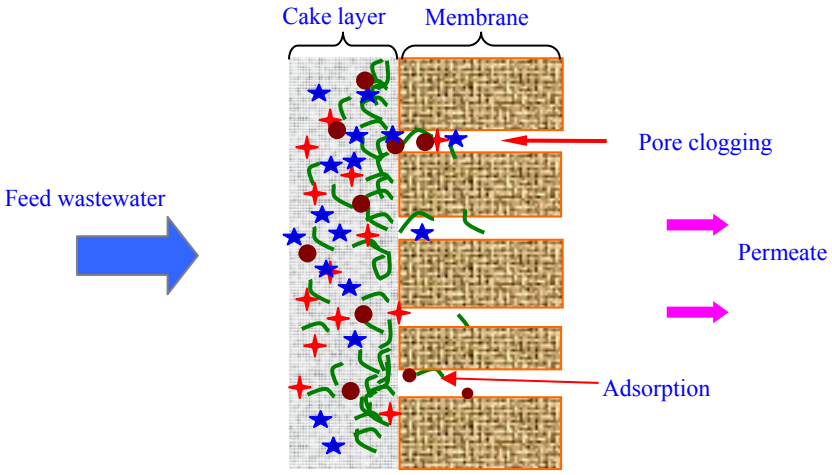


Figure 2.15 Membrane fouling behavior

b. Biomass characteristics

The mixed liquor suspension in MBR was classified into three biomass fractions, namely suspended solids, colloids and solutes. Each biomass fraction impacted on fouling in different rate. Bouhabila et al. (2001) found that colloidal fraction was significant in flux decline compared to others.

Viscosity was another important factor which affects biomass characteristics. High viscosity led to reduction of the oxygen transfer and high fouling propensity (Germain and Stephenson, 2005).

Floc size of MBR sludge was smaller than that of conventional activated sludge and often larger than the pore size of used membrane. Larger size of flocs could not directly block the pores of membrane, while the biological flocs played major role in forming cake on the membrane surface (Le-Clech et al., 2006). In sidestream MBR, the intensive recirculation of biomass resulted in flocs breakage which caused poor settleability of suspension and generated smaller flocs in the reactor which, in turn, increased the potential for fouling of membrane (Wisniewski and Grasmick, 1998).

c. Extracellular polymeric substances (EPS) and soluble microbial product (SMP)

EPS are one of the cell components whose function is microbial aggregation. EPS includes bound EPS (in cells/flocs) and soluble EPS (in bulk liquid). Further, soluble EPS and SMP are identical (Le-Clech et al., 2006). The SMP compounds consist of proteins, polysaccharides and organic colloids which are produced during substrate utilization, biomass growth and cell lysis. Due to hydraulic shock loads, low pH, nutrient deficiency and presence

of toxic compounds the SMP is formed (Rosenberger et al., 2006; Jarusutthirak and Amy, 2007). SMP level in the MBR sludge is high due to retention of large amount of macromolecules on the membrane surface. Liang et al. (2007) found that majority of the SMP were hydrophobic. The accumulation of SMP was high when the SRT was short which caused an increase in fouling potential. Currently, several authors realize that SMP plays an important role in membrane fouling.

d. Operating conditions

Aeration in membrane system has several functions including providing oxygen to the biomass, mixing suspension and mitigating fouling. The aeration causes shear at the membrane surface which prevents particle deposition.

The biomass characteristics are controlled by SRT which is one of the most important parameters impacting on degree of membrane fouling. Long SRT increases nitrogen and organic removal efficiency and lowers fouling rate (Bouhabila et al., 1998). However, high viscosity associated with very high biomass concentration leads to excessive fouling (Chang et al., 2002).

As a general trend it is now accepted that the shorter the HRT, the longer the SRT and the higher the MLSS concentration. It is clear that SRT and HRT are not the direct fouling causes but rather parameters influencing factors like EPS (or SMP), particle size distribution and MLSS. Therefore, Chang et al. (2002) proposed that HRT and SRT only indirect impacted on membrane fouling.

Previously, membrane fouling was proposed to be due to the deposition of suspended solids/flocs (cake/gel formation, pore blocking), colloids (Bouhabila et al., 2001) and solutes (Trussell et al., 2006; Jarusutthirak and Amy, 2006). Recently it has been found that the fouling mechanism of conventional submerged MBR is mainly caused by the deposition and/or accumulation of SMP or soluble extracellular polymeric substances (sEPS) on membrane if reversible fouling (cake formation) is well controlled. The sEPS mainly comprises of soluble polysaccharide (sPS) and soluble protein (sPN). The fouling potential of sPS, sPN or both of them is still unclear. The total sEPS (sPS and sPN) is tone of the factors which influence membrane fouling (Trussell et al., 2006; Liang at al., 2007) where sPS plays a major role as membrane foulant (Rogenberger at al., 2006; Jarusutthirak and Amy, 2006; Kim and Digiano, 2006).

2.2.4 Measures of fouling mitigation

Membrane fouling can be reversible or irreversible depending on the degree of fouling. There are two main groups of fouling control, namely removal of fouling and limitation of fouling as described in Table 2.13.

Table 2.13 Measures of membrane fouling mitigation (Modified from Le-Clech et al., 2006)

Removal of fouling	Methods	Details
Physical cleaning	Water BW (similar to BF) Air BW Membrane relaxation Air scouring	+ Less frequent but longer BW is more efficient than more frequent BW (Ex: Frequency: 600 s filtration/45 s BW); + Amount of 5-30 % of produced permeate is often used; + Membrane breakage and rewetting are potential issues (not applicable for flatsheet membrane); + Attached foulants can diffuse away from membrane surface (on/off filtration cycle); + Air scouring applied during relaxation could enhance efficiency;
Chemical cleaning	Chemically enhanced BW Maintenance cleaning with higher chemical conc. Intensive chemical cleaning Combination of sonication, BW and chemical cleaning	+ Daily basis; + Weekly, about 30 minutes for a complete cycle; + Carried out when filtration is not sustainable due to an elevated TMP; + CIL (by 0.01% NaOCl, citric acid); + Once or twice a year; + CIP (0.2-0.5% NaOCl and 0.2-0.3% citric acid or 0.5-1.0% oxalic acid) + Effective but difficult to apply for full-scale;
Limitation of fouling	Improving of anti-fouling properties of membrane Operating MBR under less fouling conditions Modification of biomass suspension	+ Increase the hydrophilic characteristics of membrane by introducing polar group on membrane surface (NH_3 and CO_2 plasma treatments of membrane materials for hollow fibre polyethylene); + Addition of TiO_2 nanoparticles to the casting solution and precoating of TiO_2; + Precoating is more efficient than entrapment; + Aeration of 24-50 m^3 air/m^3 permeate; + Increase CFV does not reduce fouling rate when the deposition layer starts; + Pulsing air at a frequency of 1 s on/ 1 s off (still not economic); + SRT control (long SRT less fouling) + Reactor design (addition of spiral flocculator; vibrating membrane; helical baffles; suction mode; compact reactor; airlift configuration; addition of carrier; SBR-MBR, + Operated under sustainable flux (sub-critical flux) + Pre-coagulation/sedimentation limits fouling potential; + Increase floc size by addition of coagulant/flocculent or zeolite which adsorbs colloids, solutes and SS (Fe is more effective than Al); + Precipitates (Ferric phosphate and K-jarosite ($K\text{-}Fe_3(SO_4)_2(OH)_6$)) stimulates fouling rate; + Regular addition of adsorbent such as PAC (1 g/L) + Use of cationic polymer (MPE50 with conc. of 1.0-2.2 g/L) + Use of aerobic granular sludge in MBR

Note: BW: Backwashing; BF:Backflushing; CIP: Cleaning in place; CIL: Cleaning in line;

2.4 Aerobic granular sludge and its coupling with Membrane Bioreactor

2.4.1 Combining conditions and granule stability of granular sludge MBR

The integration of granular sludge into MBR promises a novel technology for wastewater treatment. Thus, some researchers start focusing on this trend. The stability and the coupling method of granule in MBR are the most concerned issue as follows:

Tay et al. (2007) operated a granular sludge MBR based on SBR operating conditions. Aerobic granule was previously cultivated in a sequencing aerobic sludge blanket rector. When matured granule formed, membrane module was submerged in the reactor. Effluent was discharged both by membrane filtration (3/8 reactor volume) and by gravity through an outlet valve (1/8 reactor volume). Those aimed to create feast-famine period and to wash out a light fraction of biomass, respectively. Membrane filtration was started after 1 h of aeration cycle and stopped before the cycle 3 minutes. The direct discharge of light biomass was to maintain the granule stability in the system.

Wang et al. (2008) studied the stability of aerobic granule in a semi-continuously submerged MBR in which matured granular sludge were seeded in reactor and waste sludge was removed after each 15 days (SRT = 35-45 d). By this operation mode, the feast and famine condition was as long as 15 days. The granule was found to be less disintegration after 24 days of operation. The size distribution of biomass in the MBR was wider compared to that of a granulation SBR. The percentage of granular biomass was 56-62% of total sludge concentration during the operation. The granules with small particle size could be formed in granular sludge MBR.

Li and co-workers (2005; 2007 & 2008) operated a submerged MBR with granular sludge taken from a batch granulation reactor to study on the stability and characteristics of granule in the MBR. The system was operated in a short period of 55 days (SRT = ∞). Aerobic granule was possibly maintained in reactor during 55 days. However, the granules had smaller size, poorer settleability and sludge activity during operation. The size reduced from 3 mm on the first day to 2 mm on day 55. The reduction was thought to be the overgrowth of filamentous microorganisms which occurred due to the combined effect of high aeration shear stress, reduced DO, and long SRT.

Interestingly, Trigo et al. (2006) found that granules could be formed contingently in the MBR conducting the anammox process. The formation of granule was thought to be due to the tendency of anammox microorganisms to grow into biofilms or granules.

2.4.2 Fouling behavior in MBR

Aerobic granule combines with MBR could develop a novel MBR. Granules minimize the concentration of flocs, consequently alleviating fouling (Li et al., 2005; Le-Clech et al., 2006; Tay et al., 2007; Trigo et al., 2007). It was observed that the fouling propensity of granular sludge MBR was less than that of floc MBR. The specific cake resistance of granular sludge was about 6 times less than that of flocs (Table 2.14) which indicates that the granules were much less compressed compared to that of flocs. The resistance of suspended solids (SS) fraction was insignificant compared to that of colloids (CL) and solutes (SL) fraction. Further, Tay et al. (2007) reported that the constant

pressure test showed that when TMP increased by 8 fold, the membrane permeability loss in granular sludge MBR mixed liquor was 1.68 fold lower than that of floc MBR. Constant flux test indicated that when flux increased by 3 fold, the loss of membrane permeability in granular sludge MBR mixed liquor was 21 fold lower than that of floc MBR. During operation, the TMP in the floc MBR increased periodically to 50–60 kPa with regular physical cleaning. In the granular sludge MBR, TMP of 3–6 kPa was maintained without any physical cleaning.

The flux of MBR containing granules was more 50% higher than that of MBR containing sludge flocs during the operation. The foulants of granular sludge MBR were thought to be mainly from colloids and solutes fractions. In addition, it was obviously that the pre-cultivated granules were slowly broken and partly preserved when operated in normal operating conditions of MBR (Li and co-workers, 2007)

The short term filtration test of sludge samples taken from a granular sludge SBR and activated sludge process was conducted to study on their fouling tendency. After 15 minutes of filtration, the final flux of granule sludge sample was twice compared to that of activated sludge one. The fouling mechanism of granular sludge was noted due to the cause of pore-blocking while that of activated was caused by cake layer resistance. Cake resistance of AS sample occupied 72.68% while fouling resistance of granular sludge sample was 44.2% (Zhou et al., 2007).

Table 2.14 Resistance of biomass fractions and cake resistance of granule and floc

Sludge	R_c (m^{-1}) x10^{11}	R_f (m^{-1}) x10^{11}	$\propto$ (m/kg) x10^{12}	$R_{SS\text{-}CL\text{-}SL}$ (m^{-1}) x10^{11}	R_{CL} (m^{-1}) x10^{11}	R_{SL} (m^{-1}) x10^{11}	Reference
Granules	-	-	7.86	3.17	4.00	3.81	Tay et al., 2007
Flocs	-	-	45.6	31.6	4.36	4.81	
Granules	3.04	3.9	16.0	8.76	-	-	Zhou et al., 2007
Flocs	12.29	2.8	49.1	16.91	-	-	

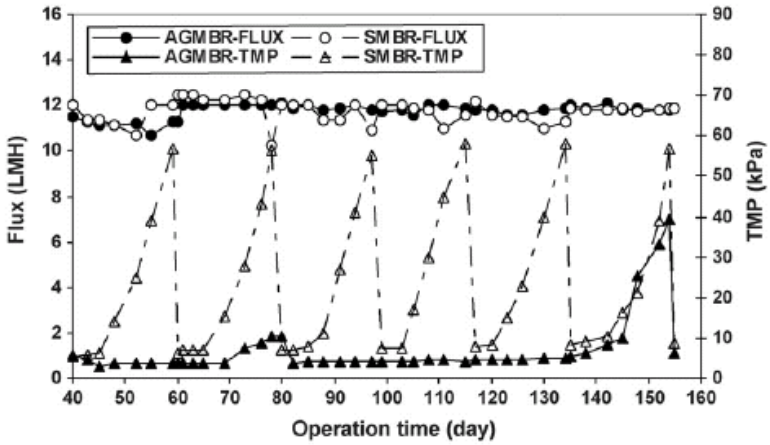


Figure 2.16 TMP profile of MBR with granular sludge (AGMBR) and floc (SMBR) (Tay et al., 2007)

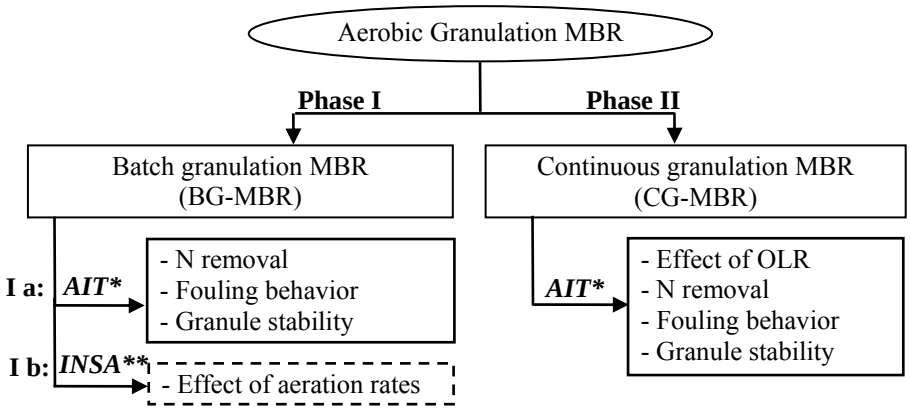
Generally, the development of MBR treatment is limited due to problems such as fouling, energy cost, cleaning cost, etc. Hence, the MBR is coupled with aerobic granulation to overcome these problems. The research on this aspect was untaken to investigate the fouling behaviour and nitrogen removal of aerobic granulation MBR which could be an attractive solution for water reuse in near future.

Chapter 3

Methodology

3.1 Introduction

This experimental study comprised of two phases, namely Phase I: Batch Granulation MBR (BG-MBR) and Phase II: Continuous Granulation MBR (CG-MBR) (Figure 3.1). The research work focused on possibility of coupling of aerobic granular sludge reactor (Sequencing batch airlift reactor-SBAR) with MBR configurations. The fouling potential, nitrogen removal, granule stability and organic loading rate (OLR) were investigated for both systems. Further, the effect of various aeration rates on characteristics of effluent was examined to compare the fouling characteristics between conventional SBAR and granulation SBAR (Phase I b).



** at Asian Institute of Technology(AIT), ** at Institut National des Sciences Appliques (INSA), France*

Figure 3.1 Flowchart of overall research study

3.2 Materials and microorganisms

3.2.1 Synthetic wastewater

Glucose and ammonium chloride were the carbon and nitrogen source respectively for granule cultivation. Table 3.1 describes the components of wastewater used for research phases. For Phase I b and phase II, the composition of OLR of 2 kg COD/m^3.d and NLR of 1 kg N/m^3.d (COD = 700 mg/L, NH_4^+-N = 325 mg/L) was used. When OLR was increased to 4 and 8 kg COD/m^3.d, glucose and phosphorus were proportional to OLR. Nitrogen component was kept constant for different OLRs. Concentration of $NaHCO_3$ in the feed varied with OLR to maintain reactor pH in the range of 8.0±0.2. Other components were not changed during experiment.

3.2.2 Seed sludge

Seed sludge with the concentration of 4 gSS/L was taken from a conventional activated sludge process.

3.2.3 Shell support media

Shell carrier was used as support media to cultivate aerobic granules. The support was made up of bivalve shell of rose cockle which was produced in AIT laboratory. First, the selected shells were dried to remove all organic constituents. Second, the dried shells were ground into powder form and 150-300 μm range of size was selected by sieving technique. At last, shell support was washed with tap water to remove impurities and then dried at 105°C for 24 h before use. In phase I a, 20 g/L of support was added into reactor. An additional 10 g was added every month to compensate the possible media lost due to sampling and washout. The support was only used in Phase I a. The shell carrier was a good support media for microbial adhesion and granulation due to porous structure and good settling ability. The physical characteristics of the support are presented in Table 3.2.

Table 3.1 Components of feed wastewater (mg/L)

Components	Phase Ia, II	Phase Ib
Organic	775 (glucose)	176 (glucose), 173 (C_2H_5COONa), 275 (CH_3COONa), 90 (Ethanol). Each component contributes 25% of COD.
$NaHCO_3$*	2640	50-100
NH_4Cl	1242	143
KH_2PO_4	50-100	50-100
$CaCl_2.2H_2O$	30	30
$MgSO_4.7H_2O$	12	12
$FeCl_3$	4	4 ($FeSO_4$)
Trace solution 1ml/L	H_3BO_3 0.15 g/L; $CoCl_2.6H_2O$ 0.15 g/L; $CuSO_2.5H_2O$ 0.03 g/L; $FeCl_3.6H_2O$ 1.5 g/L; $MnCl_2.2H_2O$ 0.12 g/L; KI 0.03 g/L; $Na_2Mo_4O_{24}.2H_2O$ 0.06 g/L; $ZnSO_4.7H_2O$ 0.12 g/L; (Wang et al., 2004).	$CuSO_4.5H_2O$ 0.03 g/L; $MnCl_2.2H_2O$ 0.12 g/L; $ZnSO_4.7H_2O$ 0.12 g/L
COD (mg/L)	700	700-1000

Note: The $NaHCO_3$ concentration was changed to control reactor pH in the range of 7.8-8.2

Table 3.2 Physical characteristics of shell support media

Characteristics	Value
Density	1.45 g/cm^3
Settling velocity	55-300 m/h
Colour	White
Size	0.15-0.30 mm
Components	Ca, Fe, Mg
Loss weight (550°C, 20 min)	2%

Figure 3.2 Cockle shell (a), shell support (b) and support morphology (c) (distance between two lines is 1 mm)

3.3 Experimental set-up and operating conditions

3.3.1 Overall experimental plan

This study includes two main phases (Phase Ia, Phase II) investigating fouling behavior and nitrogen removal of granulation MBR, namely the BG-MBR and the CG-MBR. An additional study on the effect of aeration rates on fouling ability of effluent of SBAR (Phase I b) was conducted at INSA, Toulouse, France. In this experiment, it was aimed to understand the characteristics of SBAR effluent from the conventional to granulated system and its effect on filterability.

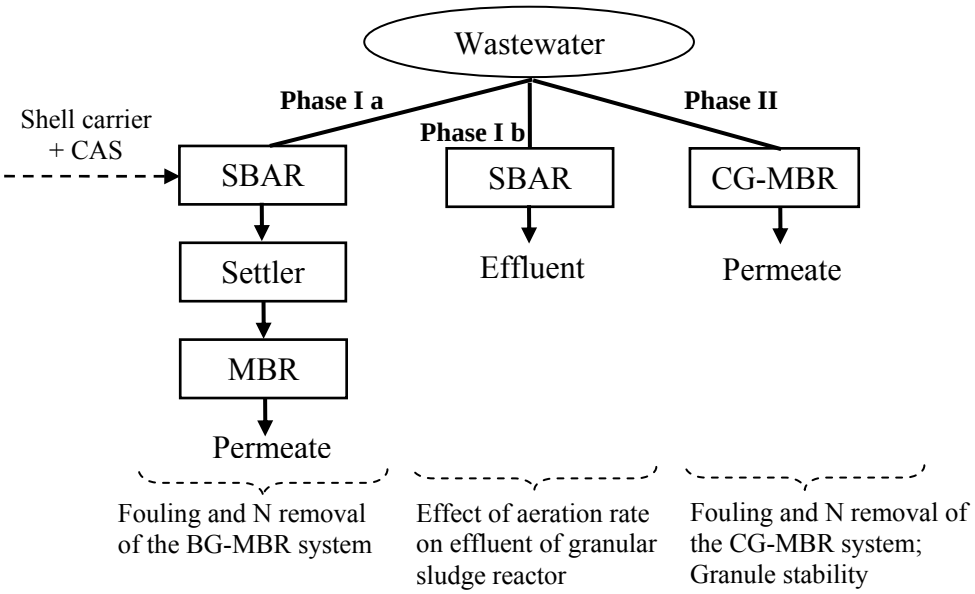


Figure 3.3 Overall experimental plan

3.3.2 Experimental set-up of BG-MBR (Phase I a)

Figure 3.4 describes the BG-MBR system including a SBAR (granulation reactor), a settler and an external submerged MBR. The second unit is a dual purpose tank which functions as both holding and settling tank (denoted as settler). The SBAR effluent was transferred into the settler which was fed into the MBR in continuous mode of operation. Settled sludge of 500 mL (twice a day with each time 250 mL) from the settler was removed daily. The final unit, the external submerged MBR was used for separation of liquid and solid fractions. The remaining substrate, unsettled colloids and biomass could be biologically degraded in the MBR which was especially operated in endogenous condition with very low incoming substrate. Air was just supplied at low flowrate of 1.2 L/min (0.1 cm/s) by a stone diffuser. All these systems were controlled automatically by programmable logic controller. This experimental set-up is referred to Appendix A, Figure A-1 to A-4.

Nitrogen removal and fouling ability of the BG-MBR was investigated through first four scenarios as presented in Table 3.3. In this phase, aerobic granule was cultured in SBAR by synthetic wastewater with organic and nitrogenous sources (OLR of 2 kg

COD/m^3.d, NLR of 1 kg N/m^3.d). The SBAR was operated at 1.0 kg N/m^3.d during the study to find the optimum scenario (day 1-136). After achieving the optimum scenario, system was operated at NLR of 0.6 kg N/m^3.d to avoid the excessive pH reduction due to nitrification (day 137-215).

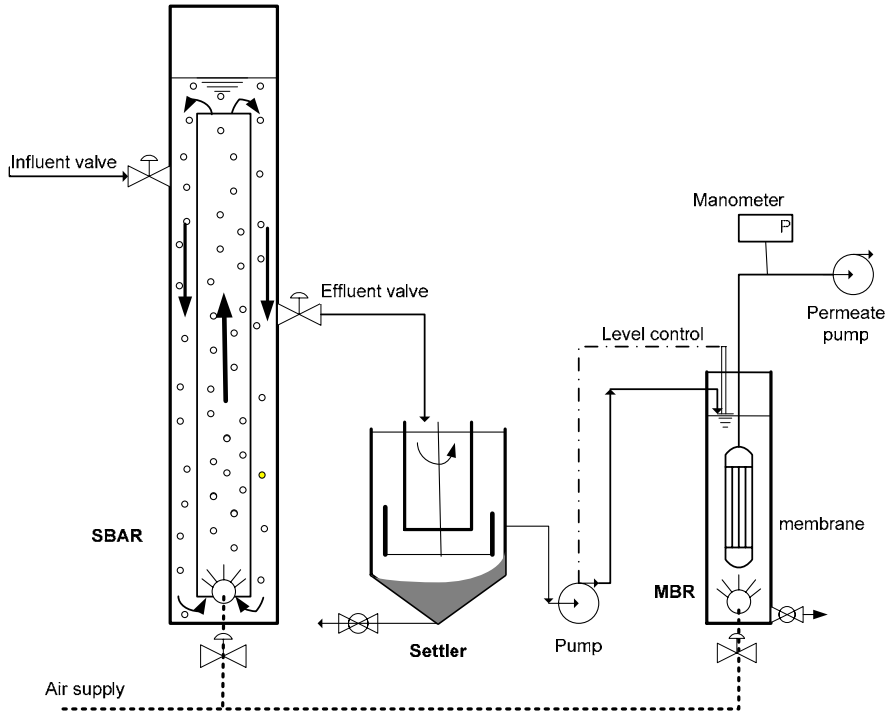


Figure 3.4 Batch granulation MBR (Phase I a – at AIT)

The SBAR was run with 6 cycles per day (4 h/cycle). Each cycle includes 4 stages (feeding, reaction, settling and withdrawal). All scenarios had similar number of cycles and stages. There was only stage 2 (reaction stage) which was different from others. Table 3.4 shows operating conditions of various scenarios. The reaction stage (stage 2) was changed between aerobic/anaerobic conditions. The anaerobic condition in the SBAR was achieved by the recirculation pumping from the top to the bottom of the reactor with the flowrate of 2.2 L/min. The screened liquid was pumped through the raiser tube where the granular sludge bed existed. In this manner, the nitrified liquid passed through the granule bed, thus denitrification process could happen effectively. Another measure of denitrification enhancement was an application of low aeration rate following high aeration rate. This solution could help save energy due to less aeration and enhance nitrogen removal. The low aeration rate created lower gradient of oxygen concentration in the SBAR, which was favorable for the denitrification inside the granule core. Low gradient of oxygen concentration could limit the diffusivity depth into the granule cores.

Table 3.3 Time duration of a batch of the SBAR

Stage	Feeding	Reaction*	Settling	Withdrawal
Duration	6 min	228 min	3 min	3 min

* The reaction stage (stage 2) is different for each scenario

Table 3.4 Operating condition of reaction stage of scenarios

Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5*
198 min aeration 30 min low aeration	40 min recirculation 90 min aeration 40 min reciculation 58 min aeration	48 min recirculation 180 min aeration	198 min aeration 30 min recirculation	180 min aeration 48 min low aeration
Day 1-71	Day 72-93	Day 94-112	Day 113-136	Day 137 – 215
Total duration of recirculation without aeration:				
0 min	80 min	48 min	30 min	0 min

* Scenario 5 was selected for further investigation on fouling behavior.

Table 3.5 Operating conditions of the BG-MBR

Reactor	SBAR	Settler	MBR
Size (cm x cm)	Down comer : LxD = 130x11.5 Raiser: LxD = 90x7	DxH = 20x35	DxH = 10x53
Working volume (L)	9.7	8	4
HRT (h)	7.3	6	3.4
SRT (d)	24*	NA	20
Aeration rate (cm/s)	Aeration: 1.7 ± 0.05 Low aeration: 0.1 ± 0.01	NA	0.3 ± 0.01
Sludge removal (mL/d)	None	500	200
Flowrate	5.3 L/batch, 4h/batch	NA	28 mL/min, 7on/3off
Flux ($L/m^2.h$)	NA	NA	2.8
Volume exchange ratio	55%	NA	NA

* Note: SRT of SBAR was calculated based on the ratio of sludge washed out over sludge in reactor

Table 3.6 Membrane module specifications

Membrane type	Submerged hollow fibre, Mitsubishi
Materials	PE
Size (DxL)	4.5 cm x 38 cm
Pore size	0.1 μm
Surface area	0.42 m^2
Outer diameter of a fibre	0.04 cm

3.3.3 Effect of aeration rate on fouling ability of SBAR effluent (Phase I b – INSA, Toulouse, France)

The effect of aeration rates and anoxic/aerobic conditions on characteristics of SBAR effluent was investigated for three aeration rates (0.8 cm/s, 2.2 cm/s and 0.6 cm/s with anoxic/aerobic stage) to understand the fouling ability of effluent from conventional activated sludge operation to granulation process. The characteristics of effluent such as MWCO, hydrophobicity and filterability were examined during the operation. This study was conducted in INSA laboratory. The reactor configuration and operating conditions are presented in Table 3.7 and Table 3.8.

A plate is positioned vertically in the middle of the SBAR for dividing the column into two zones namely raiser and down comer. The SBAR operation includes 4 batches per day with each batch consists of: filling without aeration during 30 minutes, aeration during 270 minutes, settling without aeration during 30 minutes and finally effluent withdrawal without aeration during 30 minutes. The reactor was operated at aeration rates of 0.8 cm/s (day 1-37), 2.2 cm/s (day 38-79) and 0.6 cm/s (day 80-174). During the last run from day 121 to 174, the cycle was modified by adding an anoxic mixing stage just after filling the reactor. Here, nitrogen gas instead of air was supplied for 30 minutes at the same gas

flowrate of 0.6 cm/s. This step aimed to enhance denitrification process by anoxic/aerobic condition. This SBAR configuration is presented in Appendix A, Figure A-9, A-10.

Table 3.7 Operating conditions of SBAR (Phase I b)

Parameters	Value
Diameter x Height (mm x mm)	150 x 1050 (H/D = 7)
Working volume (L)	17
Effluent (L/batch)	8
Volume exchange ratio (%)	47
TOC influent (mg C/L)	228 ± 72
COD influent (mg/L)	700 – 1000
COD/N ratio	20
Cycle (h/batch)	6
Settling time (min)	3 – 30
Air flowrate (L/h)	200 – 900
Operating temp (^oC)	20
OLR (kg COD/m^3.d)	1.7-2.0
TOC loading rate (kgTOC/m^3.d)	0.41-0.64
SRT (d)	Depends on sludge washout through effluent

Table 3.8 Experimental runs for SBAR

Aeration rate	Duration	Feeding	Aeration	Settling	Withdrawal
0.8 cm/s	day 1-38	30 min	270 min	30 min	30 min
2.2 cm/s	day 39-79	30 min	270 min	30 min	30 min
0.6 cm/s + anoxic stage (by introducing N_2 gas)	day 80 onward	30 min	30 min anoxic + 240 min aeration	30 min	30 min

3.3.4 Continuous Granulation MBR (CG-MBR) (Phase II – at AIT)

In this phase, it was aimed to study the granule stability, fouling and nitrogen removal of the continuous granulation MBR at three OLRs (2, 4 and 8 kg COD/m^3.d). The nitrogen loading rate was fixed at 0.6 kg N/m^3.d during this phase. The CG-MBR system was modified from the BG-MBR which was used in the Phase I a (Figure 3.5). The airlift reactor and membrane chamber were connected by two pipes to achieve convection movement at the two openings. Matured granules without shell media which were cultivated in SBAR were added into the CG-MBR system.

Table 3.9 Operating conditions of CG-MBR system (Phase II - AIT)

Parameters	Value
Working volume	13.5 L (airlift: 10 L, MBR chamber: 3.5 L)
Influent flowrate	29.2 L/d
Discharge flowrate in each 4 h	2.9 L/d
Total flowrate	32.1 L/d
Gross flux	2.9 (L/m^2.h)
Membrane cycle	7 min on / 3 min off
Air flowrate	60 m^3/m^2.h (airlift) and 9.2 m^3/m^2.h (MBR chamber)
HRT	10 h
SRT*	Depends on sludge discharge (~5.5 d)

* Note: SRT was calculated based on the amount of wasted sludge over the sludge in the CG-MBR

Airlift reactor was fed continuously by a signal from level control sensor installed in MBR and air is supplied at the bottom of airlift reactor with velocity of 1.7 cm/s to

create hydraulic shear force. To maintain granule formation in the CG-MBR, a certain amount of light fraction of biomass (suspended solids) was removed. Each 4 h, system was stopped to settle for 30 seconds then 440 mL of supernatant was removed by pump during one minute. The sludge removal was to create a gradient of organic concentration in the CG-MBR which is favorable for granulation process. The reactor volume and operating conditions of membrane modules were maintained as same as the BG-MBR system.

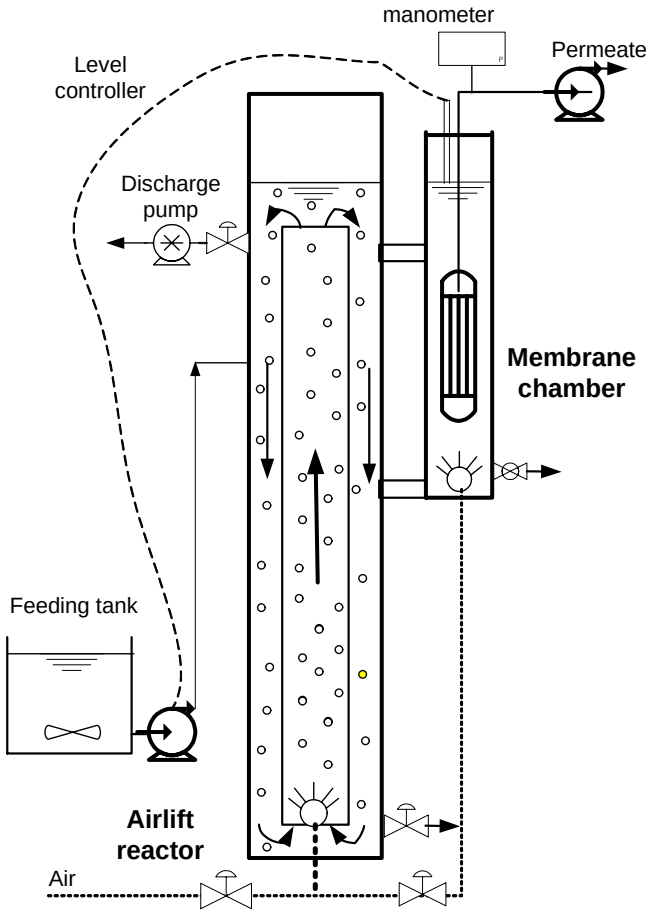


Figure 3.5 Continuous granulation MBR (CG-MBR) – Phase II

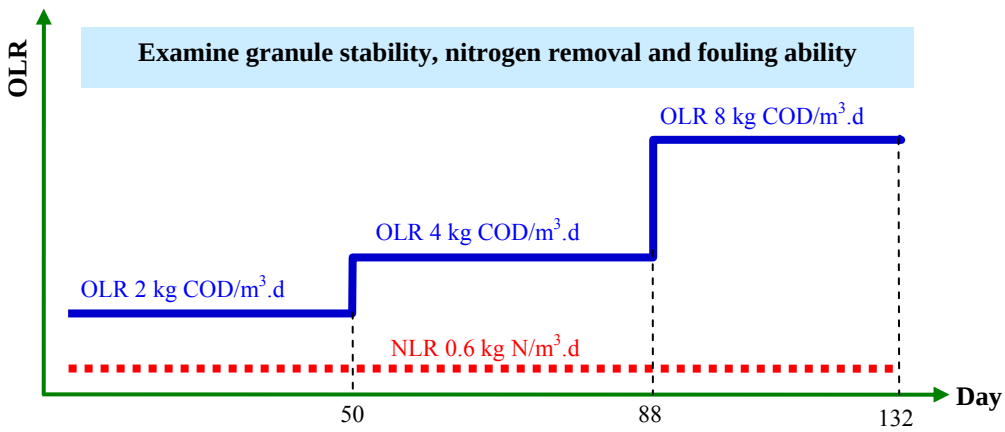


Figure 3.6 Operating conditions of CG-MBR (Phase II - AIT)

3.4 Analytical methods

3.4.1 Modified fouling index and fouling behavior of sludge fractions

Membrane fouling potential of sludge fractions, namely suspended solids (SS), colloids (CL) and solutes (SL) was quantified by measuring modified fouling index (MFI) and resistances of sludge fractions. The procedure is presented in Figure 3.7.

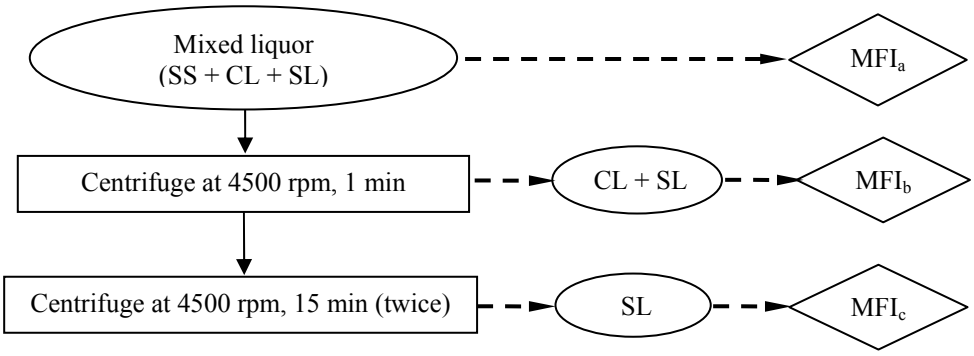


Figure 3.7 Methods of sludge fractionation

The MFI and cake resistance was measured by a stirred cell (AMICON 8400 USA, diameter 67 mm, area = 41.8 cm^2) with stirring speed 500 rpm and flat sheet membrane of pore size of 0.22 µm under a constant trans-membrane pressure (TMP) of one bar. The raw experimental data including accumulated permeate volume (V) and time (t) was used to plot t/V versus V graph to get the slope (s/L^2) which represents the MFI of the sample. The MFI is defined as the gradient of the linear region found in the well-known cake filtration equation (Boerlage et al., 2002). Cake resistance was then calculated as follows:

$$\frac{t}{V} = \frac{\mu.\alpha.C}{2.A^2.TMP}V + \frac{\mu.R_m}{A.TMP} \qquad \text{(Eq. 3.1)}$$

Fouling index I ($1/m^2$) is related to specific cake resistance α (m/kg) and cake mass C (kg/m^3):

$$I = \alpha.C \qquad \text{(Eq. 3.2)}$$

Resistance of each fraction was calculated as follows:

$$R_t = R_m + R_f + R_c \qquad \text{(Eq. 3.3)}$$

Whereas,

R_t, R_m, R_f, R_c are total, membrane, fouling and cake resistance (1/m), respectively.

3.4.2 Extraction and measurement of EPS in sludge and fouled membrane

The PN and PS were analyzed by methods of Lowry et al. (1959) and Dubois et al. (1951) respectively (EPS = PS + PN). The bound EPS (bEPS) of granular sludge, MBR sludge and fouling layer sample were extracted using the cation exchange resin technique (Dowex HCR-S/S, 16-50 mesh, sodium form, Dow Chemical Company) according to Frølund et al. (1996). For granular sludge sample, it was ground by the Ultra-Turrax equipment for one minute before carrying out resin extraction. The buffer solution was prepared with the concentrations (Na_3PO_4 2 mM, NaH_2PO_4 4 mM, NaCl 9 mM and KCl 1mM). The EPS extraction process is shown in Figure 3.8.

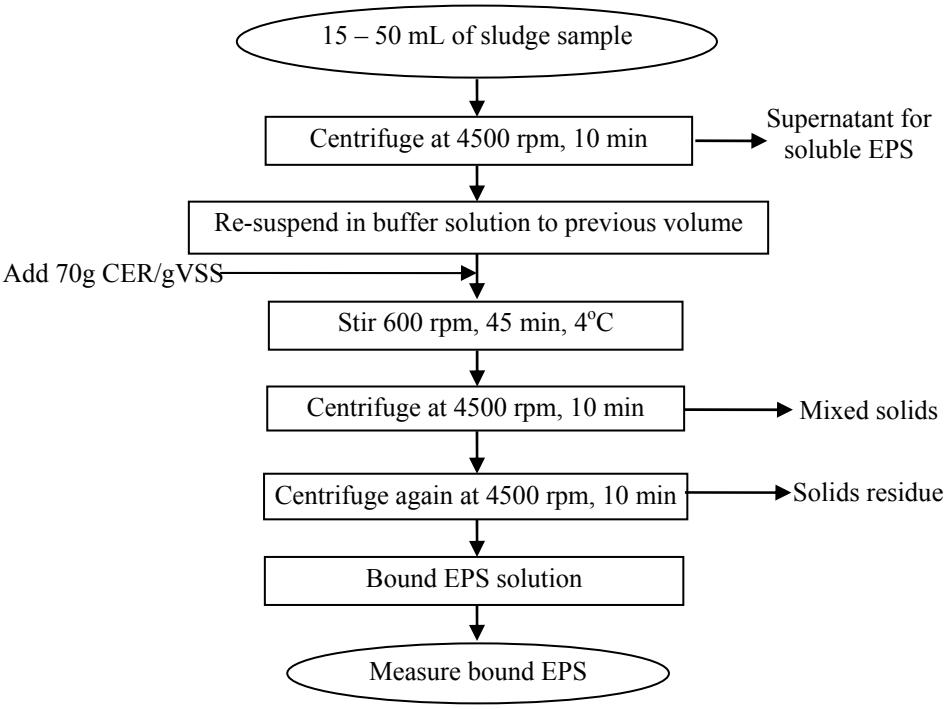


Figure 3.8 Extraction of EPS from sludge and fouling layer

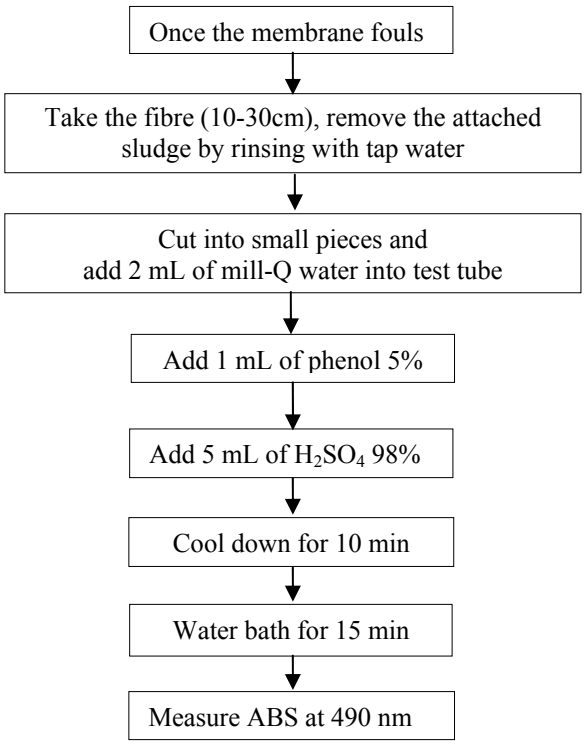


Figure 3.9 Extraction of EPS in fouled membrane

The amount of EPS deposition on membrane was quantified with the similar method adopted by Cho and Fane (2002); Kim and DiGiano (2006). Two typical fibre lengths (about 10-30 cm) were cut off from the fouled membrane and washed with tap

water until the membrane fibre becomes white/clean like initial (clean) membrane (removal of entire fouling layer attached on the membrane). The fibres were cut into small segments and immerged into test tube containing 2 mL milli-Q water. After that the color reagent (1 mL of 5% phenol and 5 mL of concentrated H_2SO_4) was added into the test tube which is similar to the measurement procedure of Dubois. The EPS deposition on membrane was measured at wavelength of 490 nm and converted to the unit of μg EPS/cm^2 of fibre.

3.4.3 Behavior of MBR supernatant test

The test was conducted to understand the behavior of characteristics of MBR supernatant which was operated under endogenous condition. The operating conditions of a separate reactor were simulated similar to that of MBR of the BG-MBR system (MLVSS of 1 g/L, working volume of 4 L, air flowrate of 0.3 cm/s). The settled sludge from effluent of SBAR and sludge taken from the operating MBR (MLVSS ratio 3:1) were mixed in the separate reactor. Parameters such as DO, pH, VSS, soluble EPS, DOC and TN were measured at the interval of one hour for total 10 h duration. The rates of consumption and release of each soluble species (sEPS, sTN and DOC) were calculated.

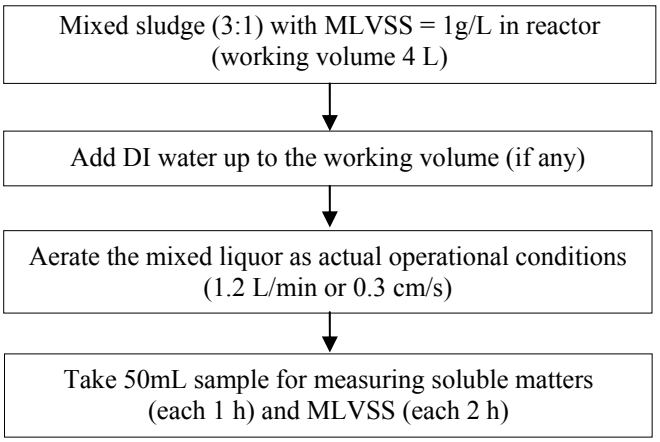


Figure 3.10 Procedure of MBR supernatant behavior test

3.4.4 Membrane resistances

The fouled membrane was taken out of the reactor for cleaning when it fouled (15-20 kPa). Membrane resistances were measured by using the resistance-in-series model (Choo and Lee, 1996) according to equation 3.4. Membrane resistances were measured by filtrating with distilled water at different fluxes and corresponding TMP were recorded. Membrane resistances were derived from the slope of the linear curve of TMP versus flux from the equation. Membrane resistance measurement procedure is as follows:

- Take out the membrane from the reactor;
- Measure total membrane resistance (R_t);
- Wash the membrane by spraying tap water;
- Dip membrane in distilled water container (2 L) and gently shake it for 10 minutes to remove the attached cake layer;
- Measure membrane resistance ($R_f + R_m$);
- Clean membrane by chemicals (NaOH 4%, chlorine 3000 mg/L) for 6-24 h;

- Measure membrane resistance (R_m);
- Resistance of cleaned membrane (R_m) should be close to the initial resistance (recovery higher than 90%). If any, the cleaning time by chemical should be longer.

$$J = \frac{\Delta P}{\mu * R_t} \Rightarrow \Delta P = J * \mu * R_t \qquad \text{(Eq. 3.4)}$$

Where:

J: Permeate flux ($L/m^2.h$)

ΔP: TMP (kPa)

μ: Viscosity of the permeate (Pa.s)

R_t: Total resistance (1/m) ($R_t = R_m + R_c + R_f$)

R_m: Intrinsic membrane resistance

R_c: Cake resistance caused by the cake layer

R_f: Fouling resistance caused by adsorption of colloids/solutes into the membrane pores.

3.4.5 Other parameters

Other measurement parameters are presented in Table 3.10. Further detailed information is referred to Appendix B; Appendix D, Table D-4 and Figure D-6.

Table 3.10 Measured parameters

Parameters/unit	Method/equipment	Interference and measurement range	References
TOC, DOC, TN (mg/L)	Total organic carbon analyser (TOC-V_{CSN}, Shimadzu, Japan)	TOC = TC - IC TC: 0-500 mg/L; IC: 0-200 mg/L	-
NH_4^+-N (mg/L)	Distiller	-	APHA, 1998
NO_2^--N	Spectrophotometer U2001, Hitachi, Japan	Range: 0-25 µg/L	APHA, 1998
NO_3^--N (mg/L)	Spectrophotometer U2001, Hitachi, Japan	Range: 0-5 mg/L	APHA, 1998
DO (mg/L), pH	DO and pH meter	-	APHA, 1998
SVI (mL/g)	-	-	APHA, 1998
Viscosity	Viscometer DV II+	-	-
UVA_{254} (1/cm)	Spectrophotometer U2001, Hitachi, Japan	Nitrite	Her at al., 2007
Bound EPS extraction	Cation exchange resin	-	Frølund et al, 1996
Polysaccharides (mg/L)	Spectrophotometer U2001, Hitachi, Japan	Nitrite; Range: 0-80 mg/L Add sulfamic acid:NO_2-N =15 (6-12 h prior to measurement);	Dubois et al., 1951
Protein (mg/L)	Spectrophotometer U2001, Hitachi, Japan	Range : 0-60 mg/L	Lowry et al, 1959
CST	Capillary suction method	Triton Electronics Ltd., England	APHA, 1998
Granule/sludge morphology	Microscope Olympus BH2-RFCA; Digital camera	-	-
Granule settling velocity	Free settling test	-	Etterer and Wilderer, 2001
$SOUR_H$, $SOUR_{NH3}$, $SOUR_{NO2}$	Batch respirometer	-	Cech et al., 1984; Liu et al., 2004
MFI (s/L^2)	Stirred cell (Amicon 8400 USA) with plate membrane 0.22 µm, 1 bar, 500 rpm	-	Boerlage et al., 2002
Membrane resistance (1/m), resistance rate	Resistance-in-series model	-	Choo and Lee, 1996
Critical flux analysis ($L/m^2.h$)	Flux step method	-	Le Clech et al., 2003
Particle size distribution (µm)	Mastersizer S, Malvern UK	Range: 0.05-900 µm	-
Nano size (nm)	Zetasizer, nano ZS	Range: 0.6-6,000 nm	
MLSS (mg/L)	Gravitational method	-	APHA, 1998
MLVSS (mg/L)	Gravitational method, TOC method	-	APHA 1998; Tijhuis et al, 1994; & Beun et al., 2002
Trans-membrane pressure (kPa)	Digital pressure gauge, PG30, Japan	0-100 kPa (negative pressure)	Monitoring TMP change
Molecular weight (kDa)	HPLC, Akta purifier (UV210, UV254, UV280) equipped with fluorescence excitation–emission matrix (EEM) detector.	Size exclusion column superpose 6 (1-40,000 kDa)	-
Hydrophobic interaction chromatography (HIC)	HPLC (Akta); Column HiTrap Octyl 1mL, Amersham, Sweden	-	Lienqueo et al., 2003

Chapter 4

Results and Discussions

4.1 Optimum scenario of BG-MBR

4.1.1 Organic matter and nitrogen removal at different scenarios

This set of experimental runs was conducted to select the best operational scenario for the aerobic granulation coupled membrane filtration system. The operation criteria were mainly based on fouling tendency and high organic/nitrogen removal rate. The system was operated at OLR of 2 kgCOD/m^3.d and NLR of 1 kgN/m^3.d.

Feed synthetic wastewater was maintained at pH 8.0±0.3. The pH of SBAR and MBR was in the range of 8.0±0.2. This pH range was favorable for SND and nitrification to take place in the SBAR and MBR respectively. In this experiment, nitrogen removal capacity and fouling behavior were investigated by changing the aeration rate and aerobic/anoxic conditions in reaction stages of SBAR cycle. The anaerobic condition was created by the recirculation flow (2.2 L/min) from the top to the bottom of the SBAR. The low aeration rate in the SBAR was introduced through an air diffuser with lower flowrate as described in Table 3.3 and Table 3.4. In addition, lower aeration rate at the end of the batch leads to significant reduction in aeration energy cost.

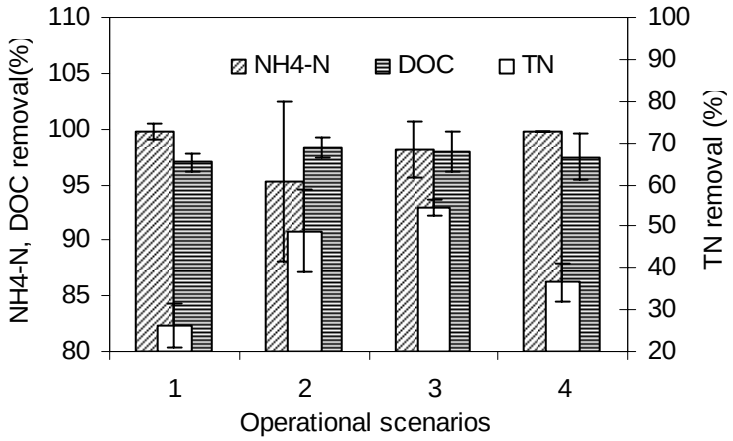


Figure 4.1 Organic and nitrogen removal in SBAR at various scenarios

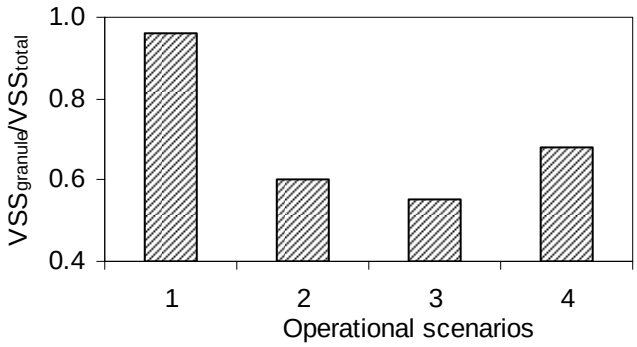


Figure 4.2 Ratio of granulated biomass to total biomass in SBAR at various scenarios

Figure 4.1 shows organic and nitrogen removal performance of different treatment scenarios, as presented in Table 3.4. Organic matter removal is not limited for the aerobic granular sludge system at the range of operating conditions tested. The ammonia removal efficiency is slightly lower in scenario 2 and 3 compared to others but the TN removal is inverse. The lower ammonia removal in the scenarios is because of shorter aerating duration which is necessary for nitrification process. By contrast, the TN removal is higher in scenario 2 and 3 as anaerobic condition is maintained for a longer period (Appendix C, Table C-1, C-2).

Moreover, in the scenario 2 and 3 the ratio between granulated biomass and total biomass ratio is less than 0.6 indicating that shell granules were disintegrated (Figure 4.2). The anaerobic condition caused granule disintegration because of the low shear stress and high free ammonia (FA) concentration in the reactor. It was recognized that the behavior of aerobic granules in this study was somehow similar to activated sludge which deflocculated under anaerobic condition (Wilen et al., 2000). Further, less shear stress was due to less aeration intensity in the reactor. Shear stress is a prerequisite for granule formation (Tay et al., 2001b). Furthermore, FA was found to be an inhibitor of granule formation at concentration of 23.5 mg/L (Yang et al., 2004). In scenario 2 and 3, longer duration of anaerobic condition led to higher amount of FA and slower nitrification. Basically, the reactor needs continuous aeration of 3 hours to oxidize ammonia completely. Hence, the nitrification was slower in these scenarios. The FA in the reactor was estimated according the Eq. 4.1 given below as described by Yang et al. (2004). The FA concentration in reactor varied from 6 to 16 mg/L at pH 7.8-8.2 and 340 mg/L of ammonia. Even though the FA concentration did not reach the inhibition level for granule formation yet, it could partially affect the granules and caused disintegration. However less granule breakage was noted in conditions of scenario 4 as compared to those of scenario 2 and scenario 3 because of less contact time under anaerobic condition. Further, shear stress was sufficient enough (192 min) to somehow sustain the granules. In general, it can be concluded that anaerobic conditions with high free ammonia and low shear stress are the influencing factors for the granule disintegration (Refer to Table 3.4).

$$FA(mg/L) = \frac{[NH_4 - N] \times 10^{pH}}{\exp(\frac{6334}{273+T}) + 10^{pH}} \qquad \text{(Eq. 4.1)}$$

4.1.2 Fouling propensity of different scenarios

Figure 4.3 presents the fouling rate in MBR under different operational conditions. The fouling rate was derived from the linear slope of trans-membrane pressure profile (Appendix C, Figure C-1), which indicates the membrane fouling propensity. The fouling rates were much higher for the scenario 2 and scenario 3 compared to scenario 1 and scenario 4. In other words, the anaerobic condition resulted in rapid fouling and granule breakage. Kang et al. (2003) reported that the mean particle size of a submerged MBR reduced from 30 μm (DO = 7 mg/L) to 13 μm (DO = 0.3 mg/L), resulting in an increased specific cake resistance. Jin et al (2006) also reported that critical filtration time (t_{cri}) was 7.5 times earlier when operating at a DO equal to 0.1 mg/L compared to 3.0 mg/L. It was due to an increase in concentration of fine particles ranging from 2 to 5 μm. Concurrently, low DO concentration also produced more SMPs which was considered to be membrane foulants (Kang et al., 2003). All these previous findings support the present study.

Furthermore, F/M ratio was high with the scenarios having low granulated biomass to total biomass ratio (Figure 4.4). At the same OLR, the biomass concentration in the SBAR was decreased due to granule breakage and washed out with effluent, thus made the F/M ratio increased. At the scenario 4, the F/M increases further because at that operating condition, shell granule stopped disintegration and started recovery. Biomass started accumulation in the granulation reactor but the $VSS_{granule}/VSS_{total}$ was still high. The increase in F/M ratio might contribute to the increment of the fouling rate. In addition, the reduction of bound PS/PN ratio due to granule disintegration (release into bulk liquid) could induce fouling (Appendix C, Table C-3) because polysaccharides have been identified to be the linking factor of cell in aerobic aggregate (Tay et al., 2001).

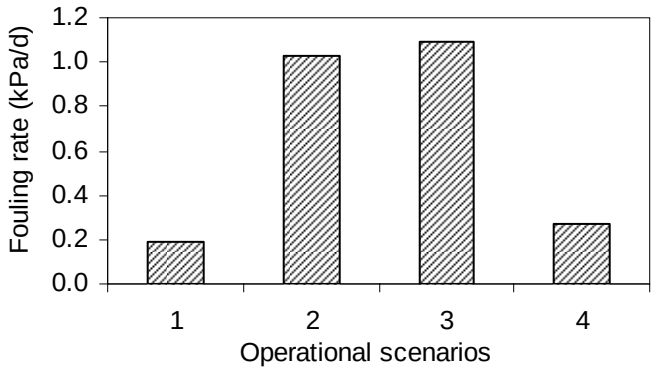


Figure 4.3 Fouling rates of MBR at different scenarios

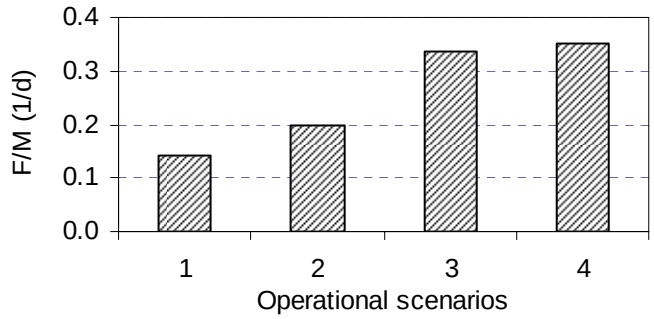


Figure 4.4 F/M ratios at different scenarios

Finally, the best coupling condition of the BG-MBR system is to maintain high aeration at the initial stage and low aeration at the end of SBAR cycle. This operating mode makes the aerobic granule stable and produces the effluent with less fouling propensity. Therefore, operating conditions of scenario 1 is selected to be the best coupling condition for the BG-MBR.

The next section studies the details of fouling behavior and nitrogen removal of the BG-MBR system with the operating conditions similar to scenario 1. The scenario was denoted as scenario 5 in which the NLR was reduced to 0.6 $kgN/m^3.d$ to avoid extreme pH fluctuation in the SBAR.

4.2 Fouling behavior and nitrogen removal of BG-MBR

4.2.1 Granule characteristics and treatment performance of BG-MBR

4.2.1.1 Dissolved oxygen and pH profile in SBAR

As shown in Figure 4.5, pH of SBAR started decreasing after 60 minutes of operation. This happened because nitrification process took place and consumed alkalinity. In the next batch, pH increased again because the new feeding wastewater was introduced. Dissolved oxygen (DO) was 7.1±0.8 mg/L for high aeration stage (1.7 cm/s). Then, it was reduced to 4.0-5.0 mg/L for the low aeration stage (0.1 cm/s). The DO of bulk liquid was high but it was low in the core of shell granule due to the limitation of oxygen transfer. This phenomenon could allow the denitrification process to occur in the core of granule simultaneously. Further, the anaerobic condition occurred in the core of granule at the depth of 300 μm from the granule surface (Tijhuis et al., 1994). In the study under discussion, average granule size was approximately 4.7 mm whose radius was almost 8 times longer than the diffusion depth. Therefore, the anaerobic condition definitely existed in the granule core. In other words, the spherical structure of granule allowed the SND to happen even under fully aerobic operating condition in the granulation reactor.

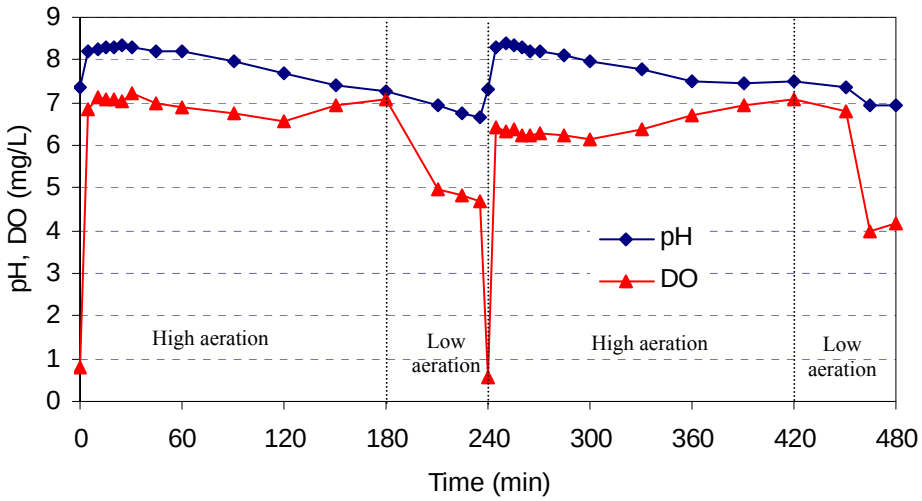


Figure 4.5 Typical pH and DO profile of SBAR during two consecutive batches

4.2.1.2 Organic and nitrogen removal of SBAR

Figure 4.6 shows that organic matter in term of TOC was significantly removed at the early stage of aeration within 30 minutes. The DO concentration was saturated at about 6 mg/L during first 3 h and then it gradually reduced to 4 mg/L during the next 48 minutes due to the application of low aeration rate.

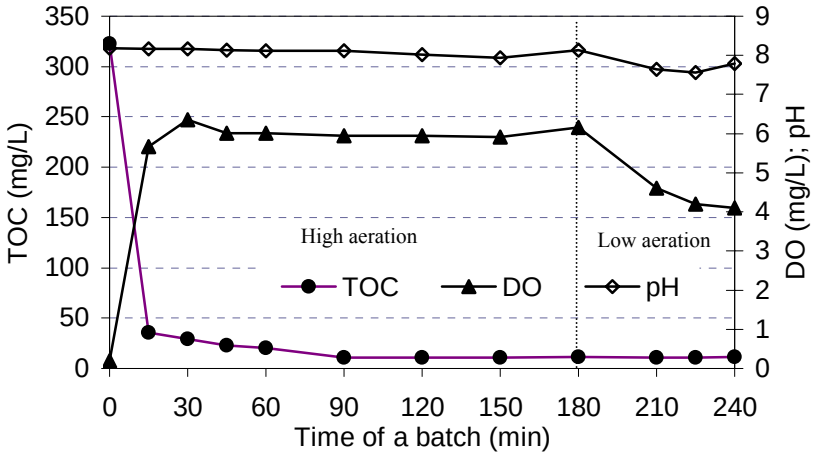


Figure 4.6 Typical TOC, DO and pH profile of a SBAR batch

Figure 4.7 presents that the nitrification process was complete during the first three hours. Nitrate concentration was not significant during a batch. Nitrite concentration increased according to the time which was from the partial nitrification process. Total nitrogen did not fluctuate during the last 3.5 hours. It shows that the SND reached maximum efficiency during first 30 minutes which organic substrate was available in the bulk liquid. The nitrite concentration was dominant in the SBAR effluent. It could be explained that at the high nitrogen loading rate, the nitrite-oxidizing bacteria was inhibited due to the high toxic concentration of nitrite generated, thus caused inhibition to nitrate formation. Tsunenda et al. (2003) observed the similar results that the nitrite-oxidizing bacteria exhibited minor population in the structure of granule. Majority of microorganisms existed in the granule surface (200 μm from the granule surface). They included heterotrophs, ammonia-oxidizing bacteria and nitrite-oxidizing bacteria while nitrite-oxidizing bacteria appeared a minor population compared to ammonia-oxidizing bacteria and heterotrophs. This observation could be explained for the absence of nitrate nitrogen in the SBAR effluent in this study. As a conclusion, the simultaneous organic and nitrogenous removal indicated that there were co-existence of heterotrophs, nitrificants and denificants in the granulation reactor. The nitrogen removal occurred in the single aerobic granular sludge SBAR that promises its widespread application in the future because the complicated anaerobic/aerobic system could be integrated in the aerobic granulation reactor.

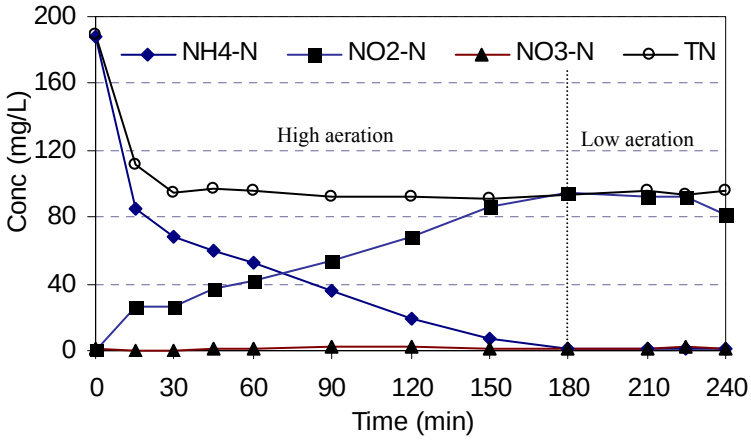


Figure 4.7 Nitrogen species profile of SBAR in a batch

4.2.1.3 Nitrogen balance in SBAR

To balance the total nitrogen (TN) of SBAR, the data set of nitrogen species was used for mass balance (Appendix C, Table C-4, C-5). The TN for cell yield was inferred from the theoretical ratio (COD/N = 30).

$$TN = \text{ammonia} + \text{nitrite} + \text{nitrate}$$

$$TN_{inf} = TN_{eff} + TN_{removal} = TN_{eff} + TN_{assimilation} + TN_{denitrification}$$

Where,

TN_{inf} and TN_{eff} are total nitrogen in the influent and effluent respectively.

Figure 4.8 shows nitrogen balance in the SBAR. The nitrogen removal due to SND process in the SBAR was 47% or the denitrification rate was 22.2 mgN/L.h (1.76 mgN/gVSS.h).

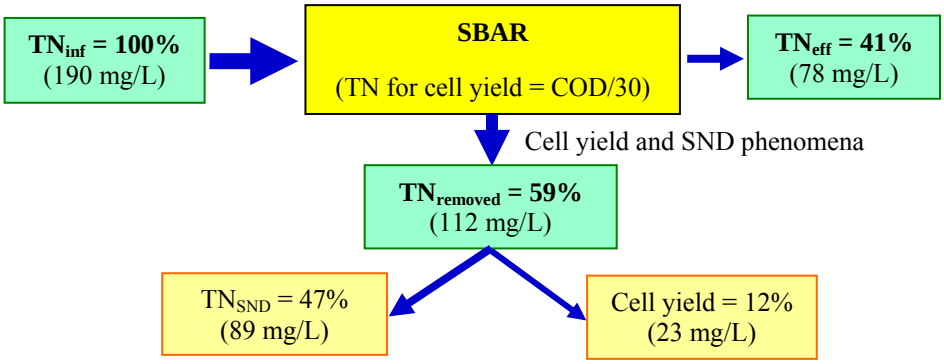


Figure 4.8 Nitrogen balance in granulation reactor (SBAR)

4.2.1.4 Treatment performance of the BG-MBR

Table 4.1 briefly shows the treatment performance of the BG-MBR system. Figure 4.9 shows that most of the organic substrate in terms of TOC (> 97%) was removed in the SBAR.

Most of the suspended solids from SBAR effluent were retained in settler (61-90%). The removal efficiency of SS depends on the settling time, loading rate, feeding wastewater characteristics and operating conditions of the SBAR. At the OLR of 2 kgCOD/m^3.d, the VSS concentration in the SBAR effluent and the settler were 239±42 and 35±15 mg/L respectively. The remaining soluble matters and SS were further aerobically treated in MBR. In MBR, the nitrites were converted to nitrates while TN almost remained constant (Figure 4.10). The SBAR effluent was rich in nitrite (75±18 mg/L). Thus, the role of MBR in the BG-MBR system was filtration and partial nitrification.

Table 4.1 Treatment performance of the BG-MBR system

Parameters	SBAR	MBR
pH	7.97 (±0.22)	8.07 (±0.24)
TOC removal (%)	97.7 (±1.4)	51.7*
Ammonia removal (%)	99.2 (±0.3)	60.3*
TN removal (%)	57 (±10)	2*

SVI (mL/g)	25 (±5)	93 (±26)
Viscosity at 180 rpm (cP)	-	1.1
CST (s)	10 (±1)	14 (±4)
F/M (d^{-1})	0.18 (±0.05)	-
TOC (kgTOC/ m^3.d)	0.86 (±0.22)	-
OLR (kgCOD/m^3.d)	2 (±0.2)	-
NLR (kgNH_4^+-N/m^3.d)	0.6 (±0.1)	-

* As compared with influent ammonia/TOC (from settler)

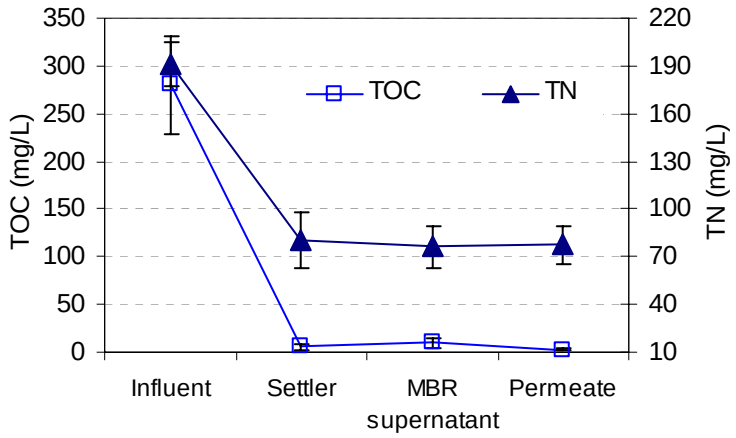


Figure 4.9 Organic and nitrogen removal in the BG-MBR

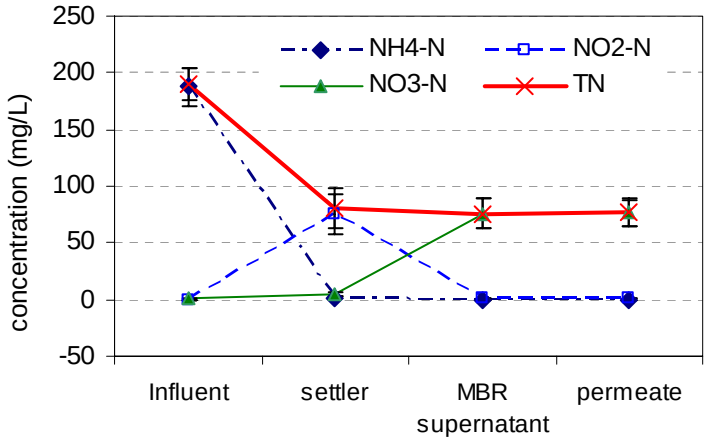


Figure 4.10 Nitrogen species in the BG-MBR

4.2.1.5 Biomass concentration in SBAR and MBR

Figure 4.11 shows that from day 40 to 100, a biomass reduction occurred which was caused by the anaerobic conditions of scenarios 2 and scenario 3. The operation of these scenarios made the granules disintegrated, thus excessive amount of biomass was washout. The maximum biomass concentration of the SBAR was approximately 18 g/L. It reached maximum when the level of settled granule (at the end of settling stage) was almost equal to the effluent discharge valve of the SBAR. At that point, it was the ideal steady state of the granulation reactor. At that moment, a part of granule was washed out together with suspended solids. Moreover, the phenomena only happened in the scenario 1 and scenario 5 in which granules were under the steady conditions. Additionally, the biomass concentration of granulation reactor was higher than that of conventional SBR (2–

6 g/L) which made it able to operate at high loading rates and to resist shock loading (Appendix C, Table C-6).

There was no manual sludge withdrawal in the SBAR. The SRT was automatically controlled by the natural washed-out biomass of the reactor which was calculated based on the sludge in reactor and the daily sludge discharge with effluent. It was about 24 days for scenario 1 (stable operation). However, the actual SRT of granulation reactor was much higher than the calculated value because the washed-out sludge was in fact the new sludge (from biological assimilation). The old granules still retained in reactor until they were disintegrated into flocs and debris. Granules were retained because they had higher settling velocity and density than the newly generated biomass. Therefore, all of the slow-growing bacteria could exist in the granulation reactor which could perform well the SND process and recalcitrant degradation.

In MBR, the biomass concentration was stable in the range of 1600 – 3200 mg/L at the 20 d SRT. The biomass growth in MBR was originated from the unsettled SS, organic residue (from the settler supernatant) and yield from partial nitrification.

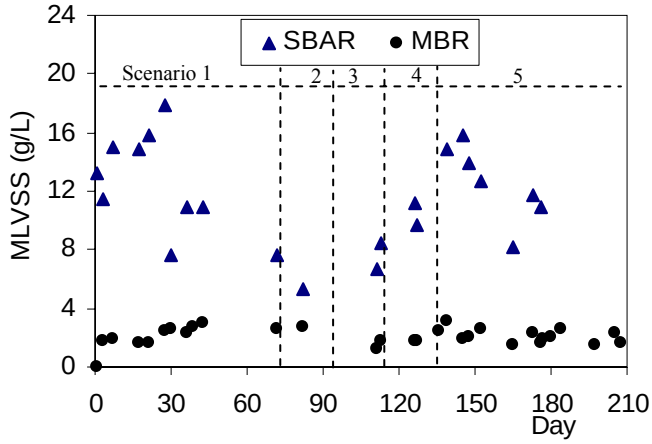


Figure 4.11 MLVSS of SBAR and MBR with time course

4.2.1.6 Size and settling velocity of granules

The average granule size increased gradually with time. Figure 4.12 shows that the average granule size was 4.9 ± 1.0 mm. The settling velocity was bimodal distribution which was due to the difference between shell and non-shell granules (sample size of 400 granules). The settling velocity of shell granules was always greater than 180 m/h while that of non-shell granule was lower. The average settling velocity was 260 ± 124 m/h which was much higher than that of activated sludge (1-2 m/h). This reveals the advantage of shell media in aerobic granule formation.

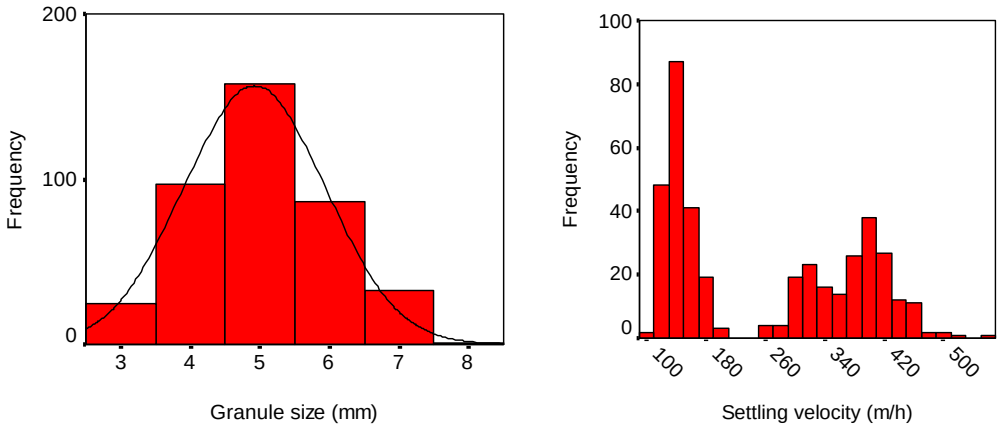


Figure 4.12 Distribution of size and settling velocity of granules in the SBAR

4.2.1.7 Organic and nitrogenous removal of settler

The settler worked as a storage tank in the BG-MBR system. However denitrification process occurred due to anoxic conditions during storage. The settler supernatant was monitored for 4 h (equal to a SBAR cycle) to observe the evolution of substrate concentrations. pH within the settler was about 8 during the whole period. Figure 4.13 shows that soluble PS and DOC did not change significantly. However, TN and DO reduced with time. The DO concentration decreased from 2.5 to 0.2 mg/L which favored the denitrification of nitrite and nitrate present in the settler. The TN removal rate in settler was 4 mg N/L.h with the removal efficiency of 19% after 4 h. The electron donor for the process was taken from the organic residue or/and soluble PS. Furthermore, soluble protein was negligible during this experiment.

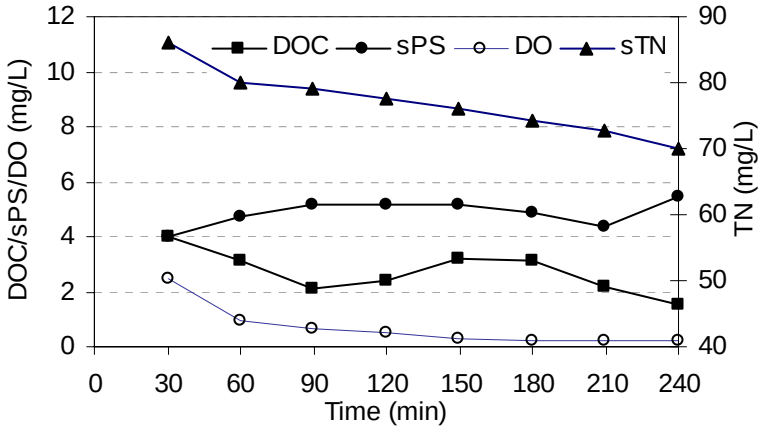


Figure 4.13 Typical substrate removal profile of the settler

4.2.2 Fouling behavior of the BG-MBR

4.2.2.1 Behavior of MBR supernatant test

MBR was operated under endogenous condition with low biomass concentration and low incoming substrates, thus there was a possibility for cell lysis in the reactor. The test was conducted to understand the fate of substrates and biomass in the MBR. Reactor pH varied from 7.8-7.9 during the experimental duration with DO of 6.1 mg/L. The MLVSS was gradually reduced (2% reduction) during 10 h of aeration, presumably as the net loss due to the cell lysis and synthesis processes instantly. DOC which was the result of organic matters of soluble polysaccharides (sPS), soluble protein (sPN) and other organic byproducts reduced during first 3 h while the concentrations of sPS and soluble total nitrogen (sTN) increased with time. The variation in sPN concentration was insignificant and the sPS was always much higher compared to sPN during this experiment (sPS = 86-100% sEPS) (Figure 4.14, Appendix C, Table C-7). The reduction and release rate of soluble matters are presented in Table 4.2.

Table 4.2 Release and reduction rate of soluble matters in the cell lysis test

Parameters	Slope	Unit	R^2	Calculated duration
sTN	0.59	Mg sTN/gVSS.h	0.83	10 h
sPS	0.33	Mg sPS/gVSS.h	0.75	10 h
DOC	-0.95	Mg DOC/gVSS.h	0.68	3 h

Note: DOC was reduced during first 3 h

The reduction of volatile biomass plus the production of sTN, sPS and sPN were probably the result of lysis and/or deflocculation of cells which was caused by the famine condition in MBR. The reduction of DOC and sPN was due to the biodegradation and assimilation process while the biochemical reactions like conventional activated sludge process also existed. The bound protein (bPN) had been reported much higher than bound polysaccharides (bPS) in submerged MBR sludge with the bPS/bPN ratio of 0.25-0.50 (Massé et al., 2006) and 0.33 (Le-Clech et al., 2006) while in this study it the bPS/bPN ratio was as high as 0.7. Thus, if bound EPS (bEPS) are released from cells, sPN is probably higher than sPS in the bulk liquid. However, protein compounds are easily degradable that can be used for cell assimilation process. Massé et al. (2006) noticed that protein compounds were more easily degradable than polysaccharides. The insignificant value of sPN concentration in the bulk liquid was due to its consumption at a rate faster than the rate of production. In addition, the sPN could be broken down into smaller molecules easily, thus release of total nitrogen into bulk liquid was increased. Again, the increase of TN and sPS could be the evidence for the release of soluble microbial products (SMPs) from the biomass. However, the sPS was not readily degradable as sPN, thus it accumulated with time within the reactor. The products of cell lysis are usually refractory which include cell wall (peptidoglycan) and cell membrane (lipopolysaccharides) (Le-Clech et al., 2006).

In addition, the deflocculation phenomena occurred in the reactor due to its operating conditions. This caused the production of SMPs due to the release of bridging polymers which were the components of flocs structure (Wisniewski and Grasmick, 1998). The deflocculation makes the particle size smaller which results in the increase of smaller sludge particles in MBR compared to settler as shown in Figure 4.15a. Based on these results, it can be concluded that the range of HRT of 2-5 h is suitable for treating the granulation effluent. At this range, the membrane permeate can have better quality (low DOC and sTN), possibly less fouling propensity (low sPS, sPN, TN, DOC) and aeration cost.

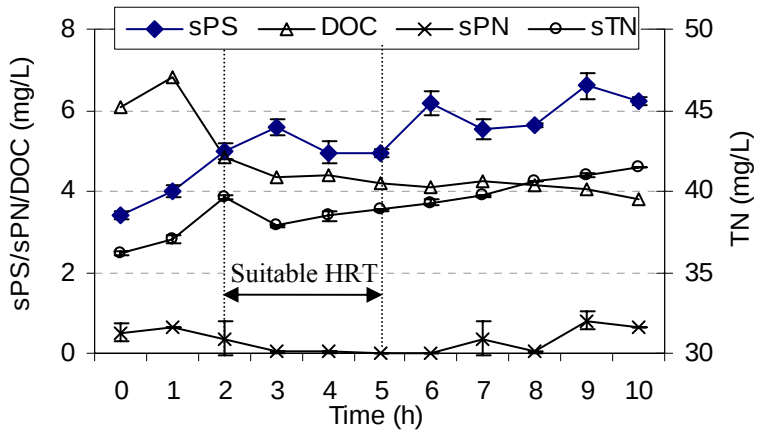


Figure 4.14 Behavior of MBR supernatant under endogenous condition

4.2.2.3 Particle size distribution in MBR

There are two modes of measurement for particle size distribution (PSD), namely volume distribution and number distribution. In terms of volume distribution, the particle size of settler and MBR mixed liquor was 98 μm and 158 μm respectively. However in

terms of number distribution it was 0.53 μm and 0.20 μm (Figure 4.15a, Appendix C, Table C-8). By the light scattering technique, the volume distribution did not provide the representative size of majority of particles because there was a large distribution range in the sludge samples. The volume of all small particles made only a little volume percentage. Therefore, the number distribution mode could reflect the actual size of the measured samples more accurately. The colloidal size measurement confirmed that the nanosize of MBR was 262 nm (0.26 μm) which was almost similar to the result achieved for mixed sludge sample (0.20 μm) (Figure 4.15b). For the colloidal size measurement, the number and volume distribution were rather identical because the centrifugation step had removed all the large particles and made the two distribution curves narrow and comparable.

MBR sludge showed wider distribution and smaller size than settler sample (number distribution). Again, this indicates that the sludge flocs/particles were degraded and/or deflocculated in MBR due to endogenous respiration in MBR. The shear stress of aeration, again, could break the linkage of flocs structure and generate smaller particles, debris and SMPs as stated above. The destruction was certainly due to erosion strengths or ruptures of the network of polysaccharides fibrils which was the support of the different compounds and particularly of the cells. Wisniewski and Grasmick (1998) found a decrease in the settleable fraction of flocs and consequently, observed an increase in the non-settleable fraction that is in line with the results of our present research in which the particle size reduced.

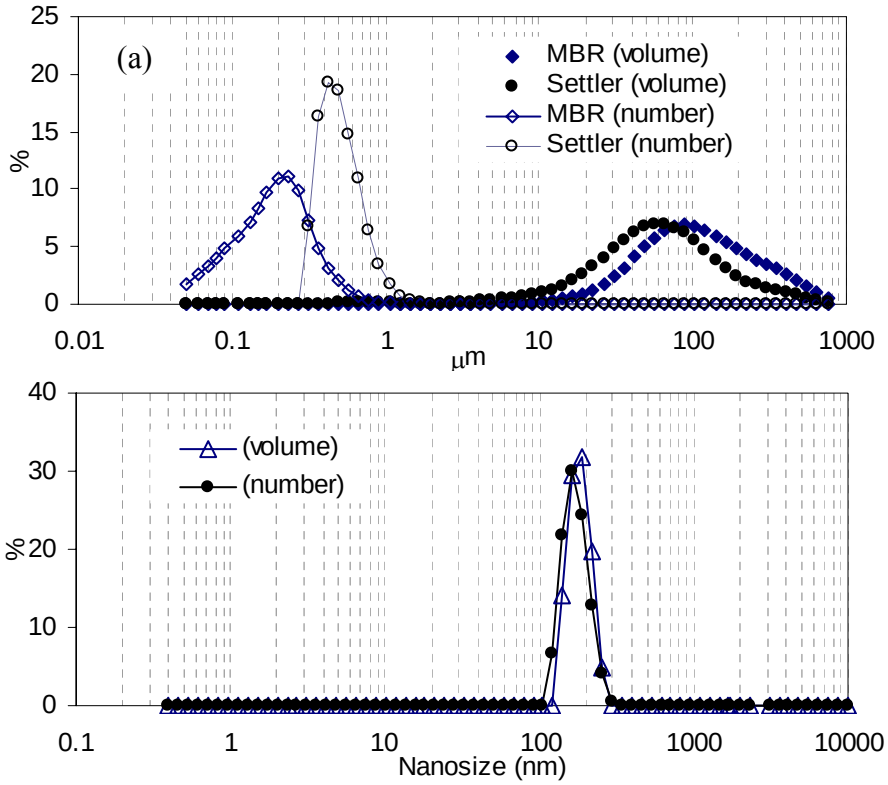


Figure 4.1 5 A typical particle size distribution: (a) microsize of settler supernatant and MBR sludge; (b) colloidal fraction of MBR sludge

As far as fouling sense is concerned, the particle size of MBR sludge was larger than the pores of the membrane, thus the particles had less possibility to infiltrate through the membrane pores. If the effect of fouling was due to suspended solids, it can be

considered as reversible fouling which could be avoided by physical cleaning techniques. However, the fouling contribution of suspended solids fraction was observed to be insignificant among the sludge fractions, which is described in section 4.2.2.4.

4.2.2.4 Fouling behavior of biomass fractions

Figure 4.16 illustrates the fouling behavior of three different biomass fractions, namely suspended solids, colloids and solutes (SS, CL and SL). The fouling potential (MFI) of SS, CL and SL fractions constituted 12, 39 and 49% of the total fouling potential respectively in MBR mixed liquor. The resistance of SS, CL and SL fractions was $0.01x10^{12}$ m^{-1}, $0.33x10^{12}$ m^{-1} and $2.38x10^{12}$ m^{-1} (inferred from Table 4.3) which made up 2, 12 and 86%, respectively (Appendix C, Table C-9, C-10). This supported the notion that the SS and CL fractions did not influence flux decline significantly or soluble fraction was the main fouling contributor among the biomass fractions in the case of granulation effluent. The comparison of fouling potential of biomass fractions with other findings is presented in Table 4.4. This study supported that soluble fraction or SMPs were the major foulants in the MBR treating granulation effluent.

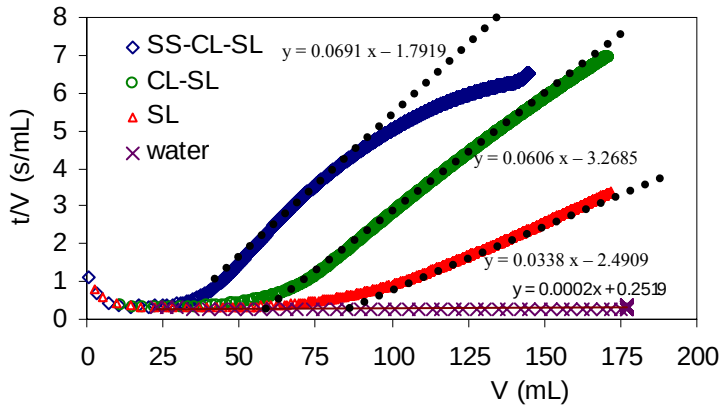


Figure 4.16 Filtration time vs permeate volume of sludge fractions (dotted line: fouling rate)

Table 4.3 Fouling behavior of sludge fractions

Fractions of sludge	SS-CL-SL	CL-SL	SL
MFI_{20} (10^3 s/L^2)	86.7 (0.986)	76 (0.999)	42.4 (0.996)
$\alpha*C$ ($1/m^2$)	$3.02*10^{14}$	$2.65*10^{14}$	$1.48*10^{14}$
α (m/kg)	$1.37*10^{14}$	-	-
R_t (m^{-1})	$2.83*10^{12}$	$2.82*10^{12}$	$2.49*10^{12}$
R_m (m^{-1})	$1.12*10^{11}$	$1.12*10^{11}$	$1.12*10^{11}$
$R_f = R_t-R_m$ (m^{-1})	$2.72*10^{12}$	$2.71*10^{12}$	$2.38*10^{12}$

Note: The number in the brackets are R^2 of the linear segments in the time to volume profile

Table 4.4 Comparison of fouling potential of sludge fractions (%)

Fraction/sludge type	SS	CL	SL	Remark	Reference
MBR treating granulation effluent	2	12	86	No backwash, HF, PE	This study
MBR sludge	24	50	26	Backwash, HF	Bouhabila et al., 2001
MBR sludge (solute separation)	23	25	52	Backwash, ceramic membrane	Wisniewski et al., 1996

Note: HF: Hollow fibre, PE: Polyethylene

In addition, it was observed that the formation of cake layer took long time (70 days) to form on the membrane surface even without backwashing or air scouring. Membrane fouling occurred on day 78 with low fouling rate (0.027 kPa/d) where there was no complete cake layer formation on the membrane surface during operation. The white color (original) of membrane fibres still was seen at almost all area of fibers which was very different compared to the same membrane module operating as conventional submerged MBR (Khan and Visvanathan, 2008). This is the advantage of the low substrate and low biomass submerged MBR in which biomass is always under endogenous condition due to low substrate supplied. In general, based on the fouling potential and result of membrane resistance of sludge fractions it can be stated that the fouling of the MBR is mainly caused by the soluble fraction of MBR sludge.

4.2.2.5 EPS deposition on membrane

Figure 4.17 shows increase in concentration of sPS in MBR supernatant compared to settler (MBR influent) and then slight reduction in permeate (after passing through the membrane). On the other hand, the trend of sPN shows a slight increase in MBR supernatant as compared to sPS and its concentration is negligible in permeate. As observed, sPS is always much higher than sPN in the filtration system (the ratio of sPS/sEPS = 0.72-0.98). The concentrations of sPS in the settler, MBR supernatant and MBR permeate were 7.2±1.1 mg/L, 14.9±2.6 mg/L and 10.3±2.2 mg/L respectively during a membrane fouling cycle (78 days) while sPN concentration was 2.9±2.1 mg/L, 3.5±1.3 mg/L and 0.2±0.2 mg/L (Appendix C, Table C-11).

The sPS in the settler are generated from the granular sludge activity in the SBAR as the byproducts of substrate metabolism (or biomass growth). The increase in concentration of sPS in MBR supernatant as compared to settler could be caused by two facts. Firstly, it could be due to the rejection of PS by membrane causing its accumulation in soluble and/or colloidal forms (Liang et al., 2007). Secondly, it could be caused by the lysis of cells and deflocculation, thus leading to the lysis of dispersed bacteria and/or floc structure which generated smaller size particles as mentioned earlier. The cell lysis or the floc rupture also caused the release of sPS. The bound EPS were hydrolyzed to be soluble EPS which were called biomass associated products (Barker and Stuckey, 1999). In this MBR, the endogenous decay could happen because of the low or refractory DOC concentration in the settler and MBR supernatant. DOC of settler, MBR supernatant and permeate was 6.0±3.1 mg/L, 9.8±5.2 mg/L and 2.9±1.9 mg/L respectively during the operating duration. The insignificant increase of sPN in MBR compared to settler could be hypothesized that the sPN was quickly degradable or less released from the cells/flocs into the MBR. However, the amount of bPN in MBR mixed liquor was twice higher than that in granule and fouling layer (Table 4.5). Therefore, the insignificant increase of sPN in the MBR was due to its rapid degradation.

The decrease in concentrations of sPS and sPN in permeate compared to MBR supernatant proclaims that they were both trapped on the surface and inside of the pores of the membrane. The amount of sPS and sPN adsorbed in the membrane was about 31% and 94% of that in MBR supernatant respectively. This indicates that the sPS in the MBR supernatant was partially deposited while the sPN was completely retained on the membrane surface. The specific deposition loading on membrane surface was calculated by the loss of concentration after passing through the membrane which was 11 mg sPS/L.$m^2_{membrane}$ and 8 mg sPN/L.$m^2_{membrane}$. In addition, the sPN due to its readily

biodegradable characteristics could be further degraded in MBR and/or in membrane pores during the operation. The difference of deposition percentage of soluble macromolecules (sPS and sPN) shows that they possess different characteristics. The partial deposition of sPS on membrane could hypothesize that there were two main fractions of sPS existing in MBR supernatant (large and small molecules relative to membrane pore size). The large ones were deposited on membrane and the smaller ones were passed through it.

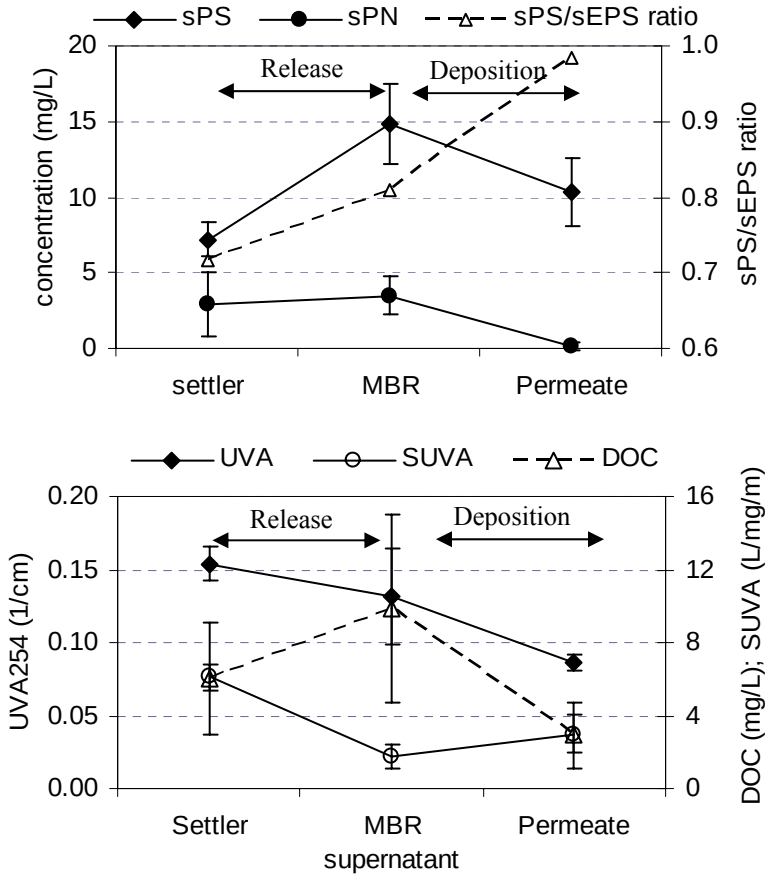


Figure 4.17 Soluble organic matters characteristics profile in the BG-MBR

Although the DOC in MBR supernatant increased compared to that in settler, the UVA_{254} and SUVA showed decreasing trend. This indicates that there is reduction of double-bond substances (such as humic-like materials, protein) which are prone to absorb UV light (Jarusutthirak and Amy, 2006). This correlates with the majority of sPS present in the MBR which are usually long chain macromolecules with less double-bond linkages. DOC reduction in permeate again, confirms that DOC (i.e., mainly sPS and sPN) sludge was deposited on membrane surface. UVA_{254} reduction of permeate means that the double-bond compounds (mostly high MW protein) were trapped on membrane. The passage through pore size of membrane could be equal to the size of small MW organic matter such as low molecular weight sPS portion and humic-like materials.

4.2.2.5 Bound EPS of fouling layer and EPS deposition on membrane

The bound EPS (bEPS) of fouling layer were extracted to understand their characteristics and fouling behavior, and to compare them with those of MBR and granular sludge. The bEPS of the fouling layer were similar to those of granular sludge and

approximately half of MBR sludge (Table 4.5). This result implies that the biomass in fouling layers on membrane started lysis due to the dense biomass concentration and limitation of substrate transfer from the bulk liquid. The speed of lysis in that layer became faster compared to the mixed liquor biomass. This phenomenon could cause SMP release from the bEPS (bound exocellular substances being a major constituent of the floc complex structure) and might add more release of SMP to membrane surface and pores along with SMP from the bulk liquid. The sludge particles did not contribute significantly to fouling propensity when moving in bulk liquid as mentioned but when attached on the membrane as a fouling layer they could accelerate the fouling process. This explains the reason for the so-called TMP "jump" observed in this study which is similar to another lab-scale submerged MBR (Zhang et al., 2006). The TMP profile was usually steeply changed after an operating duration which was suspected to occur by the cake layer formation in this study.

Table 4.5 Bound EPS of fouling layer, mixed liquor and granule

Bound EPS	Bound PS (mgPS/gVSS)	Bound PN (mgPN/gVSS)	Bound EPS (mgEPS/gVSS)	bPS/bPN
Bound EPS of fouling layer (n = 2)	10.5 (±0.4)	19.9 (±1.9)	30.4	0.5
Bound EPS of mixed liquor (n = 7)	18.4 (±7.7)	39.9 (±11.5)	58.3	0.7
Bound EPS of granules (n = 7)	10.7 (±1.4)	17.0 (±2.4)	27.7	0.6

Note: n is number of measurements during experimental period

Table 4.6 Comparison of total EPS deposition on membrane

No	This study	Kim and Digiano, 2006	Cho and Fane, 2002
Amount of EPS deposition ($\mu g/cm^2$)	20 ± 1	3 - 10	20 – 70 (200 – 700 mg/m^2)
Cumulative volume per unit of membrane area (L/m^2)	524 (78 days)	1000 – 3600	NA
Operating flux ($L/m^2.h$)	2.8	50	80 - 20
Membrane pore size	MF 0.1 µm	UF 150 kDa	MF 0.22µm
Module configuration	Submerged HF	Pressurized filtration HF (two fibres)	Flatsheet, pressurized filtration
Filtration	Granulation effluent	Secondary effluent (pretreated by sand filter and MF 150 µm)	UASB effluent
Backwashing	No	Yes	No

The explanation for the low EPS content in granule is similar to that of fouling layer because granule is also a kind of spherical biofilm with limitation of substrate and nutrient transfer to the granule core. The lower bPS/bPN ratios of fouling layer and granule compared to MBR sludge indicate that the lysis process can release more sPS than sPN (Zhang et al., 2006).

Table 4.6 presents the comparison of EPS deposition on membrane and inside the pores for various operating modes of MBR (Appendix C, Table C-12). It appears that the deposition of sEPS on MF membrane is high because sEPS can penetrate into and adsorb on the surface and pores of membrane. This result confirms the evidence of sEPS deposition on/inside the membrane pores.

4.2.2.6 Critical flux analysis (CFA)

According to Le-Clech et al. (2003) a critical flux (J_c) for each MBR condition was defined from dTMP/dt limit of 0.1 mbar/min (0.01 kPa/min). The critical flux of MBR in this study is 18 L/m^2.h (Figure 4.18, Appendix C, Table C-13). This value was bit lower than other researches listed in Table 4.7. At this flux the fouling rate starts steeply increasing. The low value of the critical flux compared to other researches is because of the presence of several fibres in the HF membrane module used in this study. It has large actual surface area but in fact not active surface area. During suction, the surface area of only the outer bundles is active. The inner fibres are filled up by the sludge and then become inactive.

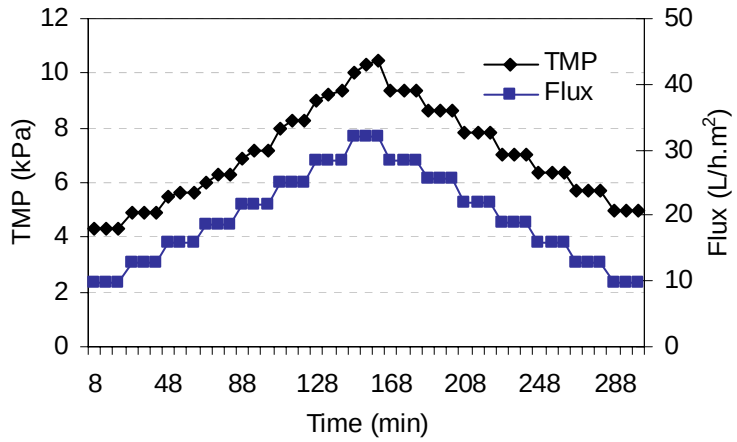


Figure 4.18 Critical flux of MBR

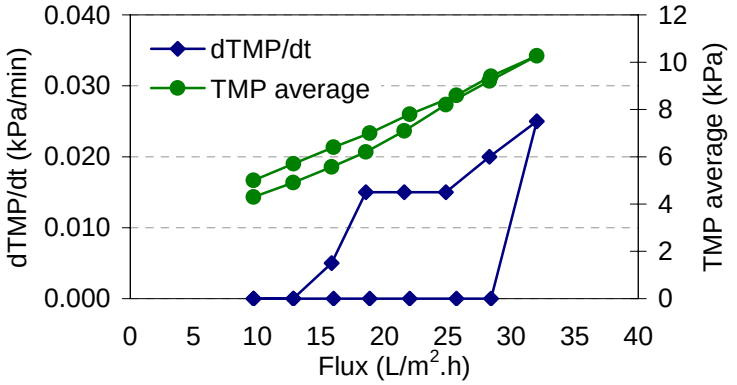


Figure 4.19 Fouling rate and pressure versus MBR flux

Table 4.7 Comparison of critical flux (J_c) analysis (modified from Bacchin et al. 2006)

Suspension	pH	Conc. (g/L)	Re/aeration rate	Membrane	J_c ($L/m^2.h$)	References
MBR sludge	8	2.18	0.3 cm/s	HF, PE 0.1 µm, Mitsubishi	18	This study
MBR sludge	-	10±0.5	0.048 m/s	Full scale, HF, PVDF 0.04 µm, Zenon ZW500c	30	Guglielmi et al., 2007
Bentonite	-	0.1	0.47 $cm^3/cm^3.min$	Single fibre, Polysulfone, HF 0.1 µm (KOLON, Korea)	80-98	Kim and DiGiano, 2006
Latex	-				105-130	
AS	-	10	4 m/s	Kerasep 0.1 µm	115	Defrance and Jaffrin, 1999
AS	-	3-10	2300	Millipore plane membranes	65	Madaeni et al., 1999
Fermentation broths (lactic acid)	6.2	2.6 (bacteria)	4 m/s	Kerasep 0.1 µm	50	Milcent and Carrere, 2001
Skimmed milk	-	-	3.8-5.4 m/s	Kerasep 0.1 µm	60	Gésan-Guisiou et al., 1999
Sillica X30	9.7	0.5%	580	PS 0.2 µm	50	Wu et al., 1999
Yeast	-	5	Bubbling	PP, HF	10	Chang and Fane, 2000

4.2.2.7 Metabolic activities of MBR sludge

Figure 4.20 shows the heterotrophic and nitrifying bioactivity in terms of specific oxygen uptake rate (SOUR). The maximum $SOUR_H$, $SOUR_{NH4}$ and $SOUR_{NO2}$ were 30, 15 and 25 $mgO_2/gVSS.h$ respectively. This result shows that the $SOUR_{NO2}$ is quite high as compared to the rest of SOURs. This indicates that there was only partial nitrification (conversion from nitrite to nitrate) in MBR. Nitrite nitrogen was 48-97 $mgNO_2$-N/L in the settler whereas organic matter and ammonia were less (6 mg DOC/L; 1.5 mg NH_4-N/L). The partial nitrification efficiency was more than 99% in MBR. In other words, significant amount of nitrifying microorganisms existed in MBR.

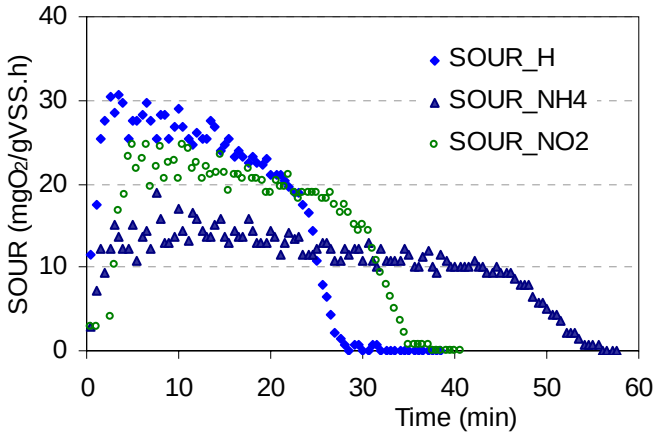


Figure 4.20 Microbial activity of MBR sludge

4.3 Stability of aerobic granule and its effect on fouling ability

4.3.1 Granule stability and its effect

In order to obtain additional nitrogen removal through denitrification, the original BG-MBR system was modified. Here, the settler and the MBR units were replaced by a single baffled MBR unit which is known as membrane airlift bioreactor (MABR) (refer to Appendix A, Figure A-5, A-6, Table A-1). This MABR configuration consists of aerobic and anoxic zones. In addition, it was reported that the MABR showed less direct contact with substrate supply, low biomass concentration in the reactor, low aeration requirement to achieve the required aeration shear stress and presence of aerobic and anoxic zones within the reactor when compared with the conventional MBR (Kimura et al., 2008). Moreover, the granules were found to disintegrate after certain of operation (Liu and Liu, 2006) which was observed in this study as well. However, there was little information on the effect of fouling behavior of the SBAR effluent while the granule breakage. In this experimental operation, it was aimed to investigate the effect of granule stability on fouling propensity and nitrogen removal by coupling the SBAR with the MABR. This combined system was named as the Batch Granulation Membrane Airlift Bioreactor (BG-MABR).

The SBAR which was previous operated for 250 days with the stable conditions was coupling with MABR. In this study, the first day was counted when the MABR was connected as the BG-MABR. After 20 days of operation with the BG-MABR, the granules started to disintegrate and the filamentous and fungal granules appeared in the SBAR. Similar observations were reported by Schwarzenbeck et al. (2005); Liu and Liu (2006). The fungal granules were white in colour with weak settleability when compared to the dark brown bacterial granules. The long SRT causes excessive growth of filamentous microorganisms in the reactor. It encourages the growth of the slow growing filamentous microorganisms over the floc forming microorganisms (Liu and Liu, 2006). The instability of granules in this study could be explained due to the long retention of aged granules in the SBAR. The granules were settled and retained in the reactor for more than 250 days. In the SBAR the granules were allowed to settle for 3 minutes and the effluent was pumped into the MABR. Hence, the new sludge (from assimilation) was being washed out with effluent while the accumulation of aged sludge in the reactor was occurring in every batch of operation. The long retention of granules (more than 270 days) could have resulted in disintegration due to excessive growth of filamentous microorganisms in the reactor and lack of substrate and nutrient diffusion into the core of the granules. This led to cell lysis in the core of the granules which caused breakage of granules.

The average granule size and the settling velocity in Run 1 (before granule disintegration) and Run 2 (after granule disintegration) were 5.8±1.3 mm and 135±17 m/h, and 5.2±1.3 mm and 125±22 m/h for OLR 2 kgCOD/m^3.d respectively. The granule size reduced from 5.8 to 5.2 mm which concluded that the matured and large granules progressively disintegrated. Similarly, the granule settling velocity reduced from 142 to 125 m/h because the broken granules were disintegrated into flocs and smaller particles.

Furthermore, the MLSS of the granular sludge in the SBAR was 12000 mg/L at the initial stages of this experiment (day 10). Later it decreased gradually and reached 7500 mg/L by day 40 due to the detachment phenomena of granules caused by long SRT. During this period, the bed volume of granular sludge was reduced from 3.5 L to 2.0 L. The average SVI_{15} of the granular sludge were maintained at 29±4 mL/g in the reactor.

According to other researchers, the SVI was maintained in between 10–140 mL/g (Jang et al., 2003; Qin et al., 2004, Tay et al., 2004, Thanh, 2005 and Kim et al., 2008). In SBAR, the biomass concentration reduced while the settled volume of biomass was almost similar for the periods before and after granule disintegration as presented in Figure 4.21. This observation shows the excellent settling ability and densification of granular sludge.

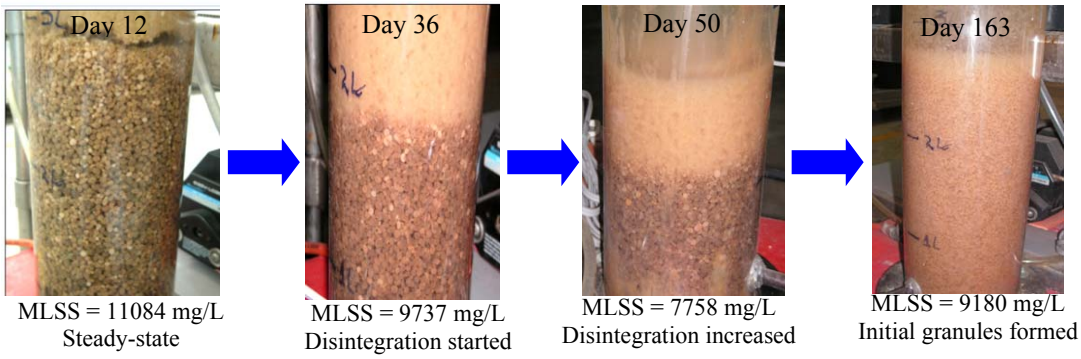


Figure 4.21 Disintegration of granular sludge in SBAR

Figure 4.22a shows black spots on some of the big and matured granules which imply that there exists the anoxic zone due to limited diffusion of oxygen in the core of the granules. Hence, simultaneous nitrification in the surface and denitrification in the core of the granules was achieved at the beginning of the research. However, at later stages, due to granule disintegration the big and matured granules disappeared and small granules were dominant in the reactor. For the day 6 and 34, it was seen that the percentage of small granules (size < 5 mm) increased by 20%. Also, at day 34, white granules were dominant in the reactor which showed that less number of nitrifiers and denitrifiers were present in the reactor (Figure 4.22b).

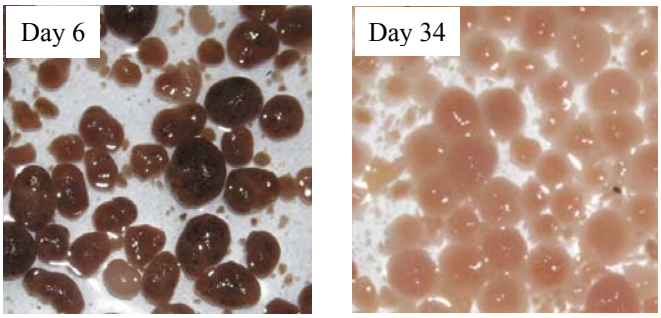


Figure 4.22 Granule morphology in the SBAR on day 6 and day 34

Hence, the decrease in nitrogen removal through denitrification in the SBAR is evident from 34 days of operation. The denitrification was 47% during the stable operation before granule breakage and it was reduced to 27% after granule breakage. Moreover, the ammonium nitrogen level in SBAR effluent started to increase and as a result the complete nitrification was reduced (Figure 4.23, Appendix C, Table C-14). In addition, the white granules (filamentous and fungal granules) appeared in the reactor after 20 days of operation due to the long SRT (Schwarzenbeck et al., 2005; Liu and Liu, 2006). The granule instability at high SRT in this study reveals a necessity of SRT control for granulation system. The pre-set SRT will be much effective than the method of controlling SRT based on the biomass wash out gravitationally and biomass in reactor. The SRT should be controlled in the range of 10-15 days to avoid the overgrowth of filamentous

microorganisms which causes failure in granulation system. As a conclusion, the suitable selection of SRT and sludge removal method can maintain the performance and stability of the granulation reactor.

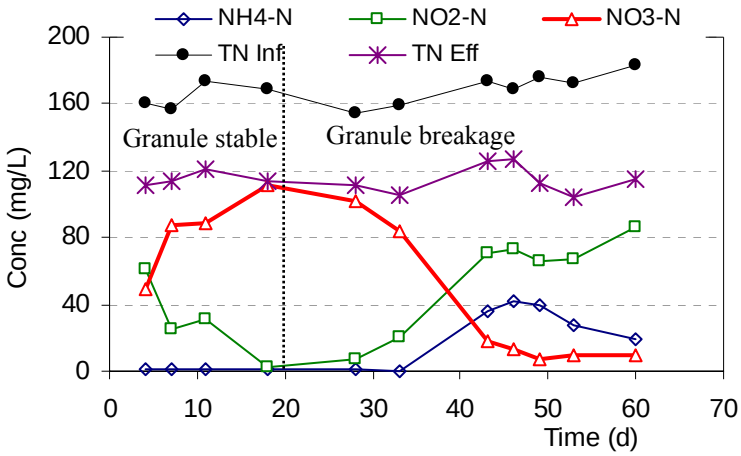


Figure 4.23 Evolution of nitrogen species in SBAR effluent

The fouling rate profiles for the two runs are plotted in Figure 4.24. In run 2 (during granule breakage), the fouling was rapid when compared with run 1 (matured granule). The fouling rate of membrane for run 1 and run 2 at OLR 2 kgCOD/m^3.d were 0.105 and 0.475 kPa/d respectively. The granule breakage in the SBAR led to rapid fouling in the MABR. The disintegration generated flocs, debris and SMPs in SBAR effluent which was fed to the MABR. As mentioned earlier the granule breakage resulted in high sPS, sPN and DOC in SBAR effluent which was susceptible for membrane fouling. Figure 4.25 shows more production of sPS and sPN during the granule breakage when compared with the steady state. Whereas DOC was slightly higher at the steady state that attributed to other organic products rather than PS and PN as it includes all organic matters. Moreover, sPS and DOC in MABR supernatant was higher at the disintegration duration when compared with the matured granule duration (Appendix C, Table C-15). Further the granule disintegration also produced more SS in SBAR effluent that was fed into the MABR which caused an increment of around 1 g/L in MLSS in the MBAR. However, the suspended solids did not contribute significantly for fouling (result from previous section of the BG-MBR) so the release of SMPs from granule disintegration was accountable for membrane fouling. Similarly, Tay et al. (2007) found that the fouling frequency of granular sludge MBR was three fold higher than that of conventional MBR. Hence, it can be concluded that the granular sludge can enhance the fouling control in membrane if the granule stability is maintained in the reactor. Operational guideline for granulation systems is presented in Appendix C, Table C-16.

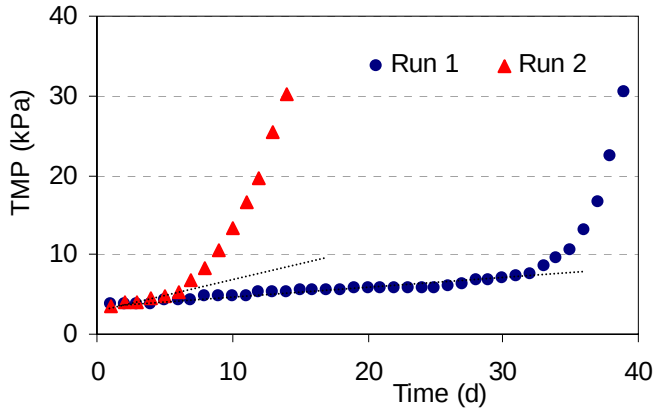


Figure 4.24 Fouling rate of the MABR during stable granule stably (Run 1) and granule breakage (Run 2)

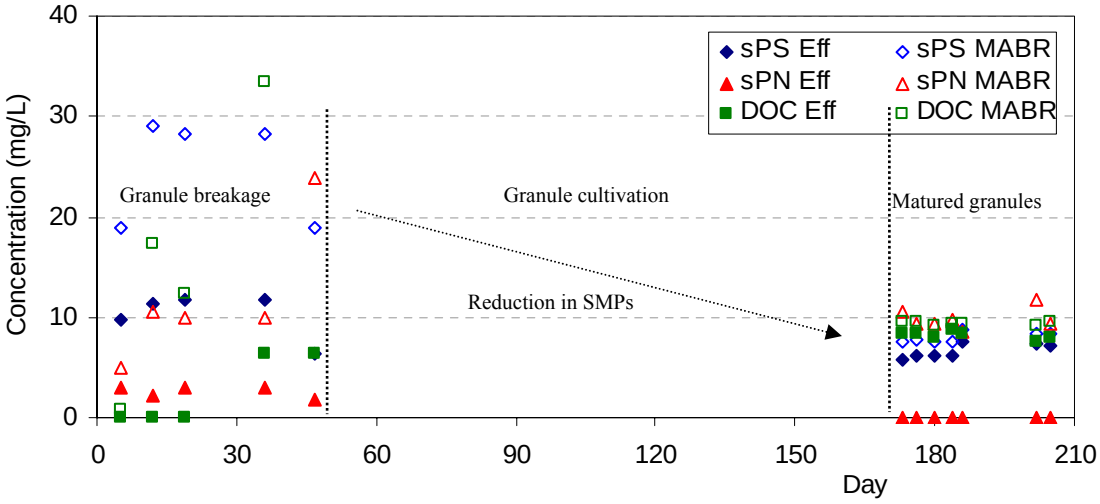


Figure 4.25 Change of soluble microbial products from granule disintegration in SBAR effluent and MBAR supernatant

4.4 Effect of aeration rates on characteristics of SBAR effluent (INSA)

This experiment investigates the characteristics of sludge and SBAR effluent at various aeration rates when fed with high nitrate containing wastewater. It is aimed to understand the characteristics of sludge and effluent at the aeration rates which are representative for conventional and granulation processes. The anoxic/aerobic condition improves the sludge characteristics in terms of increased biomass retention, density, settling ability and minimizes the fouling potential.

4.4.1 SBAR performance at various aeration rates

The nitrogen removal and sludge characteristics of the SBAR are summarized in Figure 4.26. During the first two runs (0.8 and 2.2 cm/s) no significant nitrate removal was observed. The sludge characteristics in terms of SVI and MLSS were almost similar to conventional SBR. In contrast at aeration rate of 0.6 cm/s and pre-anoxic mixing with nitrogen gas introduced from day 121 onward, the nitrogen removal and sludge quality was

found to improve significantly. During this period, an improved denitrification of nitrate was observed. In parallel, the biomass concentration and settling ability in reactor were found to increase impressively while the effluent suspended solids reduced. MLSS and SVI could reach 9 g/L and 44 mL/g respectively at the aeration rate of 0.6 cm/s under anoxic/aerobic condition. In conventional SBR, MLSS and SVI were maintained in the range of 1.5-5.0 g/L and 80-150 mL/g respectively (Metcalf and Eddy, 1991). Hence, the sludge retention or biomass density increased under anoxic/aerobic condition compared to the previous operating aeration rates. Similarly, it was observed that existence of pre-anoxic stage in a SBAR could enhance structure of aggregate (McSwain et al., 2004). This shows the positive effect of anoxic/aerobic condition on the performance of the SBAR.

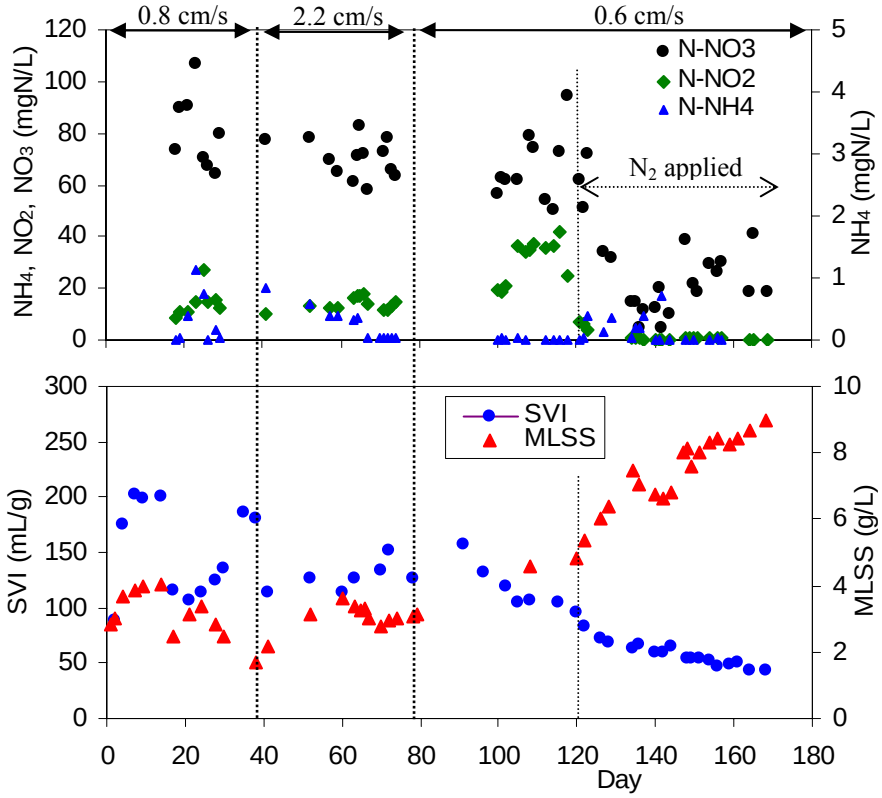


Figure 4.26 Nitrogen removal and sludge characteristics of the SBAR

4.4.2 Effect of aeration rate and anoxic/aerobic condition on resistance rate

Figure 4.27 represents the fouling rates of the three sludge fractions, namely suspended solids (SS), colloids (CL) and solutes (SL) of the SBAR effluent at the different aeration rates. At the aeration rate of 0.8 cm/s of conventional SBAR, the resistance rates contributed by SS, CL and SL fractions were of the same order of magnitude. Operation at this aeration rate resulted in the lowest resistance rate (Figure 4.27a). The increase in aeration rate from 0.8 cm/s to 2.2 cm/s resulted in augmentation of resistance rates for all three sludge fractions of SBAR effluent (Figure 4.27a, b). Furthermore, it was also noted that the resistance rate was distributed among the biomass fractions. The resistance rate of SS fraction was significantly larger than the CL and SL fractions. This can be explained that the flocs structure was disrupted by aeration shear stress producing small particles and SMPs. It was observed that the particle size in term of volume distribution for SBAR effluent was averagely from 87.7, 79.5, 68.7 and 61.2 µm at aeration rates of 0.8, 2.2, 0.6

and 0.6 cm/s under anoxic/aerobic condition respectively. In addition, the production of macromolecules was observed at aeration rate of 2.2 cm/s (granulation condition) (Figure 4.29) was responsible for the increase in resistance rate of soluble fraction.

At the reduced aeration rate of 0.6 cm/s under anoxic/aerobic condition, the resistance rate of SS fraction decreased compared to that of aeration rate of 2.2 cm/s (Figure 4.27c). However, resistance rates of three sludge fractions were similar and slightly higher than those at aeration rate of 0.8 cm/s (Appendix D, Table D-1, D-2, D-3, D-4). In addition, the SS in the effluent slightly increased during second phase and decreased at the third phase (Table 4.8), showing that shear stress due to strong aeration probably releases more small particles influencing the fouling potential. In general, the resistance rate at low aeration rates (0.6-0.8 cm/s) the fouling potential of SS and CL fraction was not significant compared to that of SL fraction. This observation is similar to the case of the BG-MBR in which fouling potential of SL fraction was 86% of all sludge fractions (section 4.2.2.4).

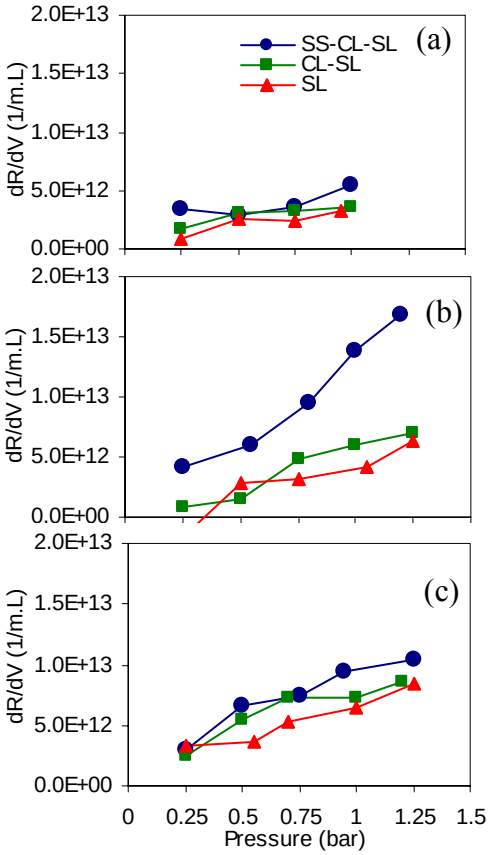


Figure 4.27 Resistance rates at various aeration rates; (a) 0.8 cm/s (day 28), (b) 2.2 cm/s (day 65) and (c) 0.6 cm/s under anoxic/aerobic operation (day 145)

The fouling trend was again confirmed by the specific resistances of sludge fractions (Figure 4.28a,b,c) and fouling resistances such as reversible (R_{rev}) and irreversible resistances (R_{ir}) (Figure 4.28d,e,f). Specific resistances were calculated based on the resistance over the filter volume. Specific resistances of SL and SS increased significantly at high aeration rates and slightly at anoxic/aerobic conditions. The high aeration rate resulted in higher irreversible fouling.

Again, the increase of the resistance of soluble fraction at aeration rate of 2.2 cm/s is evident from the production of large MW protein-like materials (Figure 4.29). For a short period, high aeration shear stress caused floc breakage, thus releasing large molecule weight (MW) substances (bound exocellular substances being a major constituent of the floc complex structure). Most of the released substances had MW of 30-50 kDa and the remaining was of 800-1050 kDa. The same phenomena was observed by Ji and Zhou (2006) that the increase in aeration rate led to the breakage of microbial flocs, the decrease in floc size and the release of EPS into supernatant. This proves that the characteristics of soluble fractions of SBAR effluent significantly changed when the SBAR was operated at high aeration shear stress (2.2 cm/s) which is similar to aeration rate of granulation reactor.

The resistance rates of sludge fractions at 0.8 cm/s and 0.6 cm/s show similar trend (Figure 4.27a,c). This indicates that at low aeration rate the fouling resistance of SL is much significant when compared with that of SS and CL. In other words, the fractions of SS and CL did not have significant influence on fouling rate at low aeration rate. However, the resistance rates at aeration rate of 0.6 cm/s under anoxic/aerobic operation were slightly higher than those at 0.8 cm/s and lower than those at 2.2 cm/s aeration rates. This could be due to the certain impact of anoxic growth on quality of sludge and supernatant.

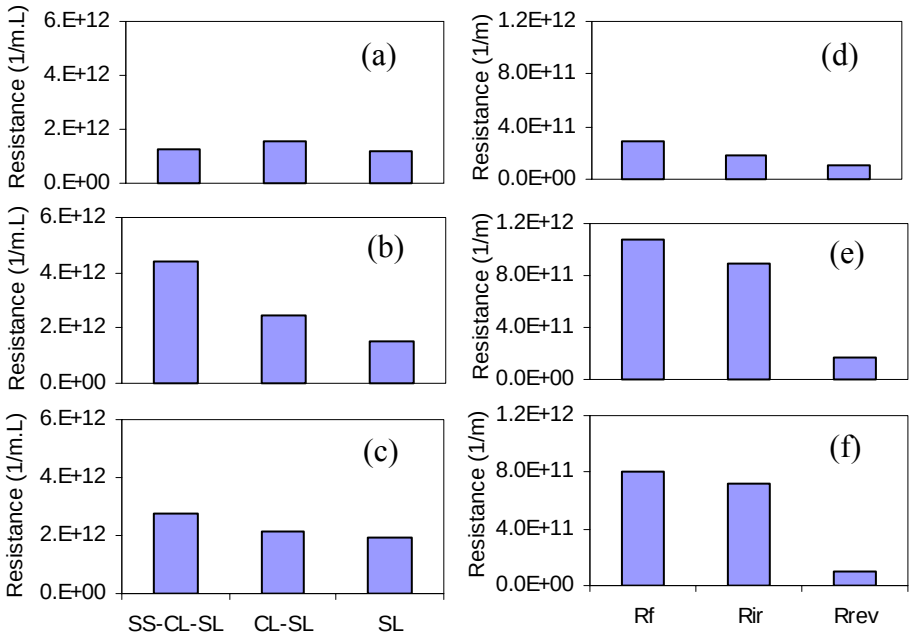


Figure 4.28 Typical values of specific resistance (left) and resistance of SBAR effluent (right) at aeration rates of 0.8 cm/s - day 28 (a,d), 2.2 cm/s – day 65 (b,e) and 0.6 cm/s under anoxic/aerobic operation – day 145 (c,f)

In addition, the resistance rate of SBAR mixed liquor (Appendix D, Figure D-1) was always less than that of SBAR effluent under all the conditions despite the fact that the suspended solids concentration was much greater. This shows that the quality of biomass has a stronger impact on fouling rate than its concentration. The particle size distribution of mixed liquor and effluent was almost similar. Again, the particle size for SBAR effluent was averagely from 87.7, 79.5, 68.7 and 61.2 μm at aeration rates of 0.8, 2.2, 0.6 and 0.6 cm/s under anoxic/aerobic condition respectively. Similarly it was 86.6, 84.3, 53.0 and 63.3 respectively for mixed liquor. This shows that floc size does not play a significant role

in resistance rate in this work. Furthermore, the effluent and mixed liquor suspended solids were different in terms of settling velocity, density and retention. The washed out of biomass through effluent was mainly from the biological assimilation (and small particles detached from granules) while the retained biomass was endogenously aerated due to long retention. The longer contact of mixed liquor with endogenous condition compared to that of effluent is probably the reasons for the lower resistance rate. The endogenous condition has certain influence on quality and fouling of biomass in SBAR effluent and mixed liquor.

The specific cake resistances (α) of SBAR effluent decreased as the aeration rate was increased and was high under anoxic/aerobic operation. The suspended solids concentration did not influence specific cake resistance. It can be seen in Table 4.8 that compares the results for the effluent and the mixed liquor. The specific cake resistance of mixed liquor was lower than that of effluent despite higher suspended solids in mixed liquor. The irreversible resistance of mixed liquor (Table 4.9) was larger for high aeration rate probably due to more release of large MW substances.

Table 4.8 Typical specific cake resistances of sludge sources at various aeration rates

Source	SBAR effluent			SBAR mixed liquor	
Aeration rate	0.8 cm/s	2.2 cm/s	0.6 cm/s + anoxic/aerobic	2.2 cm/s	0.6 cm/s
Day	28	65	142	79	120
C ($kgSS/m^3$)	0.334	0.474	0.097	3.160	4.750
$\alpha.C$ (10^{12} $1/m^2$)	9.1	9.6	20.8	23.7	7.4
α (10^{12} m/kg)	27.2	20.1	214.0	7.5	1.6

Table 4.9 Resistance of SBAR mixed liquor at various aeration rates

Aeration rate	2.2 cm/s	Percentage	0.6 cm/s	Percentage
R_m (m^{-1})	1.52×10^{12}		1.49×10^{12}	
R_t (m^{-1})	2.40×10^{12}		2.25×10^{12}	
R_f (m^{-1})	8.79×10^{11}		7.64×10^{11}	
R_{ir} (m^{-1})	6.85×10^{11}	78 %	4.27×10^{11}	56 %
R_{rev} (m^{-1})	1.94×10^{11}	22 %	3.37×10^{11}	44 %

It can be concluded that the anoxic/aerobic operation with existence of nitrate in the feed was found to enhance denitrification and biomass retention and to control fouling. Anoxic growth seems to have a negative impact on effluent but a positive role on mixed liquor sludge during filtration. Fouling propensity of sludge fractions of SBAR effluent increased with aeration rate in the reactor. The release of large MW substances from the floc structures occurred at high aeration intensity increased fouling potential of soluble fraction. The longer contact under endogenous condition of sludge could alleviate fouling potential.

4.4.3 Effect of aeration rates on molecular weight distribution of SBAR effluent

Figure 4.29 shows the size exclusion chromatography (SEC) profile of macromolecules of SBAR effluent at various aeration rates. The distribution is bimodal with two main groups of protein-like materials, namely 4-7 kDa and colloidal aggregate (left hand peaks). The colloidal aggregate peaks were not noticed when samples were filtered though membrane with pore size of 0.1 µm. This indicates that the colloidal aggregates were larger than 0.1 µm. The maximum MW which can be detected by the column superpose6 is 669,000 kDa. Thus it could be stated that the colloidal aggregates

had the size larger than 0.1 µm and smaller than 669,000 kDa. The SEC profiles of other substances have been shown in Appendix D, Figure D-2, D-3, D-4.

The high aeration rate (2.2 cm/s) in the SBAR produced macromolecules of the size of 30-50 kDa and 620 kDa. The large MW could be released from the cells as a result of their lysis due to intensive aeration stress under endogenous respiration. The released products were found to be the reason of increase in resistance rate and irreversible fouling as mentioned above. Furthermore, Jarusutthirak and Amy (2006) postulated that the high MW compounds play an important role in creating high resistance of membrane. The SEC profile of effluent at aeration rate of 0.6 cm/s was almost similar to that at 0.8 cm/s. However, MW of 6 kDa was present in stead of 4-5 kDa (Figure 4.29). This might be the response from the anoxic condition or denitrification process. Drews et al. (2007) reported that the nitrate and nitrite had a certain effect on the formation of SMPs during the biological treatment process. In this work, bigger protein-like materials could be generated at high aeration rate (granulation condition of the SBAR) and anoxic/aerobic operation (conventional SBAR). This result supports the increase of the fouling potential due to SMPs at aeration rates of 2.2 cm/s and 0.6 cm/s under anoxic/aerobic condition in previous section.

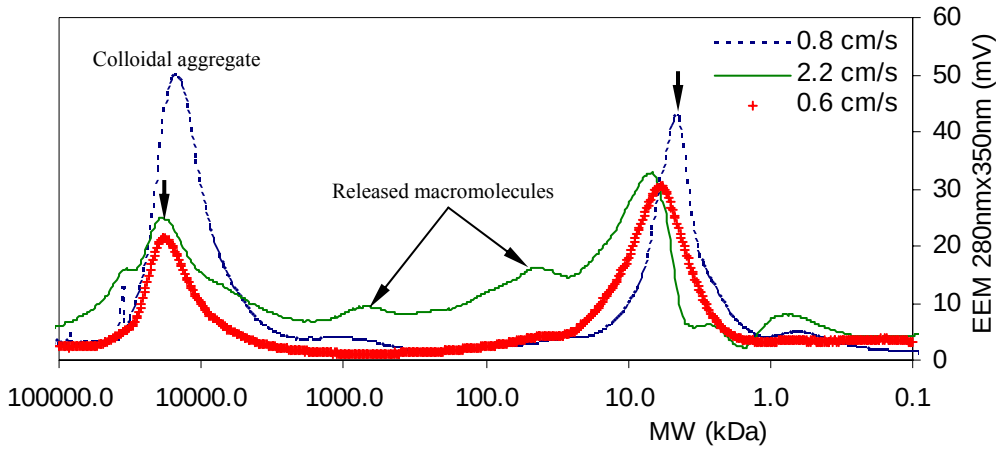


Figure 4.29 Size exclusion chromatograph of SBAR effluent at various aeration rates (day 28, 74 and 102)

Table 4.10 Molecular weight of SBAR effluent at various aeration rates

0.8 cm/s (day 28)	2.2 cm/s (day 74)	0.6 cm/s (day 102)
+ Colloidal aggregates + 4-5 kDa	+ Colloidal aggregates + 5-6 kDa + 30-50 kDa + 620 kDa (small amount)	+ Colloidal aggregates + 5.7-6.2 kDa + 38 kDa (small amount)

4.4.4 Effect of aeration rates on hydrophobicity of SBAR effluent

Figure 4.30 shows that the SBAR effluent had a large hydrophilic peak and small hydrophobic peaks for all three aeration rates. This indicates that the SBAR effluent contains significant amount of hydrophilic macromolecules at all aeration rates. The relative importance of the hydrophobic and non-hydrophobic fractions as the value of the intensity of hydrophobicity (Dimensionless Retention Time - DRT) has been shown in Figure 4.31 and Table 4.11. The calculation of DRT is referred to Appendix D, Figure D-

6). Based on these results it was found that at high aeration rate, the effluent contained more hydrophobic fractions (13-67%) with less hydrophobic intensity (DRT = 0.08–0.22). In contrast, at low aeration rate (0.6-0.8 cm/s), effluent was favorable to lesser hydrophobic fraction (mostly < 20%) with high hydrophobic intensity (DRT > 0.8). The presence of nitrate in the feed wastewater made the SBAR effluent hydrophilic at various aeration rates. Moreover, the hydrophobic percentage and intensity of feeding without nitrate addition was much higher (43%, DRT > 0.8) at aeration rate of 0.8 cm/s (Appendix D, Figure D-7). Additionally, the hydrophilic character of soluble fraction is responsible for its low resistance rate and fouling potential of SBAR effluent.

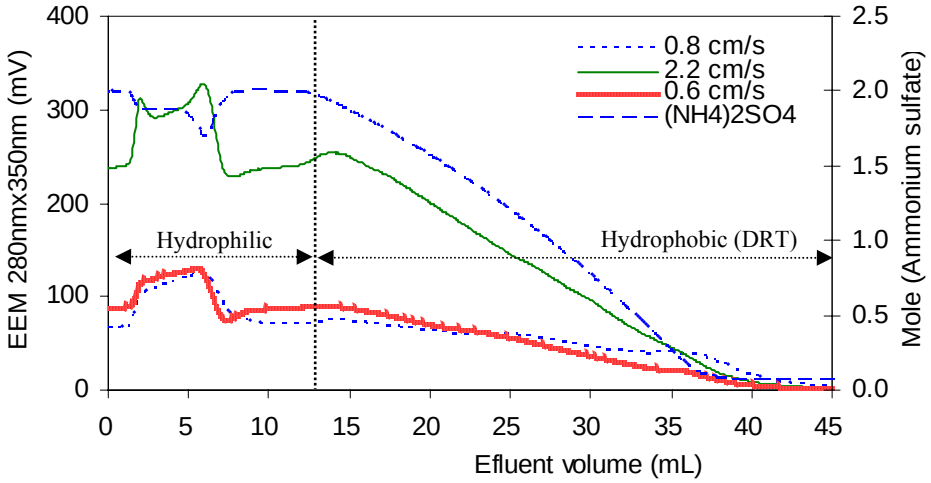


Figure 4.30 Hydrophobicity chromatograph of soluble fraction of SBAR effluent at aeration rates (day 28, 74, 92)

Table 4.11 DRT of peaks of macromolecules (SEC-EEM, Ex: 280nm/Em: 350nm)

Aeratiion rate	0.8 cm/s	2.2 cm/s			0.6 cm/s				
Day No.	28	60	72	74	81	86	88	92	101
DRT	0.91; 0.92	0.08; 0.13	0.11; 0.22	0.1	0.1	0.06; 0.14	0.81	0.79; 0.87	0.85

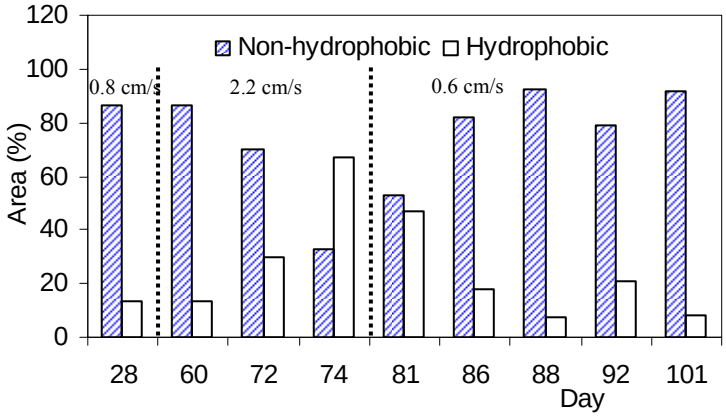


Figure 4.31 Evolution of hydrophobicity of soluble fraction of SBAR effluent

In short, high sludge settling and retention were maintained in the SBAR under anoxic/aerobic operation. For SBAR effluent, resistance rate and irreversible resistance increased at high aeration rate (granulation aeration rate) due to the release of SMPs (30-50

kDa) and small particles. Soluble fraction plays a major role in fouling potential at low aeration rates (conventional aeration rate). At high aeration rate, SBAR effluent contains larger (60%) hydrophobic fraction with low hydrophobic intensity. At low aeration rate it contains less hydrophobic fraction (20%) with high hydrophobic intensity. Furthermore, the specific cake resistance of SBAR effluent was significantly higher when compared to that of mixed liquor, thus confirming the crucial role of biomass quality and history of fouling mechanism. High aeration rate and anoxic phases generate different types and amounts of SMPs which influence the filtration behavior. Therefore, the anoxic/aerobic operation causes certain impacts on filterability and biomass characteristics. The fouling behavior is different for the effluents of granulation and conventional reactors.

4.5 Fouling behavior and treatment performance of CG-MBR at various OLRs

This experiment was aimed to investigate the stability of granule in the continuous granulation MBR system (CG-MBR) at OLR of 2, 4 and 8 kgCOD/m^3.d. The solid/liquid characteristics and membrane fouling of the system were examined during the operational period.

4.5.1 Organic and nitrogen removal of the CG-MBR at various OLRs

Matured granules were added into the CG-MBR with the granulated biomass to total biomass ratio of 0.75. The granules had the size of 1.6±1.0 mm and settling velocity of 55±12 m/h (refer to Appendix E, Figure E-1). Figure 4.32 presents that the TOC removal efficiency of the CG-MBR system was 97.8±2.0, 99.0±0.5 and 99.4±0.3 % at OLR of 2, 4 and 8 kgCOD/m^3.d (Equivalent to 0.8, 1.4 and 3.0 kgTOC/m^3.d) respectively. The respective F/M ratio was 0.72, 1.06 and 1.37 d^{-1}. Overall the TN removal increased with the operating OLR. At OLR 2 kgCOD/m^3.d, the TN removal was about 40% on day 0-20 and then reduced to 20% on day 21-51 (at the end of this OLR). The added granules were worn completely after 20 days of operation. The nitrogen removal reduced as granules disintegrated and the denitrification process was inactive in the system. In other words, the TN removal from day 21 to 51 was mainly due to the assimilation. The instability of granules could be explained by the two reasons: (1) The semi-continuous feeding of the CG-MBR could not create the feast and famine condition effectively. The periodical starvation was considered as one of the granule forming condition. (2) Part of granules was gradually stuck within the membrane fibres which restricted the granule movement. As shear force was a prerequisite for granule cultivation. The granules stuck within fibres progressively disintegrated. Similarly the TN removal efficiency was 30±8 and 53±15% at OLR of 4 and 8 kgCOD/m^3.d respectively. The increase in removal at high OLR was due to the augmentation of organic substrate in the feed which required more nitrogen for assimilation. In addition, the nitrate in permeate reduced while nitrite and ammonia increased at OLR 8 kgCOD/m^3.d. The nitrification reactions reduced at OLR 8 kgCOD/m^3.d. The nitrate concentration in permeate significantly decreased at this OLR. It was 108±25, 122±9 and 62±27 mg/L at the respective OLRs. The nitrite concentration in permeate during operation was 6±4, 5±3 and 16±11 mg/L (Appendix E, Table E-1, E-2). This shows that the activity of nitrifying microorganisms decreases with increase in OLR. In other words, the nitrifiers were unable to compete with heterotrophs at high OLR.

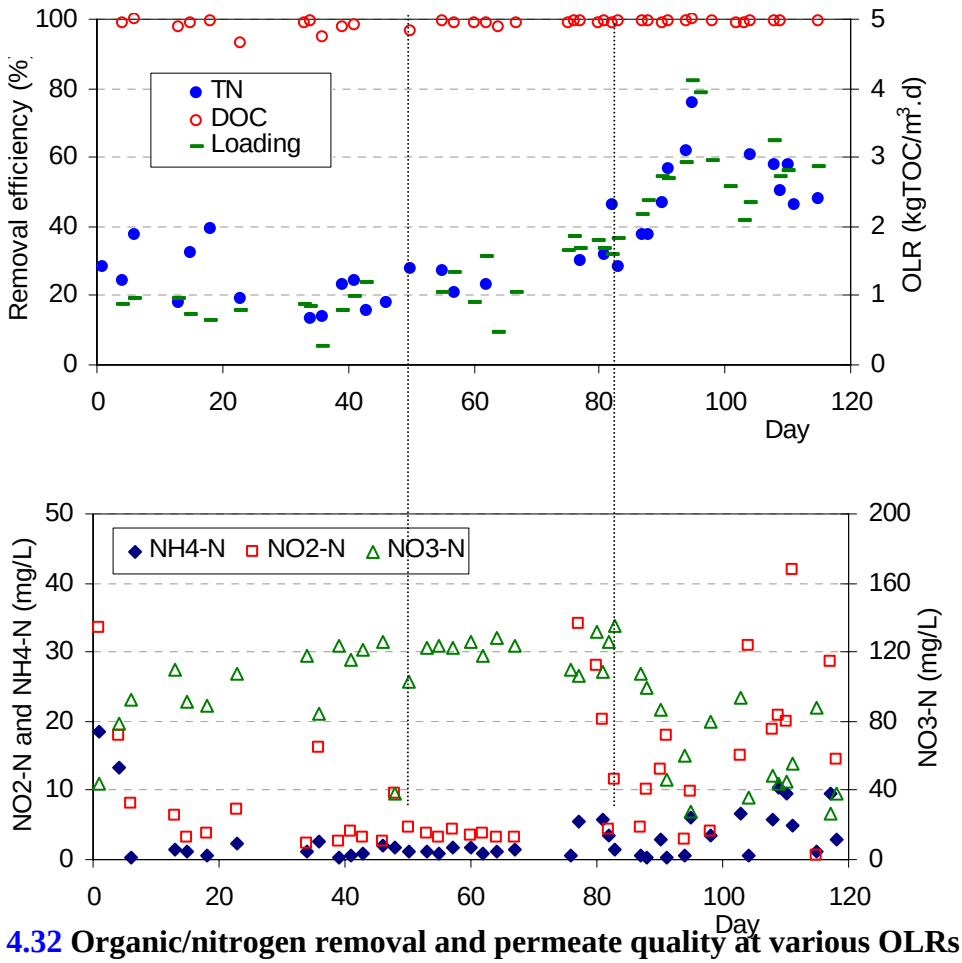


Figure 4.32 Organic/nitrogen removal and permeate quality at various OLRs

*Remark: at OLR of 8 kgCOD/m^3.d, system experienced operational problems so the OLR was reduced slightly at day of 100-107.

4.5.2 Sludge characteristics of CG-MBR at various OLRs

4.5.2.1 Biomass concentration and SVI

The MLVSS/MLSS ratio ranged from 0.85 to 0.89 for all OLRs. Sludge settling ability in terms of SVI was proportional to MLSS concentration. Figure 4.33 presents SVI was stable in the range from 64±11, 87±30 and 64±18 mL/g at 2, 4 and 8 kgCOD/m^3.d. High biomass concentration in the beginning of OLR 8 kgCOD/m^3.d created the hindered settling effect in SVI measurement, thus it made SVI to increase. While granules were maintained in the system, the SVI was low due to compactness of granules. In addition, the MLSS increased with the applied OLR and progressively reduced as the membrane fouled. A thick cake layer attached to membrane was removed during membrane cleaning. Cake layer was less in the case of the BG-MBR system which was operated at the biomass concentration as low as 2000 mg/L (refer to Appendix E, Table E-3).

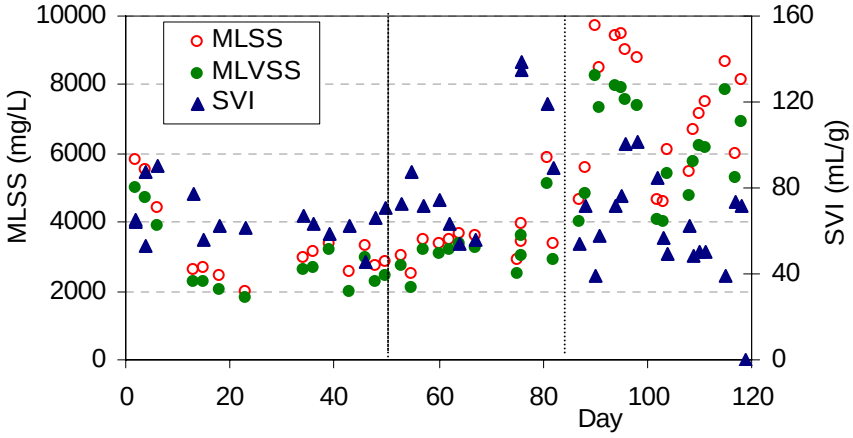


Figure 4.33 Biomass concentration and SVI at various OLRs

4.5.2.2 Sludge dewatering ability (CST)

The CST was 7.7±0.5, 41.9±1.6 and 27.2±14.0 s at 2, 4 and 8 kgCOD/m^3.d respectively. The increase in CST with OLR indicates that heterotrophic microorganisms which were dominant (or nitrifying microorganisms was minor) under high loading condition could deteriorate the sludge dewatering ability. At OLR of 8 kgCOD/m^3.d, the CST was highly fluctuated (it was high in the beginning and lower in the end of operating period). At this high OLR, we have observed progressive disintegration of the granules, which led to the situation of the CG-MBR to behave like the conventional activated sludge process. At this operational conditions severe foaming problems was noticed. Whereas, granular sludge reactor was maintained steadily at OLR range as high as 9-15 kgCOD/m^3.d (Moy et al., 2002; Tay et al., 2003, Thanh, 2005)

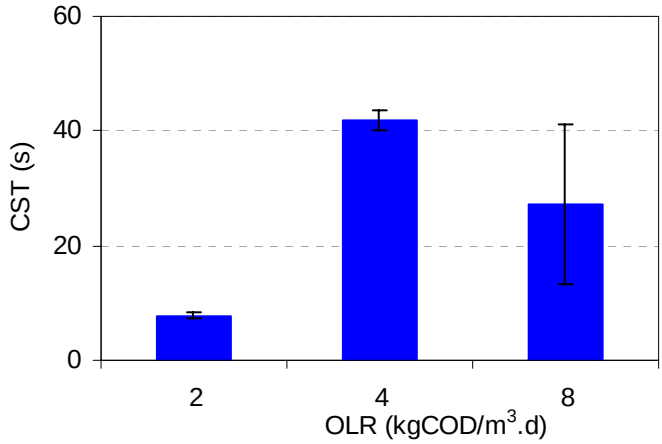


Figure 4.34 Sludge dewatering ability (CST) at various OLRs

4.5.2.3 Bound EPS of mixed liquor and fouling layer

Table 4.12 presents that bound EPS (bPS and bPN) concentration in both mixed liquor and fouling layer likely increased with elevated OLR. This result is in line with the result of Barker and Stuckey (1999). In other words, the bound EPS was low in the CG-MBR compared to that of the BG-MBR which was under the endogenous condition (refer to Table 4.5). It could be inferred that at high F/M ratio, microbes produced less bound

EPS. That could be the reason why granule could not be formed. Bound EPS was noticed to be a linking factor for aggregation of granules.

Moreover, the bound EPS of the fouling layer sludge had same trend with mixed liquor sludge (higher at higher OLRs). However it was lower compared to mixed liquor sludge at the same loading rate. The bound EPS of sludge in fouling layer was lower than that in mixed liquor that could be explained due to the dense biomass cake which needed more substrates. However limitation of substrate diffusion created endogenous condition inside the cake layer thus storage polymers were consumed. Hence, bound EPS of fouling layer was lower (Appendix E, Table E.4).

Table 4.12 Bound EPS of mixed liquor and fouling layer

bEPS (mg/gVSS)	OLR (kgCOD/m^3.d)		
	2	4	8
Mixed liquor:			
bPS	6.4 ± 0.6	13.8	7.3 ± 1.3
bPN	21.6 ± 3.6	27.2	21.3 ± 4.6
Fouling layer:			
bPS	3.9 ± 0.1	5.4	3.4 ± 0.5
bPN	11.0 ± 3.9	8.2	11.0 ± 0.4

4.5.3 Fouling propensity of CG-MBR at various operating OLRs

4.5.3.1 Fouling propensity of CG-MBR system

Figure 4.35 indicates that the in-situ fouling rate of the CG-MBR increased with the applied OLR. The typical fouling rates were 0.168, 0.292 and 0.818 kPa/d for OLR of 2, 4 and 8 kgCOD/m^3.d at the corresponding fouling duration of 52, 23 and 7 days (The cycle 2 of OLR 4 kgCOD/m^3.d and cycle 1 of OLR 8 kgCOD/m^3.d were not representative values because of the reactor operational problems which affected the fouling rate of membrane system, see Table 4.13). In this study, the fouling rate was found to be proportional to both organic loading rate and F/M ratio (Figure 4.36). As mentioned, the F/M ratio was 0.72, 1.06 and 1.37 d^{-1} for OLR of 2, 4 and 8 kgCOD/m^3.d respectively. The correlation between F/M ratio and loading rate was linear while that between F/M and fouling rate was seemingly a parabolic which has a minimum fouling rate between F/M ratio of 0.72 and 1.06 d (Appendix E, Table E.5). At high F/M ratio the membrane was indirect contact with high amount of organic matters which could have accelerated the fouling process. Likewise, Trussell et al. (2006) found that the specific flux (flux over TMP) was reduced at high F/M ratio.

Table 4.13 Fouling rates (kPa/d) of CG-MBR at various operating OLRs

Cycle	OLR (kgCOD/m^3.d)		
	2	4	8
Cycle 1	0.168 (0.914)	0.292 (0.792)	5.500 (0.791)*
Cycle 2		0.793 (0.710)*	0.818 (0.938)
Cycle 3			1.350 (0.977)
Cycle 4			7.254 (0.477)*
Cycle 5			2.014 (0.635)

Note: numbers in brackets are the R^2 of the linear slopes for TMP profiles; *Cycles were affected by operational problem (level sensor inactivated and membrane exposed to air) and caused serious fouling.

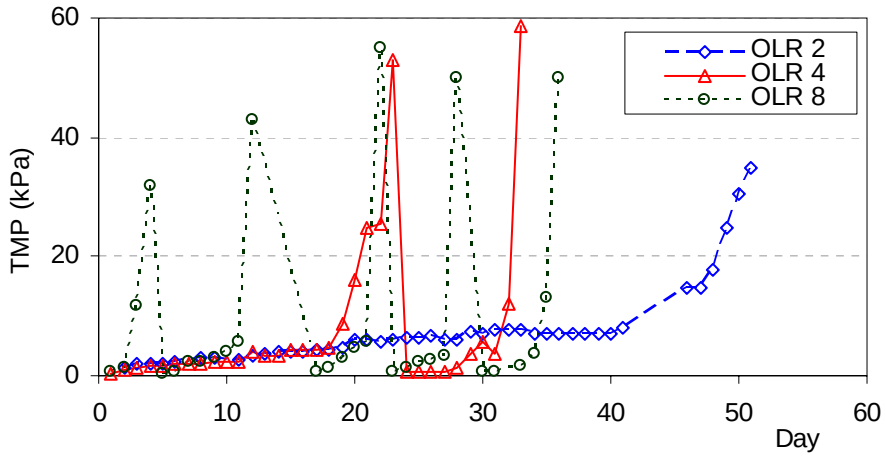


Figure 4.35 TMP profile of CG-MBR at various operating OLRs

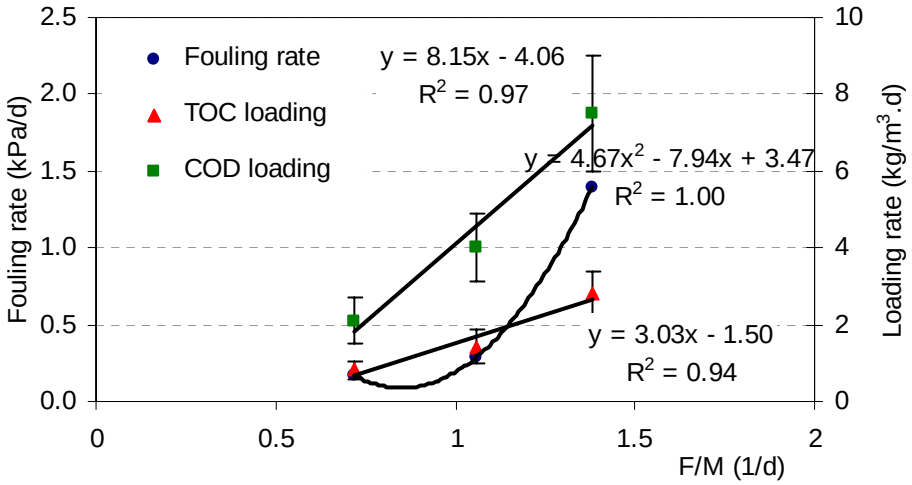


Figure 4.36 Correlation between F/M ratios vs fouling rate and loading rates

The resistances of sludge sample in the CG-MBR were caused mainly by cake deposition in which the cake resistance contributed more than 87.5% for all OLRs (Appendix E, Table E-6 & E-7). In other words, the fouling effect of soluble and colloidal fraction was found to be less at higher loading. In this study, no backwashing techniques were applied. The cake deposition on membrane module increased at the high loading rate. As observed, the cake layer covered fully the membrane module which was very different from the case of the BG-MBR. As a note, the white color of membrane was still seen in the membrane in the BG-MBR. This could be due to the difference in F/M ratio in the systems.

4.5.3.2 Characteristics of soluble matters in CG-MBR (DOC, sPS, sPN, UVA, SUVA)

Similar to the case of the BG-MBR, the soluble matters in terms of DOC, sPS and sPN in supernatant were higher in permeate of CG-MBR which is indicated by their deposition on membrane (Figure 4.37). The specific deposition rates of sPS, sPN and DOC are presented in Table 4.14. This observation reveals that the macromolecules probably were trapped and/or adsorbed by membrane during filtration (refer to Appendix E, Table E-8, E-9).

The UVA_{254} indicates the higher amount of double-bond compounds such as humic substances, protein-like materials, etc while SUVA stands for the presence of aromatic compounds such as humic substances (Her et al., 2007). In this system, UVA_{254} is slightly higher in supernatant compared to permeate at all OLRs but SUVA is reversed (Figure 4.38). This supports the fact that most of humic materials passed through membrane while the SMPs were rejected because the SUVA was high in permeate. Hence, the deposited substances were mostly sEPS (sPs, sPN). The specific deposition rates of soluble matters are presented in Table 4.15. The deposition rates of DOC increases with OLRs.

Table 4.14 Specific deposition rate of SMPs on membrane of CG-MBR at various OLRs

Deposition rates ($mg/L.m^2_{membrane}$)	OLR ($kgCOD/m^3.d$)		
	2	4	8
F/M (d^{-1})	0.72	1.06	1.37
DOC	16.3	18.2	17.2
sPS	13.1	11.3	13.7
sPN	5.6	5.0	6.7
sEPS	18.7	16.3	20.5

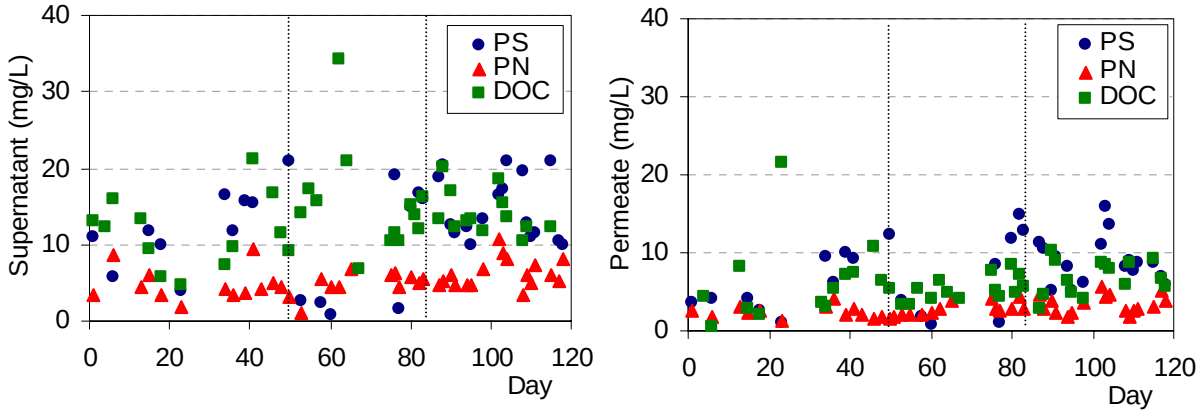


Figure 4.37 Soluble matters in supernatant and permeate of CG-MBR

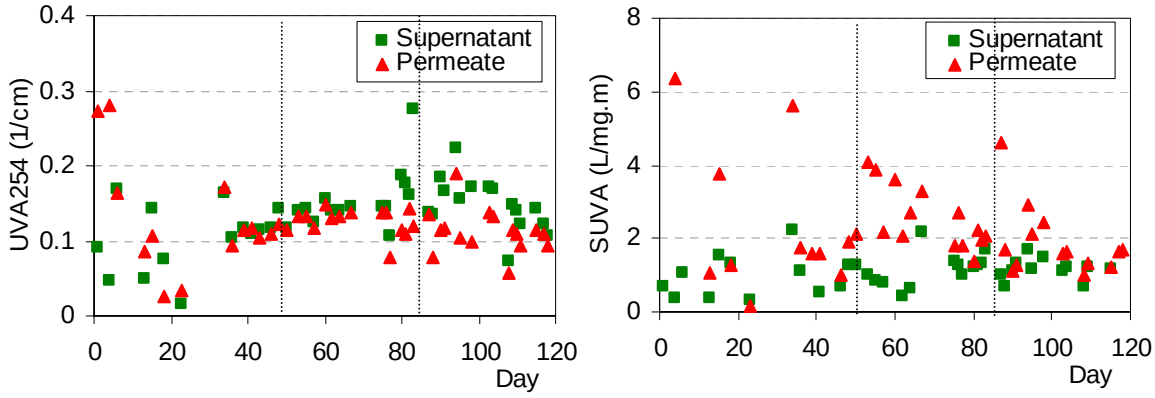


Figure 4.38 UVA_{254} and SUVA in supernatant and permeate of CG-MBR

4.5.4 Comparison of treatment performance between CG-MBR and BG-MBR systems

At the same OLR of 2 kgCOD/m^3.d, the granules were stable in the BG-MBR but broken after two weeks of operation in CG-MBR. Therefore, the CG-MBR functioned as same as the conventional submerged MBR. The organic removal capacity was similar in both systems. The BG-MBR shows the advantages in terms of high nitrogen removal and slow fouling rate as presented in Figure 4.39 and Table 4.15. The high nitrogen removal in BG-MBR was due to the simultaneous nitrification and denitrification of granules. The TN removal was 60 % and 20 % for the BG-MBR and the CG-MBR respectively. Moreover, the fouling rate was 6 times lower than that of the BG-MBR. The low F/M ratio in the BG-MBR caused less fouling. The F/M ratio was low due to the high biomass retention in the granulation reactor (SBAR). Granulated biomass could be maintained up to 18 gVSS/L while it reached a maximum of only 5 gVSS/L in the CG-MBR system at the same OLR. Thus, the low F/M ratio can be the reason for slow fouling rate in the BG-MBR system.

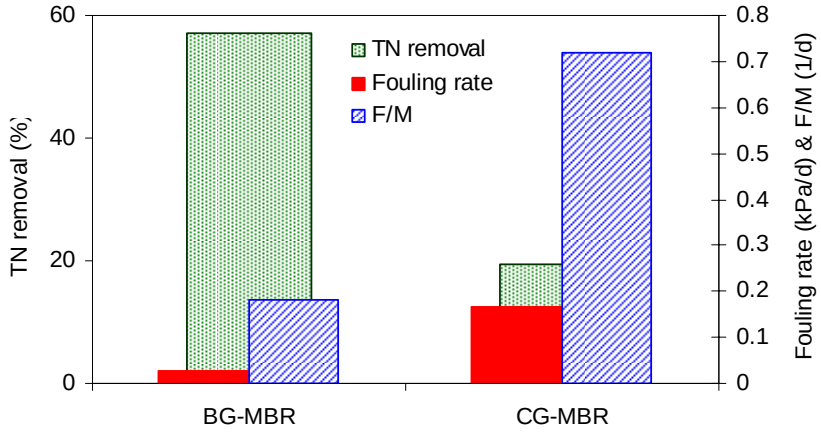


Figure 4.39 Treatment performances of BG-MBR and CG-MBR

The sludge characteristics and fouling behavior of the two systems are presented in Table 4.15. The bound EPS of the BG-MBR sludge was higher than that of the CG-MBR. This was due to the production of SMPs in MBR which treated SBAR effluent. The MBR coupling with the SBAR forming the BG-MBR system was always operated under endogenous condition due to low incoming substrate concentration.

Table 4.15 Comparison between BG-MBR and CG-MBR at OLR of 2 kgCOD/m^3.d

Parameters/systems	BG-MBR	CG-MBR
Granule stability	Yes	No
TOC (kgTOC/m^3.d)	0.86 ± 0.22	0.82 ± 0.24
F/M (d^{-1})	0.18 ±0.05	0.72 ± 0.28
TOC removal (%)	97.7 ± 1.4	97.8 ± 2.0
Ammonia removal (%)	99.2 ± 0.3	99.1 ± 0.6
TN removal (%)	57 ± 10	19 ± 5
Fouling rate (kPa/d)	0.027	0.168
Deposition rate (mgsPS/L.m^2)	11 ± 3	13 ± 7
Deposition rate (mgsPN/L.m^2)	8 ± 3	6 ± 5
Deposition rate (mgDOC/L.m^2)	17 ± 11	16 ± 10
SVI (mL/g)	93 ± 26	64 ± 11
CST (s)	14 ± 4	7.7 ± 0.5
Bound EPS: Mixed liquor		
mgPS/gVSS	18.4 ± 7.7	6.4 ± 0.6
mgPN/gVSS	39.9 ± 11.5	21.6 ± 3.6
mgEPS/gVSS	58.3	28.0
Bound EPS: Fouling layer		
mgPS/gVSS	10.5 ± 0.4	3.9 ± 0.1
mgPN/gVSS	19.9 ± 1.9	11 ± 3.9
mgEPS/gVSS	30.4	14.9

Table 4.16 summarizes the characteristics of different treatment systems. The BG-MBR shows better performance in terms of effluent quality and operating conditions compared to anaerobic reactor and submerged MBR. However its specific energy requirement is still high (1.6 kWh/m^3) which is almost double than that of submerged MBR. However, if the operated OLR is taken into account, the energy cost of the BG-MBR becomes comparable due to its operation at higher organic loading. The MBR was operated steadily at OLR range of 2-4 kgCOD/m^3.d while it was 9-15 kgCOD/m^3.d for the BG-MBR system.

Table 4.16 Sumary of operating conditions of various treatment systems

Items	Conventional anaerobic reactor	Submerged MBR	Granulation MBR (BG-MBR)
Operating mode	Continuous or batch operation	Continuous operation	+ Batch operation (granulation unit); + Continuous operation (MBR unit)
Operating temperature (^oC)	30-55	25-35	8-55* (de Kreuk et al., 2005; Zitomer et al., 2007)
Necessity of post-treatment	+ High NH_3, COD, SS in effluent; + Need to coupling with other aerobic processes.	+ Can be reused in processes; + Complete C removal and nitrification.	+ Can be reused in processes; + Complete C, N removal.
Odor generation	Yes	No	No
Specific energy requirement (kWh/m^3)**	0.1 (potential energy production from biogas)	0.9 (calculated at OLR of 2-4 $kgCOD/m^3.d$)	1.6 (calculated at OLR of 8 $kgCOD/m^3.d$)
Microbial population	Anaerobes	Aerobes	Anaerobes & Aerobes
Sludge failure	Possible	-	At high SRT, overgrowth of filaments (Liu and Liu, 2006)
Shock loading resistance	Possible for attached growth system	-	Yes
Start-up time (days)	100	10	30
Effluent SS (mg/L)	100-500	~ 0	~ 0
MLSS (g/L)	2-60	8-15	+ 10-18 g/L (granulation unit) + 2-4 g/L (MBR unit)
SRT (day)	10-300	15-30	10-100***
SVI (mL/g)	10-280	120-250 mL/g	10-40 mL/g
Settling velocity (m/h)	< 10	< 10	20-110 (higher for shell support granule)
Particle size (μm)	0.5- 8.0 mm (anaerobic granules) 0.3-200 (flocs)	1-250 (flocs)	0.5-9.0 mm (granules in granulation units) 0.3-301.7 (flocs in MBR)
Loading ($kg\ COD/m^3.d$)	Up to 40	< 8	2-30
Simultaneous nitrification/denitrification	No nitrification	Possible at high SRT & combined anoxic/aerobic MBR	1.76 mgTN/gVSS.h (due to spherical structure of granule)
Fouling potential	-	0.168 kPa/d	0.027 kPa/d

* The operating temperature range was applicable for granular sludge which exists in the granulation reactor (SBAR).
** Energy requirement is referred to Appendix F.
***The SRT in granulation reactor was a relative concept. Granules can be retained in reactor till disintegration.

Chapter 5

Conclusions and Recommendations

This study investigated the fouling behavior and nitrogen removal of granulation MBR systems, namely the batch granulation MBR (BG-MBR) and the continuous granulation MBR (CG-MBR). For the BG-MBR system, the simultaneous organic and nitrogen removal, granule stability and membrane fouling characteristics of granulation effluent were examined at the OLR of 2 kgCOD/m^3.d. In addition, SBAR sludge and effluent were characterized at 0.8 cm/s, 2.2cm/s and 0.6 cm/s under anoxic condition to understand their characteristics while operating under conventional and granulation operating conditions. For the CG-MBR, the treatment performance, granule stability and membrane fouling were investigated at the OLR of 2, 4 and 8 kgCOD/m^3.d. The treatment performance of both systems was comparatively evaluated. All the proposed research objectives were achieved and the conclusions drawn from this research are as follows:

5.1 Conclusions

Aerobic granules disintegrated under anaerobic operational condition in the BG-MBR system due to free ammonia production and low shear intensity. As a result, the system faced biomass reduction and serious membrane fouling. The batch granulation mode of the SBAR with high aeration rate followed by low aeration rate was found to be suitable for coupling with MBR. The BG-MBR system possessed both organic and nitrogen removal which almost occurred in the granulation reactor. The simultaneous nitrification and denitrification at OLR of 2 kgCOD/m^3.d was 47% or 22 mgTN/L.h (1.76 mgTN/gVSS.h) under aerobic operation (DO in reactor greater than 4 mg/L).

In the BG-MBR system, the release of soluble matters in MBR unit depends on the HRT which influences the fouling propensity and supernatant quality in the system. Soluble microbial products were found to be the main cause for fouling in the MBR of the system where polysaccharides were the dominant substances. The specific deposition loading on membrane surface during membrane filtration was 11 mg/L.m^2 and 8 mg/L.m^2 for soluble polysaccharides and soluble protein respectively. The amount of EPS deposited on membrane fibres during 78 days of operation was 20 μg/cm^2. Furthermore, the granule disintegration resulted in the release of soluble microbial products and increased the fouling propensity of the BG-MBR system.

The bound EPS and the ratio bPS/bPN of fouling layer was less than that of MBR sludge in the BG-MBR. The release of soluble matters from the fouling layer could contribute to the fouling phenomena as well. In addition, the particle size of MBR sludge was bigger than the nominal pore size of the membrane. Further, the big particles (158 μm) constituted the major volume of the mixed liquor sludge. The contribution to fouling due to suspended solids of sludge fractions was insignificant compared to colloidal and soluble fractions.

The results from the effect of aeration rates and anoxic operation on sludge and SBAR effluent indicate that aeration rate and anoxic/aerobic condition have certain impact on characteristics of sludge and SBAR effluent. The anoxic/aerobic conditions in the

SBAR enhanced the biomass retention, settling ability, denitrification and filterability of sludge. Resistance rate and specific cake resistance of SBAR effluent were higher than that of mixed liquor in anoxic/aerobic operation despite higher suspended solids in mixed liquor.

The resistance and irreversible resistance rates of SBAR effluent were increased at high aeration rate (2.2 cm/s) due to release of macromolecules (30-50 kDa) and small particles while the soluble microbial products were released at lower aeration rate (0.8 cm/s). Moreover, around 60% of the hydrophobic fraction was found at high aeration rate (2.2 cm/s) in the soluble fraction of SBAR effluent with low hydrophobic intensity. On the other hand, at the low aeration rate (0.6 cm/s) with anoxic growth, 20% of the hydrophobic fraction was found with high hydrophobic intensity.

The results of the CG-MBR system show that lack of shear stress in the MBR chamber disturbed the granule stability of the system where granule disintegration occurred due to stagnation of granules within membrane fibres. Fouling rate showed a second order increment with the increase in F/M ratio and first order with organic loading rate. Comparing the two systems, the BG-MBR exhibited better operational performance than CG-MBR in terms of granule stability, biomass retention, nitrogen removal and fouling propensity. Fouling rate of the BG-MBR system was 6 times lower than that of the CG-MBR or conventional MBR. The high biomass retention of granular sludge in the BG-MBR system (18 gVSS/L in the BG-MBR and 5 gVSS/L in the CG-MBR) decreases F/M ratio and thus results in low fouling propensity. Moreover, the BG-MBR is becoming attractive in terms of energy requirement at high OLR operation.

5.2 Recommendations

Based on the extensive experimental data obtained, several recommendations for future studies can be outlined:

* *Formation and stability of granule:*

1. Locate a method to form the aerobic granule with optimum size (0.3 – 1.2 mm) to enhance simultaneous nitrification and denitrification.

2. Carbon audit and bioactivity for granules and suspended solids can be examined through kinetic parameters.

3. To accelerate and stabilize the granulation process, methods namely support media addition, bridging polymer addition, aeration rates, cycle length, etc should be investigated and optimized.

4. Granule stability plays an important role to control fouling and to enhance simultaneous nitrification denitrification in the system. Thus it is important to further study the effect of sludge retention time on stability of granular sludge.

5. Granule stability based on various SRT of granular sludge can be investigated to maintain the stable operation of the SBAR. Besides the sludge removal through gravitational washout of the SBAR, the addition of periodical sludge removal is important in the granulation reactor to maintain the actual SRT. The sludge removal

methods can be further studied such as periodical removal of (a) mixed sludge (during aeration); (b) top sludge (after sludge bed settled) and (c) bottom sludge (after sludge bed settled).

6. Investigation of technically economical method to culture the granules should be done, especially with low aeration rate in the range of 1.0-1.5 cm/s. At present the energy usage for granule cultivation is high due to high aeration requirement. The duration between high aeration and low aeration in batch granulation system could be interesting. The percentage of low aeration rate could be 10, 20, 40, 60, and 80 % of the total cycle length of batch.

7. Granule formation in continuous or semi-continuous reactors to eliminate storage tank, transfer tank and pump can be focused. The semi-continuous granulation could be possible because it can maintain the cyclic feast and famine condition which is required for aggregation process.

8. Study on nitrogen removal through anamox granules for old leachate wastewater which contains high ammonia concentration.

9. Study on the necessary time of aerobic/anoxic to achieve complete nitrogen removal through ASM1 model. For this study, it needs to investigate specific kinetic data (maximum specific growth rates, decay constants, yield coefficients, and half-saturated constants), mass transfer constants and active biomass for granular sludge at various organic and nitrogen loading conditions.

** The Granulation MBR:*

1. The BG-MBR was found to be the potential process to couple granular sludge and MBR. The hydraulic retention time of MBR treating SBAR effluent, affects the release of soluble microbial products and as a result influences the fouling propensity of the system. Thus, the study on fouling and sludge characteristics needs further investigation.

2. Study on the possibility of granulation and fouling characteristics in sequencing batch MBR in which membrane functions as an ideal decanter in a sequencing batch reactor. The light fraction of sludge is removed periodically. This operating mode could be attractive because the feast/famine condition is always maintained in the system.

3. The membrane submerged in a specific zone of settling tank could be an interesting idea in terms of aeration energy and reduction in number of unit processes. In this integrated BG-MBR, the settling tank and MBR will be combined into one unit as “settler-combined MBR” (Figure 5.1). In this system, the granular sludge SBAR will be operated in batch operation. Its effluent flows to settler-combined MBR which includes settling chamber and membrane chamber. The settleable solids will be mostly settled at the bottom of the settling chamber and removed periodically. In the membrane chamber, air is supplied to avoid the anaerobic condition to control fouling. Furthermore, denitrification can be enhanced with a recirculation flow from membrane chamber to settling chamber. The ratio of recirculation and sludge removal could be optimized operating conditions with fouling propensity. This system is probably compact and less fouling compared to the investigated Batch granulation MBR.

4. In this research, hollow fibre membrane module was used to investigate the objectives where stagnation of granules in between the fibres occurred frequently. Hence, the CG-MBR with the submerged flat sheet membrane module should be used to avoid the blockage of granules and to maintain the shear stress in the reactor. This configuration might help to maintain granule stability in the semi-continuous system such as the proposed CG-MBR (see Figure 5.2).

5. The coupling between anamox process and MBR could form anamox granules as observed by Trigo et al. (2006). The further study on the fouling characteristics and nitrogen removal capacity of anamox granules in MBR could be attractive.

6. Soluble microbial products were found to play an important role in fouling of the granulation MBR. The study on the quality and quantity of soluble fraction through SEC-EEM and SEC-DOC would be useful for understanding the nature of foulants at certain operating conditions.

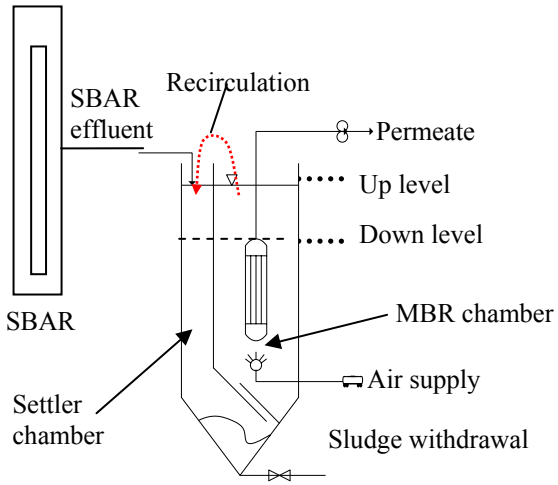


Figure 5.1 Proposed compact granulation MBR

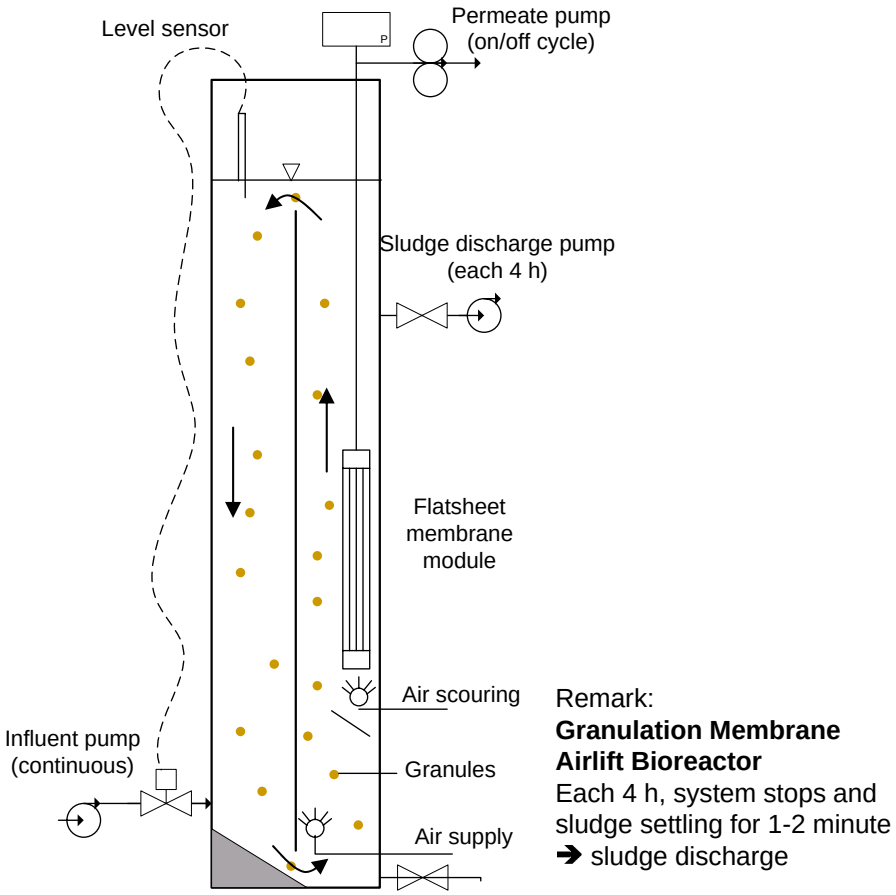


Figure 5.2 Proposed granulation membrane airlift bioreactor system

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Appendix A

Experimental Details

Figure A-1 Experimental set-up of the Batch Granulation MBR system (BG-MBR)

Figure A-2 From left to right (settler supernatant – MBR mixed liquor – membrane permeate)

Figure A-3 Matured granule in SBAR (left) and size measurement (right) of the BG-MBR

Figure A-4 Clean membrane (left) and fouled membrane (right) of the BG-MBR

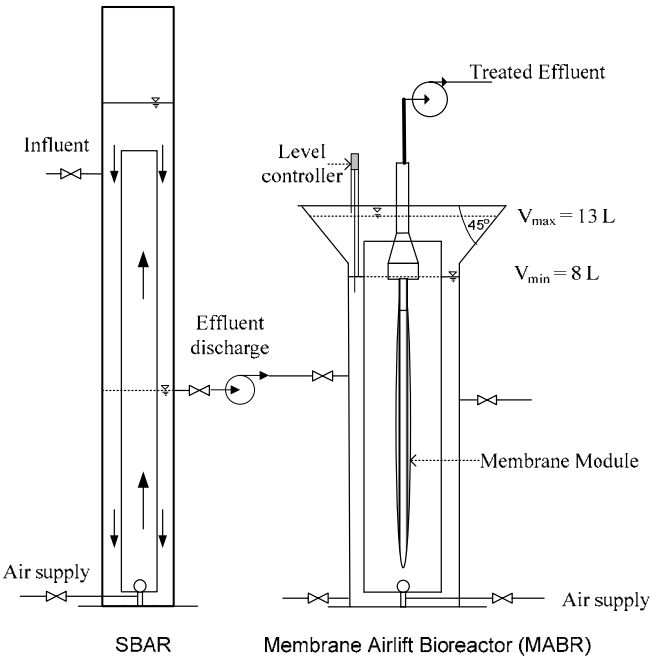


Figure A-5 Schematic diagram of the Batch Granulation Membrane Airlift Bioreactor (BG-MABR) (Experiment: Effect of granule stability on membrane fouling behaviour)

Figure A-6 View of SBAR (left) and MABR (right)

Table A-1 Operation and design details of SBAR and MABR

Reactor	Unit	SBAR	MABR
Size	mm x mm	Raiser: LxD = 1300 x 115 Down comer: lxd = 900 x 70	Height = 620 Refer Figure 3.5
Working volume	L	9.7	13.0
HRT	h	7.3	10.4
SRT	d	Depends on OLRs	20
Air flow rate	cm/s	Aeration: 1.7 & Low aeration: 0.1	0.3
Sludge removal	mL/d	Automatic removal with effluent	375
Flow rate	-	5.3 L/batch, 4 h/batch, 6 batch/d)	28 mL/min (7 on/ 3 off)
Flux	$L/m^2.h$	-	2.8

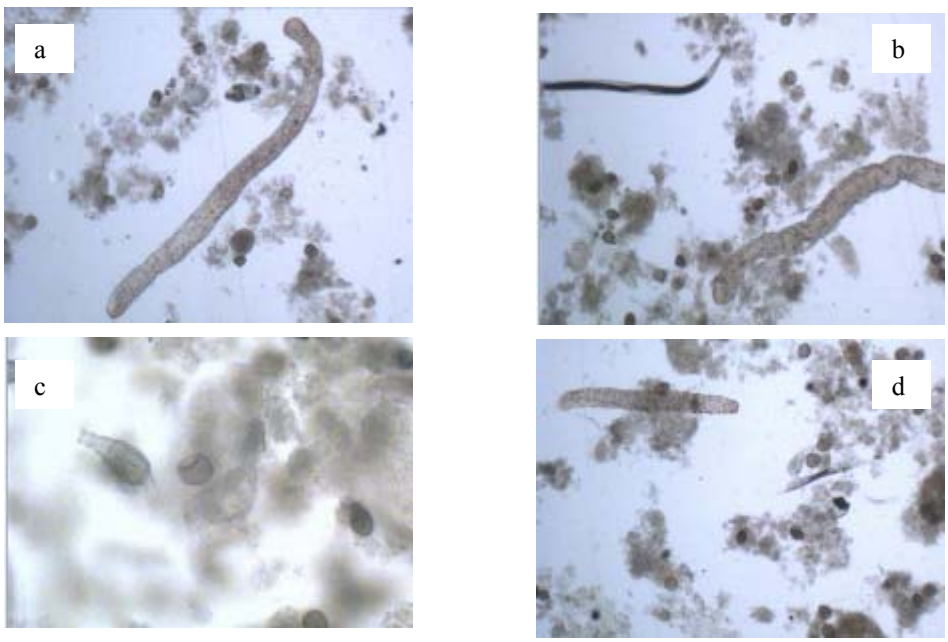


Figure A-7 Microbial community in MABR sludge (x10) (a) *Aeolosoma hemprichii* (b) *nematodes* (c) *rotifer* (d) Combined community

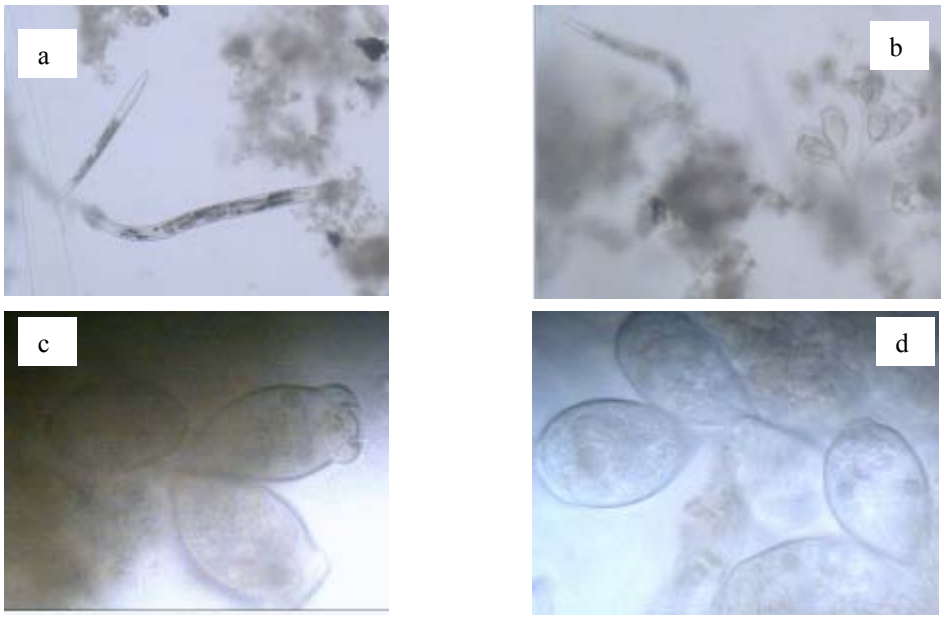


Figure A-8 Microbial communities in SBAR (granulation reactor) (a) *nematodes* (b) *rotifers & nematodes* (x10) (c) & (d) *rotifer* (x40)

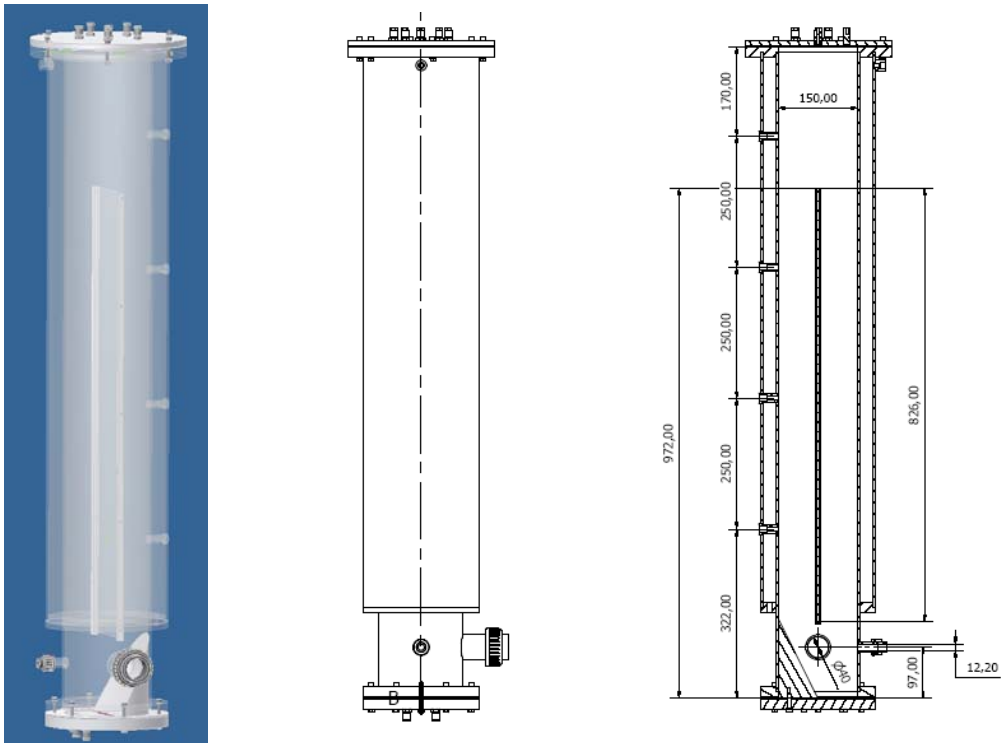


Figure A-9 SBAR configuration
(Study on effect of aeration rates on characteristics of SBAR effluent at INSA-Toulouse-France)

Figure A-10 Experimental setup at INSA-Toulouse-France

Figure A-11 Resistance rate measurement at INSA-Toulouse-France

Figure A-12 HPLC system for size exclusion and hydrophobic interaction chromatography at INSA-Toulouse-France

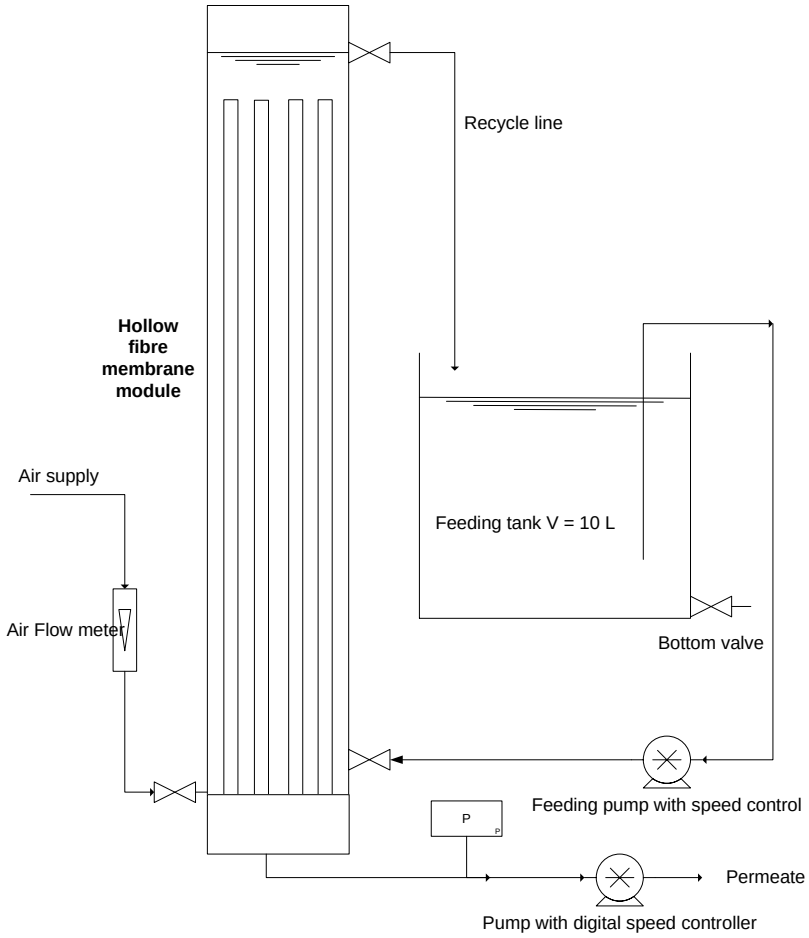
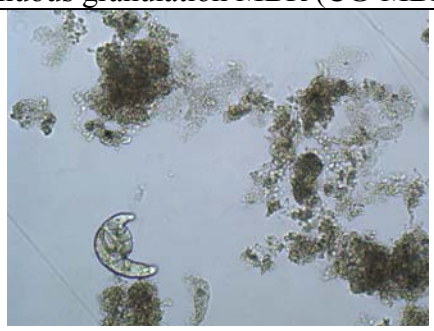


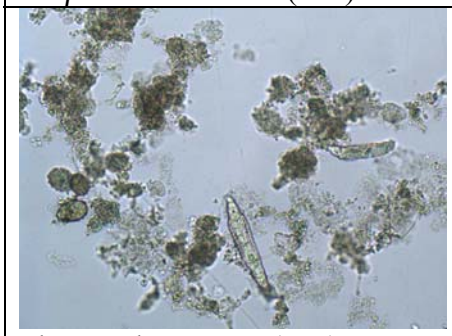
Figure A-13 Critical flux measurement system at INSA-Toulouse-France (Air scouring 250 L/h.$m^2_{membrane}$, recycle flow 5 L/h)

Figure A-14 Experimental setup of the continuous granulation MBR (CG-MBR) at AIT

rotifers and cilliates (x10)



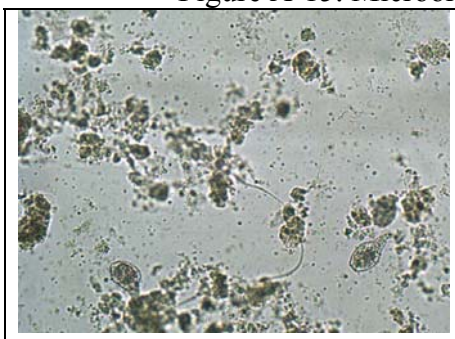
Broken granules, flocs and *rotifers* (x10)



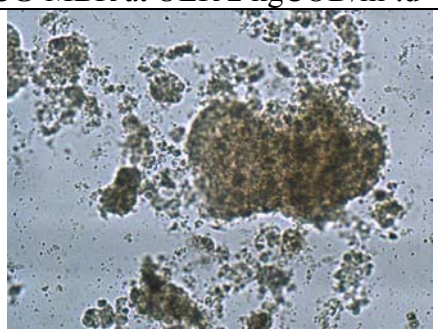
Flocs and protozoa (x10)

Aeolosoma hemprichii (x10)

Figure A-15. Microorganisms in CG-MBR at OLR 2 kgCOD/m^3.d



Debris and protozoa (less protozoa compared to mixed liquor sludge)



Lyzed granule and debris

Figure A-16 Microorganisms in fouled membrane of the CG-MBR at OLR 2 kgCOD/m^3.d

Appendix B

SOUR Measurement and Standard Curves

Procedure of SOUR measurement:

The configuration of respirometer was shown below. It had working volume of 0.9 L and a fluorescent DO meter which could measure at each 10 second interval was inserted into bulk liquid. Dissolved oxygen was recorded automatically and extracted after each test.

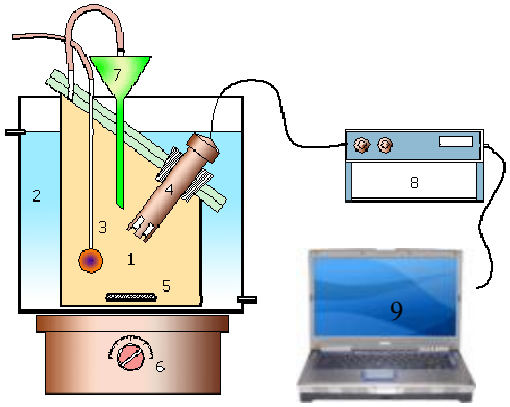


Figure B-1. Respirometer configuration

Experimental procedure was as follows:

a. DO probe preparation.

b. Filtered effluent of reactor is used to dilute liquid media.

c. Mixed liquor of granule/sludge is aerated for 30 minutes prior to testing to oxidize residual substrate remained. Test is conducted at temperature of 20^{o}C.

d. Biomass concentration in respirometer is measured in term of mgVSS/L.

e. Granular sludge is aerated at least one hour until endogenous respiration reached. DO in respirometer is saturated in range of 6-8.5 mg/L.

f. An accurate amount of concentrated substrate is added to obtain ration of So/Xo=0.01 -0.2 mg COD/mgVSS (Cech et al., 1984). DO in respirometer must be maintained higher than 2 mg/L.

g. DO reduction due to microbial respiration is monitored every ten seconds by DO meter. OUR, SOUR are calculated from DO change with time. When one dose of substrate finishes, the new one is injected into the cell and the new respirogram is recorded.

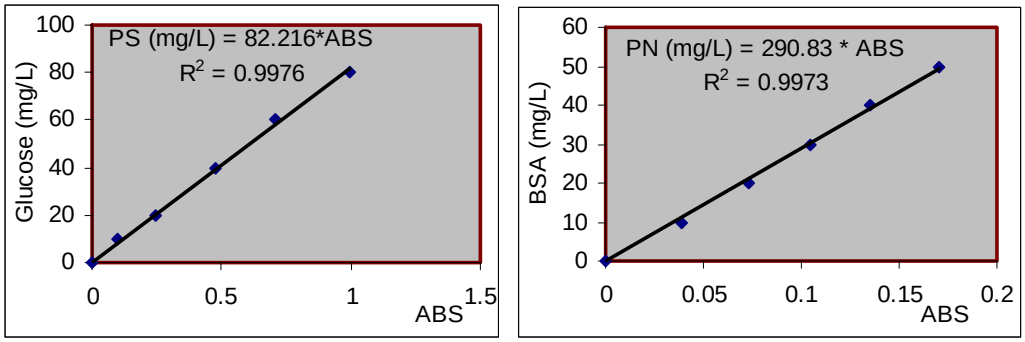


Figure B-2. Standard curves of Polysaccharides (left) and protein (right)

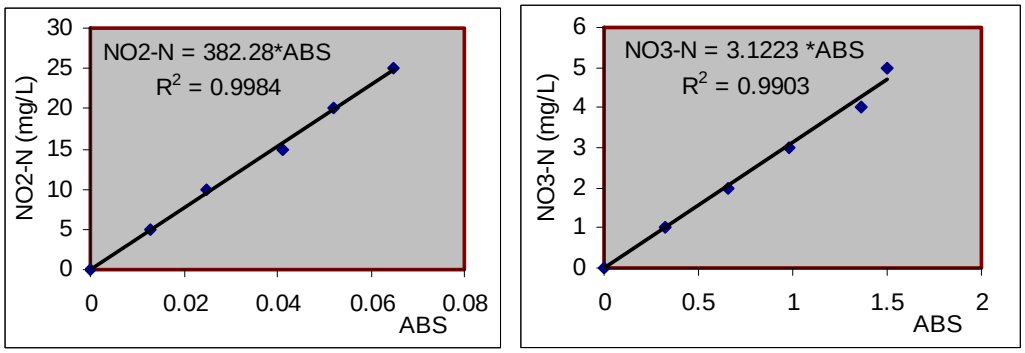


Figure B-3 Standard curves of nitrite (left) and nitrate (right)

Appendix C

Experimental Data
of
the Batch Granulation Membrane Bioreactor Systems (BG-MBR)
(Results at AIT)

Table C-1 Nitrogen removal of SBAR at scenarios (1,2,3,4)

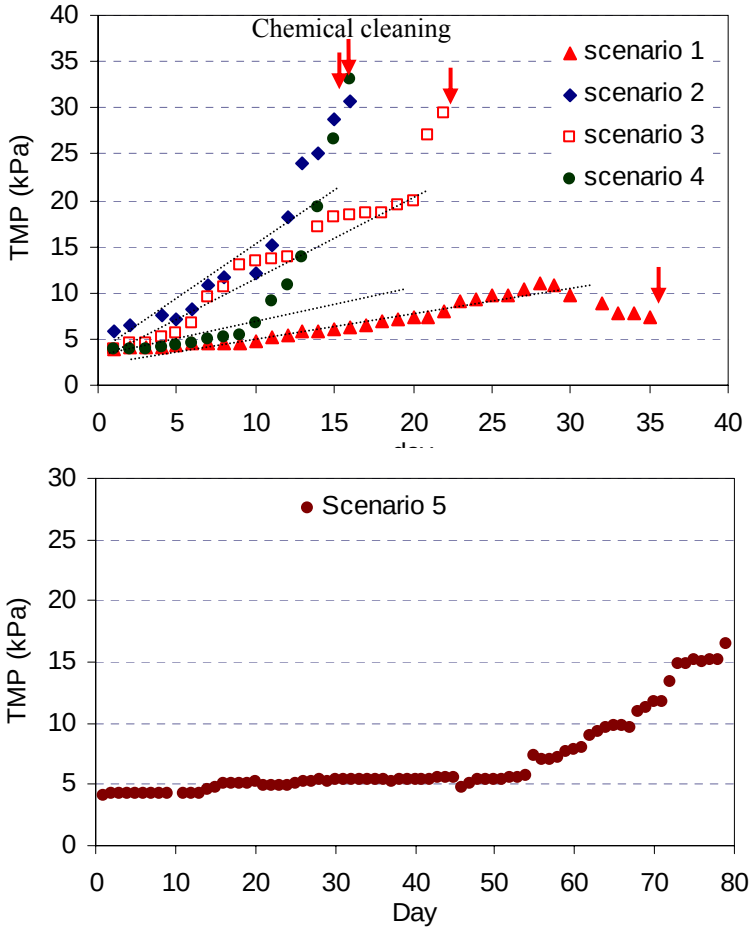
Day	NH_4-N (mg/L)					NO_2-N (mg/L)				NO_3-N				TN = NH_4-N + NO_2-N + NO_3-N			
	Inf	Settler	MBR sup	Per	NLR ($kgN/m^3.d$)	Inf	Settler	MBR sup	Per	Inf	Settler	MBR sup	Per	Inf	Settler	MBR sup	Per
Scenario 1																	
20	242.2	-	4.2	4.25	0.8	-	172.39	97.4	94.2	-	50.17	55.49	55.49	-	-	-	-
24	292.6	-	-	-	1.0	-	-	-	-	-	-	-	-	-	-	-	-
31	330.0	15.4	9.1	9.1	1.1	0.08	185.2	101.92	98.72	1.26	31.87	112.01	110.95	331.34	232.47	223.03	218.77
35	280.0	0.8	1.0	0.4	0.9	1.23	169.18	0.77	2.37	0.65	45.84	181.10	186.73	281.88	215.87	182.85	189.53
38	327.6	2.2	0.6	0.4	1.1	0.38	188.40	3.99	7.91	0.52	48.60	203.62	203.25	328.50	239.24	208.17	211.58
48	316.4	0.7	0.4	0.1	1.0	0.49	82.70	0.23	3.51	0.56	164.59	216.51	212.26	317.45	247.99	217.16	215.91
53	289.8	5.9	1.4	1.1	1.0	0.01	127.55	2.08	5.82	0.00	94.90	185.62	203.13	289.81	228.33	189.09	210.08
56	357.0	0.8	0.4	0.1	1.2	0.22	114.73	0.38	0.38	12.50	137.76	245.99	248.49	369.72	253.33	246.79	249.01
64	375.2	5.6	1.3	0.3	1.2	0.25	137.15	2.36	0.83	1.44	118.67	236.29	250.68	376.89	261.43	239.92	251.79
66	358.4	0.3	0.7	0.3	1.2	0.31	101.92	0.67	0.80	0.26	137.13	251.62	250.99	358.97	239.33	252.99	252.07
70	294.0	0.8	0.4	0.3	1.0	0.09	95.52	0.22	0.41	0.00	146.52	234.73	225.97	294.09	242.87	235.37	226.66
Scenario 2																	
73	-	-	-	-	-	0.01	162.78	4.96	6.08	0.00	74.88	188.12	242.23	-	237.66	193.08	248.31
74	350.0	3.4	0.3	0.1	1.1	0.05	140.36	0.12	0.67	0.00	82.11	202.82	223.78	350.05	225.82	203.22	224.59
77	414.4	0.6	0.1	0.1	1.4	0.08	92.31	0.35	0.41	0.00	80.82	162.78	175.61	414.48	173.70	163.27	176.16
88	333.2	43.4	1.4	1.1	1.1	0.49	112.88	3.17	0.28	0.00	0.00	166.85	147.14	333.69	156.28	171.41	148.47
Scenario 3																	
93	350.0	0.3	0.1	0.3	1.1	0.01	150.80	13.76	4.67	0.00	4.19	157.78	195.00	350.01	155.27	171.68	199.95
101	345.8	2.1	0.6	0.3	1.1	0.00	156.89	41.45	1.97	0.00	3.56	109.92	162.16	345.80	162.55	151.93	164.41
106	327.6	2.2	0.7	0.3	1.1	0.05	146.96	0.63	0.31	-	-	-	-	327.65	149.20	1.33	0.59
111	319.2	19.6	2.2	0.8	1.0	0.14	124.86	2.15	1.03	0.00	5.89	56.94	56.91	319.34	150.35	61.33	58.78
Scenario 4																	
127	344.4	1.0	1.1	0.4	1.1	0.01	199.90	53.75	56.96	0.76	5.12	139.32	153.40	345.17	206.00	194.19	210.77
131	338.8	1.3	0.4	0.3	1.1	0.03	194.81	79.79	86.19	1.04	13.68	31.74	31.43	339.87	209.74	111.95	117.90
133	330.4	1.1	0.6	0.4	1.1	0.02	204.70	-	-	0.89	20.93	31.41	31.37	331.31	226.76	-	-

Note: Inf: Influent; Settler: Settler supernatant; MBR sup: MBR supernatant; Per: membrane permeate

Table C-2 Organic and nitrogen removal of SBAR at scenarios (1,2,3,4)

Scenarios	NH_4-N influent (mg/L)	NH_4-N effluent (mg/L)	NO_2-N in effluent (mg/L)	NO_3-N effluent (mg/L)	NH_4-N removal (%)	TN removal (%)	DOC removal (%)
Scenario 1 (n=8)	324.8	2.2	127.1	111.8	99.7	26.2	97.0
Scenario 2 (n=3)	365.9	15.8	115.2	54.3	95.3	48.9	98.4
Scenario 3 (n=4)	335.7	6.1	144.9	4.5	98.1	54.3	97.9
Scenario 4 (n=3)	334.6	1.2	197.4	17.3	99.7	36.7	97.5

* n: number of me asurement



Scenarios	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5
Fouling rate (kPa/d)	0.188	1.029	1.091	0.269	0.027
R^2	0.956	0.900	0.958	0.841	0.767
Operating day	1-77	78-93	94-112	113-136	137-215

Figure C-1. TMP profile of various scenarios in MBR (dotted line: fouling rate)

Table C-3 Bound EPS ratio of sludge in SBAR and MBR

	SBAR		MBR		PS/PN ratio	
Bound EPS (mg/gVSS)	PS	PN	PS	PN	SBAR	MBR
Scenario 1	8.5 (1.4)	10.2 (2.4)	13.3 (2.9)	34.1 (13.2)	0.8	0.4
Scenario 2	5.6 (3.6)	9.2 (2.6)	11.3 (4.9)	34.8 (5.5)	0.6	0.3
Scenario 3	-	-	-	-	-	-
Scenario 4	10.1 (1.1)	25.4 (12.2)	12.2 (4.4)	61.3 (15.7)	0.4	0.2

Note: The number in brackets is SD. Date of Scenario 3 was missing.

Table C-4 Data set at steady state of nitrogen species in the BG-MBR (scenario 5)

Day	NH_4-N (mg/L)				NLR	NO_2-N (mg/L)				NO_3-N (mg/L)			
	Inf	Settler	MBR sup	Per	($kgN/m^3.d$)	Inf	Settler	MBR sup	Per	Inf	Settler	MBR sup	Per
139	198.8	0.7	0.3	0.1	0.7	0.17	113.42	1.12	1.05	1.01	8.22	100.37	98.31
141	204.4	0.8	0.1	0.1	0.7	0.05	97.40	0.35	2.01	1.26	3.55	103.53	102.40
143	204.4	2.1	0.3	0.1	0.7	0.01	87.80	0.13	1.28	0.58	1.67	76.50	76.25
146	154.0	0.3	0.1	0.1	0.5	0.02	97.40	-	-	0.26	0.05	-	-
150	201.6	1.7	0.3	0.3	0.7	0.05	86.19	1.23	0.38	0.87	0.42	79.50	81.25
151	204.4	1.7	0.6	0.4	0.7	0.16	47.76	0.13	0.01	0.46	3.18	51.22	57.23
152	190.4	2.2	0.1	0.1	0.6	0.07	68.58	0.05	0.05	0.57	6.59	71.99	80.63
155	196.0	2.5	0.3	0.1	0.6	0.01	54.16	0.17	0.11	0.01	6.03	69.62	73.62
156	196.0	2.2	0.3	0.1	0.6	0.01	54.16	0.14	0.11	0.01	3.72	67.11	69.62
157	168.0	1.4	0.3	0.3	0.6	0.01	66.98	0.02	0.38	0.52	4.97	64.24	65.99
158	198.8	1.1	0.3	0.3	0.7	0.03	79.79	0.22	0.17	0.98	5.76	75.55	79.07
169	170.8	1.7	0.0	0.0	0.6	0.01	71.78	0.18	0.16	0.00	6.93	79.33	75.29
176	170.8	1.4	0.3	0.3	0.6	0.11	66.98	0.01	0.01	1.24	5.83	74.25	70.47
179	176.4	1.7	0.3	0.3	0.6	0.23	66.98	0.01	0.42	1.34	5.17	75.42	73.73
183	179.2	1.7	0.1	0.1	0.6	0.02	68.69	0.42	0.41	0.01	4.52	68.55	69.66

Table C-5 Data set at steady state of organic/nitrogen removal in the BG-MBR (scenario 5)

Day	Inf	Settler	MBR sup	Perm	Inf	Settler	MBR sup	Per	Inf	Settler	MBR sup	Per	TN assimilation
	TN (mg/L)				TOC (mg/L)				COD (mg/L)				
139	206	102	104	105	182.6	6.7	16.7	3.1	494	24	51	15	16
141	205	92	77	78	138.0	6.6	19.7	1.9	375	24	59	11	13
150	203	88	81	82	250.0	8.8	16.2	2.7	675	30	50	13	22
151	205	53	52	58	282.0	13.2	6.1	2.0	760	42	23	12	25
152	191	77	72	81	297.6	7.2	9.4	2.6	802	26	31	13	27
155	196	60	68	70	217.0	6.4	10.6	5.4	586	23	35	21	20
157	169	73	65	67	269.5	7.1	8.5	2.0	727	25	29	12	24
158	200	87	76	80	332.7	8.4	16.0	6.1	896	29	49	23	30
169	171	80	80	75	207.9	3.9	6.3	0.8	562	17	23	8	19
176	172	74	75	71	327.9	4.8	5.5	1.2	883	19	21	10	29
183	178	74	76	74	345.3	1.9	3.1	1.3	929	11	15	10	31
Aver	190	78	75	76	259.1	6.8	10.7	2.6	699	25	35	13	23
SD	(14)	(13)	(12)	(11)	(63.7)	(2.8)	(5.3)	(1.6)	(170)	(7)	(14)	(4)	(6)

Note: Assume the nitrogen for cell yield is at COD:N:P =150:5:1 (COD = 1.055*(2.5337*TOC+5.9835))

Table C-6 SVI, MLSS and CST of the BG-MBR

Day	SVI (mL/g)		MLSS (mg/L)		CST (s)		
	SBAR	MBR	SBAR	MBR	SBAR	settler	MBR
30	29.0	66.8	10990	1760	8.8	8.3	15.5
36	21.2	57.4	13184	2440	-	-	-
40	-	-	13184	1950	-	-	-
45	38.3	63.8	9911	1880	9.7	7.3	19.8
51	-	-	10705	1920	8.5	7	90.2
59	-	54.2	-	1660	10.7	7.8	27.4
65	32.8	-	12212	-	11.9	10.8	8.8
72	27.6	20.7	10862	4340	8.2	7.5	12.0
106	37.6	81.7	5711	3060	11.1	-	14.1
141	25.5	73.8	8821	2440	11.2	8.8	12.5
150	22.7	84.7	10923	2480	8.8	9.2	12.8

157	16.9	71.4	11843	2100	10.9	7.4	15
164	24.5	77.2	11031	2460	8.3	13.9	6.5
171	21.3	74.6	14118	2840	10.1	7	13.5
178	21.1	153.8	18016	2100	9.8	7.9	13.9
185	27.1	116.4	11433	2920	11.6	8.4	23.2
194	30.5	105.0	10810	3620	11.7	8.6	15.7
198	28.9	85.6	10860	4440	10.6	8.2	16.2
206	31.3	85.8	10440	4440	10.4	7.9	13.6

Table C-7 Behavior of MBR supernatant under endogenous condition

Time (h)	0	1	2	3	4	5	6	7	8	9	10
sPS	3.5	4.1	4.9	5.7	5.1	4.9	5.9	5.7	5.7	6.4	6.3
sPS	3.3	3.9	5.1	5.4	4.8	5.0	6.4	5.4	5.6	6.8	6.2
Average sPS (mg/L)	3.4	4.0	5.0	5.6	5.0	4.9	6.2	5.5	5.6	6.6	6.2
SD	0.1	0.2	0.2	0.2	0.3	0.1	0.3	0.3	0.1	0.3	0.1
sPN	0.4	0.7	0.1	0.0	0.0	0.0	0.0	0.7	0.0	0.7	0.7
sPN	0.7	0.7	0.7	0.1	0.1	0.0	0.0	0.1	0.1	1.0	0.7
Average sPN (mg/L)	0.5	0.7	0.4	0.0	0.0	0.0	0.0	0.4	0.0	0.8	0.7
SD	0.2	0.0	0.4	0.0	0.0	0.0	0.0	0.4	0.0	0.2	0.0
pH (mg/L)	7.80	7.87	7.60	7.83	7.91	7.92	7.92	7.92	7.89	7.90	7.85
DOC (mg/L)	6.09	6.81	4.83	4.36	4.42	4.20	4.12	4.25	4.16	4.07	3.79
TN_{TOC} (mg/L)	36.2	37.0	39.7	37.9	38.5	38.8	39.3	39.7	40.6	41.0	41.5
SD	0.1	0.2	0.2	0.0	0.3	0.0	0.2	0.0	0.0	0.1	0.0
VSS_{TOC} (mg/L)	771		724		764		813		754		757
sPS/sEPS (%)	87	86	93	99	99	100	100	94	99	89	90

Table C-8 Typical particle size distribution of the BG-MBR system

MBR sludge			Settler supernatant			MBR sludge (nanosize)		
Size Low (um)	% volume	% number	Size Low (um)	% volume	% number	Size	% volume	% number
0.05	0	1.73	0.05	0	0	0.4	0	0
0.06	0	2.69	0.06	0	0	0.4632	0	0
0.07	0	3.37	0.07	0	0	0.5365	0	0
0.08	0	4.04	0.08	0	0	0.6213	0	0
0.09	0	4.84	0.09	0	0	0.7195	0	0
0.11	0	5.85	0.11	0	0	0.8332	0	0
0.13	0	7.05	0.13	0	0	0.9649	0	0
0.15	0.01	8.4	0.15	0	0	1.117	0	0
0.17	0.01	9.79	0.17	0	0	1.294	0	0
0.2	0.02	10.94	0.2	0	0	1.499	0	0
0.23	0.03	11.18	0.23	0	0	1.736	0	0
0.27	0.04	9.81	0.27	0	0	2.01	0	0
0.31	0.05	7.23	0.31	0.01	6.73	2.328	0	0
0.36	0.05	4.79	0.36	0.05	16.34	2.696	0	0
0.42	0.06	3.19	0.42	0.08	19.28	3.122	0	0
0.49	0.06	2.1	0.49	0.13	18.51	3.615	0	0
0.58	0.06	1.27	0.58	0.16	14.84	4.187	0	0
0.67	0.05	0.77	0.67	0.19	10.97	4.849	0	0
0.78	0.05	0.42	0.78	0.17	6.35	5.615	0	0
0.91	0.04	0.23	0.91	0.15	3.46	6.503	0	0
1.06	0.04	0.13	1.06	0.11	1.66	7.531	0	0

1.24	0.03	0.07	1.24	0.08	0.71	8.721	0	0
1.44	0.03	0.04	1.44	0.05	0.29	10.1	0	0
1.68	0.02	0.02	1.68	0.03	0.13	11.7	0	0
1.95	0.02	0.01	1.95	0.03	0.08	13.54	0	0
2.28	0.03	0.01	2.28	0.05	0.08	15.69	0	0
2.65	0.04	0.01	2.65	0.09	0.08	18.17	0	0
3.09	0.05	0.01	3.09	0.15	0.09	21.04	0	0
3.6	0.07	0.01	3.6	0.22	0.08	24.36	0	0
4.19	0.09	0	4.19	0.31	0.07	28.21	0	0
4.88	0.11	0	4.88	0.39	0.06	32.67	0	0
5.69	0.14	0	5.69	0.48	0.05	37.84	0	0
6.63	0.16	0	6.63	0.58	0.03	43.82	0	0
7.72	0.19	0	7.72	0.68	0.03	50.75	0	0
9	0.22	0	9	0.82	0.02	58.77	0	0
10.48	0.27	0	10.48	1	0.01	68.06	0	0
12.21	0.34	0	12.21	1.25	0.01	78.82	0	0
14.22	0.45	0	14.22	1.59	0.01	91.28	0	0
16.57	0.62	0	16.57	2.04	0.01	105.7	0	0
19.31	0.88	0	19.31	2.61	0.01	122.4	0	6.646
22.49	1.26	0	22.49	3.29	0.01	141.8	14.03	21.68
26.2	1.77	0	26.2	4.06	0	164.2	29.58	29.91
30.53	2.42	0	30.53	4.86	0	190.1	31.92	24.31
35.56	3.2	0	35.56	5.63	0	220.2	19.72	12.91
41.43	4.08	0	41.43	6.28	0	255	4.75	4.01
48.27	4.96	0	48.27	6.72	0	295.3	0	0.5332
56.23	5.75	0	56.23	6.92	0	342	0	0
65.51	6.38	0	65.51	6.87	0	396.1	0	0
76.32	6.77	0	76.32	6.63	0	458.7	0	0
88.91	6.91	0	88.91	6.29	0	531.2	0	0
103.58	6.79	0	103.58	5.54	0	615.1	0	0
120.67	6.46	0	120.67	4.68	0	712.4	0	0
140.58	5.97	0	140.58	3.83	0	825	0	0
163.77	5.41	0	163.77	3.06	0	955.4	0	0
190.8	4.79	0	190.8	2.44	0	1106	0	0
222.28	4.27	0	222.28	1.98	0	1281	0	0
258.95	3.84	0	258.95	1.67	0	1484	0	0
301.68	3.46	0	301.68	1.44	0	1718	0	0
351.46	3.1	0	351.46	1.25	0	1990	0	0
409.45	2.67	0	409.45	1.04	0	2305	0	0
477.01	2.15	0	477.01	0.82	0	1669	0	0
555.71	1.6	0	555.71	0.6	0	3091	0	0
647.41	1.07	0	647.41	0.38	0	3580	0	0
754.23	0.57	0	754.23	0.17	0	4145	0	0
	99.98	100.00		99.95	100.00	4801	0	0
						5560	0	0
						6439	0	0
						7456	0	0
						8635	0	0
						1.00E+04	0	0

Table C-9 Fouling behavior of biomass fractions (Scenario 5, Day 190, BG-MBR system)

Fraction of biomass	SS-CL-SL	CL-SL	SL
MFI (10^{-3} s/L^2)	69.1	60.6	33.8
%	12	39	49

Table C-10 Resistances of sludge fractions in MBR (Scenario 5, day 190, BG-MBR)

Biomass fractions	R_t	R_m	R_t-R_m
SS+Colloids+ Solutes	$3.1*10^{12}$	$1.4*10^{11}$	$2.9*10^{12}$
Colloids+Solute	$3.1*10^{12}$	$1.4*10^{11}$	$2.9*10^{12}$
Solutes	$2.7*10^{12}$	$1.4*10^{11}$	$2.6*10^{12}$

Table C-11 Soluble and bound EPS of the BG-MBR system (Scenario 5)

Day	VSS (mg/L)		Settler		MBR sup		Per		Bound EPS of SBAR sludge (mg/gVSS)		Bound EPS of MBR sludge (mg/gVSS)	
	SBAR	MBR	sPS (mg/L)	sPN (mg/L)	sPS (mg/L)	sPN (mg/L)	sPS (mg/L)	sPN (mg/L)	mgPS/g	mgPN/g	mgPS/g	mgPN/g
171	14907	3159	7.7	6.4	12.0	2.8	6.9	0.1	11.5	13.3	12.9	9.9
177	15771	1913	5.9	2.8	14.8	3.7	11.5	0.1	9.8	15.0	18.2	38.7
180	13859	2054	6.2	3.1	13.9	3.7	10.4	0.4	12.0	19.5	19.5	48.4
184	12702	2635	6.2	4.6	13.7	4.3	9.1	0.7	8.5	17.1	13.9	32.2
197	8206	1492	8.3	0.4	20.3	5.8	14.2	0.1	10.0	20.3	16.6	59.2
205	11677	2374	8.3	0.7	15.8	2.2	10.4	0.0	12.5	17.2	13.3	34.5
208	10938	1660	7.9	2.2	13.8	2.2	10.0	0.0	10.8	17.0	34.3	56.7
Aver.	12580	2184	7.2	2.9	14.9	3.5	10.3	0.2	10.7	17.0	18.4	39.9
SD	2370	582	1.1	2.1	2.6	1.3	2.2	0.2	1.4	2.4	7.7	11.5

Table C-12 Test of EPS deposition on membrane of the BG-MBR after 78 days (scenario 5)

No	ABS	EPS (mg/L)	Fibre length (cm)	Diameter (cm)	Membrane surface area (cm^2)	EPS deposition (μg/cm^2)
Fibre 1	0.638	47.54	38.7	0.04	4.861	20
Fibre 2	0.221	17.16	13.0	0.04	1.633	21
Fibre 3	0.188	14.76	13.0	0.04	1.633	18
Aver						20 ± 1

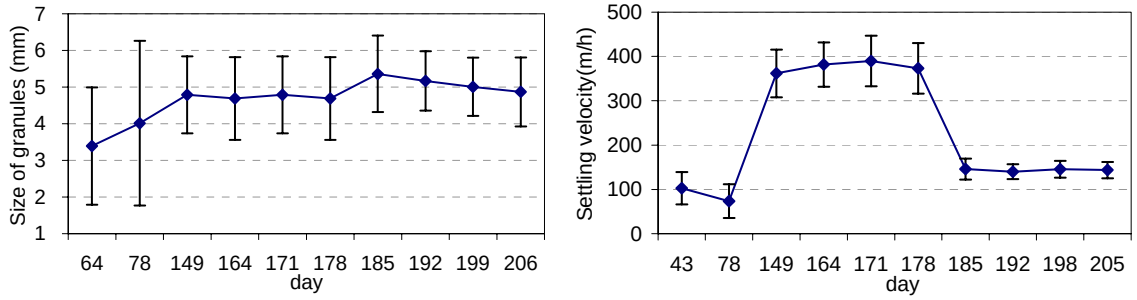


Figure C-2. Size and settling velocity of granules of with time

Table C-13 Critical flux analysis

Speed (rpm)	Flow rate Q (mL/min)			TMP (kPa)			TMPave (KPa)	dTMP/dt (kPa/min)	K ($L/m^2.h.bar$)	Flux ($L/h.m^2$)			Time (min)	Average flux ($L/h.m^2$)	Rt (m^{-1})
	Q1	Q2	Q3	1	2	3				F1	F2	F3			
30	68.0	68.0	68.0	4.3	4.3	4.3	4.3	0.000	225.9	9.71	9.71	9.71	20	9.7	2.00E+12
40	90.0	89.5	90.0	4.9	4.9	4.9	4.9	0.000	261.9	12.86	12.79	12.86	40	12.8	1.72E+12
50	111.0	111.0	111.0	5.5	5.6	5.6	5.6	0.005	284.9	15.86	15.86	15.86	60	15.9	1.58E+12
60	**130.0**	**129.5**	**130.0**	**6.0**	**6.3**	**6.3**	**6.2**	**0.015**	**299.2**	**18.57**	**18.50**	**18.57**	**80**	**18.5 (Critical flux)**	**1.51E+12**
70	151.0	151.0	151.0	6.9	7.2	7.2	7.1	0.015	303.8	21.57	21.57	21.57	100	21.6	1.48E+12
80	174.0	174.0	174.0	8.0	8.3	8.3	8.2	0.015	303.1	24.86	24.86	24.86	120	24.9	1.49E+12
90	198.0	198.0	198.0	9.0	9.2	9.4	9.2	0.020	307.5	28.29	28.29	28.29	140	28.3	1.47E+12
100	224.0	224.0	224.0	10.0	10.3	10.5	10.3	0.025	311.7	32.00	32.00	32.00	160	32.0	1.45E+12
90	199.0	198.5	199.0	9.4	9.4	9.4	9.4	0.000	302.2	28.43	28.36	28.43	180	28.4	1.49E+12
80	180.0	180.0	179.5	8.6	8.6	8.6	8.6	0.000	298.7	25.71	25.71	25.64	200	25.7	1.51E+12
70	154.0	154.0	154.0	7.8	7.8	7.8	7.8	0.000	282.1	22.00	22.00	22.00	220	22.0	1.60E+12
60	132.0	132.0	132.0	7.0	7.0	7.0	7.0	0.000	269.4	18.86	18.86	18.86	240	18.9	1.67E+12
50	112.0	112.0	112.0	6.4	6.4	6.4	6.4	0.000	250.0	16.00	16.00	16.00	260	16.0	1.80E+12
40	90.0	90.0	90.0	5.7	5.7	5.7	5.7	0.000	225.6	12.86	12.86	12.86	280	12.9	2.00E+12
30	68.0	68.0	68.0	5.0	5.0	5.0	5.0	0.000	194.3	9.71	9.71	9.71	300	9.7	2.32E+12

Table C-14 Nitrogen species from granule disintegration in SBAR effluent and MBAR supernatant (BG-MABR system)

	NH_4-N		NO_2-N		NO_3-N		TN		TOC	
Run 1: Granule disintegration										
Day	Inf	SBAR eff	Inf	SBAR eff	Inf	SBAR eff	Inf	SBAR eff	Inf	SBAR eff
4	159.6	1.1	0.03	61.01	1.12	49.53	160.8	111.7	282	0
7	156.8	1.4	0.02	25.25	0.00	86.99	156.8	113.6	278	0
11	173.6	1.1	0.08	30.62	0.32	88.90	174.0	120.6	279	0
18	168.0	0.8	0.03	1.83	0.63	110.91	168.7	113.6	236	0
28	154.0	1.1	0.01	7.37	0.00	102.30	154.0	110.8	233	9
33	159.6	0.3	0.01	20.95	0.26	84.26	159.9	105.5	258	6
Run 2: Granule formation										
43	173.6	36.4	0.00	70.81	0.48	18.08	174.1	125.3	372	6
46	168.0	42.0	0.00	72.47	0.68	12.80	168.7	127.3	420	6
49	173.6	39.2	0.02	65.82	1.98	7.51	175.6	112.5	402	8
53	170.8	28.0	0.02	67.49	1.48	9.23	172.3	104.7	414	4
60	182.0	19.6	0.16	85.77	1.05	9.48	183.2	114.8	379	10

Table C-15 Change of soluble microbial products from granule disintegration in SBAR effluent and MBAR supernatant (BG-MABR system)

Day	SBAR effluent			MABR		
	sPS (mg/L)	sPN (mg/L)	DOC (mg/L)	sPS (mg/L)	sPN (mg/L)	DOC (mg/L)
Run 1: Granule disintegration						
5	9.8	3.1	0.0	19.0	4.9	0.8
12	11.4	2.2	0.0	29.0	10.6	17.4
19	11.8	3.1	0.0	28.2	10.0	12.4
36	11.8	3.1	6.4	28.2	10.0	33.3
Run 2: Granule formation						
47	6.3	1.7	6.3	19.0	23.8	65.9
173	5.8	0.0	8.3	7.5	10.6	9.6
176	6.1	0.0	8.3	7.7	9.4	9.5
180	6.2	0.1	7.9	7.6	9.4	9.1
184	6.1	0.0	8.7	7.5	9.7	9.4
186	7.5	0.0	8.3	8.7	8.5	9.4
202	7.3	0.0	7.6	8.3	11.8	9.2
205	7.2	0.0	7.9	8.4	9.4	9.6

Table C-16 Operational guideline for granulation system

OPERATIONAL GUIDELINES FOR GRANULATION SYSTEM

*** Potential Operational Problems:**

- Granule disintegrates.
- White granules (fungus/filamentous granules) and black granules (has no biomass in the cores) appear in the SBAR.
- Granule is not stable (sudden breakage after formed).

*** Possible reasons for granule disintegration in SBAR:**

- Reduction in aeration rate less than 43 m/h (Tay et al., 2001)
- Long sludge retention time: Granule, itself has very long sludge age in SBAR due to its good settling velocity. This special operating condition makes fungi, actinomycet, filaments, yeast, and etc. outgrowth in the sludge microbial components. The fungus/filamentous granules appear dominant in reactor instead of bacterial granules (Liu and Liu, 2006).
- Feeding is not cyclic or regular. The operating organic loading rate (OLR) is less than 1 kgCOD/m^3.d.
- Shock of pH during batches: pH could be varied significantly at the beginning (high pH due to alkalinity production of denitrification process) and the end of batch (low pH due to alkalinity consumption of nitrification process)
- Deficiency of nutrient (N,P). The N and P constituents should be provided as required condition of conventional biological process (BOD:N:P=100:5:1).
- Free ammonia of higher than 23 mg/L inhibit granulation process (Yang et al., 2004).
- Long settling time.

*** Operational Modifications:**

- Aeration rate plays an important role in granulation process. The shear stress should be sufficient to achieve stable operation (aeration rate > 43 m/h).
- When white granule appears, the characteristics of granular sludge are changed. It might need to change the new seed sludge and restart the granulation process.
- In practice, it is necessary to control the sludge retention time by withdrawing the granular sludge (remove extra sludge while reactor is homogenously mixed). This should be done periodically like CASP. Moreover, the discharge of the light sludge fraction of the granulation process is maintained as usual.
- Control pH in the range of 7.2-8.2 depends on the operating purposes (by adjusting the amount of $NaHCO_3$ in the feeding solution or other alkaline/acid solutions). The pH higher than 9 causes system disturbance and granule breakage.
- Add more phosphorus if it is deficient. The ratio: P/COD > 1/100.
- Control the OLR higher than 1 kgCOD/m^3.d.
- Feeding should be introduced regularly for batch reactor. Shorter cycle length could enhance granulation process.
- Maintain settling time which is much higher than that of conventional activated sludge (v > 10 m/h). This will decide the duration of settling of the batch granulation reactors.

Appendix D

Experimental Data
Of
Effect of Aeration Rates on Characteristics of SBAR Effluent
(Results at INSA-Toulouse-France)

Table D-1 Resistance data of SBAR effluent at 0.8 cm/s (date 10/12/07)

SS-CL-SL				CL-SL				SL			
P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/s.L)	P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/s.L)	P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/m*L)
0.25	1.46E+08	5.28E+12	3.46E+12	0.25	5.78E+07	1.61E+12	1.68E+12	0.25	4.04E+07	7.37E+12	8.27E+11
0.50	2.42E+08	9.09E+12	2.92E+12	0.50	2.77E+08	1.18E+13	3.08E+12	0.50	2.42E+08	2.53E+12	2.48E+12
0.75	3.84E+08	1.05E+13	3.51E+12	0.75	3.37E+08	1.32E+13	3.33E+12	0.75	2.98E+08	5.69E+12	2.41E+12
1.00	6.93E+08	1.50E+13	5.43E+12	1.00	4.38E+08	1.14E+13	3.54E+12	0.95	4.50E+08	9.03E+12	3.33E+12
C (kg/m^3)	0.334										
α (m/kg)	2.72E+13										

	SS-CL-SL	CL-SL	SL
R_m	1.45E+12	1.71E+12	1.35E+12
R_t	1.74E+12	2.04E+12	1.67E+12
R_f	2.89E+11	3.25E+11	3.20E+11
R_{ir}	1.79E+11		
R_{rev}	1.09E+11		
V filtered, L	0.230	0.206	0.266
R_f/V (1/m.L)	1.26E+12	1.57E+12	1.20E+12

Table D-2 Resistance data of SBAR effluent at aeration rate of 2.2 cm/s (date 16/01/08)

SS-CL-SL				CL-SL				SL			
P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/m.L)	P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/m.L)	P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/m.L)
0.25	1.65E+08	1.31E+13	4.19E+12	0.25	3.69E+07	3.60E+11	9.16E+11	0.25	6.65E+07	1.04E+13	1.59E+12
0.55	4.95E+08	9.55E+12	5.97E+12	0.50	1.44E+08	3.93E+12	1.52E+12	0.50	2.43E+08	4.83E+12	2.86E+12
0.80	8.69E+08	2.14E+13	9.53E+12	0.75	5.17E+08	1.31E+13	4.77E+12	0.75	4.16E+08	9.43E+12	3.15E+12
1.00	1.17E+09	4.11E+13	1.38E+13	1.00	7.39E+08	1.57E+13	5.96E+12	1.05	5.60E+08	1.17E+13	4.15E+12
1.20	1.27E+09	5.35E+13	1.68E+13	1.25	8.61E+08	1.93E+13	7.08E+12	1.25	8.13E+08	1.88E+13	6.35E+12
C (kg/m^3)	0.474										
α (m/kg)	2.01E+13										

	SS-CL-SL	CL-SL	SL
R_m	1.60E+12	1.47E+12	1.49E+12
R_t	2.67E+12	2.25E+12	2.03E+12
R_f	1.07E+12	7.75E+11	5.36E+11
R_{ir}	8.98E+11		
R_{rev}	1.72E+11		
V filter (L)	0.241	0.314	0.346
R_f/V	1.07E+12	7.75E+11	5.36E+11

Table D-3 Resistance data of SBAR effluent at aeration rate of 0.6 cm/s with anoxic/aerobic stage (date 03/04/08)

SS-CL-SL				CL-SL				SL			
P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/m.L)	P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/m.L)	P (bar)	dR/dt (1/m.s)	α*C ($1/m^2$)	dR/dV (1/m.L)
0.25	1.37E+08	8.95E+12	2.99E+12	0.25	1.09E+08	0.00E+00	2.4E+12	0.25	1.56E+08	8.73E+12	3.23E+12
0.5	6.27E+08	2.08E+13	6.6E+12	0.5	4.73E+08	1.54E+13	5.52E+12	0.55	3.64E+08	9.14E+12	3.64E+12
0.75	7.86E+08	2.17E+13	7.38E+12	0.7	7.67E+08	2.09E+13	7.2E+12	0.7	6.11E+08	1.67E+13	5.35E+12
0.95	1.03E+09	2.63E+13	9.42E+12	1	8.90E+08	2.12E+13	7.35E+12	1	8.27E+08	1.85E+13	6.52E+12
1.25	1.15E+09	3.22E+13	1.04E+13	1.2	1.05E+09	2.68E+13	8.65E+12	1.25	1.04E+09	2.50E+13	8.42E+12
C (kg/m^3)	0.097										
a (m/kg)	2.14E+14										

	SS-CL-SL	CL-SL	SL
R_m	1.46E+12	1.42E+12	1.39E+12
R_t	2.27E+12	2.07E+12	2.03E+12
R_f	8.12E+11	6.50E+11	6.38E+11
R_{ir}	7.20E+11		
R_{rev}	9.21E+10		
V filtered (L)	0.292	0.307	0.331
R_f/V	2.78E+12	2.12E+12	1.92E+12

Table D-4 Typical resistance rate data and calculation for sample SS-CL-SL at filtration pressure of 0.25 bar and 0.5 bar (date 03/04/08)

viscosity 20°C, mPa.s			1.002		
viscosity at T°C			0.962		
pressure, bar			0.25		
t°C			21.6		
t, sec	V*, ml	V, mL	t/V, s/mL	J 20oC	Rm+Rf, 1/m
0	0.38	0		48.27	1.86E+12
5	0.61	0.23	21.74	55.51	1.62E+12
10	0.82	0.44	22.73	50.69	1.77E+12
15	1.06	0.68	22.06	57.93	1.55E+12
20	1.29	0.91	21.98	55.51	1.62E+12
25	1.52	1.14	21.93	55.51	1.62E+12
30	1.74	1.36	22.06	53.10	1.69E+12
35	1.96	1.58	22.15	53.10	1.69E+12
40	2.21	1.83	21.86	60.34	1.49E+12
45	2.44	2.06	21.84	55.51	1.62E+12
50	2.67	2.29	21.83	55.51	1.62E+12
55	2.9	2.52	21.83	55.51	1.62E+12
60	3.13	2.75	21.82	55.51	1.62E+12
65	3.36	2.98	21.81	55.51	1.62E+12
70	3.6	3.22	21.74	57.93	1.55E+12
75	3.83	3.45	21.74	55.51	1.62E+12
80	4.06	3.68	21.74	55.51	1.62E+12
85	4.3	3.92	21.68	57.93	1.55E+12
90	4.52	4.14	21.74	53.10	1.69E+12
95	4.76	4.38	21.69	57.93	1.55E+12
100	4.99	4.61	21.69	55.51	1.62E+12
105	5.22	4.84	21.69	55.51	1.62E+12
110	5.45	5.07	21.70	55.51	1.62E+12
115	5.68	5.3	21.70	55.51	1.62E+12
120	5.91	5.53	21.70	55.51	1.62E+12
125	6.15	5.77	21.66	57.93	1.55E+12
130	6.39	6.01	21.63	57.93	1.55E+12
135	6.61	6.23	21.67	53.10	1.69E+12
140	6.84	6.46	21.67	55.51	1.62E+12
145	7.07	6.69	21.67	55.51	1.62E+12
150	7.31	6.93	21.65	57.93	1.55E+12
155	7.55	7.17	21.62	57.93	1.55E+12
160	7.77	7.39	21.65	53.10	1.69E+12
165	8	7.62	21.65	55.51	1.62E+12
170	8.23	7.85	21.66	55.51	1.62E+12
175	8.46	8.08	21.66	55.51	1.62E+12
180	8.7	8.32	21.63	57.93	1.55E+12
185	8.92	8.54	21.66	53.10	1.69E+12
190	9.15	8.77	21.66	55.51	1.62E+12
195	9.38	9	21.67	55.51	1.62E+12
200	9.61	9.23	21.67	55.51	1.62E+12

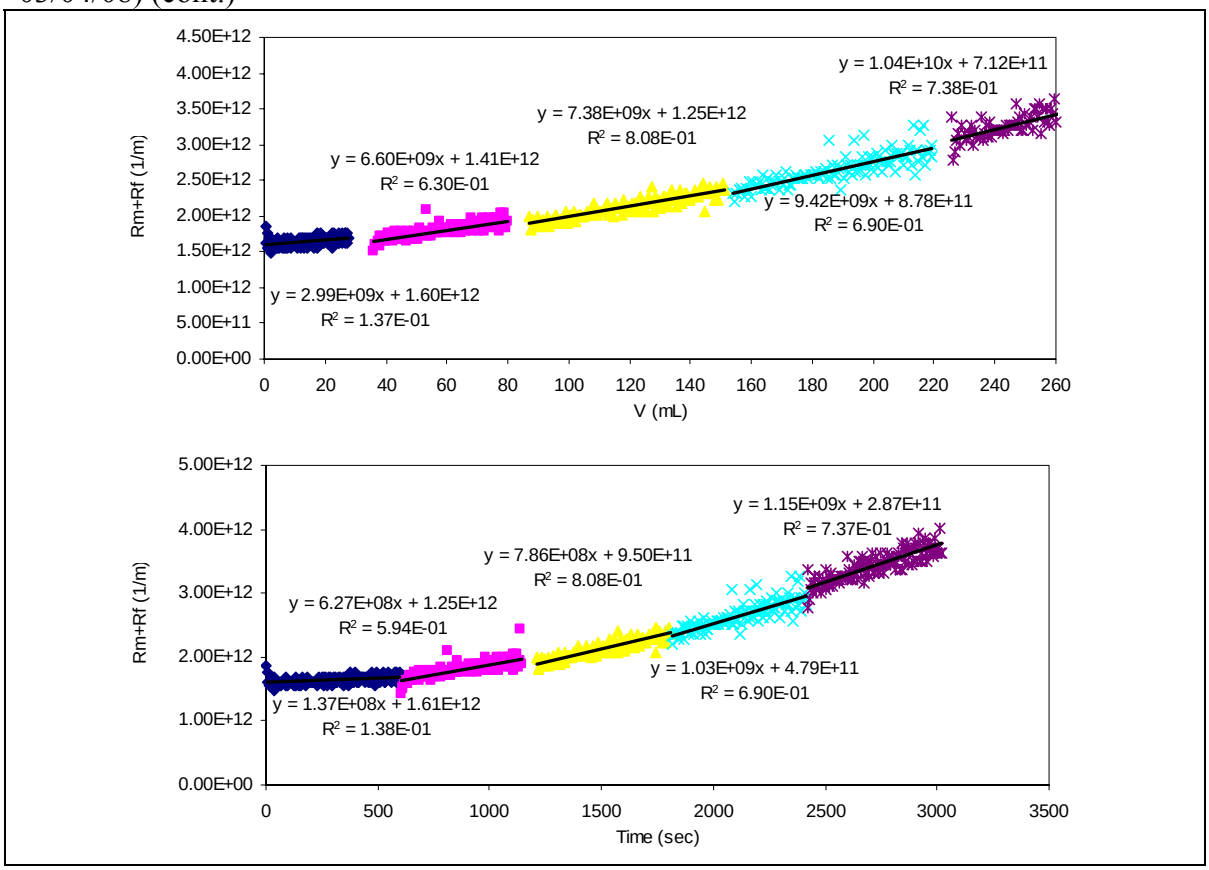
viscosity at T°C			0.951		
pressure, bar			0.5		
t°C			22.1		
t, sec	V*, ml	V, mL	t/V, s/mL	J 20oC	Rm+Rf, 1/m
605	35.02	0		126.41	1.42E+12
610	35.51	0.49	10.20	116.87	1.54E+12
615	36.01	0.99	10.10	119.26	1.51E+12
620	36.48	1.46	10.27	112.10	1.60E+12
625	36.95	1.93	10.36	112.10	1.60E+12
630	37.41	2.39	10.46	109.72	1.64E+12
635	37.89	2.87	10.45	114.49	1.57E+12
640	38.33	3.31	10.57	104.95	1.71E+12
645	38.77	3.75	10.67	104.95	1.71E+12
650	39.23	4.21	10.69	109.72	1.64E+12
655	39.69	4.67	10.71	109.72	1.64E+12
660	40.15	5.13	10.72	109.72	1.64E+12
665	40.58	5.56	10.79	102.56	1.75E+12
670	41.03	6.01	10.82	107.33	1.67E+12
675	41.47	6.45	10.85	104.95	1.71E+12
680	41.92	6.9	10.87	107.33	1.67E+12
685	42.38	7.36	10.87	109.72	1.64E+12
690	42.8	7.78	10.93	100.18	1.79E+12
695	43.24	8.22	10.95	104.95	1.71E+12
700	43.69	8.67	10.96	107.33	1.67E+12
705	44.13	9.11	10.98	104.95	1.71E+12
710	44.58	9.56	10.98	107.33	1.67E+12
715	45	9.98	11.02	100.18	1.79E+12
720	45.44	10.42	11.04	104.95	1.71E+12
725	45.88	10.86	11.05	104.95	1.71E+12
730	46.32	11.3	11.06	104.95	1.71E+12
735	46.78	11.76	11.05	109.72	1.64E+12
740	47.2	12.18	11.08	100.18	1.79E+12
745	47.63	12.61	11.10	102.56	1.75E+12
750	48.06	13.04	11.12	102.56	1.75E+12
755	48.51	13.49	11.12	107.33	1.67E+12
760	48.93	13.91	11.14	100.18	1.79E+12
765	49.36	14.34	11.16	102.56	1.75E+12
770	49.79	14.77	11.17	102.56	1.75E+12
775	50.22	15.2	11.18	102.56	1.75E+12
780	50.67	15.65	11.18	107.33	1.67E+12
785	51.08	16.06	11.21	97.79	1.84E+12
790	51.5	16.48	11.23	100.18	1.79E+12
795	51.93	16.91	11.24	102.56	1.75E+12
800	52.36	17.34	11.25	102.56	1.75E+12
805	52.8	17.78	11.25	104.95	1.71E+12
810	53.16	18.14	11.30	85.87	2.09E+12

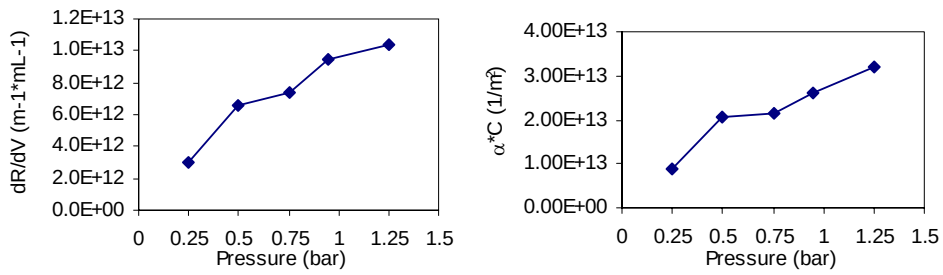
205	9.84	9.46	21.67	55.51	1.62E+12	815	53.6	18.58	11.30	104.95	1.71E+12
210	10.07	9.69	21.67	55.51	1.62E+12	820	54.02	19	11.32	100.18	1.79E+12
215	10.3	9.92	21.67	55.51	1.62E+12	825	54.45	19.43	11.32	102.56	1.75E+12
220	10.53	10.15	21.67	55.51	1.62E+12	830	54.89	19.87	11.32	104.95	1.71E+12
225	10.76	10.38	21.68	55.51	1.62E+12	835	55.3	20.28	11.34	97.79	1.84E+12
230	11	10.62	21.66	57.93	1.55E+12	840	55.72	20.7	11.35	100.18	1.79E+12
235	11.22	10.84	21.68	53.10	1.69E+12	845	56.15	21.13	11.36	102.56	1.75E+12
240	11.45	11.07	21.68	55.51	1.62E+12	850	56.58	21.56	11.36	102.56	1.75E+12
245	11.68	11.3	21.68	55.51	1.62E+12	855	57.01	21.99	11.37	102.56	1.75E+12
250	11.9	11.52	21.70	53.10	1.69E+12	860	57.4	22.38	11.39	93.02	1.93E+12
255	12.14	11.76	21.68	57.93	1.55E+12	865	57.83	22.81	11.40	102.56	1.75E+12
260	12.36	11.98	21.70	53.10	1.69E+12	870	58.25	23.23	11.41	100.18	1.79E+12
265	12.58	12.2	21.72	53.10	1.69E+12	875	58.68	23.66	11.41	102.56	1.75E+12
270	12.81	12.43	21.72	55.51	1.62E+12	880	59.1	24.08	11.42	100.18	1.79E+12
275	13.05	12.67	21.70	57.93	1.55E+12	885	59.51	24.49	11.43	97.79	1.84E+12
280	13.27	12.89	21.72	53.10	1.69E+12	890	59.92	24.9	11.45	97.79	1.84E+12
285	13.49	13.11	21.74	53.10	1.69E+12	895	60.34	25.32	11.45	100.18	1.79E+12
290	13.72	13.34	21.74	55.51	1.62E+12	900	60.76	25.74	11.46	100.18	1.79E+12
295	13.96	13.58	21.72	57.93	1.55E+12	905	61.19	26.17	11.46	102.56	1.75E+12
300	14.18	13.8	21.74	53.10	1.69E+12	910	61.59	26.57	11.48	95.41	1.88E+12
305	14.41	14.03	21.74	55.51	1.62E+12	915	62	26.98	11.49	97.79	1.84E+12
310	14.63	14.25	21.75	53.10	1.69E+12	920	62.42	27.4	11.50	100.18	1.79E+12
315	14.86	14.48	21.75	55.51	1.62E+12	925	62.85	27.83	11.50	102.56	1.75E+12
320	15.09	14.71	21.75	55.51	1.62E+12	930	63.26	28.24	11.51	97.79	1.84E+12
325	15.31	14.93	21.77	53.10	1.69E+12	935	63.66	28.64	11.52	95.41	1.88E+12
330	15.54	15.16	21.77	55.51	1.62E+12	940	64.07	29.05	11.53	97.79	1.84E+12
335	15.76	15.38	21.78	53.10	1.69E+12	945	64.5	29.48	11.53	102.56	1.75E+12
340	15.99	15.61	21.78	55.51	1.62E+12	950	64.9	29.88	11.55	95.41	1.88E+12
345	16.23	15.85	21.77	57.93	1.55E+12	955	65.31	30.29	11.55	97.79	1.84E+12
350	16.45	16.07	21.78	53.10	1.69E+12	960	65.71	30.69	11.57	95.41	1.88E+12
355	16.67	16.29	21.79	53.10	1.69E+12	965	66.12	31.1	11.58	97.79	1.84E+12
360	16.9	16.52	21.79	55.51	1.62E+12	970	66.54	31.52	11.58	100.18	1.79E+12
365	17.12	16.74	21.80	53.10	1.69E+12	975	66.95	31.93	11.59	97.79	1.84E+12
370	17.36	16.98	21.79	57.93	1.55E+12	980	67.36	32.34	11.60	97.79	1.84E+12
375	17.57	17.19	21.82	50.69	1.77E+12	985	67.74	32.72	11.61	90.64	1.98E+12
380	17.79	17.41	21.83	53.10	1.69E+12	990	68.15	33.13	11.62	97.79	1.84E+12
385	18.02	17.64	21.83	55.51	1.62E+12	995	68.57	33.55	11.62	100.18	1.79E+12
390	18.25	17.87	21.82	55.51	1.62E+12	1000	68.98	33.96	11.63	97.79	1.84E+12
395	18.48	18.1	21.82	55.51	1.62E+12	1005	69.38	34.36	11.64	95.41	1.88E+12
400	18.69	18.31	21.85	50.69	1.77E+12	1010	69.77	34.75	11.65	93.02	1.93E+12
405	18.92	18.54	21.84	55.51	1.62E+12	1015	70.17	35.15	11.66	95.41	1.88E+12
410	19.15	18.77	21.84	55.51	1.62E+12	1020	70.59	35.57	11.67	100.18	1.79E+12
415	19.38	19	21.84	55.51	1.62E+12	1025	71	35.98	11.67	97.79	1.84E+12
420	19.6	19.22	21.85	53.10	1.69E+12	1030	71.39	36.37	11.69	93.02	1.93E+12
425	19.82	19.44	21.86	53.10	1.69E+12	1035	71.77	36.75	11.70	90.64	1.98E+12
430	20.04	19.66	21.87	53.10	1.69E+12	1040	72.19	37.17	11.70	100.18	1.79E+12
435	20.26	19.88	21.88	53.10	1.69E+12	1045	72.59	37.57	11.71	95.41	1.88E+12
440	20.49	20.11	21.88	55.51	1.62E+12	1050	72.99	37.97	11.72	95.41	1.88E+12
445	20.71	20.33	21.89	53.10	1.69E+12	1055	73.39	38.37	11.73	95.41	1.88E+12
450	20.93	20.55	21.90	53.10	1.69E+12	1060	73.77	38.75	11.74	90.64	1.98E+12
455	21.15	20.77	21.91	53.10	1.69E+12	1065	74.18	39.16	11.75	97.79	1.84E+12
460	21.38	21	21.90	55.51	1.62E+12	1070	74.58	39.56	11.75	95.41	1.88E+12
465	21.61	21.23	21.90	55.51	1.62E+12	1075	74.98	39.96	11.76	95.41	1.88E+12
470	21.83	21.45	21.91	53.10	1.69E+12	1080	75.37	40.35	11.77	93.02	1.93E+12
475	22.04	21.66	21.93	50.69	1.77E+12	1085	75.75	40.73	11.78	90.64	1.98E+12
480	22.26	21.88	21.94	53.10	1.69E+12	1090	76.16	41.14	11.79	97.79	1.84E+12
485	22.5	22.12	21.93	57.93	1.55E+12	1095	76.56	41.54	11.80	95.41	1.88E+12
490	22.73	22.35	21.92	55.51	1.62E+12	1100	76.94	41.92	11.81	90.64	1.98E+12
495	22.94	22.56	21.94	50.69	1.77E+12	1105	77.36	42.34	11.81	100.18	1.79E+12

500	23.16	22.78	21.95	53.10	1.69E+12
505	23.38	23	21.96	53.10	1.69E+12
510	23.61	23.23	21.95	55.51	1.62E+12
515	23.83	23.45	21.96	53.10	1.69E+12
520	24.05	23.67	21.97	53.10	1.69E+12
525	24.26	23.88	21.98	50.69	1.77E+12
530	24.49	24.11	21.98	55.51	1.62E+12
535	24.72	24.34	21.98	55.51	1.62E+12
540	24.94	24.56	21.99	53.10	1.69E+12
545	25.16	24.78	21.99	53.10	1.69E+12
550	25.37	24.99	22.01	50.69	1.77E+12
555	25.59	25.21	22.02	53.10	1.69E+12
560	25.82	25.44	22.01	55.51	1.62E+12
565	26.04	25.66	22.02	53.10	1.69E+12
570	26.26	25.88	22.02	53.10	1.69E+12
575	26.47	26.09	22.04	50.69	1.77E+12
580	26.7	26.32	22.04	55.51	1.62E+12
585	26.91	26.53	22.05	50.69	1.77E+12
590	27.14	26.76	22.05	55.51	1.62E+12
595	27.35	26.97	22.06	50.69	1.77E+12
600	27.57	27.19	22.07	53.10	1.69E+12

1110	77.73	42.71	11.82	88.25	2.04E+12
1115	78.14	43.12	11.83	97.79	1.84E+12
1120	78.53	43.51	11.84	93.02	1.93E+12
1125	78.9	43.88	11.85	88.25	2.04E+12
1130	79.31	44.29	11.85	97.79	1.84E+12
1135	79.7	44.68	11.86	93.02	1.93E+12
1140	80.01	44.99	11.89	73.94	2.43E+12
1145	80.41	45.39	11.90	95.41	1.88E+12

Table D-4 Typical calculation for resistance rate for SBAR effluent (SS-CL-SL) (date 03/04/08) (cont.)





P (bar)	dR/dt (resistance normalized with time)	Slope from t/V vs V curve at t°C	Slope from t/V vs V curve at 20°C	α*C (1/m²)	dR/dV (1/m.L) (resistance normalized with filtrate volume)
0.25	1.37E+08	0.0209	0.0218	8.95E+12	2.99E+12
0.5	6.27E+08	0.0240	0.0253	2.08E+13	6.60E+12
0.75	7.86E+08	0.0166	0.0176	2.17E+13	7.38E+12
0.95	1.03E+09	0.0159	0.0168	2.63E+13	9.42E+12
1.25	1.15E+09	0.0148	0.0157	3.22E+13	1.04E+13
C (kg/m³)	0.097	(Specific cake resistance was calculated at reference pressure of 0.5 bar)			
α (m/kg)	2.14E+14				

Resistance (m^{-1})		%
Rm	1.46E+12	64
Rt (of sample SS+CL+SL)	2.27E+12	
R after rinse (with 200 mL of DI water)	2.17E+12	
Rf	8.12E+11	
Rir	7.20E+11	89
Rrev	9.21E+10	11

Note: Clean membrane resistance (Rm) was measured similarly by this method with DI water instead sludge sample

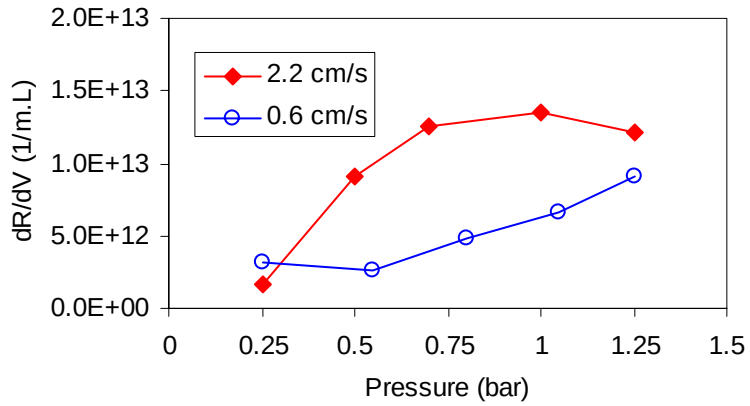


Figure D-1 Resistance rate of mixed liquor sludge (reactor sludge) at various aeration rates

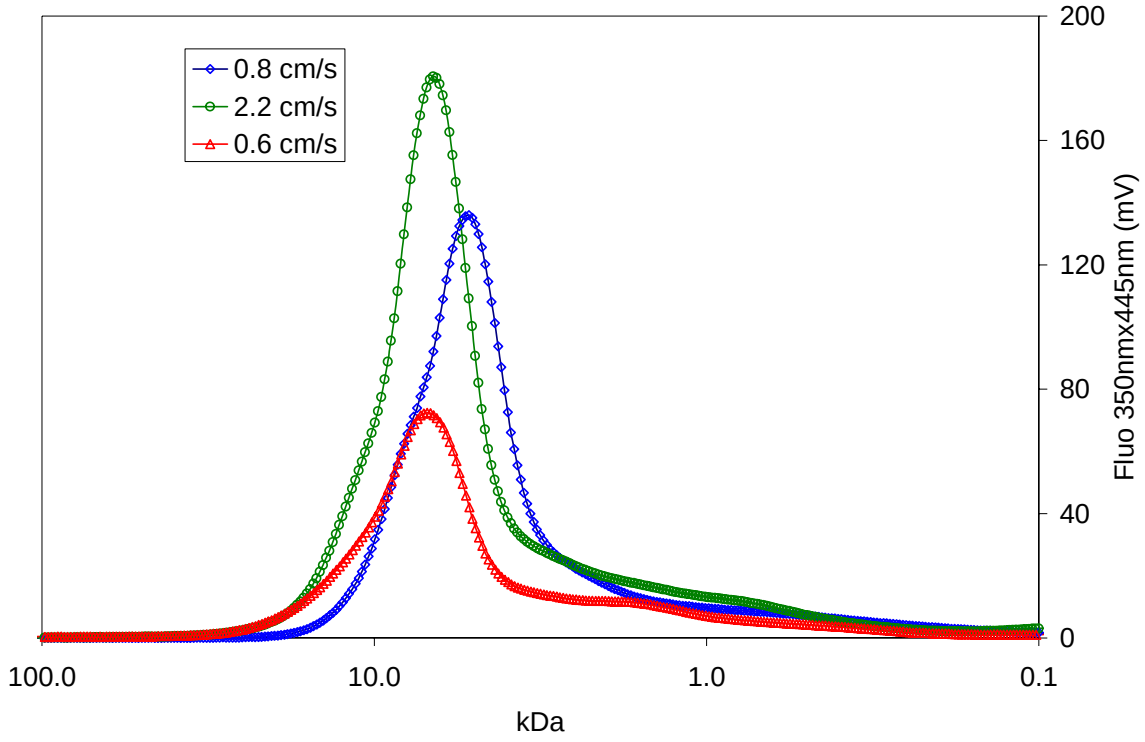


Figure D-2 Humic-like materials of SBAR effluent at various aeration rates (EEM 350 nm x 445 nm)

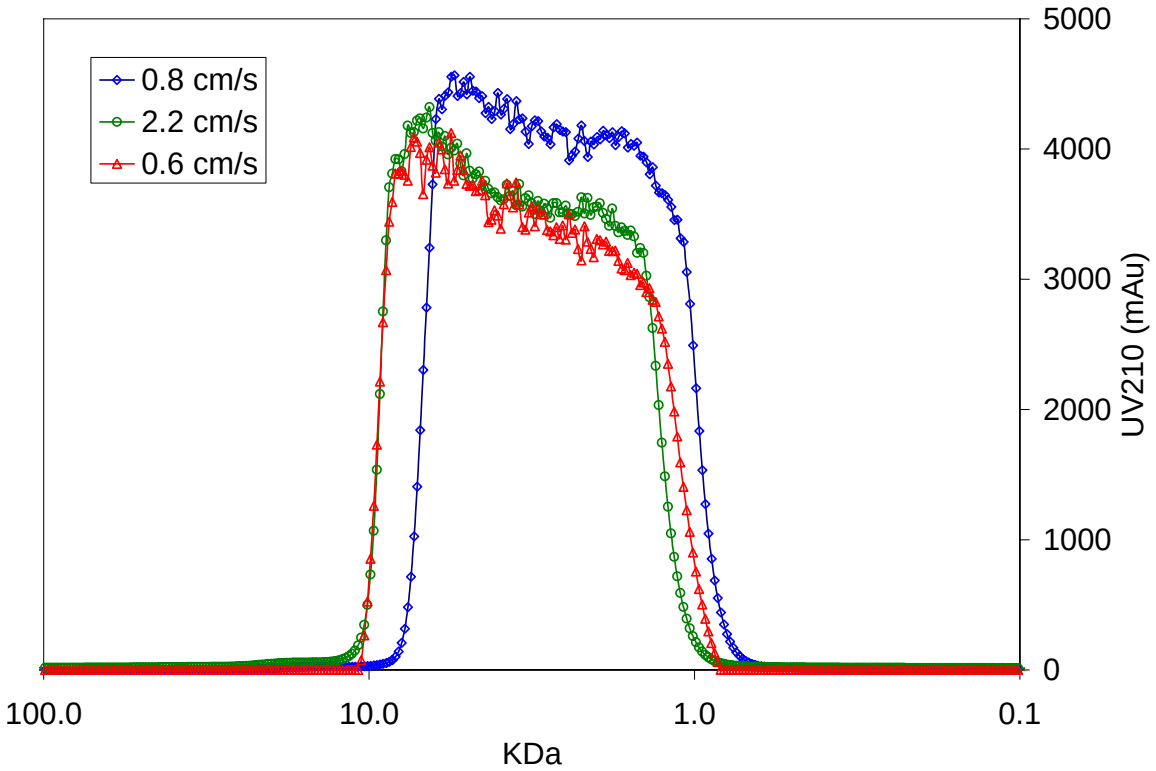


Figure D-3 Organic materials of SBAR effluent at various aeration rates (UVA 210 nm)

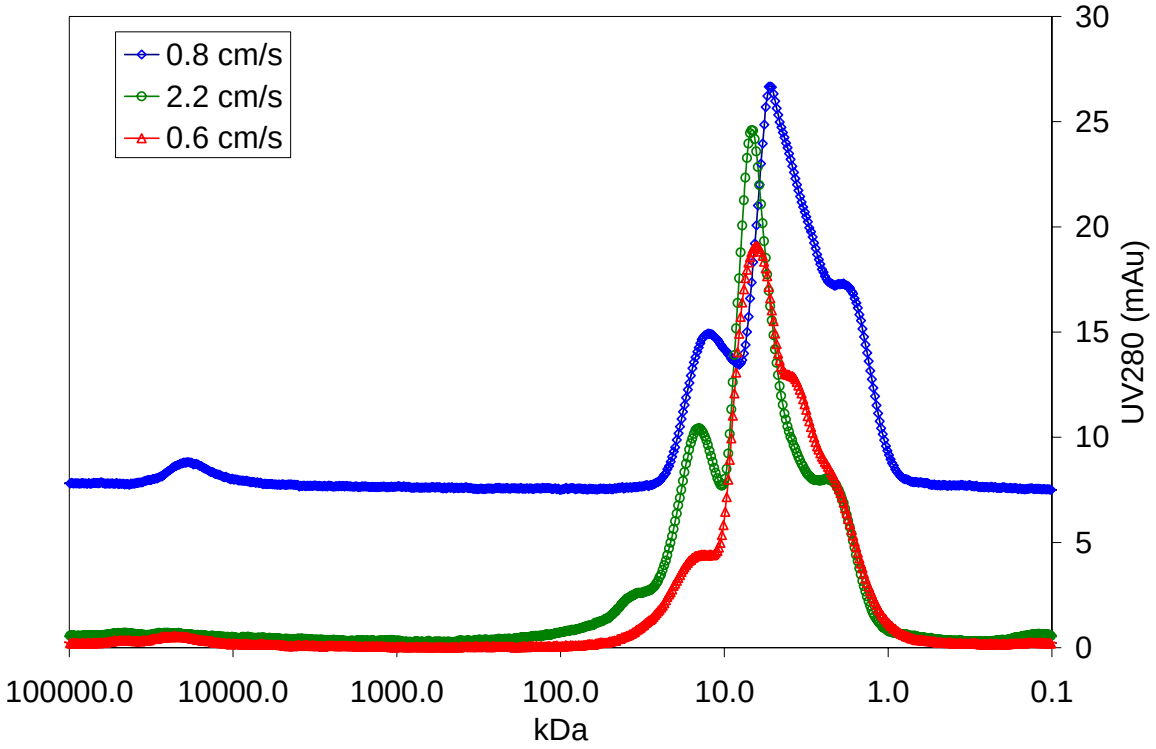
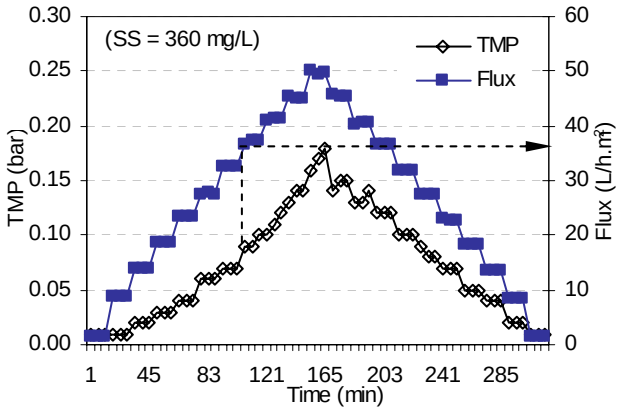


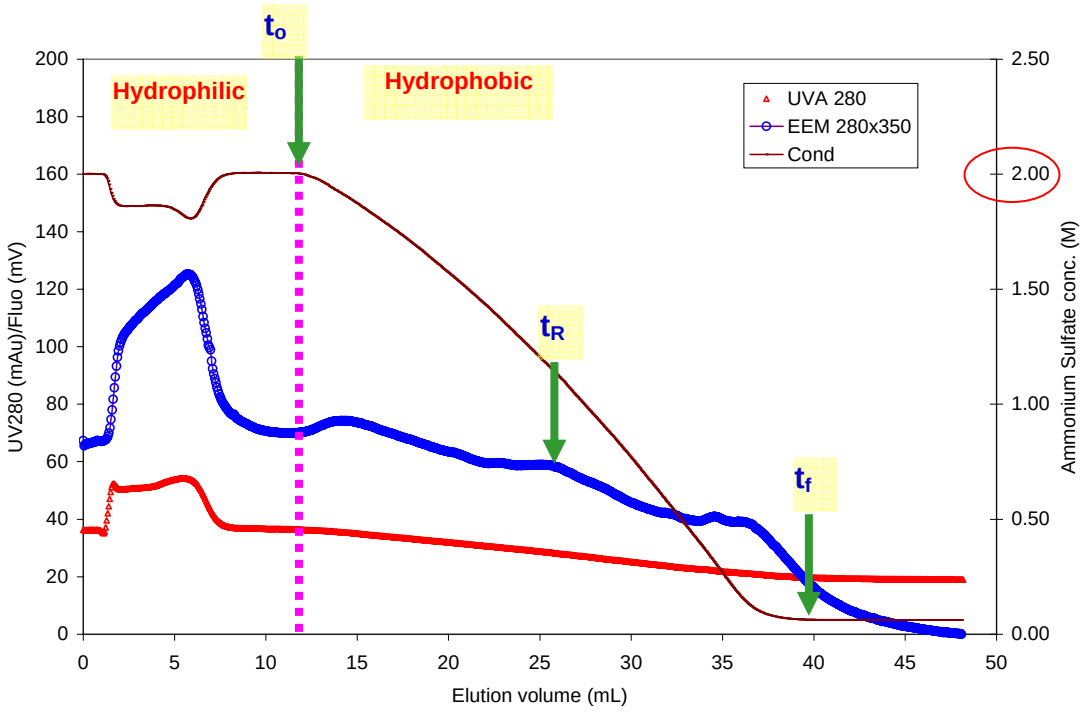
Figure D-4 Protein-like materials of SBAR effluent at various aeration rates (UVA 280 nm)



Time (min)	Pump frequency	TMP1 (bar)	TMP2 (bar)	TMP3 (bar)	Q1, (mL/min)	Q2, (mL/min)	Flux aver (mL/min)	TMP aver (bar)	dTMP/dt (mbar/min)
0	2	0.01	0.01	0.01	5.4	5.2	1.6	0.010	0.000
15	6	0.01	0.01	0.01	29	29	8.7	0.010	0.000
30	10	0.02	0.02	0.02	46	45.75	13.8	0.020	0.000
45	14	0.03	0.03	0.03	62	62	18.6	0.030	0.000
60	18	0.04	0.04	0.04	77.5	77.5	23.3	0.040	0.000
75	22	0.06	0.06	0.06	92	93	27.8	0.060	0.000
90	26	0.07	0.07	0.08	108	109	32.6	0.073	0.667
105	30	0.09	0.09	0.1	122.5	124	37.0	0.093	0.667
120	34	0.1	0.11	0.12	136.5	138	41.2	0.110	1.333
135	38	0.13	0.14	0.14	151	150	45.2	0.137	0.667
150	42	0.16	0.17	0.18	167	165	49.8	0.170	1.333

165	38	0.14	0.15	0.15	152	151	45.5	0.147	0.667
180	34	0.13	0.13	0.14	134	135.5	40.4	0.133	0.667
195	30	0.12	0.12	0.12	122	122	36.6	0.120	0.000
210	26	0.1	0.1	0.1	106	106	31.8	0.100	0.000
225	22	0.08	0.08	0.08	91.5	92	27.5	0.080	0.000
240	18	0.07	0.07	0.07	76.7	76	22.9	0.070	0.000
255	14	0.05	0.05	0.05	61	61	18.3	0.050	0.000
270	10	0.04	0.04	0.04	45.5	45.5	13.7	0.040	0.000
285	6	0.02	0.02	0.02	28	28	8.4	0.020	0.000
300	2	0.01	0.01	0.01	4.8	4.8	1.4	0.010	0.000

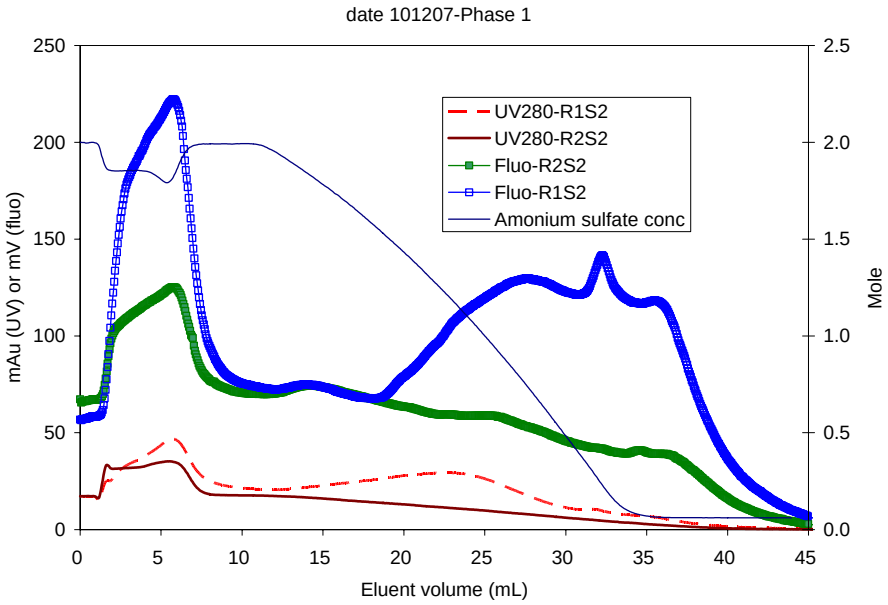
Figure D-5 Critical flux analysis of SBAR effluent at aeration rate of 0.6 cm/s with anoxic/aerobic condition (at INSA, Toulouse, France)



$$DRT = \frac{t_R - t_o}{t_f - t_o}$$

DRT = 0: less hydrophobic
DRT = 1: extreme hydrophobic

Figure D-6. Calculation of hydrophobic intensity (DRT)



Hydrophobic intensity of R1 effluent (no nitrate in the feed wastewater)									
Aeration rate	0.8 cm/s	2.2 cm/s			2.8 cm/s				
Day No.	28	60	72	74	81	86	88	92	101
Peak 1	0.57	0.12				0.09	0.71	0.56	0.88
Peak 2	0.82	0.14				0.12	0.89	0.89	
Peak 3	0.87								
Note: The blank cell means that there is no hydrophobic peak									

Figure D-7 Comparison of hydrophobicity of effluent of R1 (without nitrate in the feeding, named R1S2) and R2 (with nitrate in the feeding, name R2S2) at aeration rate of 0.8 cm/s

Appendix E

Experimental Data
of
the Continuous Granulation Membrane Bioreactor systems (CG-MBR)
(Results at AIT)

Table E-1 Organic and nitrogen removal at various OLR of the CG-MBR system

	Inf	MBR sup	Per	NLR	Inf	MBR sup	Per	Inf	MBR sup	Per	Inf	MBR sup	Per	Inf	MBR sup	Per	TOC removal (%)	TN removal (%)	COD inf (mg/L)	TOC	COD
Day	NH4-N (mg/L)				NO2-N (mg/L)			NO3-N (mg/L)			TN = NH4-N+NO2-N+NO3-N (mg/L)			DOC (mg/L)						$kg/m^3.d$	$kg/m^3.d$
OLR 2 $kgCOD/m^3.d$																					
1	134.4		18.5	0.4		35.7	33.6		49.8	44.5	134.4	85.5	96.6		13.0			28.1			
4	145.6	11.8	13.2	0.5	0.1	19.9	18.0	0.1	77.3	78.8	145.8	108.9	110.0	365.6	12.2	4.4	98.8	24.6	984	0.9	2.3
6	159.6	3.4	0.4	0.5	0.1	8.2	8.2	2.4	92.9	92.3	162.1	104.5	100.9	404.5	15.9	0.6	99.8	37.8	1088	1.0	2.6
13	142.8	0.6	1.4	0.5	0.1	9.0	6.5	1.1	83.9	110.4	144.0	93.4	118.3	405.1	13.3	8.3	98.0	17.8	1089	1.0	2.6
15	140.0	1.4	1.1	0.5	0.1	4.8	3.2	0.9	105.7	91.2	141.0	111.9	95.5	299.1	9.4	2.8	99.1	32.2	806	0.7	1.9
18	154.0	0.6	0.6	0.5	0.1	4.6	3.8	1.0	92.2	89.5	155.1	97.4	93.9	263.7	5.7	2.0	99.2	39.5	711	0.6	1.7
23	142.8	0.8	2.4	0.5	0.2	10.3	7.3	2.2	106.0	108.0	145.1	117.2	117.7	323.3	4.7	21.6	93.3	18.9	871	0.8	2.1
33														363.9		3.6	99.0		979	0.9	2.3
34	137.2	0.6	1.1	0.4	0.6	4.0	2.3	1.6	137.8	117.6	139.3	142.3	121.1	357.5	7.3	3.1	99.1	13.1	962	0.9	2.3
36	117.6	2.0	2.5	0.4	0.2	14.7	16.1	2.0	87.8	84.6	119.9	104.5	103.1	105.2	9.6	5.3	95.0	14.0	287	0.3	0.7
39	162.4	0.3	0.3	0.5	0.0	2.3	2.7	2.3	125.9	123.3	164.7	128.5	126.2	329.0		7.1	97.8	23.4	886	0.8	2.1
41	156.8	0.8	0.6	0.5	0.2	3.1	4.0	2.3	123.1	116.0	159.3	127.0	120.6	415.8	21.2	7.5	98.2	24.3	1118	1.0	2.7
43	145.6	1.1	0.8	0.5	0.2	3.4	3.2	2.4	118.4	120.9	148.2	123.0	125.0	503.5				15.7	515	1.2	
46	156.8	2.0	2.0	0.5	0.1	3.1	2.7	2.3	124.5	126.4	159.2	129.5	131.0		16.8	10.7		17.7			
48	154.0	0.6	1.7	0.5	0.1	8.2	9.6	1.7			155.8				11.5	6.5					
50	148.4	0.8	1.1	0.5	0.2	4.0	4.6	1.9	101.2	103.2	150.5	106.0	108.9	158.3	9.1	5.4	96.6	27.7	429		
OLR 4 $kgCOD/m^3.d$																					
53	151.2	1.4	1.1	0.5	0.2	4.0	3.6	2.3	115.0	122.5	153.6	120.4	127.3		14.1	3.2					
55	173.6	0.6	0.8	0.6	0.2	3.6	3.1	2.2	120.9	123.9	176.0	125.1	127.8	436.4	17.3	3.4	99.2	27.4	1173	1.0	2.8
57	159.6	1.1	1.7	0.5	0.2	4.2	4.4	2.3	125.1	122.5	162.1	130.5	128.6	557.1	15.7	5.4	99.0	20.7	1495	1.3	3.6
60	145.6	1.1	1.7	0.5	0.2	3.6	3.4	2.5	123.1	125.8	148.3	127.9	130.9	382.0		4.1	98.9		1027	0.9	2.4
62	156.8	0.8	0.8	0.5	0.2	4.2	3.6	2.3	120.6	118.4	159.3	125.7	122.9	659.3	34.2	6.4	99.0	22.9	1769	1.6	4.2
64	154.0	0.8	1.1	0.5	0.2	3.4	3.2	2.4	126.4	128.1	156.6	130.7	132.5	190.4	21.0	4.9	97.4		515	0.5	
67	137.2	1.4	1.4	0.4	0.2	3.1	3.1	2.4	125.1	123.5	139.8	129.6	128.0	441.8	6.7	4.2	99.0		1187	1.1	2.8
75														688.9	10.5	7.7	98.9		1848	1.6	4.4
76	165.2	0.3	0.6	0.5				3.3	107.7	109.3				778.7	11.5	5.1	99.3		2088	1.9	5.0

77	204.4	19.3	5.6	0.7	0.1	37.1	34.0	4.8	101.5	106.2	209.3	157.9	145.8	707.9	10.5	4.4	99.4	30.3	1898	1.7	4.5
80	162.4				0.7	22.6	27.9	4.2	121.9	132.2	167.2	144.5	160.1	758.7	15.2	8.4	98.9		2034	1.8	4.8
81	193.2	12.0	5.9	0.6	0.2	22.2	20.3	4.8	90.7	109.0	198.1	124.9	135.1	710.6	13.9	4.9	99.3	31.8	1906	1.7	4.5
82	240.8	19.3	3.4	0.8	0.1	19.9	4.2	6.5	98.7	125.8	247.4	137.9	133.4	671.4	12.1	7.2	98.9	46.1	1801	1.6	4.3
83	198.8	8.7	1.4	0.7	0.2	13.0	11.5	7.8	136.9	135.2	206.8	158.6	148.1	770.1	16.2	5.8	99.3	28.4	2065	1.8	4.9
OLR 8 kgCOD/m^3.d																					
87	176.4	5.3	0.7	0.6	0.3	4.2	4.6	5.1	77.7	107.7	181.8	87.3	113.0	907.1	13.4	2.9	99.7	37.8	2431	2.2	5.8
88	173.6	2.7	0.3	0.6	0.2	15.5	10.1	3.5	94.1	99.8	177.3	112.3	110.2	992.7	20.2	4.6	99.5	37.9	2660	2.4	6.3
90	186.2	7.6	2.8	0.6	0.0	15.7	13.0	7.3	81.5	87.0	193.5	104.7	102.8	1137.2	16.9	10.3	99.1	46.9	3046	2.7	7.2
91	142.8	7.8	0.3	0.5	0.1	26.0	18.0	5.1	45.0	45.7	148.0	78.8	64.0	1125.4	12.4	9.1	99.2	56.8	3014	2.7	7.2
94	159.6	2.5	0.6	0.5	0.1	8.0	2.9	7.5	57.1	59.9	167.2	67.7	63.4	1229.3	13.0	6.5	99.5	62.1	3292	2.9	7.8
95	173.6	1.7	6.2	0.6	0.1	6.7	9.7	7.9	25.1	27.8	181.5	33.5	43.7	1720.0	13.4	4.9	99.7	75.9	4604	4.1	10.9
96	170.8			0.6										1648.4					4413	3.9	10.5
98	159.6	4.2	3.4	0.5	0.1	15.1	4.0	9.6	64.0	80.1	169.3	83.3	87.5	1241.6	11.6	4.0	99.7		3325	3.0	7.9
101														1083.7					2903	2.6	6.9
102														952.3	18.5	8.8	99.1		2552		6.1
103	168.0	6.4	6.7	0.6	0.2	15.1	14.9	4.9	103.0	93.5	173.1	124.6	115.1	874.9	15.5	8.5	99.0		2345	2.1	5.6
104	162.4	2.5	0.6	0.5	0.2	32.1	31.0	10.7	42.3	36.4	173.3	76.9	67.9	989.4	13.7	8.0	99.2	60.8	2651	2.4	6.3
108	170.8	6.7	5.9	0.6	0.0	17.4	18.9	3.2	42.6	48.7	174.1	66.7	73.5	1363.5	10.4	5.9	99.6	57.8	3651	3.2	8.7
109	145.6	10.4	10.4	0.5	0.1	26.0	20.8	3.7	40.1	43.4	149.4	76.5	74.6	1138.2	12.2	8.7	99.2	50.1	3049	2.7	7.2
110	170.8	8.4	9.5	0.6	0.1	27.5	19.9	6.3	41.8	45.4	177.2	77.8	74.8	1184.9				57.8	3174	2.8	7.5
111	184.8	5.6	5.0	0.6	0.1	50.1	42.1	4.8	50.0	55.1	189.6	105.6	102.2					46.1			
115	165.2	1.4	1.1	0.5	0.2	1.1	0.6	6.7	84.3	87.6	172.1	86.8	89.3	1209.2	12.4	9.2	99.2	48.1	3239	2.9	7.7
117	168.0	8.4	9.5	0.6	0.3	20.6	28.7	6.9	37.5	26.9	175.1	66.5	65.0	1020.6	11.6	6.6	99.4	62.9	2734	2.4	6.5
118	156.8	2.5	2.8	0.5	0.5	8.0	14.5	2.3	43.6	38.7	159.6	54.1	56.0			5.5		64.9			

Table E-2 UVA_{254} and SUVA of the CG-MBR at various OLRs

	Inf	MBR sup	Per	Inf	MBR sup	Per	Inf	MBR sup	Per
Day	DOC (mg/L)			UVA254 (1/cm)			SUVA (L/mg/m)		
OLR 2 kgCOD/m^3.d									
1		13.0		0.535	0.091	0.274		0.70	
4	365.6	12.2	4.4	0.430	0.048	0.280	0.12	0.39	6.38
6	404.5	15.9	0.6	1.405	0.169	0.164	0.35	1.06	26.13
13	405.1	13.3	8.3	0.730	0.049	0.086	0.18	0.37	1.04
15	299.1	9.4	2.8	0.492	0.143	0.106	0.16	1.52	3.75
18	263.7	5.7	2.0	0.540	0.075	0.026	0.20	1.32	1.28
23	323.3	4.7	21.6	0.085	0.015	0.034	0.03	0.32	0.16
33	363.9		3.6						
34	357.5	7.3	3.1		0.163	0.172		2.24	5.61
36	105.2	9.6	5.3		0.104	0.093		1.09	1.76
39	329.0		7.1	0.160	0.116	0.114	0.05		1.61
41	415.8	21.2	7.5	0.166	0.109	0.117	0.04	0.51	1.57
43	503.5			0.203	0.114	0.104	0.11		
46		16.8	10.7	0.192	0.118	0.109		0.70	1.02
48		11.5	6.5	0.274	0.144	0.123		1.26	1.90
50	158.3	9.1	5.4	0.206	0.116	0.114	0.13	1.27	2.11
OLR 4 kgCOD/m^3.d									
53		14.1	3.2	0.268	0.141	0.132		1.00	4.10
55	436.4	17.3	3.4	0.239	0.142	0.133	0.05	0.82	3.89
57	557.1	15.7	5.4	0.212	0.124	0.118	0.04	0.79	2.19
60	382.0		4.1	0.242	0.156	0.148	0.1		3.6
62	659.3	34.2	6.4	0.251	0.139	0.131	0.04	0.41	2.06
64	190.4	21.0	4.9	0.268	0.139	0.132		0.66	2.69
67	441.8	6.7	4.2	0.236	0.146	0.137	0.05	2.19	3.26
75	688.9	10.5	7.7	0.236	0.146	0.137	0.03	1.39	1.78
76	778.7	11.5	5.1	0.236	0.146	0.137	0.03	1.27	2.69
77	707.9	10.5	4.4	0.229	0.107	0.078	0.03	1.02	1.78
80	758.7	15.2	8.4	0.452	0.187	0.114	0.06	1.23	1.36
81	710.6	13.9	4.9	0.997	0.177	0.109	0.14	1.27	2.21
82	671.4	12.1	7.2	0.453	0.162	0.142	0.07	1.34	1.97
83	770.1	16.2	5.8	0.577	0.275	0.119	0.07	1.70	2.06
OLR 8 kgCOD/m^3.d									
87	907.1	13.4	2.9	1.184	0.137	0.135	0.13	1.02	4.60
88	992.7	20.2	4.6	2.267	0.135	0.079	0.23	0.67	1.71
90	1137.2	16.9	10.3	0.609	0.185	0.115	0.05	1.09	1.12
91	1125.4	12.4	9.1	1.285	0.165	0.117	0.11	1.33	1.29
94	1229.3	13.0	6.5	1.263	0.223	0.189	0.10	1.72	2.90
95	1720.0	13.4	4.9	0.784	0.157	0.104	0.05	1.18	2.12
98	1241.6	11.6	4.0	0.873	0.171	0.098	0.07	1.47	2.43
102	952.3	18.5	8.8						
103	874.9	15.5	8.5	1.995	0.172	0.137	0.23	1.11	1.60
104	989.4	13.7	8.0	1.365	0.170	0.132	0.14	1.24	1.64
108	1363.5	10.4	5.9	0.265	0.074	0.058	0.02	0.71	0.98
109	1138.2	12.2	8.7	1.103	0.149	0.113	0.10	1.22	1.30
110	1184.9			0.875	0.141	0.108	0.07		
115	1209.2	12.4	9.2	1.419	0.144	0.113	0.12	1.16	1.23
117	1020.6	11.6	6.6	1.011	0.122	0.109			1.64
118			5.5	2.139	0.107	0.093			1.68

Table E-3 SVI, CST and biomass concentration of the CG-MBR at various OLRs

Day	CST (s)	SVI (mL/g)	MLSS (mg/L)		MLVSS (mg/L)		VSS/SS (reactor)
			Reactor	Wasted sludge	Reactor	Wasted sludge	
OLR 2 kgCOD/m^3.d							
2		65	5836		5025		0.86
4		53	5529		4737		0.86
6		90	4436	2526	3874	2295	0.87
13		77	2596	2022	2288	1801	0.88
15		56	2678	2853	2275	2419	0.85
18		62	2420	2633	2059	2514	0.85
23		61	1963	2348	1792	2036	0.91
34	8.1	67	2981	2895	2631	2546	0.88
36	7.5	63	3153	3009	2694	2606	0.85
39	8.2	59	3399	3095	3205	2750	0.94
43	7.1	63	2550	2137	1988	1721	0.78
46		46	3286	2457	2964	2240	0.90
48		66	2733	2413	2267	1941	0.83
50		70	2842	2225	2466	1863	0.87
OLR 4 kgCOD/m^3.d							
53		73	3030	2593	2747	2146	0.91
55		87	2520	2473	2083	2260	0.83
57		72	3479	2878	3224	2704	0.93
60		75	3352	3023	3053	2805	0.91
62		63	3494	3111	3209	2860	0.92
64		54	3676	3216	3345	2931	0.91
67		55	3612	3167	3240	2872	0.90
75			2906		2506		0.86
76		139	3438		3033		0.88
76		134	3980	3591	3582	3232	0.90
81	41	119	5892	6105	5100	5442	0.87
82	40.9	89	3366	5528	2883	4760	0.86
83	43.8						
OLR 8 kgCOD/m^3.d							
87		54	4658	6049	3994	5282	0.86
88	58.9	72	5553	5986	4806	5112	0.87
90	32.0	39	9694	9924	8262	8526	0.85
91	31.0	58	8470	9104	7303	7850	0.86
94	26.3	72	9399	9420	7992	7922	0.85
95	40.4	76	9456	9913	7920	8338	0.84
96	51.9	101	9033	10288	7567	8353	0.84
98		101	8783	8533	7357	7263	0.84
102	37.9	84	4632	4812	4047	4246	0.87
103	17.8	57	4593	5052	4004	4415	0.87
104	16.9	49	6098	3607	5378	3152	0.88
108	14.9	62	5445	5075	4770	4888	0.88
109	15.6	48	6675	6833	5751	5876	0.86
110	16.1	50	7149	7175	6247	6275	0.87
111		50	7527	7638	6145	6316	0.82
115	14.9	39	5584	4925	4865	4333	0.87
117	12.4	74	5983	6224	5280	5545	0.88
118	22.1	71	8130	7880	6946	6865	0.85
119	25.7	56	7543	7491	6752	6692	0.9

Table E-4 EPS of fouling layer of the CG-MBR at various OLRs

OLR (kgCOD/m^3.d)	Sample	MLSS (mg/L)	MLVSS (mg/L)	mgPS/gVSS	mgPN/gVSS
2	1	2861	2424	3.9	13.7
	2	3784	3361	3.8	8.2
Average				**3.9±0.1**	**11.0±3.9**
4	1	5104	4523	5.4	8.2
Average				**5.4**	**8.2**
8	1	17822	15900	3.1	10.7
	2	17432	15534	3.7	11.3
Average				**3.4±0.5**	**11.0±0.4**

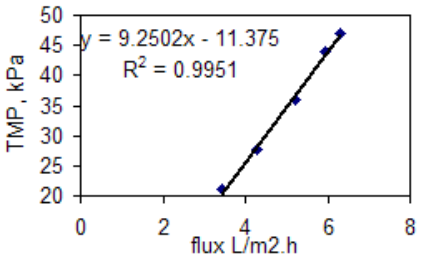
Table E-5 Correlation between F/M ratio and other factors

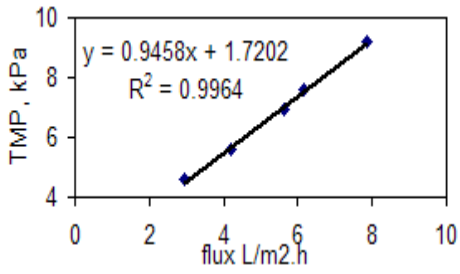
F/M (1/d)	SD-F/M	TOC (kgCOD/m^3.d)	SD-TOC	OLR (kgCOD/m^3.d)	SD-COD	Fouling rate (kPa/d)	SD
0.72	0.3	0.82	0.24	2.1	0.6	0.168	
1.06	0.3	1.42	0.43	4.0	0.9	0.292	
1.38	0.4	2.83	0.57	7.5	1.5	1.394	0.599

SD: Standard deviation

Table E-6 Typical resistance at OLR 2 kgCOD/m^3.d of the CG-MBR system

Fouled membrane after taking out from MBR			
P (kPa)	flowrate (mL/min)	Flux (L/m^2.h)	
21.1	24	3.43	
27.6	30	4.29	
35.9	36.5	5.21	
44	41.5	5.93	
47.1	44	6.29	
Rt=Rm+Rf+Rc	4.17E+13		at 30oC
After removing cake by tap water spray and shaking for 10 minutes			
P, kPa	Flowrate (mL/min)	Flux (L/m^2.h)	
4.6	20.5	2.93	
5.6	29.5	4.21	
6.9	39.3	5.61	
7.6	43	6.14	
9.2	55	7.86	
Rf+Rm	4.27E+12		at 30oC
Membrane after chemical cleaning (NaOH 4%, Chlorine 3 g/L):			
P (kPa)	Flowrate (mL/min)	Flux (L/m^2.h)	
3	28.8	4.11	
3.2	42	6.00	
3.5	60.5	8.64	
3.9	80	11.43	
4.1	100	14.29	
Rm	5.07E+11		
Rc+Rf+Rm	4.17E+13		
Rf+Rm	4.27E+12		
Rm	5.07E+11		
Rc	3.75E+13	89.8 %	
Rf	3.76E+12	9.0 %	
Rm	5.07E+11	1.2 %	





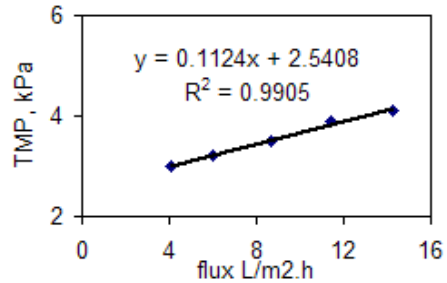


Table E.7 Typical membrane resistance of the CG-MBR at various OLRs

Cycle/resistance	OLR (kgCOD/m^3.d)					
	2		4		8	
Cycle 1		(%)		(%)		(%)
R_c (m^{-1})	3.75E+13	89.8	1.37E+14	97.3	1.11E+13	87.5
R_f (m^{-1})	3.76E+12	9.0	3.05E+12	2.2	1.14E+12	9.0
R_m (m^{-1})	5.07E+11	1.2	7.57E+11	0.5	4.50E+11	3.6
Cycle 2				(%)		
R_c (m^{-1})			8.99E+13	98.7		
R_f (m^{-1})			4.48E+11	0.5		
R_m (m^{-1})			7.51E+11	0.8		

Table E-8 SMP concentration of the CG-MBR at various OLRs

	Biomass concentration		MBR sup		Per		bound EPS	
Day	MLSS (mg/L)	MLVSS (mg/L)	sPS (mg/L)	sPN (mg/L)	sPS (mg/L)	sPN (mg/L)	mgPS/gVSS	mgPN/gVSS
OLR 2 kgCOD/m^3.d								
1	5025		10.9	3.5	3.6	2.6		
6			5.8	8.7	4.1	1.7		
13				4.4		3.2		
15			11.7	6.1	4.0	2.3		
18			10.0	3.5	2.5	2.6		
23			4.0	1.7	0.9	1.2		
34			16.4	4.1	9.5	3.2		
36			11.8	3.5	6.1	4.1		
39			15.6	3.8	9.9	2.0		
41			15.5	9.3	9.3	2.9		
43	2549	2103		4.1		2.0	7.08	24.79
44	2549	2103					6.32	22.37
46				4.9		1.5		
48				4.4		1.7		
50			20.9	3.2	12.3	1.5		
51	2104	1991				1.7	5.88	17.61
OLR 4 kgCOD/m^3.d								
53			2.7	1.2	3.8	2.0		
58			2.5	5.5	1.8	2.0		
60			0.8	4.4	0.7	2.3		
62				4.4		2.9		
65				6.7		3.8		
74	2906	2506					13.78	27.19
75				6.1		4.1		
76			19.2	6.4	8.5	2.9		
77			1.6	4.4	1.2	2.6		
80			15.0	5.8	11.8	2.9		
82			16.8	4.9	14.9	4.4		
83			15.9	5.5	12.7	2.9		
OLR 8 kgCOD/m^3.d								
87			18.9	4.7	11.3	4.7		
88			20.3	5.2	10.5	2.9		
90			12.6	6.1	5.0	3.8		
91			11.5	4.7	9.5	2.3		
94			12.3	4.7	8.2	1.7		
95			10.0	4.7	5.1	2.3		

98			13.3	6.7	6.2	3.5		
102			16.4	10.8	11.1	5.5		
103	4593	4004	17.3	9.0	15.9	4.4	7.72	23.84
104	6098	5378	20.9	8.1	13.6	4.7	6.41	17.41
108			19.7	3.5	8.2	2.6		
109	6675	5751	12.8	6.1	9.0	1.7	6.03	17.49
110	7149	6247	10.9	4.9	7.7	2.6	8.91	26.57
111			11.6	7.3	8.7	2.9		
112	10481	9172					1.63	8.70
115			20.9	6.1	8.7	3.2		
117			10.4	5.2	6.8	5.2		
118			9.9	8.1	6.0	3.8		

Table E-9 SMPs deposition rate on membrane of the CG-MBR at various OLRs

OLR 2 kgCOD/m^3.d			
Average conc.	MBR sup (mg/L)	Permeate (mg/L)	mg/L.m^2
PS	11.3	5.6	13.7
PN	4.7	2.3	5.6
EPS			19.3
DOC	12.1	5.2	16.5
OLR 4 kgCOD/m^3.d			
PS	16.7	12.0	11.3
PN	5.0	2.9	5.0
EPS			16.3
DOC	13.1	5.4	18.2
OLR8 kgCOD/m^3.d			
PS	14.7	8.9	13.7
PN	6.2	3.4	6.7
EPS			20.5
DOC	14.1	6.9	17.2

Note : Specific deposition rate = [MBR supernatant (mg/L) – Permeate (mg/L)]/membrane surface area (m^2). Refer to data of Table E-1 and Table E-5.

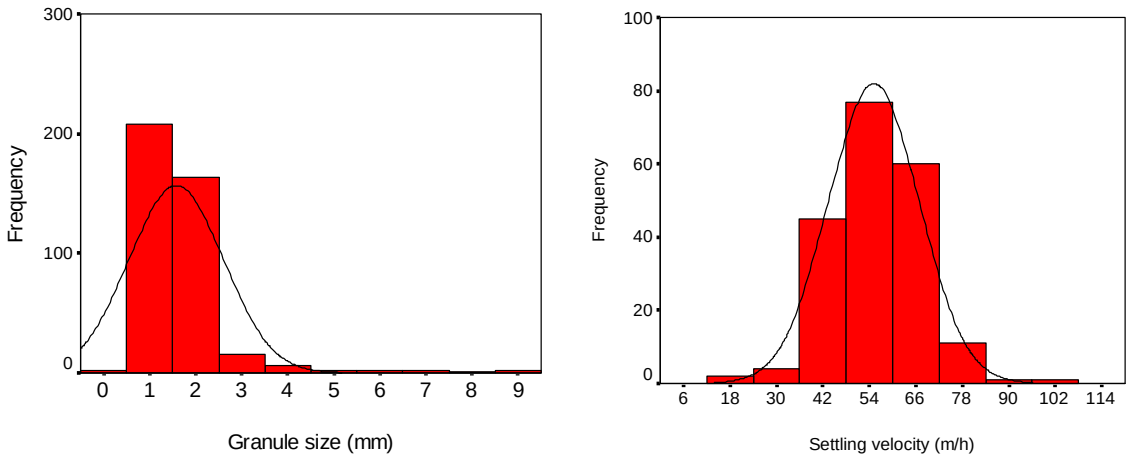


Figure E-1 Particle size distribution of granules (1.6±1.0 mm) and settling velocity distribution of granules used in the CG-MBR (20-107 m/h, average of 55±12 m/h)

Appendix F

Calculation of Energy Requirement for Different Systems
&
List of Publications

Energy Requirement for Treatment Systems

1. Energy requirement for wastewater treatment by the BG-MBR system:

Scale-up the BG-MBR system: Including SBAR, Settler and MBR (similar to Figure 3.4)

Q = 1000 m^3/d = 41.7 m^3/h = 0.01158 m^3/s
Q_{batch} = 250 m^3/batch
S_o = 800 mg/L = 0.8 kg/m^3
S = 64 mg/L (SBAR effluent)

a. Design for SBAR:
Design cycles of granulation reactor (SBAR): 4 batches/d, 6h/batch
+ Feeding = 60 min
+ Aeration = 240 min
+ Settling = 10 min
+ Discharge = 50 min

OLR of conventional activated sludge process = 2 kgCOD/m^3.d

$$OLR = \frac{Q * S_o}{V} \Rightarrow V = \frac{Q * S_o}{OLR} = \frac{1000m^3/d * 0.8kg/m^3}{2kg/m^3.d} = 400m^3$$

Check F/M ratio

$$F/M = \frac{Q * S_o}{V * X} = \frac{1000m^3/d * 0.8kg/m^3}{400m^3/d * 4kg/m^3} = 0.5d^{-1}$$

The granulation reactor can operate at OLR from 9-15 kgCOD/m^3.d due to the high biomass retention in reactor. It can reach 16g/L (X =16 g/L).

When X = 16 g/L ➔ F/M = 0.5 d^{-1} (const) ➔ Influent COD = S_o = 3.2 kg/m^3

Recalculate OLR for granulation system:

$$OLR = \frac{Q * S_o}{V} = \frac{1000m^3/d * 3.2kg/m^3}{400m^3} = 8kgCOD/m^3.d$$

Q = 250 m^3/batch
Volume exchange ratio (VER) = 250/400 = 63%
Select the height H = 8 m (In the granulation reactor, the height of reactor should be sufficient to create the shear stress in reactor)
➔ D = 5 m, A = 3.14*5*5/4 = 19.63 m^2
The SBAR is separated two zones (riser and down comer) by a stainless steel baffle:
L x W = 6 m x 5 m, thickness 8 mm.

Choose aeration rate for SBAR: v = 2.5 cm/s
Aeration duration = 4 batch/d * 4 h aeration/batch = 16 h/d

Air flowrate:
w = 19.63 m^2 *2.5 cm/s * m/100cm = 0.491 m^3/s = 0.491 m^3/s*1.2 kg air/m^3 = 0.589 kg/s

Power requirement for air blower:

$$P_b = \frac{w*R*T}{29.7*n*e}\left[\frac{P_2^{0.283}}{P_1}-1\right] = \frac{0.589kg/s*8.314*(30+273)}{29.7*0.283*0.8}\left[\frac{(1+1)^{0.283}}{1}-1\right] = 47.8kW \text{ (a)}$$

Where,
w: air flowrate (kg/s)
T: temperature (^oK)
R: 8.314 kJ/k mol.^oK
P_1: absolute inlet pressure (atm) (1 atm)
P_2: absolute outlet pressure (reactor height = 8 m ➔ relative pressure ~ 1 atm)
n = 0.283 for air
e = efficiency (0.7-0.9)

Oxygen requirement from biological reaction:
Oxygen required = $Q*(S_o-S)$ = 0.01158 $m^3/s*(3200-64)g/m^3*10^{-3}kg/g$ = 0.0363 kg/s

Sludge production:

$$Y_{obs} = \frac{Y}{1+k_d*SRT} = \frac{0.5}{1+0.06*15} = 0.263$$

$P_x = Y_{obs}*Q(S_o-S)$ =0.263*0.01158 $m^3/s*(3200-64)g/m^3*10^{-3}kg/g$=0.00955 kg/s

Net Oxygen required = 0.0363 – 1.42 * 0.00955 = 0.0228 kg/s
Oxygen transfer 8%, air = 23.2% by weight, air density 1.2 kg/m^3

$$\text{Air flowrate required} = \frac{0.0228kg/s}{1.2kg/m^3*0.232} = 0.0817m^3/s$$

Air transfer efficiency of 8% = 0.0817/0.08=1.0213 m^3/s

Check velocity (from biological air flowrate)
v = 1.0213 m^3/s /(19.63 m^2) *100 cm/m = 5.2 cm/s

The power requirement for air blower:

$$P_b = \frac{w*R*T}{29.7*n*e}\left[\frac{P_2^{0.283}}{P_1}-1\right] = \frac{1.0213kg/s*8.314*(30+273)}{29.7*0.283*0.8}\left[\frac{(1+1)^{0.283}}{1}-1\right] = 82.9\text{kW (b)}$$

Compare between (a) and (b), so the power requirement for SBAR = 82.9 kW
Average power requirement for air blower (16h/24h) = 82.9*16/24 **= 55.27 kW**

b. Design for Settler (store two batches)**:**

V = 500 m^3
LxWxH = 10 m x 10 m x 5 m

c. Design for external submerged MBR:

HRT = 2-5 h ➔ chose HRT = 3 h
V = 41.7 m^3/h * 3 h = 125 m^3
Size: LxWxH = 6 m x 6 m x 4 m (A = 36 m^2)

+ Aeration rate for MBR in BG-MBR system: Air flow rate of MBR = 0.3 cm/s

w = 36 m^2 *0.3 cm/s * m/100cm = 0.108 m^3/s = 0.108 m^3/s*1.2 kg air/m^3
= 0.1296 kg/s

+ Power requirement for air blower:

$$P_b = \frac{w*R*T}{29.7*n*e}\left[\frac{P_2^{0.283}}{P_1}-1\right] = \frac{0.1296kg/s*8.314*(30+273)}{29.7*0.283*0.8}\left[\frac{(1+0.5)^{0.283}}{1}-1\right] = \mathbf{5.90\ kW}$$

+ Power for backflush (BF)
Flux = 25 L/m^2.h
Membrane surface area = 41.7 m^3/h / (25L/m^2.h) = 1670 m^2
Number of module:
n= 1670 m^2/ (46 m^2/module) = 36.2 module (choose n = 38 modules) ➔ A= 1748 m^2
Backflush 30 s for each 29.5 min; Q_{BF} = 3 Q = 75 L/m^2.h
Q_{BF} = 75 L/m^2.h *1748 m^2 *0.5/30 = 2185 L/h = 2.2 m^3/h
Backflush flowrate Q = 41.7 + 2.2 = 43.9 m^3/h = 0.0122 m^3/s

+ Energy for permeate suction (P_{ave} = 0.4 bar):

$$P_s = \frac{Q*P_{ave}}{\eta} = \frac{0.0122m^3/s*0.4bar*100kN/m^2}{0.4} = \mathbf{1.22kW}$$

+ Energy for backflush (operate 1 min, each 60 min):

$$P_s = \frac{Q*P_{ave}}{\eta} = \frac{3*0.0122m^3/s*0.4bar*100kN/m^2}{0.4} = 3.66kW$$

Average energy for Backflush average = 3.66 kW*1/60 = **0.061 kW**

+ Energy for Pump influent (pump about 10 m = 1 bar). This pump works 4 h/d, flowrate = 250m^3/h*1h/3600 = 0.0694m^3/s

$$P_{inf} = \frac{Q*\rho*g*H}{\eta} = \frac{0.0694m^3/s*1000kg/m^3*9.81m/s^2*10m*1kw/1000w}{0.4} = 17.03kW$$

Average energy for pumping influent = 17.03 kW *4/24 = **2.84 kW**

Energy for Pump from settler to MBR (H = 6 m):

$$P_{inf} = \frac{Q*\rho*g*H}{\eta} = \frac{0.01158m^3/s*1000kg/m^3*9.81m/s^2*6m*1kw/1000w}{0.4} = \mathbf{1.7\ kW}$$

Specific energy consumption of the BG-MBR system (kWh/m^3 of wastewater):

$= \Sigma P/Q = (55.27+5.90+1.22+0.061+2.84+1.70)kw/(41.7m^3/h) =$ **1.6 kWh/m^3**

2. Energy requirement for wastewater treatment by submerged MBR system:

Specific energy consumption of submerged MBR system = 0.7 – 1.0 (kWh/m^3) (Gunder, 2001)➔ choose 0.9 kWh/m^3

3. Energy requirement for wastewater treatment by anaerobic reactor:

OLR = 2-40 kgCOD/m^3.d for anaerobic system, choose OLR = 7 kgCOD/m^3.d

$$OLR = \frac{Q * S_o}{V} \Rightarrow V = \frac{Q * S_o}{OLR} = \frac{1000m^3/d * 3.2kg/m^3}{7kg/m^3.d} = 457m^3$$

Choose H = 6 m,
LxWxH = 9 m x 9 m x 6 m

+ Energy for influent pumping:

$$P_{inf} = \frac{Q * \rho * g * H}{\eta} = \frac{0.0116m^3/s * 1000kg/m^3 * 9.81m/s^2 * 6m * 1kw/1000w}{0.4} = 1.703kW$$

+ Energy for sludge mixing (Lamilar): two mixers

$$P = k\rho n^2 D^3 = 2*70*1100\ kg/m^3*0.07^2\ rpm/s*3\ m = 2264\ W = 2.26\ kW$$

Where,
P: Power requirement (W)
k: constant (k=70 for turbine, 6 blades)
n: rev/s
ρ: mass density of sludge (1100 kg/m^3)
D: diameter of impeller (m)

Specific energy consumption of anaerobic reactor = (1.703+2.26) kW/41.7 m^3/h = 0.095 ~ **0.1 kWh/m^3**

List of Publications

1. International Journals:

Thanh, B.X., Visvanathan, C., Spérandio, M., Ben Aim, R. (2008). Fouling characterization in aerobic granulation coupled baffled membrane bioreactor, *Journal of Membrane Science*, 318(1-2), 334-339.

Thanh, B.X., Visvanathan, C., Ben Aim, R. (2009). Characterization of aerobic granules at various organic loading rates, *Process Biochemistry*, 44, 242-245.

Thanh, B.X., Visvanathan, C., Ben Aim, R. (*submitted*). Fouling behavior in external submerged membrane bioreactor treating granulation effluent, *Separation Purification and Technology*.

Prasanthini, V., Thanh, B.X., Visvanathan, C. (*submitted*). Simultaneous nitrification denitrification and fouling of a batch granulation membrane airlift bioreactor, *Bioresource Technology*.

2. International Conferences:

Thanh, B.X., Sperandio, M., Guigui, C., Ben Aim, R., Wan, J.F., Visvanathan, C. (2008). Coupling sequencing batch airlift reactor (SBAR) and membrane filtration: Influence of nitrate removal on sludge characteristics, effluent quality and filterability, *Conference on Membranes in Drinking Water Production and Wastewater Treatment*, October 20th-22nd, 2008, Toulouse, France.

3. Book chapter:

Jegatheesan, V., Shu, L., Visvanathan, C., Thanh, B.X. (2008). Aerobic Environmental Processes: Chapter 23 in Advances in Fermentation Technology, Ed. Ashok Pandey et al., pp. 622-654, *Asiatech Press*, New Delhi. ISBN: 81-87680-18-0.