Development of specific membrane bioreactors for membrane fouling control during wastewater treatment for reuse

By

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CERTIFICATE OF ORIGINAL AUTHORSHIP

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Abbreviations

Symbol	Description
AC	Acylase
AFM	Atomic force microscopy
AnMBR	Anaerobic membrane bioreactor
AOB	Ammonium oxidizing bacteria
AOMBR	Anaerobic-oxic membrane bioreactor
A/O	Anoxic/oxic
A^2/O	Anaerobic/anoxic/oxic
BAP	Biomass-associated products
BNR	Biological nutrient removal
BOD	Biological oxygen demand
BPC	Biopolymer clusters
BPC_C	Polysaccharides in biopolymer clusters
BPC_P	Proteins in biopolymer clusters
CA	Cellulose acetate
CEB	Chemical enhanced backwashing
CEBs	Cell entrapping beads
CER	Cation exchange resin
CF	Concentration factor
CIOF	Combined inorganic-organic flocculant
CIP	Cleaning in place
CMBR	Conventional membrane bioreactor
COD	Chemical oxygen demand
C/N or COD/N	Carbon to nitrogen ratio
C/P	Carbon to phosphorus ratio
CST	Capillary suction time
DC	Direct current
DO	Dissolved oxygen
DOC	Dissolved organic carbon
DOC'	Deoxycholate
DOM	Dissolved organic matter

DPB	Denitrifying-phosphate-accumulating bacteria
DTPA	Diethylenetrinitrilopentaacetic acid
ECPS	Extra-cellular polymers
e-MBR	Electro-membrane bioreactor
DTPA	Diethylenetrinitrilopentaacetic acid
EDTA	Ethylene diamine tetraacetic acid
EL	Electrostatic double layer
EMPS	Extra-microcolony polymers
EPS	Extracellular polymeric substances
EPS_C	Polysaccharides in extracellular polymeric substances
EPS_P	Proteins in extracellular polymeric substances
EPS_P/EPS_C	Protein to polysaccharide ratio in extracellular polymeric substances
F/M	Food to microorganisms ratio
GBF	Green bioflocculant
GF	Glass fiber
HIS	Hydrophilic substances
HMBR	Hybrid membrane bioreactor
HRT	Hydraulic retention time
KMT	Kaldnes MijiØteknologi, TØnsberg
LB-EPS	Loosely bound extracellular polymeric substances
LMW	Low molecular weight
Mag-S-MPS	Magnetically separable mesoporous silica
MBBR	Moving bed biofilm reactor
MBR	Membrane bioreactor
MBR-G	Membrane bioreactor with bioflocculant addition
MBBR-MBR	Moving bed biofilm reactor-membrane bioreactor
MCE	Mixed cellulose ester
ME	Mixed ester
MF	Microfiltration
MLE	Modified Luzack-Ettinger
MLSS	Mixed liquor suspended solids
MLVSS	Mixed liquor volatile suspended solids

MRD	Membrane rejection degree
MW	Molecular weight
MWCO	Molecular weight cutoff
NLR	Nitrogen organic loading
NOB	Nitrite oxidizing bacteria
NPOC	Non-purgeable organic carbon
OHO	Ordinary heterotrophic organisms
OLR	Organic loading rate
OUR	Oxygen uptake rate
PAC	Powdered activated carbon
PACl	Polyaluminium chloride
PAC-MBR	Powdered activated carbon-membrane bioreactor
PAM	Polyacrylamide
PAN	Polyacrylonitrile
PAOs	Polyphosphate accumulating organisms
PC	Polycarbonate
PE	Polyethylene
PES	Polyethersulfone
PFC	Polymeric ferric chloride
PFS	Polymeric ferric sulfate
Phenex-NY	Phenex-Nylon
PN/PS	Protein to polysaccharide ratio
POC	Purgeable organic carbon
PP	Polypropylene
PSD	Particle size distribution
PSU	Polysulfone
PU	Polyurethane
PUS	polyurethane sponge
PVC	Polyvinyl chloride
PVDF	Polyvinylidene fluoride
$PVDF_H$	Hydrophobic polyvinylidene fluoride
QQ	Quorum quenching
QQMBR	Quorum quenching membrane bioreactor

RH	Relative hydrophobicity
SCMBR	Submerged conventional membrane bioreactor
SDS	Sodium dodecyl sulfate
SEM	Scanning electronic microscopy
S-MBBR	Moving bed biofilm reactor with sponge modified plastic carriers
S-MFC	Sludge microbial fuel cell
SMBR	Submerged membrane bioreactor
SMP	Soluble microbial products
SMP_C	Proteins in soluble microbial products
SMP_P	Polysaccharides in soluble microbial products
SMP_P/SMP_C	Protein to polysaccharide ratio in soluble microbial products
SND	Simultaneous nitrification and denitrification
SOUR	Specific oxygen uptake rate
SRT	Sludge retention time
SSMBR	Sponge-submerged membrane bioreactor
SSMBR-G	Sponge-submerged membrane bioreactor with bioflocculant addition
STP	Sodium tripolyphosphate
SVI	Sludge volume indexes
TB-EPS	Tightly bound extracellular polymeric substances
TCA	Trichloroacetic acid
TMP	Transmembrane pressure
TOC	Total organic carbon
TSAMBR	Thermophilic submerged aerobic membrane bioreactor
TSS	Total suspended solids
UAP	Utilization-associated products
UCT	The University of Cape Town
UF	Ultrafiltration
UTS	University of Technology, Sydney
WSW	Without sludge wasting
VFAs	Volatile fatty acids
VSS	Volatile suspended solids

Nomenclatures

Symbol	Description
ADIPAP KD 452 or KD452	Cationic polymers
$Al_2(SO_4)_3$	Aluminium sulfate
$CaCl_2 \cdot 2H_2O$	Calcium chloride
C_c	Concentration of potential cake forming particles in the bulk liquid (e.g. MLSS) which typically varies over time in membrane bioreactor
CGMS	Modified corn starch
$C_6H_{12}O_6$	Glucose
CO_2	Carbon dioxide
$CoCl_2 \cdot 6H_2O$	Cobalt chloride
CPE	Organic cationic polyelectrolyte
C_{SMP}	Time-dependent concentration of soluble particles entering the pores
$CuSO_4 \cdot 5H_2O$	Cupric sulphate
$d_{p,used}$	The initial pore diameter of the membrane in µm
dTMP/dt	Membrane fouling rate
f	The membrane's porosity
$FeCl_3$	Ferric chloride or Ferric chloride anhydrous
Fe-MBR	Electro-membrane bioreactor with stainless steel mesh as the anode
h_c	Variable depth of the cake layer expressed as a first order differential function in time, which relies on the attachment and detachment of cake layer
h_m	The membrane's effective thickness
H_2O_2	Hydrogen peroxide
H_2SO_4	Sulphuric acid
J	The permeate flux
k	The factor representing the detachment of the cake layer from the membrane surface
kDa	Unified atomatic mass unit

$K_2Cr_2O_7$	Potassium dichromate
KH_2PO_4	Potassium phosphate
$MLSS_e$	The MLSS concentration in the aqueous phase after emulsification
$MLSS_i$	The initial MLSS concentration of the mixed liquor sample
$m_{d,o}$	Outer membrane diameter
$m_{d,i}$	Inner membrane diameter
Mg	Magnesium
MGMS	Modified corn starch
$MgSO_4 \cdot 7H_2O$	Magnesium sulphate
MPE	Organic flocculant
MPE50	Organic flocculant (cationic polymers)
MPL30	Cationic polymers
$MnCl_2 \cdot 7H_2O$	Manganese chloride
NALCO MPE50	Cationic polymers
n_c	Cake fouling factor to explain the typically observed exponential rise of TMP due to the cake layer resistance especially at the final stage of operation of an MBR system
n_p	Pore fouling factor to explain the typically observed exponential rise of TMP due to the pore fouling resistance especially at the final stage of operation of an MBR system
NaCl	Sodium chlorite
NaClO	Sodium hypochlorite
$NaHCO_3$	Sodium hydrogen carbonate
$Na_2MoO_4 \cdot 2H_2O$	Sodium molybdate dehydrate
NH_2OH	Hydroxylamine
NH_4-N or NH_4^+	Ammonia nitrogen
$(NH_4)_2SO_4$	Ammonium sulphate
NO_2-N or NO_2^-	Nitrite
NO	Nitric oxide
N_2	Nitrogen gas
N_2O	Nitrous oxide
NO_3-N or NO_3^-	Nitrate
PAM-MGMS	Polyacrylamide-starch composite flocculant

pH	Power of hydrogen or potential hydrogen
PM30	A kind of polyethersulfone membrane
PO_4-P	Orthophosphate
Poly-1	Cationic polymers
Poly-2	Modified cationic polymers
r_p	Membrane pore radius
R80	Membrane bioreactor with 80 nm pore-sized ceramic membrane
R100	Membrane bioreactor with 100 nm pore-sized ceramic membrane
R200	Membrane bioreactor with 200 nm pore-sized ceramic membrane
R300	Membrane bioreactor with 300 nm pore-sized ceramic membrane
R^2	Squared value of correlation coefficient
R_C	Cake layer resistance
R_M	Intrinsic membrane resistance
R_{IR}	Irreversible fouling resistance
R_P	Pore blocking resistance
R_T	Total fouling resistance
SMP_{total}	Total soluble microbial products
t	The filtration time
T	Temperature
TATE & LYLE Mylbond 168	Starch
Ti-MBR	Electro-membrane bioreactor with titanium anodes
TN	Total nitrogen
$ZnSO_4 \cdot 7H_2O$	Zinc sulphate
x	Independent variable
y	Variable
ΔMLSS/Δt	Biomass growth rate
ΔP	Transmembrane pressure gradient
ΔTMP/Δt	Membrane fouling rate
YW 30	A kind of regenerated cellulose membrane

Greek symbols

Symbol	Description
α_c	Specific resistance of the compressible cake layer
α_p	Pore size reduction coefficient
γ	Shear rate
η_f	Average fraction of soluble particles that accumulate in membrane pores
ρ_c	Density of the cake layer
ρ_P	Density of biomass
τ	Shear stress
μ	Viscosity of the permeate

PhD DISSESRTATION ABSTRACT

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Thesis title: Development of specific integrated membrane bioreactors (MBRs) for membrane fouling control during wastewater treatment for reuse

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Abstract

In recent years, membrane fouling has become a critical issue of membrane bioreactor (MBR) in wastewater treatment. To resolve this obstacle, introducing biomass carriers or flocculants into submerged MBR (SMBR) has become one of the effective technologies for membrane fouling control. This study aims to provide an in-depth analysis on membrane fouling behaviour in SMBRs with sponge and/or the patented green bioflocculant by considering the properties of activated sludge and cake layer. A new functional media (sponge modified plastic carrier) was also developed to enhance the performance of integrated moving bed biofilm reactor-membrane bioreactor (MBBR-MBR) systems. The results suggested that sponge addition in a SMBR (SSMBR) or bioflocculant addition in a SMBR (MBR-G) reduced cake layer formation and limited pore blocking, thus effectively minimizing membrane fouling. Better sludge characteristics were obtained in both of the SSMBR and the MBR-G due to less soluble microbial products (SMP), lower biomass growth and sludge viscosity, higher protein to polysaccharide ratio in extracellular polymeric substances, higher zeta potential, greater relative hydrophobicity, larger floc size and better flocculation ability. The presence of sponge or bioflocculant in the SMBR also eliminated extracellular polymeric substances (EPS), SMP and/or biopolymer clusters (BPC) on membrane surface. Consequently, cake layer (R_C) and pore blocking resistance (R_P) were decreased in the SSMBR and the MBR-G. A modified resistance-in-series model proposed for the SMBR with and without bioflocculant could quantitatively demonstrate the impacts of sludge characteristics on membrane fouling. In the SSMBR, a longer hydraulic retention time (HRT) of 6.67 h permitted more considerably fouling reduction comparing to shorter HRTs (5.33 and 4.00 h). Moreover, lower R_P and R_C at the prolonged HRT were mainly ascribed to the elevated protein to polysaccharide ratio in SMP (SMP_P/SMP_C) of mixed liquor, together with the declined EPS and BPC in cake layer. SMP was not the primary membrane foulant when the SSMBRs were operated at different HRTs. Bioflocculant addition at the optimum HRT of 6.67 h further mitigated fouling in the SSMBR by improving activated sludge and cake layer characteristics. The integrated MBBR-MBR with the sponge modified plastic carriers showed better removal of DOC, NH_4-N, T-N and PO_4-P than the MBBR-MBR with plastic carriers only. Furthermore, the sponge modified plastic carriers also eliminated SMP of mixed liquor, and reduced SMP and BPC on membrane surface, which ameliorated membrane fouling, R_P and R_C as compared to the plastic carriers.

Keywords: Submerged membrane bioreactor (SMBR); Moving bed biofilm reactor (MBBR); Integrated MBBR-MBR; Sponge; Bioflocculant; Hydraulic retention time (HRT); Sponge modified plastic carriers; Membrane fouling; Nutrient removal; Cake layer; Modelling

Chapter 1

Introduction

Chapter 1 Introduction

1.1. Research background

1.1.1. Membrane fouling

In recent years, membrane bioreactors (MBRs) have been increasingly applied for municipal and industrial wastewater treatment. However, its widespread application is still limited by membrane fouling. Basically, membrane fouling is caused by (i) attachment or adsorption of solutes or colloids onto the membrane and/or into membrane pores, (ii) accumulation of sludge flocs on membrane surface, development of a cake layer on membrane surface, (iii) detachment of foulants inducing by shear stress, and (v) the alteration of membrane foulant composition over the operation time (including bacteria community and biopolymer components in the cake layer) (Meng et al., 2009a).

During the MBR process, membrane fouling reduces permeate yield, decreases membrane efficiency, aggravates flux decline, induces high-energy consumption, and increases membrane cleaning frequency or replacement and operational costs (Drew et al., 2010; Kimura et al., 2005).

1.1.2. Current fouling control strategies

To effectively mitigate membrane fouling, various kinds of approaches have been developed, such as physical cleaning (i.e. aeration, relaxation, backwash, the magnetically induced membrane vibration system), chemical cleaning (i.e. NaClO, NaOH), physio-chemical cleaning (i.e. chemically assisted maintenance membrane cleaning, chemical cleaning in place), biological/biochemical cleaning (i.e. enzymatic cleaning, energy uncoupling, quorum quenching (QQ)), and synergistic combinations of QQ and filtration/relaxation (Bilad et al., 2012; Chen and Columbia, 2011; Maqbool et al., 2015; Xu and Liu, 2011; Wang et al., 2014a; Weerasekara et al., 2014; Zsirai et al., 2012).

Additionally, some studies also focused on addition of media (i.e. plastic media, powdered activated carbon (PAC), sponge, clinoptilolite) directly into MBRs for fouling mitigation by modifying sludge characteristics (Jin et al., 2013; Ngo et al., 2008; Rezaei and Mehrnia, 2014; Skouteris et al., 2015). Huang et al. (2008) reported that effects of the suspended carriers (modified polypropylene (PP)) on membrane fouling in a submerged MBR (SMBR) depended on the relative intensity of two effects: 1) the positive effect induced by collision and friction between membrane surface and the moving carriers, resulting in effective cake layer removal; 2) the negative effect caused by breakup of sludge flocs, which increased the amount of small particles and supernatant total organic carbon, giving rise to severe membrane fouling. In addition, pore blocking could not be effectively eliminated by plastic media (Jin et al., 2013). It was found that PAC removed extracellular polymeric substances (EPS) and soluble microbial products (SMP), which favoured membrane fouling control. However, high PAC concentrations not only increased operational cost, but also deteriorated membrane permeability by pore blocking and gel layer formation (De Temmerman et al., 2014; Skouteris et al., 2015).

Sponge as an attached growth media has been also investigated. Yang et al. (2006) suggested that a hybrid MBR (HMBR) with sponge exhibited higher membrane permeability, lower hydraulic resistance, as well as less cake layer on membrane surface. The previous studies showed that sponge addition in the sponge-submerged membrane bioreactor (SSMBR) could improve sustainable flux and lower transmembrane pressure (TMP) development (Guo et al., 2008a; Nguyen et al., 2012a).

MBRs with flocculants (i.e. inorganic flocculants, organic flocculants) could also effectively minimize membrane fouling as they can improve sludge characteristics (e.g. less SMP, larger floc size) (Wu and Huang, 2008; Zhang et al., 2014). Nonetheless, these flocculants were detrimental to the environment and produced ‘secondary pollutants’ during wastewater treatment. A new green bioflocculant (GBF) as a safe biodegradable natural flocculant developed by Ngo and Guo (2009) remarkably decreased membrane fouling and energy consumption of a conventional submerged MBR.

Another kind of integrated MBR systems for membrane fouling reduction is a moving bed biofilm reactor (MBBR) coupled with a MBR. It could reduce membrane fouling whilst solving the settling problem of activated sludge in MBBRs. Yang et al. (2010a) reported that a sequencing batch moving bed membrane bioreactor (SBMBMBR) with a new kind of non-woven carriers limited overgrowth of filamentous bacteria and controlled membrane fouling. Contrary to polypropylene (PP) carriers, sponge in the MBBR-MBR reduced detachment of attached biomass from carrier, extended filtration time and mitigated membrane fouling when treating a synthetic phenolic wastewater (Raifei et al., 2014). However, some studies found that the MBBR-MBR exhibited more serious membrane fouling due to overgrowth of filamentous bacteria, higher cake layer resistance, and a large amount of submicron colloidal material (Sun et al., 2012; Yang et al., 2009a and 2009b).

1.2. Research motivations and scope

Although the previous studies have pointed out the capacity of sponge or bioflocculant in MBR for improvement of membrane permeability, as well as organic and nutrient removals, their effects on activated sludge and cake layer characteristics associated with membrane fouling behaviour are still scarce. Hence, the main focus of the research is to investigate the effectiveness of the specific integrated MBRs with sponge and/or the patented green bioflocculant (Gemfloc®) based on the study of Ngo and Guo (2009) for membrane fouling control during wastewater treatment for reuse. Moreover, the feasibility of a new functional media (i.e. plastic carrier modified using sponge) for enhancing the performance of the integrated MBBR-MBR systems is also explored.

The major objectives of this research are as follows:

1) To review currently representative studies in MBRs regarding biofouling mechanism, factors affecting biofouling and its control strategies;

2) To evaluate the performance of a SSMBR and a conventional MBR (CMBR) concerning their sludge and cake layer characteristics on membrane surface;

3) To assess the effectiveness of Gemfloc® for fouling reduction in MBR by modifying properties of mixed liquor and cake layer;

4) To evaluate the effects of hydraulic retention time (HRT) and Gemfloc® addition on the SSMBR performance in terms of biomass characteristics and cake layer properties; and

5) To demonstrate the performance of integrated MBBR-MBR systems with the sponge modified plastic carrier and the plastic carrier through analysing pollutant removal and evaluating membrane fouling.

1.3. Research significance

This research can provide a promising and effective way to promote the membrane fouling minimization and treatment performance for synthetic domestic wastewater treatment. The media (sponge and sponge modified plastic carrier) and bioflocculant used in this study have remarkable merits, such as low cost and being very friendly to environment. A developed membrane fouling indicator can determine membrane fouling potential by the main foulants (EPS and SMP). Hence, the information given in this research will be of significance to further studies regarding membrane fouling control and practical application of the integrated MBR systems for wastewater reclamation and reuse.

1.4. Organization and major contents of thesis

The thesis has been divided into eight chapters, of which the main contributors are displayed in Figure 1.1.

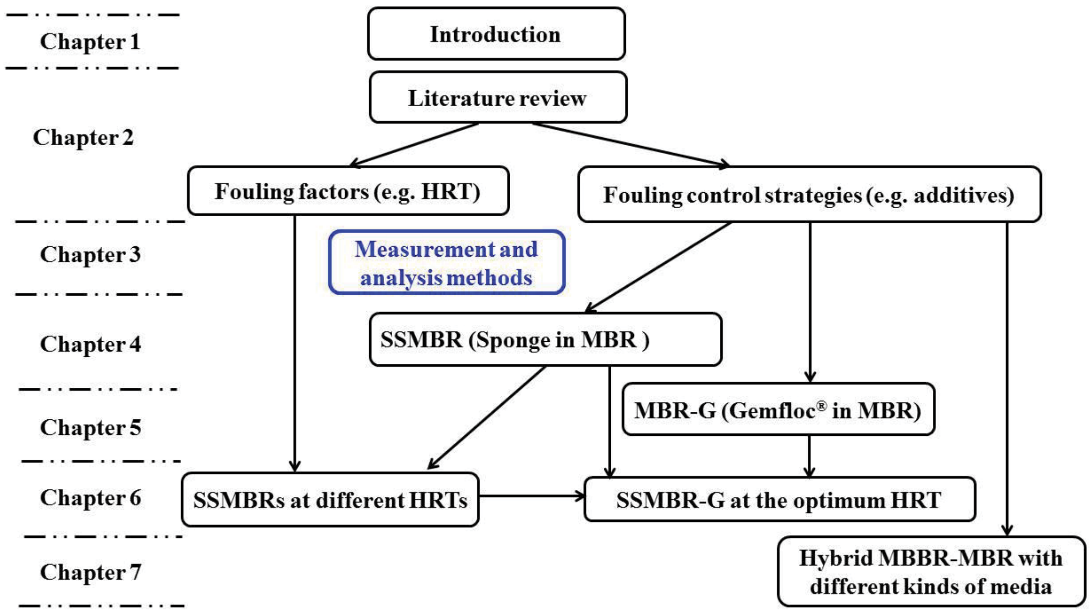

Fig. 1.1. Main structure of this research

Chapter 1 firstly presents the general information about membrane fouling and recent fouling control strategies. The research motivations, scope and significance are then highlighted. Moreover, the framework of the thesis is also given in this chapter.

Chapter 2 reviews the general aspects of MBR (e.g. their application, properties, and different configurations, membrane fouling), and the fundamental knowledge of biofouling (biofilm formation process) and their consequences. Moreover, key factors affecting membrane fouling are summarized, including biomass characteristics (e.g. EPS, SMP, biopolymer clusters (BPC), mixed liquor suspended solids, sludge floc size), operating conditions (e.g. sludge retention time (SRT), hydraulic retention time (HRT)), feedwater characteristics (e.g. organic loading rate (OLR), salinity), and membrane materials (e.g. hydrophobicity). Finally, the emerging fouling mitigation approaches are also addressed in terms of membrane cleaning, the addition of media or flocculants, and other new methods.

Chapter 3 provides the detailed materials and methods in this study. It consists of wastewater compositions, experimental set-up and operating conditions, treatment performance and membrane fouling analyses, characterization of activated sludge and cake layer samples.

Chapter 4 evaluates membrane fouling behaviour in the SSMBR and the CMBR for synthetic wastewater treatment. This chapter investigates membrane fouling by considering sludge properties in MBR with and without sponge addition. It also proposes a new fouling indicator to indicate membrane fouling potential by the main foulants (EPS and SMP).

Chapter 5 shows the experimental results of the MBR and the MBR without Gemfloc® addition (MBR-G) with respect to fouling behaviour by altering characteristics of mixed liquor and cake layer. This chapter also presents a modified resistance-in-series model to elucidate the relationship between sludge characteristics and membrane fouling.

Chapter 6 reports the experiments in the SSMBRs at various HRTs. The performance of the SSMBR with Gemfloc® addition (SSMBR-G) at the optimum HRT is then evaluated in terms of their fouling behaviour and sludge characteristics.

Chapter 7 develops a new functional media which is prepared by modifying plastic carrier using sponge. It firstly compares organic, nitrogen and phosphorus removal performance of two MBBRs with the new functional media and the plastic carrier. Treatment performance and membrane fouling behaviour in both MBBRs combined with MBR (integrated MBBR-MBR systems) are also assessed.

Finally, Chapter 8 summarizes the works and major results of studies in this thesis, and address future research needs and trends of the integrated MBR systems as well.

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Chapter 2

Literature Review

Chapter 2 Literature Review

2.1. Introduction

Membrane bioreactors (MBRs) have been extensively used for various kinds of wastewater treatment, such as industrial, domestic and municipal wastewater due to its superior merits, such as high effluent quality, small footprint, complete liquid-solid separation, high biomass content, absolute control of sludge retention time (SRT) and hydraulic retention time (HRT), low sludge production (Guo et al., 2009). However, membrane fouling, especially biofouling, presents a major hindrance for their wide application and further development (Meng et al., 2009a; Ngo et al., 2012). Recently, there have been some review papers concerning membrane fouling in MBRs for wastewater treatment. Le-Clech et al. (2006) compiled and analysed more than 300 publications on MBR for many domestic and industrial applications. However, this review did not give a thorough understanding of the effects of extracellular polymeric substances (EPS) on physico-chemical properties of activated sludge flocs that govern cake layer formation and biofouling. Biopolymer clusters (BPC) as another group of organic materials have yet to be explained in details with respect to their formation and effects on membrane fouling. It also did not offer more details on the influence of operating conditions (i.e. HRT and organic loading) on membrane fouling either. Meng et al. (2009a) addressed fouling behaviour, fouling factors and controlling strategies of MBRs for municipal and industrial wastewater treatment. However, the specific information on effects of sludge conditions (e.g. mixed liquor suspended solids (MLSS), sludge viscosity) on membrane fouling and impacts of bound EPS of mixed liquor on sludge characteristics (e.g. flocculation ability, hydrophobicity) was not clearly provided. The fouling characterization methods, results and fouling mitigation strategies were updated and critically re-evaluated by Drews (2010). Nevertheless, some key parameters affecting membrane fouling have yet to be elucidated in this review, such as EPS and HRT. Le-Clech (2010) reported MBR applications, design and removal performances, and current research trends. However, this review paper did not discuss the HRT as one of the main parameters affecting soluble microbial products (SMP) and membrane fouling. A holistic review about different membrane fouling phenomena during water and wastewater treatment was provided by Guo et al. (2012a), whereas this review only partially discussed biofouling in membrane filtration processes. An overview on current application of MBR technology for industrial wastewater treatment was presented by Lin et al. (2012), which reviewed and analysed more than 300 scientific publications on MBRs (including anaerobic MBR (AnMBR)) for industrial wastewater treatment. Nevertheless, as the objective of this review was not aimed to comprehensively present membrane fouling mechanisms and characteristics, membrane fouling (including fouling factors and membrane fouling propensity) was briefly mentioned (including two tables) together with some unique and significant findings in membrane fouling control measures. Although Mutamim et al. (2012) carried out a recent review on the performance of MBRs in treating high strength industrial wastewater, which covered the operation, constraints and fouling mitigation strategies in general views, the information was inadequate regarding the influence of membrane properties, biomass and operational conditions on membrane fouling along with membrane fouling mitigation. Similarly, a later review also did not clearly address the relationship between EPS/SMP and membrane fouling (Mutamim et al., 2013).

So far, several review papers have been published on biofouling in MBRs. Liao et al. (2004a) explained the biofouling mechanism in details, but this review simply summarized the major factors affecting membrane biofouling, including design and operating conditions of membrane modules and materials, hydrodynamic conditions, membrane materials, environmental and process conditions of a given activated sludge system, as well as partially mentioned sludge characteristics (e.g. mixed liquor suspended solids (MLSS) concentration, sludge floc size, EPS, and viscosity) related to membrane fouling. Effects of membrane and module, operating conditions (aeration, flux, and temperature), biomass (e.g. suspended solids, EPS, unassoicated fatty acid, hydrophobic or polysaccharide substances, proteins) on biofouling, especially the possible contribution of microbial products (e.g. SMP, EPS) to biofouling in MBRs were reported by Ramesh et al. (2006) during municipal wastewater treatment. However, the influence of HRT and other biomass characteristics (i.e. sludge viscosity, floc size) on biofouling have yet to be clearly discussed in this review. Wu and Fane (2012) summarized factors (feed characteristics, oxygen level, temperature, steady or unsteady-state operation) affecting microbial behaviors, characterization technologies of biofoulants and strategies for controlling microbial relevant fouling (application of additives (e.g. coagulants, ozone)) in MBRs when treating industrial and municipal wastewater. In spite of this, this review did not mention effects of HRT on characteristics of microbial flocs, and their metabolic products, which affected membrane fouling. Nguyen et al. (2012b) highlighted the underlying causes of membrane biofouling, the role of EPS in membrane biofouling, and presented the recent developments of methods for biofilm examination and biofouling control in water and wastewater treatment. However, this review did not elucidate the impacts of operating conditions (i.e. HRT) and sludge characteristics (i.e. MLSS) on membrane fouling, respectively. Later on, a comprehensive review conducted by Wang et al. (2013a) focused on membrane fouling caused by EPS, including their secretion, transformation, release, and adsorption in MBRs for municipal and industrial wastewater treatment. Nevertheless, the knowledge of sludge properties related to membrane fouling, such as MLSS and floc size, was lacking.

Based on aforementioned review papers, this literature review will be dedicated to a broader and detailed analysis of the main aspects related to biofouling in MBRs according to the current academic research achievements. Therefore, a more clear insight into biofouling mechanism, fouling factors, and control strategies during MBR operation process is provided in this chapter. It can help researchers to have an in-depth understanding on membrane fouling, especially biofouling in MBRs. Therefore, this chapter firstly presents general information about the MBRs (advantages, disadvantages, and configurations). Then it more specifically gives an overview of biofilm formation, various parameters related to biofouling, including mixed liquor characteristics, operating conditions, feed water characteristics, membrane characteristics, biofouling reduction approaches through membrane cleaning, application of additives (e.g. flocculants, media), and other methods.

2.2. Membrane bioreactor

2.2.1. Fundamental aspects of membrane bioreactor and its properties

For a traditional biological treatment system, nutrients and organic matters are removed from wastewater by microorganisms, resulting in the formation of new biomass. The application of conventional activated sludge process in wastewater treatment depends on aggregated suspended sludge with good settleability. A subsequent clarifier is required for separating the sludge from the treated water (Fig. 2.1). Membrane bioreactor (MBR) as a promising technology is referred to as the suspended-growth activated sludge biological treatment coupled with membrane filtration process for the effective solids-liquid separation. The most commonly used membranes in MBRs are low–pressure membranes, including microfiltration (MF) and ultrafiltration (UF). MF membrane can act as a physical barrier for removing particles, bacteria and protozoa cysts in size separation ranges from 100 to 1000 nm. UF membrane can not only reject particles, bacteria, protozoan cysts and some viruses ranging from 5 to 100 nm, but also remove larger organic macromolecules based on molecular weight cutoff (MWCO) levels (typical MWCO levels for UF membranes in the range of 10000–500000 daltons) (Radjenović et al., 2008; Water Environment Federation, 2006 and 2012).

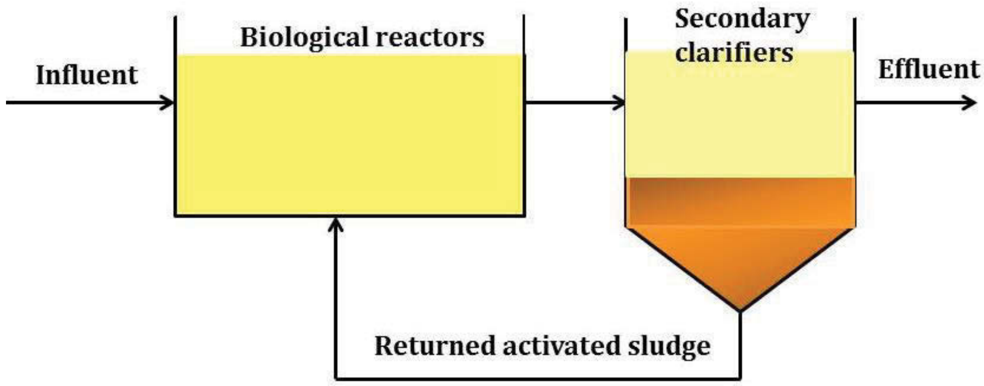


Fig. 2.1. Conventional activated sludge process
(adapted from Water Environment Federation, 2012)

In recent years, MBRs have been increasingly used for various types of wastewater treatment, including industrial wastewater (e.g. the food processing, petroleum, oily and petrochemical wastewaters), municipal or domestic wastewater due to their distinguishing merits as follows (Lin et al., 2012; Ngo et al., 2012; Wang et al., 2014b):

(1) Complete retention of all suspended substances (e.g. microorganisms, organic and inorganic matters in the influent) in an MBR reactor by membrane filtration promotes the removal of carbon and organic nutrients, which enables a high effluent quality. Therefore, the effluent can meet the water reuse requirement or stringent effluent regulations;

(2) The separation of biomass from effluent by membrane filtration allows absolute control of HRT and SRT. Hence, MBR can remove slow-degradable pollutants without sludge waste (infinite SRT). It permits the excellent performance of MBR when treating wastewater with fluctuated flow rate and feed quality;

(3) As compared to the conventional activated sludge systems, MBRs can also achieve high treatment performance in a smaller footprint due to high biomass concentration and no requirement of secondary clarifiers;

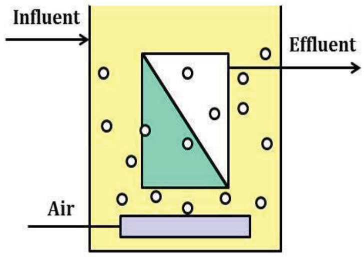
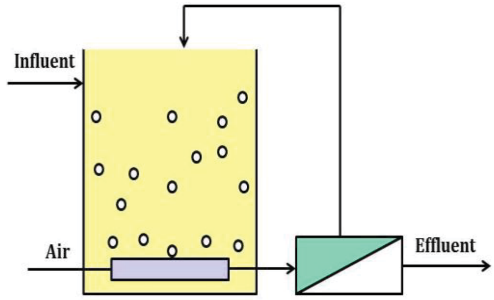
(4) MBRs have the ability to operate at high MLSS concentration. Sludge production is decreased due to the fact that limited substrate at high biomass concentration is used for microorganism maintenance.

However, the wide application of MBR is mainly limited by membrane fouling and its consequences of increased capital, operational and maintenance costs, more frequent membrane replacement and usage of chemical reagents for membrane cleaning. Moreover, when MBRs are operated at extremely high flow rate, upstream flow equalization or extra freeboard is required to be designed with MBR systems (Water Environment Federation, 2012).

2.2.2. Membrane bioreactor configurations

There are two types of membrane separation processes employed in MBRs: submerged MBR (immersed MBR) and side-stream MBR (or external MBR). Side-stream MBR contains a pressure-driven membrane unit located outside the biological tank. Submerged MBR is operated in dead-end mode with vacuum-driven membranes which are directly installed into the biological tank (Radjenović et al., 2008). Current studies focus on the application of submerged MBR because of its superior advantages when comparing with side-stream MBR, especially in terms of fouling degree and energy consumption as shown in Table 2.1.

Table 2.1. Comparison between submerged MBR and side-stream MBR (modified from Radjenović et al., 2008)

Reactors	Submerged MBR	Side-stream MBR
Illustrations		
Application (can be applied in)	• Municipal–scale systems • Various types of solids in activated sludge	• Industrial systems • High temperature of feed water
Shear provided by	Aeration	Pump
Pressure	Low	High
Energy consumption	Significantly Low	High
Fouling degree	Low	High

2.2.3. Membrane fouling

Membrane fouling takes place due to the attachment and deposition of microorganisms, colloids, solutes and cell debris within/on membranes. Based on cleaning methods, membrane fouling can be classified into three different categories, including removable fouling (reversible fouling) reduced by physical cleaning (i.e. backwashing), irremovable fouling eliminated by chemical cleaning, and irreversible fouling (permanent fouling) which cannot be removed by any methods. Cake layer formation and pore blocking contributes to removable and irremovable fouling, respectively. According to the types of foulants, membrane fouling can be divided into four categories, including particulate/colloidal fouling, organic fouling, inorganic fouling and biofouling. Particulate/colloidal fouling occurs by coverage of pores or some regions of the membrane by inorganic or organic particles/colloids, or development of a cake layer taking over the role of the membrane. Organic fouling involves the adsorption of dissolved components and colloids (e.g. humic and fulvic acids, hydrophilic and hydrophobic materials, polysaccharides, proteins) onto the membrane. Inorganic fouling is induced by: (1) precipitation of dissolved components (e.g. iron, manganese and silica) on the membrane owing to pH change (scaling) or oxidation along with generation of iron or manganese oxides; (2) co-deposition of biopolymers and metal ions on the membrane, resulting in a dense cake layer formation; and (3) the presence of coagulant/flocculant residuals. Biofouling occurs when microorganisms (e.g. bacteria) attach on the membrane (Guo et al., 2012a; Meng et al., 2009a).

MBR can be operated at constant flux or constant pressure (transmembrane pressure (TMP)). When operating at the constant TMP, membrane fouling potential or membrane permeability is indicated by the variation of fluxes. The MBR initially features a rapid flux decline, followed by a slow reduction of flux and finally an almost stable flux. In recent years, an MBR filtration experiment has more often been carried out at a constant flux. During the operation, change of membrane fouling or membrane permeability is indicated by evolution of TMP development during the operational period (Le-Clech et al., 2006; Water Environment Federation, 2012). It has been reported that membrane fouling follows a three-stage process. During Stage 1, conditioning fouling occurs because of interaction between the membrane surface and EPS in activated sludge, which yields internal fouling and pore blocking. At the later stage (Stage 2), a slow fouling along with cake formation is caused by coverage of the membrane surface by SMP and further adsorption of biomass particles, colloids and organics on the membrane. Subsequently, due to severe fouling in some regions or pores on the membrane, a significant increase in the local flux above the critical flux would be obtained within less fouled membrane areas or pores (Stage 3). It gives rise to a self-accelerating fouling as indicated by an exponential TMP rise (Braak et al., 2011; Le-Clech et al., 2006). In order to maintain sustainable membrane permeability, MBR is commonly operated at sub-critical flux. However, membrane fouling still occurs when operating at this flux. A current study carried out by Li et al. (2013) presented a new insight into subcritical flux fouling at three consecutive stages: I) a moderate linear TMP rise associated with bound EPS in activated sludge; II) an exponential TMP increment governed by SMP in activated sludge; III) a rapid linear TMP increase determined by sludge characteristics (e.g. floc size) due to a thick cake layer of flocs formed on the membrane surface.

2.3. Biofilm formation

Among six principle fouling mechanisms, biofouling is the most complicated as it is associated with undesirable deposition, growth and metabolism of bacterial cells or flocs with agglomeration of extracellular materials on the membranes (Guo et al., 2012a). Generally, there are six steps involved in biofilm formation (Bitton, 2005) (Fig. 2.2): (1) formation of a conditioning film on the surface after contacting between pristine surface and organic matters. The surface conditioning layer is comprised of proteins, glycoproteins, humic-like substances and other dissolved or colloidal organic matter; (2) transport of microorganisms (planktonic cells) to conditioned surface by diffusion, convection, turbulent eddy, and chemotaxis; (3) adhesion of microorganisms to conditioned surface due to the negatively free energy of adhesion, hydrophobic interactions, as well as the balanced force between Lifshitz-Van der Waals forces and repulsive, or electrostatic forces on both microbial and substratum surfaces; (4) continuous attachment of microorganisms on membrane surface and cohesion of microorganisms on the surface previously attached by microbes due to the presence of EPS; (5) growth and accumulation of biofilm; (6) subsequent development of a three-dimension biofilm. Therefore, attachment and growth of microorganisms play a crucial role in the biofilm accumulation on membrane (Ivnitsky et al., 2005).

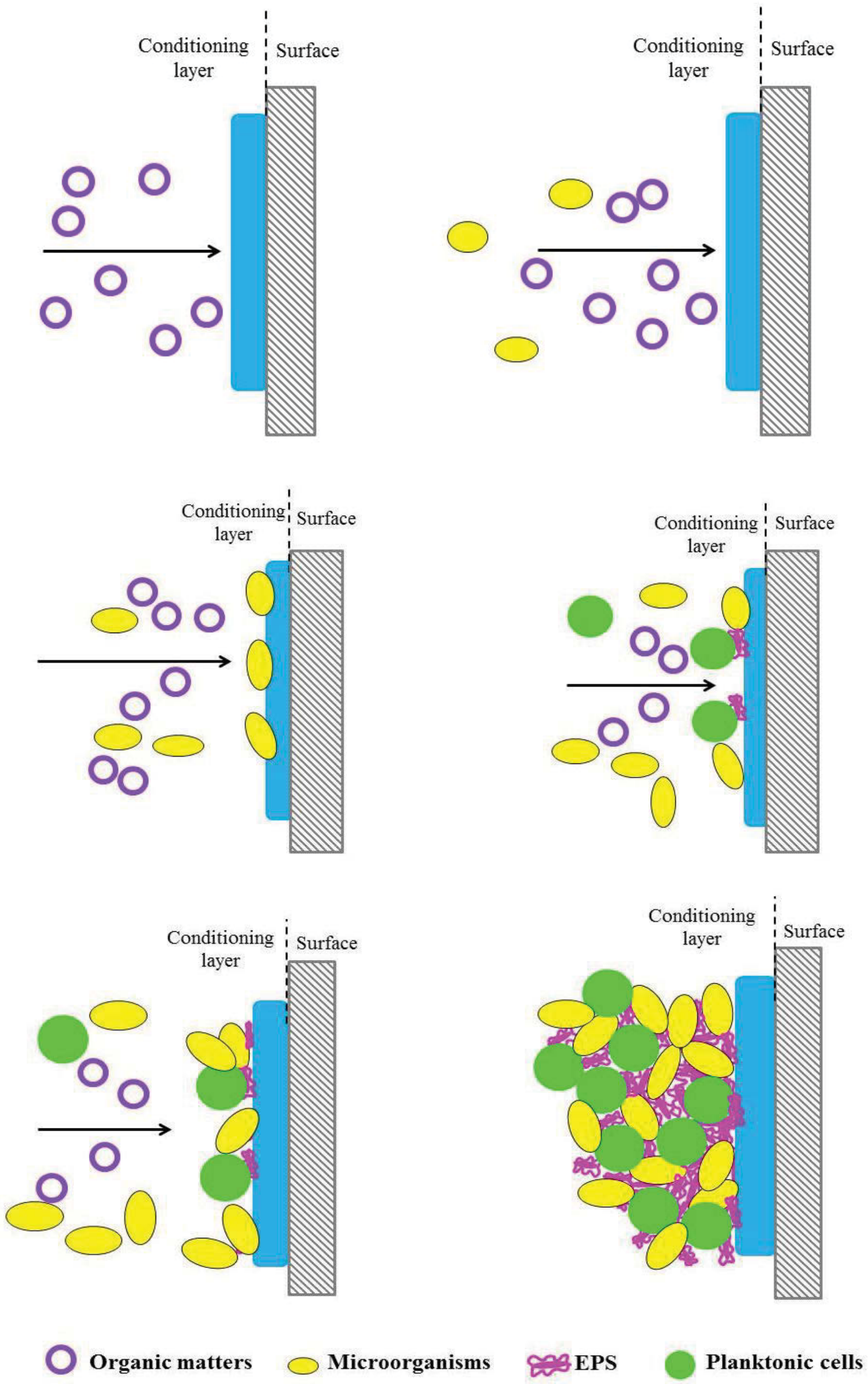


Fig. 2.2. Schematic of biofilm formation on a surface (modified from Bitton, 2005)

The effects of biofouling on the performance of membrane systems are shown as follows: (1) decreasing membrane permeability due to bacterial attachment and subsequent growth on the membrane surface; (2) causing precipitation of calcium carbonate at an elevated pH due to photosynthesis in algal biofilm; (3) inducing accumulation of abiotic particles because of microorganism adhesion or an enzymatic attack on the membrane or the glue lines; (4) leading to concentration polarization contributed by the biofilms on the membrane surface (Dreszer et al., 2013; Flemming, 2002). During the MBR process, biofouling results in decreased membrane permeability, declined flux, more frequent membrane replacement and the subsequent consequences of increased operational and maintenance costs.

2.4. Factors affecting biofouling in MBR

It has been suggested that there are four major types of factors affecting membrane fouling, including mixed liquor properties, operating conditions, feed water characteristics and membrane properties (Fig. 2.3). The last three factors could alter sludge characteristics and further fouling propensity (Le-Clech et al., 2006; Meng et al., 2009a). This section focuses on their effects on sludge properties and biofouling in MBRs.

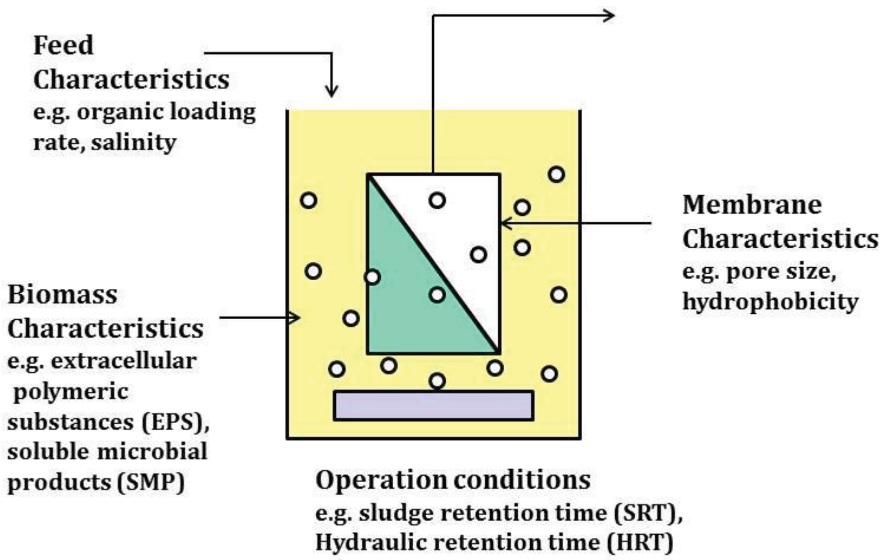


Fig. 2.3. Factors affecting fouling in the submerged MBRs (modified from Le-Clech et al., 2006)

2.4.1. Mixed liquor properties

A. Bound extracellular polymeric substances (EPS)

EPS can be found outside cells and inside microbial aggregates. The abbreviation "EPS" symbolizes various kinds of macromolecules such as polysaccharides, proteins, nucleic acids, lipids and other polymeric compounds inside the microbial aggregates (Sheng et al., 2010; Wingender et al., 1999). As the "house" of microorganisms, EPS build up a three-dimension, gel-like and highly hydrated matrix for immobilization of microorganisms. EPS promote the aggregation of sludge flocs, determine their mechanical stability as the construction materials, and allow the cells to resist the adverse environmental influences by forming a protective barrier around them. EPS are directly associated with sludge properties (e.g. hydrophobicity, surface charge, flocculation) and membrane fouling propensity. They have a dynamic double-layered structure: inner layer, consisting of tightly bound EPS (TB-EPS) which clings tightly to the surface of cells, and outer layer, comprising loosely bound EPS (LB-EPS) which is a slime layer loosely attached to outer layer (Flemming and Wingender, 2001; Lin et al., 2014; Sheng et al., 2010). It was reported that LB-EPS contents were negatively correlated with the flocculation of activated sludge, while no significant relationship was found between TB-EPS and sludge flocculation. Much higher amounts of LB-EPS would lessen cell adhesion and deteriorate the floc structure, which aggravated cell erosion, and decreased bioflocculation, settleability and dewaterability of sludge flocs (Li and Yang, 2007). However, Liu et al. (2010) suggested that both LB-EPS and TB-EPS were critical in the sludge agglomeration. LB-EPS positively affected the sludge flocculation, while the effects of TB-EPS on sludge aggregation were determined by the separation distance between sludge cells. The different results of these two studies may be attributed to different sludge and extraction methods. The former study obtained the sludge from a sewage treatment works and used a heat extraction method for LB-EPS and TB-EPS extraction, while the later study collected the sludge from a municipal treatment plant and employed the oscillation-ultrasound method followed by the cation exchange resin (CER) technology for the sample extraction. Some current studies pointed out the positive influence of TB-EPS on the gel-like and compact structure of activated sludge (Yuan and Wang, 2013; Yuan et al., 2014). In order to identify the role of EPS in the formation of activated sludge flocs, Wang et al. (2014c) distinguished the EPS in the floc level (extra-microcolony polymers, EMPS) and in the microcolony level (extra-cellular polymers, ECPS) (Fig. 2.4). The EMPS and ECPS are referred to as the exopolymers, which locate at the outer layer and the interior layer of microcolonies, respectively. They found that the floc level flocculation was mainly governed by cation bridging interactions provided by EMPS. On the other hand, polymer entanglement and hydrophobic interaction determined the microcolony level aggregation due to the presence of larger protein molecules and higher percentages of protein in ECPS. With respect to the compositions, EPS mainly consist of polysaccharides (EPS_C) and proteins (EPS_P) (Guo et al., 2012a). As EPS_C can be dissolved well in water through the strong electrostatic interactions and hydrogen bond forces, they are responsible for the hydrophilic nature of activated sludge. The amino acid with hydrophobic side groups in EPS_P plays a crucial role in the hydrophobicity of sludge flocs. Moreover, the amino groups in EPS_P can increase zeta potential or decrease net negative surface charges of sludge flocs by neutralizing some of negatively charged activated sludge due to the presence of positive charges (Liu et al., 2014).

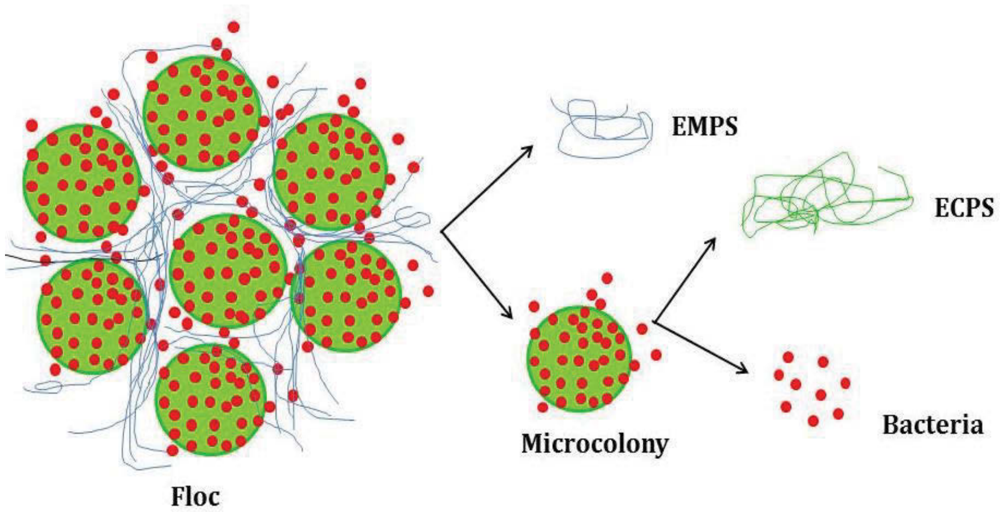


Fig. 2.4. Schematic diagram of the classification paradigm of EPS in activated sludge (adapted from Wang et al., 2014c)

During long-term operation, bound EPS show a strong relationship with membrane fouling development. The increased EPS levels not only facilitated the formation of a compressible and less porous cake layer by co-deposition of EPS and microorganisms on the membrane, but also decreased cake layer permeability, resulting in higher filtration resistance and serious membrane fouling (Chae et al., 2006). Tian et al. (2011a) reported that more severe membrane fouling occurred during filtration of bulking sludge with overgrowth of filamentous bacteria was associated with the presence of more cell-bound EPS. Moreover, these sludge flocs with higher EPS were more readily deposited on the membrane surface because of an increase in proteins/polysaccharides (PN/PS) ratio and floc hydrophobicity. An interesting relationship between foaming and fouling was found to be correlated with EPS contents, implying that the fouling rate reduction was due to entrapment of bound EPS in the foam (Cosenza et al., 2013). TB-EPS and LB-EPS also affected membrane fouling. TB-EPS made a significant contribution to membrane fouling, while LB-EPS in the sludge were mainly responsible for filtration resistances of sludge and irreversible fouling (Ramesh et al., 2007). Wang et al. (2009a) found that LB-EPS had a more remarkably positive effect on membrane fouling than TB-EPS. Liu et al. (2012) indicated that polysaccharides in LB-EPS showed a stronger relationship with physical properties of the sludge. Higher LB-EPS levels induced a poor flocculability, a low settleability, and the formation of looser sludge flocs, which increased cake layer resistance. It was also pointed out that a high level of LB-EPS promoted the generation of fine particles and floc attachment on the membrane, giving rise to the formation of a dense cake layer and serious membrane fouling, whereas TB-EPS could lower membrane fouling. A greater TB-EPS to LB-EPS ratio reflected a high flocculability of activated sludge, thus causing the generation of large flocs, less membrane fouling and lower filtration resistance (Azami et al., 2011).

It was reported that biocake on the membrane surface mainly consisted of microbial cells and EPS, with an initially reversible fouling inducing by accumulation of easily removable organic substances, followed by a gradual development of irreversible fouling at an elevated EPS levels (Domínguez et al., 2012). Hwang et al. (2012) suggested that at the subcritical flux, bio-cake was mostly comprised of EPS (especially polysaccharide) rather than bacterial cells. It induced the greatest rate of resistance increase during the fouling period (the steep TMP rise), which could be explained by inhomogeneous pore loss and alteration of percolation by EPS accumulation. Su et al. (2013a) indicated that cake sludge possessed more LB-EPS with higher PN/PS than bulk sludge, which could lead to less attachment of biomass and deterioration of cake sludge floc structure. It resulted in worse dewaterability and filterability of cake sludge, together with higher membrane fouling potential. This implied that the microbial sludge flocs demonstrating higher PN/PS ratio promoted the cake layer formation on the membrane surface. More EPS in cake sludge could be attributed to the selective accumulation of small flocs and rejection of SMP (Lin et al., 2011a). Furthermore, with the growth of bio-cake, the presence of an anoxic and endogenous environment in the deeper layer resulted in cell lysis and release of EPS and polysaccharides (Hwang et al., 2008).

B. Soluble microbial products (SMP) and biopolymer clusters (BPC)

SMP is referred to as a group of organic substances, which are dissolved into the solution. The generation of SMP is caused by requirement of concentration equilibrium for microorganisms, starvation, increased exogenous energy source, bacteria death as a result of shock load of substrate, low levels of essential nutrient, stressful conditions (e.g. sharp temperature fluctuations) as well as normal bacterial growth and metabolism (Barker and Stuckey, 1999). It was found that when compared to other sludge characteristics (e.g. MLSS, particle size distribution, bound EPS), SMP showed the strongest correlation with membrane fouling rate (dTMP/dt) (Zhang et al., 2015a). High SMP production affected by microbial community caused severe membrane fouling during the start-up period when operating a hybrid anoxic-oxic MBR (Li et al., 2012). SMP (or soluble EPS) could be readily deposited and adsorbed onto the membrane owing to the permeation drag and their low back transport velocity. Thus, they would more easily induce the formation of gel layer than cake layer, cause membrane pore blocking, and penetrate into the pores and/or spaces between particles in the cake layer. In gel layer and cake layer, SMP with macromolecular character were the major soluble organic substances. Furthermore, the gel layer possessed more SMP and smaller flocs than the cake layer. It had unusually high specific filtration resistance being almost 100 times higher than that of the cake layer. These effects led to severe membrane fouling, reduced the porosity of the gel layer, and also decreased permeate flowrate (Hong et al., 2014; Wang et al., 2012; Wu and Huang, 2009). Moreover, the molecular weight (MW) distribution of SMP also impacted membrane fouling. Arabi and Nakhla (2010) indicated that fouling showed a strongly positive relationship with SMP concentration in the range of 10-100 kDa, while lower fouling rate was related to higher concentrations of the SMP fraction < 1 kDa. Fouling in MBRs was not affected by fractions of > 100 kDa and 1–10 kDa of SMP. Shen et al. (2010) and Shen et al. (2012) indicated that the fraction of MW > 100 kDa (large MW) in hydrophilic substances

(HIS) of SMP with polysaccharides as the major fraction caused severe flux decline and pore-plugging. It suggested that instead of hydrophobic interaction, membrane fouling and irreversible fouling in the early stage of filtration was mainly determined by size exclusion in this study.

For chemical compositions of SMP, proteins (SMP_P) and polysaccharides (SMP_C) are the main components, which lead to severe membrane fouling (Guo et al., 2012a). It has been suggested that SMP_C with partially hydrophilic nature not only induced irreversible fouling, but also facilitated membrane pore blocking and gel layer formation (Jermann et al., 2007). Membrane fouling also exhibited a positive correlation with SMP_P concentration. Pore clogging of membranes through attaching SMP_P to the surface of membrane pores and their subsequent denaturation could result in serious membrane fouling (Rafiei et al., 2014). Higher SMP_P/SMP_C ratio reduced irreversible fouling and promoted the interaction of proteins and polysaccharides for cake layer formation on membrane surface (Yao et al., 2011). Gao et al. (2013a) also indicated that the chemical composition of the supernatant (PN/PS) showed a significant effect on membrane fouling in MBRs rather than their quantity. PN/PS ratio in the supernatant was positively correlated to filtration resistance. When portion of PN in the supernatant was higher than that of PS, cake layer was formed due to their higher stickiness. With respect to bacteria phase, SMP can be divided into two subgroups: biomass-associated products (BAP), which are obtained during substrate metabolism, and utilization-associated products (UAP), which come from biomass decay (Laspidou and Rittmann, 2002; Meng et al., 2009a). Tian et al. (2011b) conducted a series of stirred dead-end filtration tests to evaluate the fouling potential of BAP and UAP. The results indicated that almost complete reduction of proteins and partial decrease of polysaccharide of UAP and BAP in permeate was caused by the membrane interception effects. It implied that polysaccharides and proteins in the supernatant would induce severe membrane fouling. Moreover, the foulants on membrane surface were mainly composed of proteins and polysaccharides. The modelling work demonstrated that cake filtration and complete blockage were primary fouling mechanisms for BAP and UAP filtration, respectively, which demonstrated that fouling potential for the UAP was higher than that for BAP. The effects of SMP on membrane fouling were also found to be dependent on their molecular weight (MW). Jiang et al. (2010) pointed out that in spite of higher retention percentage of the BAP filtration, the highest specific cake resistance and pore blocking resistance were caused by the UAP filtration due to their higher percentage of low MW molecules.

Fig. 2.5 shows schematic illustration of correlation between SMP and BPC in MBRs. A pool of organic substances, classified as BPC which were larger in size than SMP, is also found to be an important foulant and exhibits a notable effect on membrane fouling. The interception of BPC by sludge layer, the aggregation and clustering of SMP and small BPC as well as attachment of loose EPS within the sludge layer are responsible for the formation and growth of BPC in sludge cake. BPC act as glue to induce the biomass deposition on membrane surface, which accelerates the formation of sludge cake with high sticky and impermeable property. It results in serious membrane fouling and high filtration resistance of cake layer. On the other hand, BPC can be detached from membrane surface into the sludge suspension by the turbulent shear caused by aeration in SMBR. Large BPC of activated sludge in MBR will be deteriorated along with the presence of smaller SMP segment due to the shear force (Fig. 2.5) (Sun et al., 2008; Wang and Li, 2008).

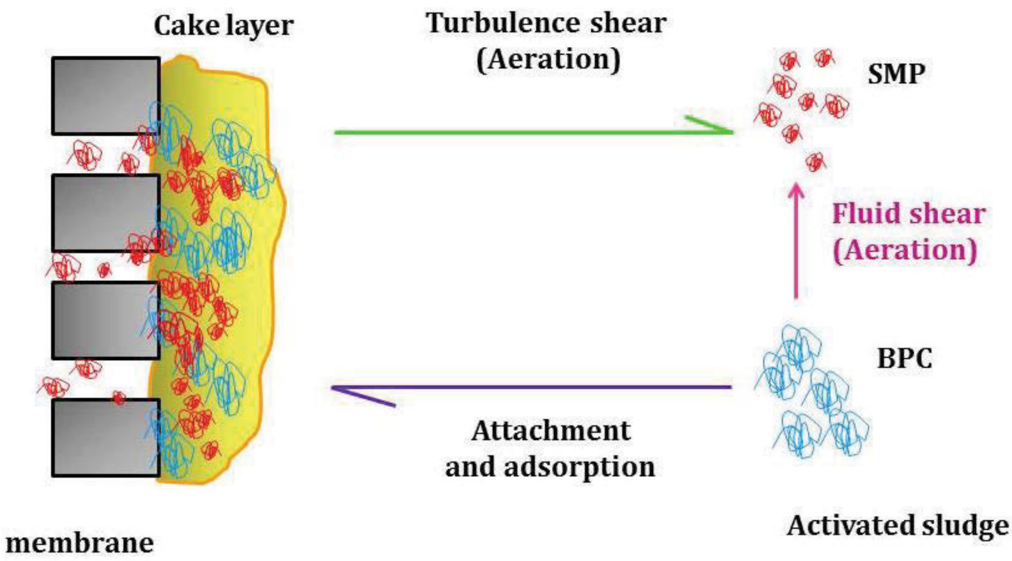


Fig. 2.5. Schematic illustration of correlation between SMP and BPC in MBR

C. Mixed liquor suspended solids (MLSS) and sludge viscosity

MLSS concentration has also been identified as one of the major factors affecting membrane fouling. Table 2.2 shows effects of MLSS concentration on biofouling in MBR. Some previous studies explored the correlation between sludge viscosity and MLSS concentration as well as their impacts on membrane fouling. Kornboonraksa and Lee (2009) demonstrated the relationship between sludge viscosity and MLSS concentration (from 9.3 to 36.1 g/L) by an exponential equation (sludge viscosity = $0.0035e^{0.00004MLSS}$). Higher MLSS levels induced rapid flux decline, lower membrane filterability, higher TMP and greater membrane fouling resistance. Moreover, higher sludge viscosity dampened the back transport effect but increased the net force towards the membrane surface, leading to the deposition of sludge flocs and deterioration of membrane performance (Kornboonraksa and Lee, 2009; Meng et al., 2007a). Wu and Huang (2009) also suggested that high MLSS concentration (> 10 g/L) caused a wide-range increase in viscosity, which showed a significant impact on membrane filterability. It implied that MLSS of 10 g/L was a critical point. It was further indicated that higher viscosity reduced the positive effect of aeration on membrane fouling reduction, depressed membrane fibre vibration, and permitted larger air bubbles to move at slow rise velocity (Lay et al., 2010; Wicaksana et al., 2006). Lousada-Ferreira et al. (2010) collected activated sludge from full-scale MBRs and submitted the sludge to cross-flow filtration employing the same membrane and operating conditions. Various MLSS concentrations (3.6–18.3 g/L) were obtained by diluting the original samples. MBR activated sludge with MLSS concentration equal to or lower than 10 g/L generated dilutions with higher filterability, while the opposite results were obtained for the original samples at MLSS concentrations above 10 g/L. Moreover, membrane fouling caused by MLSS concentration (4–25 g/L) was also found to be associated with EPS and SMP production. Higher MLSS levels could induce an increase in the concentrations of EPS and SMP (especially carbohydrate) due to reduced sludge yield, more cell decay and stressful conditions at the limited biodegradable substrate, which contributed to higher fouling rate. When increasing MLSS concentration (> 10 g/L) by two times, fouling rates increased by almost four times. Nine-fold increase in fouling rate was obtained over a three-fold increase in MLSS concentration above 15 g/L (Lee and Kim, 2013; Yigit et al., 2008). However, Dvořá et al. (2011) pointed out that as compared to low MLSS concentration of 6 g/L, EPS concentration decreased with operation time at relatively higher MLSS concentrations (10 or 14 g/L) due to utilization of EPS for metabolism of microorganisms at a low food to microorganim (F/M) ratio or at low shear force inducing by aeration at higher sludge viscosity. On the contrary, EPS levels were higher at lower MLSS concentration as the cells protected themselves from mechanical stress to release EPS at the higher shear force under constant aeration intensity.

Overall, higher MLSS levels, especially above 10 g/L, had a more significant effect on sludge viscosity, sludge filterability, and membrane performance. Therefore, the optimum MLSS is less than 10 g/L. A selection of an optimum MLSS concentration should also depend on characteristics of feed water and sludge, membrane characteristics (e.g. materials, pore size), sludge ages (or sludge retention time).

Table 2.2. Effects of MLSS concentration on biofouling

Type of wastewater	MLSS concentration	Effects on biofouling	References
Synthetic wastewater (MBR start-up)	3.0-7.9 g/L	• At low MLSS levels (< 5.0 g/L), less membrane fouling was observed at the elevated MLSS concentration, while fouling rate was almost zero when MLSS concentration increased to 7.9 g/L.	Li et al., 2012
Real wastewater	2.3-20 g/L	• A slight change of permeation flux was observed when increasing biomass concentration within low MLSS range (< 4.1 g/L). At higher MLSS concentration (< 6.75 g/L), MLSS significantly affected permeate flux decline. • Higher MLSS concentration induced higher rate of TMP change as well as higher total fouling resistance due to cake layer formation. The increase in MLSS from 5 to 20 g/L reduced the critical flux by 3 times.	Bani-Melhem et al., 2015; Damayanti et al., 2011

D. Floc size

It has been suggested that the permeate flux (membrane permeability) was positively correlated with sludge floc size (Kornboonraksa and Lee, 2009). It was pointed out that mixed liquor with mean floc diameter larger than 80 μm showed a more significant influence on membrane filterability with a lower increase rate of membrane resistance. When the mean floc size was lower than 50 μm, mixed liquor exhibited poor filterability and deteriorated membrane permeation (Meng et al., 2007b; Wu and Huang, 2009). The adhesion of sludge flocs as well as cake layer formation on the membrane is governed by two opposite forces, including permeate drag force induced by permeate flux, and back transport force which comprises Brownian diffusion, inertial lift and shear-induced diffusion (Meng et al., 2008). From the hydrodynamic viewpoint, large flocs such as suspended solids could be dragged away from the membrane due to high shear-induced diffusion and inertial force as well as lower Brownian diffusion. The deposition of some larger and looser sludge flocs on the membrane induced formation of a more porous and permeable cake layer, resulting in less fouling resistance (Dizge et al., 2011a; Pan et al., 2010). On the other hand, Brownian diffusion controlled the motions of small flocs at lower shear stress (Pan et al., 2010). Thus, the flocs with a size closer to or smaller than the size of membrane pores (e.g. colloidal substances, soluble macromolecules) could be more easily deposited on the membrane and penetrate into the membrane pores due to the permeate drag and high Brownian diffusion. It induced fouling layer formation and pore blocking. Moreover, the adsorption and accumulation of smaller flocs into cake layer resulted in the formation of a dense and less porous cake layer with a higher filtration resistance (van den Brink et al., 2011; Wang et al., 2011a). Lin et al. (2011a, 2011b) pointed out that cake layer possessed larger amounts of small sludge flocs than sludge particles due to the presence of more bound EPS in small flocs, which promoted attachment of the flocs on the membrane. Thermodynamic analyses suggested that although the reduced floc size induced a slightly higher specific energy barrier, smaller flocs promoted an increase in the attractive specific interaction energy in contact. The whole effect favoured the deposition of small flocs on the membrane, giving rise to the formation of less porous cake layer. It increased specific cake resistance, whilst retaining more biopolymer matters in pores, which led to higher osmotic pressure-induced resistance (Shen et al., 2015). Overall, small flocs made a greater contribution to membrane fouling development than large flocs.

2.4.2. Operating conditions

A. Sludge retention time (SRT)

It has been widely accepted that SRT is one of the most critical operating parameters during MBR operation as it affects sludge characteristics (e.g. MLSS, SMP, EPS) and further influences membrane fouling (Le-Clech et al., 2006). Table 2.3 summarizes the effects of SRT on biofouling in MBR. When treating synthetic wastewater, Wu et al. (2011a) operated MBRs at a high flux of 30 $L/m^2 \cdot h$ under different SRTs (10, 30 days and infinity). They found that SRTs affected the microbial community, exerting the influence on the activated sludge characteristics (e.g. MLSS concentration, EPS properties) and hydrodynamic conditions (the dynamic balance between shear force and permeate drag). It in turn determined cake layer development and membrane fouling propensity. At infinite SRT, membrane fouling (cake-fouling) was more severe because of the presence of small floc size and more EPS. It was pointed out that a moderate SRT range from 20 to 40 d could be maintained for a reliable operation in MBR (Chen et al., 2012). PS concentration in EPS accumulated from SRT of 10 to 40 d arising from endogenous respiration, cell lysis and secretion microbial death, while a prolonged SRT above 40 d induced a decrease in PS concentration due to biodegradation of PS with less available nutrients. However, PS value at higher SRT (> 40 d) was still higher than that at lower SRT (< 40 d). Reduction of supernatant COD (SMP) was observed at shorter SRT (< 40 d) as a result of biodegradation. When SRT extended, SMP accumulated due to dissoulution of intracellular substances induced by the breakdown of dead and biodegradation of less SMP as a result of limited oxygen and substrate for activated sludge flocs. During short-term filtration of activated sludge of a submerged nonwoven bioreactor with larger pore size (14 μm) at the fixed pressure of 5 kPa, Chuang et al. (2011) indicated that fully reversible fouling was obtained at SRT of 15 days with significantly high resistance percentage of suspended solids (67%), followed by colloids and solutes at 27% and 6%, respectively in the descending order. At longer SRTs of 30 and 60 d, irreversible fouling was associated with a remarkable increase in the resistance percentage of colloids (71.7% and 57.3%, respectively). In this study, irreversible fouling in large-pore materials was mainly determined by pore blocking causing by particles or colloids with size similar to that of nonwoven fabric pore.

When treating a pharmaceutical process wastewater at various SRTs (15 d, 30 d, without sludge wasting (WSW)), both permeate volumes and steady state fluxes of the submerged MBR (SMBR) were higher at increased SRTs. It could be ascribed to the decreased membrane fouling caused by lower concentrations of EPS and SMP (including PN and PS). Total fouling resistance (R_T) decreased with increasing SRTs as a result of the reduced cake resistance (R_C). Rc was the major contributor for R_T under the WSW, while for 15 and 30 days SRT, membrane pore resistance (R_P) increased when comparing to that for the WSW (Kaya et al., 2013). A later study conducted by

Zhang et al. (2015a) in an anaerobic-oxic MBR (AOMBR) at SRT of 30 d exhibited better filtration performance as compared to SRTs of 10 and 90 d when treating campus sewage. Median particle size and MLSS concentration in the MBRs followed the order of SRT = 90 d > SRT = 30 d > SRT = 10 d. However, an opposite trend was observed for bound EPS, with the highest value at SRT of 10 d. SMP concentrations were higher at SRTs of 10 and 90 d than those at SRT of 30 d. Therefore, SRT of 10 d gave rise to pore blocking and serious irreversible fouling, while cake layer formation and severe reversible fouling were induced at SRT of 90 d.

The above studies operated an MBR system with one stabilized biomass by removing some sludge for the sludge age adjustment. Thus, nutrient available for the biomass at higher sludge concentration decreased when SRT was extended, leading to the consumption of SMP as the substrate for microorganisms. A recent study conducted by Villain and Marrot (2013) dissociated the effects of SRT from those of F/M ratio on lab-scale MBRs performance through stabilizing two acclimatizations at SRT of either 20 or 50 d at a constant F/M ratio of around 0.2 $kg_{COD}\ kg_{MLVSS}^{-1}\ d^{-1}$. During stable conditions, total concentrations of SMP in the bioreactor were twice at SRT of 50 d with polysaccharides as the major fraction compared to those at SRT of 20 d with proteins as the main component. There was no significant difference in membrane fouling between SRTs of 20 and 50 d. Irremovable fouling (removed by intensive chemical cleaning) at SRT of 50 d made a greater contribution to fouling (90% of total fouling) caused by higher SMP concentration as compared to that at SRT of 20 d consisting of 85% of total fouling.

Based on investigations above and taking into account the suggestion by Meng et al. (2009a), too long or short SRT was not beneficial for fouling control. The selection of SRT for MBR operation was associated with membrane materials, types of wastewater, and F/M ratio (constant or various). For synthetic wastewater treatment, too much high SRTs (i.e. infinite) or extremely low SRTs would not only induce cake fouling and pore blocking, but also deteriorate sludge characteristics (e.g. settling property, floc structure). When treating real wastewater (e.g. municipal wastewater, domestic wastewater), poor membrane permeability was obtained at low SRTs due to deteriorated biomass properties, membrane pore blocking, along with much more EPS on membrane surface and into membrane pores. MBR at higher SRTs (i.e. WSW) exhibited better characteristics of sludge and foulants on the membrane as well as lower fouling resistance. Therefore, MBR should be operated at SRT of 20–40 d when treating synthetic wastewater, while for real wastewater treatment (e.g. domestic sewage), SRT is recommend to be controlled at 20–30 d or WSW.

Table 2.3. Effects of SRT on biofouling

Scale	Type of wastewater	SRT	Effects on sludge characteristics and biofouling[a]	References
Lab	Synthetic wastewater	3–50 d (HRT = 6–12 h)	• With SRT ranging from 3 to 10 d, shorter SRT facilitated the accumulation of carbohydrates and proteins in the submerged MBR due to higher biomass activity at higher F/M. Shorter SRT increased the amount of small molecules (MW < 1 kDa) of EPS and reduced macromolecules (MW > 30 kDa) of SMP; • At lower SRT of 15 d, MBR sludge exhibited poorer settling properties, higher RH, higher EPS levels as well as possessed more filamentous bacteria, which led to the production of sludge flocs with more loose structure and poorer stability. It generated higher population of smaller aggregates (0.5–5 μm). The sludge presented higher MLSS concentrations and sludge viscosity at higher SRTs of 30 and 50 d than those at SRT of 10 d. The carbohydrates in the supernatant were significantly higher (around 2 times) at HRT of 10 d as compared to the SRT of 50 d; • More severe membrane fouling, less porous fouling layer and a relative higher specific resistance were obtained at lower SRT (i.e. SRT of 15 d). With increasing SRTs, pore blocking resistance and cake layer resistance decreased. The reduction of pore blocking resistance was due to the cake layer which was formed as a protective layer for membrane pores to reduce SMP on the membrane.	Duan et al., 2014; Dizge et al., 2013; Tian and Su, 2012
Full and pilot	Municipal wastewater	10–50 d (HRT = 9–26 h)	• Lower SRT induced high floc-bound and soluble EPS concentrations in activated sludge, which led to poor settling and filtering properties as well as poor dewaterability. Better bioflocculation and sludge filterability, larger flocs with a smaller amount of small fragments were obtained at higher SRT. It resulted in less severe membrane fouling. • A significant decrease of both proteins and carbohydrates in EPS and SMP with	Al-Halbouni et al., 2008; Malamis and Andreadakis, 2009; Van den Broeck et al., 2012

	increasing SRT from 10 to 20 days reduced membrane fouling (reduction of membrane permeability drop) by 80%. The subsequent increase of SRT to 33 d slightly affected EPS, SMP as well as membrane fouling. Moreover, increasing SRT induced the biodegradation of some fractions of EPS and SMP (especially the smallest molecules (< 1 kDa)), but reduced biodegradation of macromolecules in the range of 0.45 μm-300 kDa due to their non-biodegradable character; • More distinct fouling layer on the membrane was formed at lower SRT with significantly higher concentrations of deposited EPS (40-fold higher for protein and 5-fold higher for carbohydrate) than those at higher SRT.

[a] RH, relative hydrophobicity; F/M, food to microorganism ratio; MW, molecular weight

B. Hydraulic retention time (HRT) and filtration flux

HRT as an important operating parameter determined hydraulic loading rates for the organic matters and nutrients, which influences sludge characteristics and membrane fouling behavior in MBRs. Previous studies have indicated that shorter HRT, resulting in higher organic loading rate (OLR), could increase biomass concentration, sludge viscosity and decreased dissolved oxygen (DO) concentration. It also induced overgrowth of filamentous bacteria, more EPS production, along with generation of larger, more irregularly and porous sludge flocs. Moreover, the abundance of filamentous bacteria and higher EPS levels facilitated adherence of membrane foulants onto membrane surface, and further promoted formation of a thicker cake layer due to bridging action among membrane foulants. In addition, the co-deposition of EPS and sludge flocs onto the membrane surface also gave rise to formation of cake layer and reduced its porosity. Therefore, deteriorated membrane permeability was obtained at shorter HRT (Chae et al., 2006; Meng et al., 2007c). When operating a thermophilic submerged aerobic MBR (TSAMBR) for the treatment of thermomechanical pulping pressate at various HRTs of 1.7 ± 0.2, 0.9 ± 0.1, and 1.1 ± 0.1 d, it was found that the contents of EPS in bulk sludge increased, but EPS of cake sludge slightly changed (Qu et al., 2013). The results also indicated that cake layer structure transformed from a thin gel layer to one or two cake layers with decreasing HRT. At shorter HRT of 1.1 ± 0.1 d, besides the formation of a homogeneous and compact cake layer, its porosity along the depth presented minor difference. The further reduction of HRT to 0.9 ± 0.1 d aggravated the development of a two-layer heterogeneous cake fouling and reduced the porosity from the surface layer to the bottom layer, which in turn increased the local flux above the critical flux of cake layer and induced TMP jump. When treating a synthetic wastewater containing toxic substances (e.g. styrene), Fallah et al. (2010) found that as compared to high HRT of 24 h, low HRT of 18 h induced higher styrene loading rate, which facilitated EPS release from bacteria cells, contributing to more SMP generation and sludge deflocculation. Therefore, more SMP, smaller mean floc size and a higher proportion of small biomass particles exacerbated membrane fouling at shorter HRT. Similar results were also reported by Shariati et al. (2011) who operated membrane sequencing batch reactors for synthetic petroleum wastewater treatment at HRTs of 8, 16 and 24 h. An increase in sludge particle size range, higher sludge viscosity and SMP concentrations (especially SMP_C) induced higher fouling rate at shorter HRTs. Therefore, less membrane fouling could be obtained at a relatively higher HRT depending on characteristics of feed water.

Concerning filtration flux, it was found that at higher flux, higher fouling rate and higher deposition rate of particles onto the membrane surface were obtained. A dense and less porous cake layer was formed with higher specific cake resistance. Moreover, the dominant fraction of the bio-cake was bacterial cells, which increased roughness and thickness of the bio-cake. When decreasing filtration flux, the bio-cake mainly contained polysaccharides of EPS (Akhondi et al., 2014; Hwang et al., 2012). Therefore, MBR should be operated at a relatively lower flux.

C. Dissolved oxygen (DO)

Aeration used in MBR is to supply oxygen for activated sludge, allow the sludge to suspend thoroughly in the bioreactor, as well as scour membrane surface for fouling control (Le-Clech et al., 2006). Air flow rate or DO is an important operating parameter, which governs the hydrodynamic conditions, affects sludge characteristics (i.e. EPS, SMP), membrane fouling and energy consumption (Faust et al., 2014). It has been suggested that the turbulence generated by air flow in the vicinity of membrane surface made the membrane move and improved the MBR filtration performance. The associated back transport of particles had a positive effect on fouling reduction. Aeration could also induce the shear stress, which physically scoured the membrane surface/removed cake layer and modified biological floc characteristics (e.g. floc size, EPS levels, EPS_P, EPS_C). When operating at low air flow rate or DO, cake layer would be formed due to the deposition of MLSS on the membrane, which could not be removed by the low shear stress. Low DO also decreased aerobic activity, which reduced production or increased anaerobic degradation of EPS, inducing the sludge deflocculation. On the other hand, a relatively high DO could promote sludge bioflocculation, enhance sludge characteristics (e.g. more stable flocs with lower EPS_P/EPS_C ratio) and membrane performance. However, an extremely high aeration intensity not only broke up sludge flocs, facilitated the release of EPS, SMP and colloids, but also deteriorated filterability. It negatively affected membrane performance. At different DO levels, the microbial community structure in biocake showed larger variations as compared to that in mixed liquor (Braak et al., 2011; De Temmerman et al., 2015; Faust et al., 2014; Gao et al., 2011). Table 2.4 exhibits effects of DO on biofouling. Fu et al. (2012) pointed out that variables affecting membrane fouling followed the order of aeration flow rate > aeration position > aeration time, with an interaction between aeration flow rate and aeration time. Reduced membrane fouling could also be achieved by decreasing the distance from membrane module to aeration pipes. De Temmerman et al. (2015) monitored a default aeration situation (reference) at DO of around 2 mg/L and an elevated aeration situation (disturbance) in two identical MBRs. The later one was obtained by manually controlling aeration at DO concentration of around 4 mg/L for 18 h, which subsequently decreased to the average setting (recovery state) with declining DO to around 2 mg/L. At high fine bubble aeration intensity, higher concentrations of SMP, biopolymers, and submicron particles (10–1000 nm) were obtained along with a shift in the particle size distribution (PSD) (3–300 μm) towards smaller sludge flocs. Thus, the system not only exhibited an increase in total fouling due to cake layer formation with less permeability, but also showed higher irreversible fouling due to pore blocking. Lowering aeration intensity caused an incomplete restoration of sludge filterability, and higher total fouling rates than the reference situation despite the reduced fouling rates. Overall, the determination of an "optimal" air flow rate or DO concentration is based on the MBR configurations, operating parameters, aeration time, position and mode (e.g. cyclic aeration on/off).

Table 2.4. Effects of DO on biofouling

Effects on biofouling	References
• No direct influence of aeration on soluble or floc-associated EPS production was found in the MBR system. • In the high shear reactor (DO > 6 mg/L), higher aeration was conductive to the selection of the predatory organisms. At low shear condition (DO 2-3 mg/L), the proliferation of large aquatic earthworms *A. hemprichi* led to an increase in floc-associated and soluble EPS production, resulting in higher membrane fouling potential when operating at the flux higher than critical flux. On the other hand, no *A. hemprichi* were observed in the high shear reactor. • As compared to lower DO concentration (1 mg/L), the improved bioflocculation was found at higher DO concentration of 4 mg/L, which was associated with higher EPS concentration and more multivalent cation concentrations (calcium, iron and aluminium) within the matrix of the flocs. Higher DO concentrations increased mean floc size, reduced supernatant turbidity, improved settleability and membrane filterability.	Faust et al., 2014; Menniti and Morgenroth, 2010

D. Temperature

Previous studies have showed that low temperature decreased flocculation ability of activated sludge, reduced floc size, increased EPS, SMP and sludge viscosity, accelerated the accumulation of particles on the membrane at lower shear stress, decreased back-transport velocity of biomass particles, and reduced biodegradation of COD, which had adverse effects on membrane performance (Krzeminski et al., 2012; Ma et al., 2013a; van den Brink, 2011). These effects contributed to severe membrane fouling and an increase in resistance to membrane filtration. Table 2.5 displays the impacts of temperature on biofouling in MBR. Some studies also were conducted in pilot-scale or full-scale MBRs for one to three years to monitor the membrane fouling behaviour and biomass characteristics. Miyoshi et al. (2009) reported that when employing pilot-scale MBRs to treat municipal wastewater, physically reversible fouling (eliminated by physical membrane cleaning) exhibited more significant development trend in the low-temperature period. It was associated with the concentration of dissolved organic matter in mixed liquor suspension, which contained more pronounced polysaccharide-like and humic acid-like substances. On the other hand, in the high-temperature period, physically irreversible fouling (eliminated by chemical membrane cleaning) was more severe, which was caused by the nature of the dissolved organic matter with an abundance of proteinaceous characteristics. The same results were obtained for the foulants inducing physically irreversible fouling. Wang et al. (2010) demonstrated that when temperature fluctuated from 26 to 8 °C, higher concentrations of bound EPS, polysaccharides and proteins in the supernatant in the MBR were achieved during low-temperature operation, which facilitated the attachment of biopolymers onto membrane surface and increased membrane fouling potential. The settleability and dewaterability of mixed liquors decreased when operating at lower temperature. It was also pointed out by Krzeminski et al. (2012) that typical seasonal fluctuations and poorer sludge characteristics (e.g. filterability, settleability) were obtained during low-temperature periods. Influent organic loading, lower MBR biomass concentrations, higher volatile suspended solids/total suspended solids (VSS/TSS) ratio and less biodegradation of wastewater in mixed liquor were responsible for the decreased filterability at low temperatures. van der Brink et al. (2011) indicated that when conducting the flux step experiments at various temperatures of 7, 15, and 25 °C, the increased membrane fouling rates with the reduced fouling reversibility were achieved at low temperature. The decreased temperature aggravated the accumulation of polysaccharides in the soluble fraction of mixed liquor and induced a shift in floc size of the soluble fraction towards smaller particles, which in turn caused pore blocking and exacerbated membrane fouling. Besides, reduction of mixed liquor temperature from 27 to 13 °C prompted serious membrane fouling and higher filtration resistance due to the increase in supernatant organics with humic substances, hydrophilic polysaccharides and large-molecular-weight proteins as dominant foulants in the full-scale MBR (Sun et al., 2014).

Overall, more severe membrane fouling behaviour obtained at low temperature could be attributed to the changes in microbial community as well as deteriorated sludge properties. It is recommended that temperature should be maintained above 15 °C but at lower than 30 °C.

Table 2.5. Effects of temperature on biofouling

T[a]	Effects on sludge characteristics and biofouling[b]	References
8 –31 °C	• During the initial stage of membrane biofouling process, *γ-Proteobacterium* (at 30 ºC), *Aquabacterium* sp. (at 20 ºC), and *β-Proteobacterium* (at 10 ºC) acted as the pioneer in the surface of membrane. With the biofilm growth, *R. bacterium* of biocake was dominated at 30 ºC, while the dominant species in biocake at 10 and 20 ºC was *β-Proteobacterium*. • The change in temperature altered the microbial community, which led to the varied compositions of EPS and SMP. In both mixed liquor and biocake, the decrease in temperature from 30 to 20 °C induced the reduction of EPS and SMP with polysaccharides as the dominant fraction. An additional decline of temperature to 10 °C resulted in a significant rise of EPS and SMP, inducing higher membrane fouling (especially irreversible fouling) as well as the total membrane resistance to permeability. EPS_C in biocake and SMP_C/SMP_P in mixed liquor showed the significant correlation with temperature. • Rich population of *α-Proteo-bacteria* and some filamentous bacteria at low temperature gave rise to serious membrane fouling, while at high temperature, the presence of *Zoogloea* diminished membrane fouling by adsorbing fine particles.	Arévalo et al., 2014; Gao et al., 2013b; Ma et al., 2013a

[a] T, temperature
[b] SMP_C/SMP_P, polysaccharide to protein ratio in SMP

2.4.3. Feed water characteristics

A. Organic loading rate (OLR) or food/microorganism (F/M) ratio

Activated sludge process can be controlled by food to microorganism ratio (F/M) according to the ratio of organic substrate (e.g. COD, BOD) available for microorganisms and biomass concentration in the bioreactor.

Alteration of F/M ratio was obtained by varying organic loadings. It changed microbial behaviours, microbial properties and biomass characteristics in mixed liquor (e.g. MLSS, particle size, viscosity, floc structure, production of EPS and SMP) as well as membrane biofilm (e.g. bacteria community), which further influenced membrane fouling. Higher OLR or F/M ratio increased EPS levels and facilitated the formation of cake fouling on membrane surface, while at high OLR within a moderate range was beneficial for fouling control (Domínguez et al. 2012; Sharghi and Bonakdarpour, 2013; Wu et al., 2011b). The effects of OLR or F/M ratio on biofouling are displayed in Table 2.6. Shane Trussell et al. (2006) employed an ultrafiltration membrane with a nominal pore size of 0.035 μm in the SMBR pilot plant at a constant membrane flux of 30 $L/m^2 \cdot h$ and found that membrane fouling increased significantly (by around 20 times) for approximately four-fold range of F/M tested (from 0.34 to 1.41 g COD/g VSS·d). There was a strong relationship between steady-state membrane fouling and total SMP as well as SMP_C. Domínguez et al. (2012) conducted two bench-scale submerged MBR experiments by employing microfiltration (MF) and UF flat sheet membranes (pore size 0.4 and 0.08 μm, respectively) within a volumetric loading rate fluctuating between 0.4 and 1.3 kg $COD/m^3 \cdot d$ at MLSS levels of 2-8 g/L. They found that at high volumetric loading rate (1.3 kg $COD/m^3 \cdot d$), the biomass growth rate for both MBRs was higher (around two times) than that at medium volumetric loading rate (0.9-1.2 kg $COD/m^3 \cdot d$), while the MF-MBR and the UF-MBR exhibited 5 and 8 times higher than those at the lowest volumetric loading rate (0.4-0.6 kg $COD/m^3 \cdot d$), respectively. Reduced cleaning frequency of MF membrane as compared to UF membrane was achieved at higher volumetric loading rates due to higher biomass growth and rapid formation of larger flocs, which inhibited pore blocking. Sharghi and Bonakdarpour (2013) assessed mixed liquor characteristics of a MBR by employing a halophilic bacterial consortium for hypersaline synthetic produced water treatment at various organic loading rates (OLRs) from 0.3 to 2.6 kg $COD/m^3 \cdot d$. An increase in OLR from 0.9 to 2.6 kg $COD/m^3 \cdot d$ induced a rise in EPS (mainly EPS_C) and zeta potential, resulting in an obviously improved bioflocculation ability of the sludge. However, it caused a decrease in compressibility of sludge flocs, which led to the occurrence of viscous bulking. The increased EPS concentrations and EPS bulking gave rise to the alteration of mixed liquor rheology and serious membrane fouling. On the other hand, a long-term starvation would induce sludge decay, release of large amounts of high-molecular-weight BAP (humic-acid substances), and serious fouling (Wu and Lee, 2011).

Based on the information above, relatively high F/M ratio or OLR within a reasonable range should be selected considering operating conditions (i.e. SRT, HRT), biomass concentrations (i.e. MLSS, EPS), membrane and influent characteristics.

Table 2.6. Effects of OLR or F/M ratio on biofouling

OLR or F/M[a]	Effects on sludge characteristics and biofouling	References
F/M = 0.33 and 0.52 g COD /(g VSS·d)	• As compared to the low loading conditions, quicker and rapid membrane fouling was obtained under the high loading conditions due to higher EPS contents in the reactor, the faster development of biofilm formation with higher thickness, as well as the dominance of two bacteria *Betaproteobacteria* and *Bacteroidetes* on membrane surface during the long-term operation.	Xia et al., 2010
OLR = 0.57, 1.14, and 2.28 g COD/L·day (filtration flux 30 L/m^2·h)	• The change in bacteria community and EPS levels at various organic loadings were affected by F/M ratios and DO levels. • Bound EPS contents (especially proteins) positively influenced membrane fouling propensity of biomass, with a slightly lower membrane fouling in the low-organic-loading MBR (0.57 g COD/L·day) but significantly higher fouling in the high-organic-loading MBR (2.28 g COD/L·day) when comparing to the medium-organic-loading MBR (1.14 g COD/L·day). It implied that cake fouling formed by deposition of the sludge flocs was the major contributor to membrane fouling.	Wu et al., 2011b

[a] VSS, volatile suspended solids

B. Carbon to nitrogen or phosphorus ratio (C/N or C/P)

Current studies focused on nutrient parameters, such as C/N, C/P ratios, which affected bacteria communities and sludge characteristics (i.e. flocculation, settleability, compositions and contents of SMP or EPS), thereby influencing membrane fouling potential. It was found that deterioration of flocculation, settleability and dewaterability of sludge flocs were more serious when C/N ratio decreased from 20 to 4 than those when C/N ratio increased from 20 to 100. The C/N ratio did not affect TB-EPS (including carbohydrates and proteins). For LB-EPS, the decrease in C/N ratio caused an increase in protein content and a decrease in carbohydrate content. When increasing C/N ratio, the protein and carbohydrate followed an adverse trend as that observed for lowering C/N ratio. The protein levels in LB-EPS positively influenced the flocculation, settleability and dewaterability of the flocs (Ye et al., 2011). Wu et al. (2012) suggested that an increased nitrogen or phosphorus loading in MBRs (low C/N or C/P ratio) induced apparent shifts of bacterial communities in a similar way. When the low C/N or C/P-MBR almost reached a steady state, comparable biomass concentrations and EPS levels were obtained as those in the control MBR, with similar dominant bacteria community in both MBRs. In addition, biomass samples from the low C/N-MBR and low C/P-MBR (increasing nitrogen or phosphorus loading 1-fold) demonstrated larger flocs and lower EPS levels, resulting in no more serious membrane fouling than the control MBR. Therefore, nitrogen or phosphorus loadings at too much high or low levels in the influent deteriorate sludge properties and membrane performance.

C. Salinity and cations

Previous studies showed that the elevated salt levels deteriorated sludge characteristics, which further impaired membrane performance. High salt concentration broke the multivalent cation bridging among EPS within sludge matrix, or reduced populations of protozoans, resulting in sludge deflocculation and poor sludge quality, mechanical integrity and structure of sludge flocs. It reduced sludge filterability, whilst exacerbating membrane fouling. Increased concentrations of foulants (e.g. SMP and/or EPS) were obtained at high salt concentration due to the following reasons: 1) protection of microbial cells from salt toxicity; 2) release of cell contents owing to the deterioration of outer layer of the cells; 3) accumulation of products caused by the incomplete degradation of organic substances and cell autolysis (De Temmerman et al., 2014; Jang et al., 2013; Sharghi et al., 2014). Current studies have investigated the influence of salt concentration on mixed liquor properties and fouling behaviour in MBRs. Table 2.7 exhibits the effects of salt on sludge characteristics and biofouling in MBR. De Temmerman et al. (2014) elucidated that an introduction of salt (NaCl 2 g/L) into MBR postponed the increase in amounts of submicron particles, SMP levels (proteins and polysaccharides), and irreversible fouling rate. Moreover, there was no time delay in the shift in the supramicron particle size distribution towards smaller floc sizes and an increase in total fouling rate. Sharghi et al. (2014) employed the moderately halophilic bacteria consortium to treat hypersaline synthetic produced water with 100–250 g/L NaCl and reported that no significant fouling occurred at any salt concentration. At elevated NaCl levels, the drop in bacterial growth rate of mixed bacterial consortium resulted in reduced EPS concentration. The mixed liquor SMP_{total} (including proteins, humic acids and polysaccharides) also decreased, which was different from activated sludge. When employing the halophilic bacteria consortium, elevated salt concentration induced lower bacterial growth rate with UAP as the main component of SMP. However, for activated sludge, only the bacteria population that could tolerate the salinity would survive, implying that BAP was the major fraction of SMP. In feed water, divalent cations (i.e. calcium, magnesium) also affects aggregation of sludge flocs, which bridged negatively functional groups within EPS and SMP, thereby improving aggregation and stabilization of matrix of biopolymers and microbes. Furthermore, monovalent to divalent cation ratio also plays a crucial role in the flocculation process. Normally, a relatively lower monovalent to divalent cation ratio but higher molar ratio of Mg:Ca through adding magnesium within the optimum range favours floc agglomeration. At constant calcium concentration, magnesium addition changes characteristics and molecular weights of EPS and SMP for membrane fouling mitigation (Arabi and Nakhla, 2009a).

Table 2.7. Effects of C/N ratio, salinity and cations on biofouling

Fouling factors	Effects on sludge characteristics and biofouling[a]	References
C/N ratio	• At lower C/N ratio of 5 (higher ammonia concentrations), higher concentrations of NH_4^+-N in the reactor as a monovalent cation replaced the polyvalent cations in EPS, which was conductive to release of EPS components to form new SMP as compared to higher C/N ratio of 10 (lower ammonia concentrations). Moreover, SMP possessed higher carbohydrate levels and higher carbohydrate/protein ratio. It gave rise to higher R_{IR} at lower C/N, which was well correlated with fouling profiles (higher TMP increase rate and shorter operational duration).	Feng et al., 2012a
Salinity	• The elevated salinity changed microbial composition, which influenced MBR performance. • With increasing salt concentrations, higher EPS concentration and the associated poor sludge characteristics (i.e. the decreased floc size) deteriorated membrane filtration as demonstrated by irreversible cake deposition on the membrane surface and higher pore blocking resistance.	Di Bella et al., 2013; Hong et al., 2013; Jang et al., 2013
Cations	Mg:Ca ratios at 1:5, 1:1, and 5:1 • Higher Mg concentration (or the highest Mg:Ca ratio of 5:1) increased membrane permeability and reduced fouling rate through magnesium bridging of negatively charged biopolymers which improved bioflocculation. • Increased magnesium levels at the highest Mg:Ca ratio of 5:1 increased MW EPS (> 10 kDa), leading to generation of larger and more permeable flocs, as well as reduced hydrophobic SMP and certain fractions of SMP (> 100 kDa and < 10 kDa).	Arabi and Nakhla 2009a

[a] MW, molecular weight; R_{IR}, irreversible fouling resistance

2.4.4. Membrane materials

Generally, some parameters should be taken into account when choosing membrane materials, such as strength, elongation, integrity, foulant-resistant ability, temperature, chemicals tolerability and lifespan. The most commonly used polymeric membranes in MBRs include polyacrylonitrile (PAN), polyethersulfone (PES), polysulfone (PSU), polyvinylidene fluoride (PVDF), polypropylene (PP), and polyethylene (PE) (Radjenović et al., 2008; Water Environment Federation, 2012). PES and PVDF have high strength and permeability. Moreover, PVDF can resist chemicals and chlorine, long life as well as higher elongation comparing to PES. For PE and PP, they are chemically and biodegradable resistant, but are easy to be dried out when conducting air-based integrity tests (Water Environment Federation, 2012; Wilf et al., 2010).

Membrane characteristics (e.g. morphology, membrane pore size, zeta potential, hydrophobicity/hydrophibicity affinity) are the important factors affecting membrane fouling. Table 2.8 shows membrane characteristics associated with biofouling. Maximous et al. (2009) reported that as compared to the hydrophilic membrane (regenerated cellulose, YW 30), the hydrophobic membrane (polyethersulfone, PM30) was more prone to pore plugging, rejected more polysaccharide and protein in SMP, and exhibited lower flux. The cake layer formed on the hydrophobic membrane with higher cake layer resistance improved the solute rejection. Membrane fouling behaviour at various MLSS concentrations was also investigated concerning different membrane materials and pore size. The impacts of MLSS levels on membrane fouling varied with the pore size (0.4 μm for MF, 0.08 μm for UF) (Domínguez et al., 2012). In the MBR, MF membranes could perform better for long-term operation at high MLSS concentration (> 2–3 g/L). At lower biomass concentration (around 2 g/L), the deposition of colloids, EPS and small particles on and/or inside the membrane gave rise to severe membrane fouling. The further increase in MLSS reduced membrane fouling due to the presence of larger sludge flocs. UF-MBR exhibited better permeability when MLSS concentration was lower. At higher MLSS levels above 6 g/L, more serious membrane fouling was obtained due to pore blocking and sludge layer formed on the membrane. Dizge et al. (2011b) investigated the type and pore size of membranes on membrane fouling by employing cellulose acetate (CA), PES, mixed ester (ME), polycarbonate (PC) membranes with three different pore sizes (0.40–0.45, 0.22, 0.10

μm) in cross flow microfiltration experiments. The results showed that a significant flux decline was obtained for CA membrane (pore size 0.45 μm) owing to its irregular and rough surface. Steady state permeate flux values for membranes were in the order of ME > PC > PES > CA. With regard to ME and PC membranes, the steady state permeate flux was higher when increasing pore size of the membrane, while the flux decreased for CA membrane with larger pore size owing to pore blocking by colloids. Types of membrane fouling were largely determined by the surface roughness and pore geometry of membranes. PC membranes exhibited the highest pore blocking resistance (R_P) for the membranes at three different pore sizes due to their straight through pore structure and/or the surface chemistry. With decreasing pore size of the membranes (e.g. PES and ME), R_P decreased due to the fact that larger microbial aggregates could not pass through the smaller pores of the membranes. Concentration polarization resistance (R_C) increased by adsorbing soluble fraction of microbial products (carbohydrate and protein) on the membrane surface. van der Marel et al. (2010) provided a more clear insight into the significant effects of membrane pore size, surface porosity, pore morphology and hydrophobicity on membrane fouling through the filtration experiments. The influence of membrane pore size on fouling behaviour (e.g. critical flux) should be closely associated with surface porosity and pore morphology (pore structure and interconnectivity). The symmetric PE membranes with larger pores exhibited more internal fouling due to pore blocking. However, the asymmetric PVDF membrane with an interconnected pore structure having relatively larger pore size and surface porosity exhibited better membrane performance with respect to critical flux, critical flux for irreversibility, and internal fouling potential. For PC membranes with straight-through track-etched pores, critical fluxes were lower due to rejection/retention of substances by the membranes comparing to those for the PVDF membranes. Better permeability for a long term filtration was obtained for the symmetric mixed cellulose ester (MCE) membranes with the tortuous path and smaller pore size. More rapid adsorption of feed water constituents on hydrophobic membrane $PVDF_H$ 0.1 μm reduced the critical flux to half of that of a hydrophilic membrane PVDF 0.1 μm. Therefore, less fouling was achieved for the membrane with higher hydrophilic property, complete asymmetric structure, and interconnected pore morphology, relatively large pore size and high porosity. It was also found that a dense cake layer with much higher cake resistance obtained on a hydrophobic membrane than that on a hydrophilic membrane was associated with the hydrophobicity of suspend sludge (Nittami et al., 2014). It should be noted that surface hydrophilicity of membranes play an important role in the interaction between membrane and foulants when the two contacting surfaces are infinite planar. However, when foulants are spherical particles as indicated by Zhang et al. (2015b), it comes to a different conclusion. The interfacial interaction between sludge foulants and membranes was more significantly affected by membrane surface zeta potential and roughness than membrane hydrophilicity/hydrophobicity. Higher membrane surface zeta potential induced higher strength of the electrostatic double layer (EL) interaction and the energy barrier. The repulsive total interaction was obtained for the membrane with a relatively higher roughness (300 nm) when the separation distance ranged from 0 to 4 nm. Therefore, membrane fouling could be achieved for membranes with relatively higher zeta potential and certain roughness. Currently, ceramic membranes, textile materials, or non-woven meshes have also been used in MBRs for wastewater treatment. They possess better chemical and physical stability, high integrity, longer lifetime and effective fouling control as compared to polymer membranes.

Considering the information mentioned above, membrane properties (e.g. membrane material, pore size, surface porosity, pore morphology, roughness, hydrophobicity, zeta potential) affect membrane fouling. The properties of foulants (e.g. hydrophilic/hydrophobic, planar/spherical particles) are also critical to effects of membrane properties on membrane fouling.

Table 2.8(a). Membrane characteristics associated with biofouling

Membrane types	**Effects of membrane characteristics on biofouling**[a]		**References**
Microporous membranes	Materials	• PVDF membrane showed more excellent performance in irreversible fouling control than PE membrane. • The increasingly sub-miron-sized organic matter that mainly consisted of carbohydrate were responsible for reversible fouling for the PVDF membrane. • The membrane polymer material played a key role in the components of the foulants on the membrane inducing irreversible fouling.	Wang et al., 2011b; Yamato et al., 2006; Zhang et al., 2008
	Hydrophobicity /hydrophobility and roughness	• The EPS adsorptive fouling degree of three polymeric UF membranes followed the order of hydrophobic PES > hydrophobic PVDF > hydrophilic PAN. A relatively high roughness and hydrophobicity of PES membrane contributed to much more severe membrane fouling for PES. • The hydrophilic PAN membrane could allow the hydrophilic SMP_C to pass through the membrane but reject the hydrophobic SMP_P, which could be attributed to the protein-membrane interaction. The PAN membrane was subjected to severe irremovable fouling due to hydrophobic protein-like substances and humic-like substances, which led to pore blocking, while for the PVDF membrane, the protein-like substances gave rise to irreversible fouling. • In terms of SMP, fouling propensity for the hydrophilic PAN and the hydrophobic PVDF was largely determined by the hydrophobic fraction and neutral hydrophilic fraction, respectively.	
	Pore size	• As compared to the PAN membrane (pore size 0.05 μm), the PVDF membrane with larger pore size of 0.15 μm showed a higher MRD of SMP_C and lower MRD of SMP_P with smaller size (small MW) based on size exclusion mechanism, leading to a higher membrane fouling propensity. • The surface foulants on the PVDF membrane exhibited larger mean particle size and consisted of larger MW molecules than those of the PAN membrane due to the size exclusion effect as the PVDF membrane had larger pore size than the PAN membrane.	

Table 2.8(b). Membrane characteristics associated with biofouling

Membrane types	Effects of membrane characteristics on biofouling[a]		References
New kinds of membranes (e.g. ceramic membranes, non-woven meshes, textile materials)	Hydrophobicity/ hydrophilicity	• The more hydrophilic materials exhibited less fouling potential.	Jin et al., 2010; Tewari et al. 2010; Zahid and El-Shafai, 2011
	Roughness	• Membrane fouling propensity was in consistent with the sequence of membrane surface roughness: R80 < R100 < R200 < R300. • A rougher membrane surface facilitated the formation of a fouling layer due to the presence of more filling-in points, resulting in a faster TMP increase. • The membrane material with more thickness and roughness surface showed more severe fouling.	
	Pore size	• The lowest and highest fouling was obtained for the ceramic membrane exhibiting the smallest and largest pore size of 80 nm and 300 nm, respectively. • The smaller pore-sized membrane (80 nm) showed higher potential for rejecting DOM in the reactor, especially smaller MW fraction (< 1 kDa), as well as a wider range of particles in mixed liquor. • The larger pore-sized membrane (200 or 300 nm) was more prone to pore blocking caused by organic and inorganic compounds, resulting in irreversible fouling. • The presence of similar PSD and MWD profiles for R80, R100, R200, and R300 was due to formation of an active layer which behaved as a sieve to reject smaller particles (i.e. colloidal particles). • MBR with the nylon mesh (30 μm pore size) and the ash filter (2–6 μm macroporous ceramic filters) exhibited high suspended solid retention, slow TMP increment (below 8 kPa), and a higher critical flux. Moreover, without a secondary filtration layer on the membrane, the ash filter could also retain suspended solids.	

[a] CA, Cellulose acetate; ME, mixed ester; MRD, membrane rejection degree; MW, molecular weight; PAN, polyacrylonitrile; PC polycarbonate; PE, polyethylene; PES, polyethersulfone; PVDF, polyvinylidene fluoride; R80, the MBR with 80 nm pore-sized ceramic membrane; R100, the MBR with 100 nm pore-sized ceramic membrane; R200, the MBR with 200 nm pore-sized ceramic membrane; R300, the MBR with 300 nm pore-sized ceramic membrane; UF, ultrafiltration

2.5. Biofouling control strategies

2.5.1. Membrane cleaning

Currently, membrane cleaning consists of four groups, including physical cleaning, chemical cleaning, physico-chemcial cleaning and biological/biochemical cleaning. The physical cleaning can remove concentration polarization and/or reversible fouling, such as the deposited particles and cake layer. The conventional physical cleaning includes air scouring, backwashing, and relaxation. Air scouring can remove reversible fouling or concentration polarization by cross flow velocity or shear stress. Backwashing employs the reversed permeate flow through the membranes, while relaxation is achieved by pausing filtration operation (no permeate flow) during the experimental period. Recent studies extended their efforts toward other hydraulic methods (i.e. cyclic aeration), vibration/rotation of membrane, or ultrasonic cleaning for more effectively preventing membrane fouling. For chemical cleaning, chemical reagents are applied for reduction of inorganic, organic and biological foulants, irremovable fouling, as well as irreversible fouling (e.g. gel layer and pore blocking) on the membrane. There are several types of chemicals, such as acids, bases, oxidants, and other chemicals (Wang et al., 2014b; Water Environment Federation, 2006). The applications of commonly used chemical reagents for membrane cleaning are shown in Table 2.9. However, the chemical cleaning methods deteriorated membrane properties (i.e. integrity, pore size, surface porosity, life span, mechanical strength). In addition, they caused release of chemical reagents and the generation of cleaning byproducts, which were detrimental to membranes, environment, microbial community and cell activity in MBR. The physico-chemical cleaning (e.g. chemical enhanced backflush (CEB), ultrasound during chemical cleaning) reduced the concentrations of the used chemical reagents, induced significant increase in flux recovery rate, permeability recovery and membrane cleaning efficiency. Nevertheless, some problems were found in MBRs when conducting CEB or cleaning in place (CIP), such as foaming, cell lysis and undesirable alteration of microbial community. Biological/biochemical cleaning as mild cleaning alternatives, mainly enzymatic cleaning, energy uncoupling and quorum quenching, has also been widely employed for the removal of membrane foulants. These methods are more efficient in decreasing biofilm formation with less adverse effects on microbial activity, microbial community, and membrane properties (Wang et al., 2014b). Some modified methods were also developed. A significant fouling mitigation could be obtained by combining QQ with physical cleaning with filtration/relaxation mode. QQMBR exhibited less biofilm formation without alteration of mixed liquor properties (Weerasekara et al., 2014). Recently, some studies attempted to immobilize QQ bacteria into individual carriers or membranes to stabilize enzymes and increased their economic feasibility for biofouling control in plant-scale MBRs or MBRs fed by real wastewater. Oh et al. (2012) successfully encapsulated a recombinant *Escherichia coli* which produced N-acyl homoserine lactonase or quorum quenching *Rhodococcus sp.* inside the lumen of microporous hollow fiber membrane as microbial-vessel, respectively, for biofouling reduction on membrane surface in MBR.

Table 2.9. The applications of different chemical reagents for membrane cleaning

Chemical reagents	**Chemicals (commonly used)[a]**	**Removal of foulants on the membrane**
Acids	Citric acid, H_2SO_4, HCl	➢ Chemical precipitation of inorganic matters, e.g. multivalent cations ➢ Biological-induced precipitants
Bases	NaOH	➢ Large organic particles, e.g. colloids and microbes ➢ Organic matters, e.g. protein- and carbohydrate-related foulants ➢ Fats and oils
Oxidants and disinfectants	NaClO, H_2O_2	➢ Organic foulants ➢ Biological foulants ➢ Colloids and microbe cells
Other chemicals	Metal chelating chemicals (e.g. EDTA, STP, DTPA), surfactants and related detergents	➢ Biopolymers associated with metal ions ➢ Macromolecules (e.g. proteins) ➢ Organic foulants ➢ Bacteria

[a] DTPA, diethylenetrinitrilopentaacetic acid; EDTA, ethylene diamine tetraacetic acid; HCl, hydrochloric acid; H_2O_2, hydrogen peroxide; H_2SO_4, sulphuric acid; NaClO, sodium hypochlorite; NaOH, sodium hydroxide; SDS, sodium dodecyl sulphate; STP, sodium tripolyphosphate.

2.5.2. Addition of flocculants

Research effort has also been extended toward adding different kinds of flocculants to alter properties of activated sludge and cake layer for enhancing membrane performance in MBRs. Table 2.10 displays flocculant addition induced membrane fouling reduction in batch tests and short-term dead end or cross-flow filtration tests. Ji et al. (2008) compared three flocculants namely aluminium sulfate ($Al_2(SO_4)_3$), polymeric ferric sulfate (PFS) and Chitosan based on long-term operation of MBRs at filtration flux of 20 L/m^2·h. The membrane fouling reduction rate of these flocculants was in the order of PFS> Chitosan> $Al_2(SO_4)_3$>Control (no flocculant added). They also concluded that flocculants addition could decrease both the concentration and molecular weight distribution of macromolecules in the supernatant, resulting in reduction of gel layer formation and membrane pore blocking. Guo et al. (2010a) investigated the impacts of flocculants addition on the short-term performance of a submerged MBR. They reported that more stable sludge volume indexes and higher specific oxygen uptake rates were obtained by adding natural organic flocculants such as chitosan, while inorganic flocculants (e.g. $FeCl_3$, Polyaluminium chloride (PACl)) reduced SMP as well as lowered membrane fouling rates. Long-term filtration experiments were conducted by Iversen et al. (2009) to investigate the effect of cationic polymers (NALCO MPE50, ADIPAP KD 452) and starch (TATE & LYLE Mylbond 168)) on the performance of a pilot-scale plant. The results suggested that two cationic polymers could mitigate membrane fouling, while starch addition led to more serious fouling phenomena. Wu and Huang (2008) reported that addition of polymeric ferric sulfate (PFS) could decrease the formation rate of gel layer on membrane surface due to the removal of high molecular weight organics, thereby retarding membrane fouling in long-term operation of the MBR system. Moreover, PFS also increased sludge floc size by supplying positive charges for organic particles and enhancing charge neutralization. PFS addition did not induce direct deposition of exotic Fe and severe inorganic fouling on membrane surface. It was suggested by Yang et al. (2011) examined the effect of PFC addition on the performance of an anoxic/oxic submerged MBR. Although polyferric chloride (PFC) could improve phosphorus removal and initially mitigate membrane fouling, more severe membrane fouling was observed in PFC-added MBR because of membrane pore blocking induced by higher concentration of PFC as well as compact gel layer formed on membrane surface by organic substances. For organic flocculants, submerged MBR with MPE50 addition exhibited significant improvement of the sustainable flux and membrane fouling reduction (Guo et al., 2008b). A more recent study conducted by Zhang et al. (2014) mentioned that the addition of organic flocculant (MPE50) was an effective approach to membrane fouling control at high salt shock due to increase in floc size, relative hydrophobicity and bound EPS (especially proteins). Additionally, combined flocculants have also been exploited recently. A new combined inorganic-organic flocculant (CIOF) of $FeCl_3$ and MPE50 prepared by Nguyen et al. (2010) was added to an aerated submerged MBR. The results indicated that the CIOF was successful in alleviating membrane fouling while maintaining stable SVI and low transmembrane pressure (TMP) development rate. Ji et al. (2014) investigated the performance of modified starch (MGMS) and its polyacrylamide–starch composite flocculant (PAM–MGMS) on fouling minimization for submerged MBRs. It was shown that the flocculant had long effect duration on reducing SMP concentration, as well as prolonged the decrease in floc size due to irreversible breakage of aggregates (de-flocculation) caused by continuous shear stress in MBR and the degradation of the modified starches. As the above-mentioned flocculants exerted adverse impacts on environment and stimulated the generation of “secondary pollutants” during wastewater reclamation and reuse, a new green bioflocculant (GBF) as the safe biodegradable natural flocculant was developed based on a natural starch-based cationic flocculant (HYDRA Ltd., Hungary) (Ngo and Guo, 2009). A significantly lower TMP development with only 2.5 kPa increase until day 70 and reduced energy by decreasing backwash frequency were obtained when supplementing GBF at very low dosage into a conventional submerged MBR.

Table 2.10. Flocculant addition induced membrane fouling reduction in batch tests and short-term dead end or cross-flow filtration tests (adapted from Deng et al., 2015)

Factors for membrane fouling reduction[a]	Flocculants[b]	References
Increasing EPS and decreasing SMP in mixed liquor	MPL30, MPE50, KD452, Poly-1 (Nalco®), Poly-2 (France Chitin®), CPE; PAM, Chitosan, Starch, CGMS, PAM-MGMS; $Al_2(SO_4)_3$, $FeCl_3$, PACl, PFS	Dizge et al., 2011a; Iversen et al., 2009; Ji et al., 2010; Ji et al., 2014; Koseoglu et al., 2008; Koseoglu et al., 2012
Enlarging floc size	MPL30, MPE50, KD452, Poly-1 (Nalco®), Poly-2 (France Chitin®), CPE; PAM, Chitosan, Starch, CGMS, PAM-MGMS; $FeCl_3$, PACl	Dizge et al., 2011a; Iversen et al., 2009; Ji et al., 2008; Ji et al., 2010; Ji et al., 2014; Koseoglu et al., 2008; Koseoglu et al., 2012
Enhancing charge neutralization	MPL30, MPE50, KD452, CPE; $Al_2(SO_4)_3$, $FeCl_3$, PAC, PFS	Dizge et al., 2011a; Ji et al., 2010; Ji et al., 2014; Koseoglu et al., 2008; Koseoglu et al., 2012
Increasing sludge hydrophobicity	CPE; $Al_2(SO_4)_3$, $FeCl_3$, PAC, PFS	Dizge et al., 2011a; Ji et al., 2010; Ji et al., 2014
Reducing gel layer and forming more porous and high permeable cake structures on membrane surface	CPE, Poly-1 (Nalco®); Chitosan	Dizge et al., 2011a; Ji et al., 2008; Koseoglu et al., 2012

[a] EPS = extracellular polymeric substances, SMP = soluble microbial products.
[b] CGMS = modified corn starch, CPE = organic cationic polyelectrolyte, MGMS = modified corn starch, PAM = polyacrylamide, PACl = polyaluminium chloride, PFS = polymeric ferric sulfate.

2.5.3. Addition of media

When choosing the proper biomass carriers for MBR operation, their properties, such as specific surface area, void space, porosity, and density, non-toxicity, chemical and mechanical stability, should be taken into account. To promote the biofilm attachment and growth, the media should have a higher specific surface area density (ratio of geometric surface area to volume), a larger void space, a smaller media size, a higher porosity, a higher void space, and a relatively lower density. Besides, media should be friendly to microorganisms and maintain their chemical and mechanical stablity (Guo et al., 2012b; Lekang and Kleppe, 2000). It has been suggested that direct supplementation of biomass carriers into MBRs could improve characteristics of mixed liquor and cake layer as well as alleviate membrane fouling. There are three main kinds of media, including plastic media, powdered activated carbon (PAC), and sponge. Table 2.11 shows the comparison on effects of different kinds of biomass carriers on membrane fouling reduction in MBR.

Siembida et al. (2010) indicated that an abrasive granular material (PP) could remarkably retard the formation of fouling layer on membrane surface in MBR, which enabled MBR to be operated for a long period (> 600 days) at high flux (< 40 L/m^2·h) without requirement of any chemical cleaning process. A high flux enhancement of more than 21% was obtained after adding the granular material. The granular material exerted a slight effect on membrane function, although brush marks were observed on the membrane surface due to its abrasion effects. Jin et al. (2013) reported that ceramic MBR with suspended carriers (AnoxKaldnes, K1 carriers) possessed relatively low density, large size presented larger biomass flocs, and less SMP and EPS. In addition, the biomass carriers could reduce cake layer on membrane surface but only slightly affect pore blocking.

The fresh PAC in a submerged MBR at relatively high concentration eliminated EPS, SMP, fine colloids, polysaccharides, total organic carbon (TOC) and planktonic cells in the supernatant of mixed liquor by synergistic effects of adsorption, decomposition, and biodegradation. It led to less membrane pore blocking, formation of higher porous, lower compact and more stable cake layer, lower fouling rate, longer lifespan of membrane, less energy consumption, and low maintenance cost in terms of membrane cleaning and/or membrane replacement. Moreover, the fresh PAC could also exert a scouring effect on membrane surface and cake layer on the membrane surface (Jamal Khan et al., 2012a; Ng et al., 2013; Yang et al., 2010b; Skouteris et al., 2015). PAC promoted formation of large, strong and dense sludge flocs, resulting in less particle deposition on the membranes, together with release of foulants and gel layer from membrane surface (Remy et al., 2010; Skouteris et al., 2015). Lin et al. (2011c) indicated that PAC-MBR process could achieve partial removal of low MW substances from the secondary effluent by membrane retention, adsorption and biodegradation. An improved organic removal and the largest fraction of organic removal were achieved in the presence of PAC. This study also demonstrated less membrane fouling, longer sustainable operation time and lower filtration resistance in the PAC-MBR process. Under stressful conditions, PAC-MBR could maintain excellent performance in terms of fouling control. In the case of salt shock, PAC could slow down the development of membrane fouling by decreasing the shifts in particle size distributions, reducing submicron particle concentrations, as well as lowering the concentrations of EPS and SMP proteins, biopolymers, humic acids, and low molecular weight (LMW) neutrals in the bulk liquid (De Temmerman et al., 2014; Remy et al., 2011). The PAC-MBR could achieve less chemical irreversible fouling at low temperature (10 °C) by adsorbing membrane foulants (e.g. polysaccharides) (Ma et al., 2013b; Remy et al., 2011). However, much higher dosage of PAC deteriorated membrane permeability due to the increased EPS levels, the elevated sludge viscosity of mixed liquor and the exacerbated membrane pore blocking. In addtion, it also gave rise to the formation of a nonporous and dense gel layer on membrane surface causing by the adhesion of organic matters and EPS to both of the PAC and the membrane surface. Moreover, it was required to replace aged biological PAC with fresh PAC for maintaining stable performance of MBR (Hu et al., 2014; Ma et al., 2012; Skouteris et al., 2015).

It has been suggested that sponge possesses excellent merits, such as high internal porosity and specific surface area, high stability to hydrolyses, and low cost (Ngo et al., 2006). Several studies have established the advantages of using sponge in MBR for membrane fouling control as sponge can promote biofilm growth, retard cake layer formed on membrane surface, and also enhance bacterial population by maintaining both attached and suspended biomass (Ngo et al., 2008). Guo et al. (2008a) found that the sponge-submerged MBR (SSMBR) with sponge volume fraction of 10% exhibited less membrane fouling, and two times of sustainable flux as compared to the SMBR alone with sustainable flux of 25 $L/m^2 \cdot h$. When compared to the conventional MBR (CMBR), sponge in the SSMBR reduced TMP development rate, improved microbial activity and maintained a stable SVI value (Nguyen et al., 2012a).

Other kinds of additives (e.g. diatomite, clinoptilolite) have also been successfully employed in MBRs for membrane fouling reduction, which enlarged sludge floc size, increased MLSS, reduced foulants (e.g. dissolved organic matter) in mixed liquor, and enhanced sludge settleability by combined effects of its adsorption and co-precipitation on fine colloids and dissolved organic matter (DOM) (Rezaei and Mehrnia, 2014; Yang et al., 2010c). Besides, Kim et al. (2013) entrapped QQ bacteria (*Rhodococcus* sp. BH4) into alginate beads to prepare cell entrapping beads (CEBs) and demonstrated their better performance in biofouling amelioration in MBRs. It was attributed to their scouring effects and biological (quorum quenching) effects of CEBs, which led to less EPS production and biofilm detachment from the membrane surface. Lee et al. (2014) obtained an effective antifouling system by immobilizing QQ enzyme (acylase, AC) into magnetically separable mesoporous silica (Mag-S-MPS) via enzyme adsorption and subsequent cross-linking processes. The system possessed high stability, magnetically separability and antifouling property for a long operation time.

Table 2.11. Comparison on effects of different kinds of biomass carriers for membrane fouling reduction in MBR

Sludge characteristics and membrane fouling behaviour	PAC[a]	Sponge	Plastic media
Membrane permeation flux	+		+
Sustainable flux		+	+
Filtration period	+	+	
Membrane fouling rate	–	–	–
Scouring effect of the membrane surface	+		+
Cake layer on the membrane surface/cake resistance	–		–
Gel deposition/gel layer on the membrane surface	–		
Sludge floc size	+		+
Floc breakage	–		
Floc strength	+		
EPS in mixed liquor	–		–
SMP in mixed liquor (causing irreversible fouling)	–		–
Internal membrane resistance caused by adsorption or pore plugging	–		±
Concentration polarization	–		
Mixed liquor viscosity			
Particle accumulation on the membranes	–		
Total membrane resistance	–	–	

Note: “+” denotes that the biomass carriers can increase the corresponding values, “-”denotes that the biomass carriers can decrease the corresponding values, “±” denotes less effects of biomass carriers on the values.
[a] up to an optimal dose

2.5.4. Other methods

It was indicated that unlike activated sludge in flocculent form, aerobic granule sludge featured an apparently larger floc size with regular and compact structure, good settleability, more biomass, varied bacteria community together with high metabolic activity. Wang et al. (2013b) employed aerobic granular sludge in a MBR for membrane fouling control at a high flux of 20 L/m^2·h and found that the good aerobic granular sludge (average diameter 903 μm) in MBR significantly extended the filtration duration as compared to the MBRs with flocculent sludge, bulking sludge and smaller aerobic granular sludge (434 μm), which led to effective membrane fouling reduction.

Membrane fouling in the aerobic granular sludge was mainly ascribed to pore blocking, while cake layer resistance dominated the filtration resistance for flocculent sludge or bulk sludge. Fouling behaviours in granules MBR and flocculent sludge MBR were determined by EPS composition of membrane foulants.

Recent studies also focused on development of electro-MBRs (e-MBRs) by applying an electric field into MBRs for fouling minimization. There are two novel types of e-MBRs with a membrane: 1) outside the electric field, which induced electro-coagulation using metal (e.g. iron) as anode to reduce deposition of foulants on the membrane; 2) inside the electric field, which gave rise to combined effects of electro-coagulation and electrophoresis when employing the cathode as a part of the membrane module to remove foulants from membrane surface (Akamatsu et al., 2012; Zhang et al., 2015c). Akamatsu et al. (2012) successfully demonstrated a novel concept of a membrane-carbon cloth assembly for submerged MBRs using carbon cloth as the anode and the assembly between two additional carbon cloths as the cathode. The results showed that the continuous or even intermittent application of an electric field reduced membrane fouling with a DC power supply. For activated sludge suspension, an intermittent electric field could promote the flux recovery. Moreover, at a relatively stronger electric field, foulants were detached from membrane surface with complete flux recovery. A later study conducted by Zhang et al. (2015c) suggested that a low-voltage electro-MBR (e-MBR) using stainless steel mesh as the anode, namely Fe-MBR, exhibited lower TMP, decreased irreversible fouling and enhanced pollutant removals as compared to another e-MBR with titanium anodes (Ti-MBR) and one MBR without an electric field. The electric field exerted the electric field force on negatively charged foulants, which reduced their tight attachment on membrane surface, promoted the removal of loose fouling layer, and induced the release of iron resulting in reduction of TOC and polysaccharides of SMPs. It led to decreased irreversible fouling and overall fouling rate. Besides, Su et al. (2013b) successfully developed a novel combined system consisting of sludge microbial fuel cell (S-MFC) stack and MBR for membrane fouling alleviation through modifying sludge characteristics, sludge reduction and energy recovery. This combined system decreased LB-EPS, especially simple aromatic proteins and tryptophan protein-like substances.

2.6. Conclusion remarks

Recent studies on fouling parameters, including sludge properties, operating conditions, feed wastewater characteristics and membrane properties, involving membrane biofouling in MBR processes were summarized as follows:

(1) With respect to sludge properties, LB-EPS negatively affect sludge characteristics and membrane performance. Besides, compositions and total concentrations of bound EPS are also essential for membrane fouling development and cake layer formation. For SMP, its polysaccharide fraction, UAP, or large MW fraction in hydrophilic substances contribute to gel layer formation, pore blocking and irreversible fouling. Besides, BPC play a crucial role in cake layer formation. It would be better for MBRs to operate at MLSS concentration below 10 g/L. From hydrodynamic and thermodynamic viewpoints, larger flocs reduce membrane fouling, while smaller particles are more readily attached to membrane and facilitated formation of less permeable cake layer;

(2) As to operating parameters, SRT, HRT and aeration intensity significantly impact sludge characteristics, membrane fouling behaviour and also cake layer properties. Too high or low SRT can deteriorate sludge characteristics, such as production of small flocs, release of EPS and/or SMP, which induce pore blocking, more severe membrane fouling or higher fouling resistance. It is recommended that SRT should be controlled in the range from 20 to 40 days for synthetic wastewater treatment. With respect to real wastewater treatment, SRT of 20-50 d or no sludge withdrawal would be better. Shorter HRT give rise to growth of filamentous bacteria, an increase in MLSS and sludge viscosity, a rise of EPS concentration, formation of thicker and less porous cake layer and severe membrane fouling. Moreover, when treating synthetic wastewater including toxic substances, more SMP and decrease in floc size are obtained at shorter HRT. It could be concluded that relatively higher HRT should be more than 10 h. The determination of the "optimal" aeration intensity or DO concentration mainly depends on aeration time, position and mode. Too high aeration breaks sludge flocs, and induces SMP/EPS release, while too low aeration cannot remove cake layer due to lower shear stress. Poorer sludge properties and membrane performance are obtained at a lower temperature, such as higher fouling rates, greater filtration resistance, higher suspended sludge concentration, more SMP and EPS, and

smaller flocs. Too high temperature also induces irreversible fouling. Temperature range from 15 to 30 °C is a desirable condition for lower membrane fouling;

(3) As compared to lower OLR or F/M ratio, higher OLR increases EPS levels, which aggravates membrane fouling, as indicated by higher fouling rate, or higher fouling resistance. On the other hand, under famine condition (low F/M ratio), cell lysis may induce higher fouling potential of the supernatant. Too much more phosphorus or nitrogen in feedwater leads to poor flocculation, settleability and dewaterability of activated sludge, more SMP release, resulting in higher filtration resistance. For normal activated sludge in MBRs, salinity may deteriorate sludge flocs, induce higher sludge density and viscosity, increase SMP and irreversible fouling, while SMP and EPS levels decrease along with a slight change of membrane fouling propensity when employing the salt-tolerable bacteria in MBRs. In the feedwater, more magnesium at a low ratio of monovalent to divalent is benefit for enhancement of membrane permeability and sludge properties;

(4) Regarding membrane properties, asymmetric membranes with more hydrophilicity, less roughness, higher zeta potential and relatively large pore size are recommended for fouling control during MBR operation. Impacts of membrane characteristics (e.g. hydrophilicity or hydrophobicity, roughness, surface charge, morphology) on membrane fouling are also determined by activated sludge properties (e.g. hydrophilicity or hydrophobicity, morphology);

(5) Recently, as compared to other cleaning methods (e.g. physical cleaning, chemical cleaning, physico-chemical cleaning), biological/biochemical cleaning as a more promising technology for biofouling control can reduce biofilm formation and slightly influence on membrane properties. Addition of biomass carriers or flocculants directly into MBRs minimizes membrane fouling by improving sludge properties. However, there are some drawbacks of plastic media and PAC. For plastic media, they cannot effectively prevent pore blocking. Besides, too high concentration of PAC would facilitate the formation of cake layer on the membrane or pore blocking inside the membrane.

UNIVERSITY OF TECHNOLOGY, SYDNEY

Chapter 3

Experimental Investigations

3.1. Introduction

This chapter presents synthetic wastewater characteristics, experimental setup, operating conditions, membrane fouling analysis, and the extraction and analysis methods of samples for the integrated systems (Chapter 4–6) during the study period and at the end of the experiments. During the operation period, the sludge samples were periodically taken from bioreactors for their characterization, while organic and nutrient of influent and effluent samples were analysed at the certain time intervals. At the end of the experiment, the foulants on membrane surface were collected for further analysis. The filtration resistance for the fouled membrane were also measured. In Chapter 7, the specific materials and methods for integrated MBBR-MBR systems were given.

3.2. Materials and methods

3.2.1. Wastewater characteristics

The experiments were conducted using a synthetic wastewater to avoid any fluctuation in the feed concentration and provide a continuous source of biodegradable organic pollutants such as glucose, ammonium sulphate and potassium dihydrogen orthophosphate. It was used to simulate domestic wastewater just after primary treatment. The synthetic wastewater has dissolved organic carbon (DOC) of 100–130 mg/L, chemical oxygen demand (COD) of 330–360 mg/L, ammonium nitrogen (NH_4-N) of 12–15 mg/L and orthophosphate (PO_4-P) of 3.3–3.5 mg/L. $NaHCO_3$ or H_2SO_4 was used to adjust pH to 7 (Deng et al., 2014). Components of the synthetic wastewater could be found in Table 3.1 (Lee et al., 2003).

Table 3.1. Compositions of synthetic wastewater

Compounds	Molecular weight (g/mol)	Concentration (mg/L)
Organics and nutrients		
Glucose ($C_6H_{12}O_6$)	180.0	280
Ammonium sulphate ($(NH_4)_2SO_4$)	132.1	72
Potassium phosphate (KH_2PO_4)	136.1	13.2
Trace nutrients		
Calcium chloride ($CaCl_2{\cdot}2H_2O$)	147.0	0.368
Magnesium sulphate ($MgSO_4{\cdot}7H_2O$)	246.5	5.07
Manganese chloride ($MnCl_2{\cdot}7H_2O$)	197.9	0.275
Zinc sulphate ($ZnSO_4{\cdot}7H_2O$)	287.5	0.44
Ferric chloride anhydrous ($FeCl_3$)	162.2	1.45
Cupric sulphate ($CuSO_4{\cdot}5H_2O$)	249.7	0.391
Cobalt chloride ($CoCl_2{\cdot}6H_2O$)	237.9	0.42
Sodium molybdate dehydrate ($Na_2MoO_4{\cdot}2H_2O$)	242.0	1.26
Yeast extract		30

3.2.2. Experimental setup and operating conditions of a submerged membrane bioreactor (MBR)

A hollow fiber polyvinylidene fluoride (PVDF) membrane used in this study had a pore size of 0.2 μm and surface area of 0.1 m^2 (Fig. 3.1). The membrane module was directly submerged into MBR with working volume of 8 L (Fig. 3.2). The seed sludge was firstly taken from a local Wastewater Treatment Plant (Sydney Olympic Park). Subsequently, the sludge was added into the MBRs and fed by synthetic wastewater for acclimatization. There was no sludge waste in both MBRs during the operation period, corresponding to infinite sludge retention time (SRT). The feed water was supplied for the MBRs through pumping the synthetic wastewater into reactors. A suction pump was used to withdraw permeate from the membrane. Membrane fouling potential was monitored by transmembrane pressure (TMP) value every day with a pressure gauge. 9–10 L/min air was supplied using a soaker hose air diffuser at the bottom of the reactor. Chemical cleaning for the membrane module was carried out in 0.5% citric acid for 6 h for inorganic matters removal (i.e. Ca^{2+}, Mg^{2+}), 0.4% sodium hydroxide for 6 h for organic foulant removal, and subsequent 0.8% sodium hypochlorite for 6 h for organic and biological foulant removal when terminating the filtration experiments at TMP of 35 kPa.

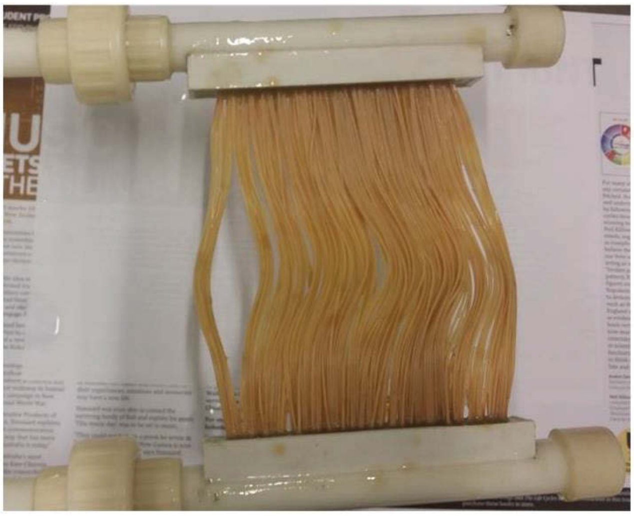


Fig. 3.1. Hollow fiber PVDF membrane module

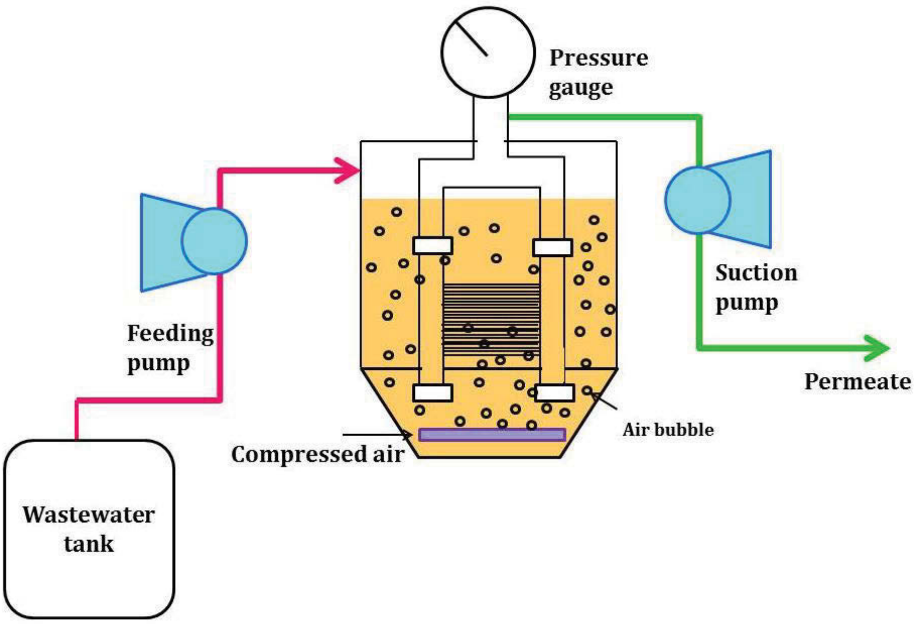


Fig. 3.2. Experimental set-up of MBR

3.2.3. Organic and nutrient analyses

Total organic carbon (TOC) describes the materials originating from decaying vegetation, bacterial growth, and metabolic activities of living organisms or chemicals in water. It reflects the concentration of all organic carbon atoms covalently bonded in the organic molecules in the aquatic system (i.e. water), which can be used to indicate water quality during wastewater treatment process.

COD can measure oxygen consumption when decomposing organic matter and oxiding inorganic chemical (e.g. ammonia, nitrite) in water. This study employed COD reagents from Hanna Instrument, Hanna reactor and Hanna spectrophotometer for COD analysis according to Standard Methods (APHA, AWWA, WEF, 1998). The measurement of COD is carried out on the basis of a commonly used oxidant (potassium dichromate ($K_2Cr_2O_7$)) and boiling sulphuric acid (H_2SO_4) under high temperature of 150 ºC and certain period of 2 h, which allows almost complete oxidation of organic and inorganic matters into CO_2, H_2O, NH_4, and NO_3.

As the presence of excessive nitrogen and phosphorus in aquatic environment induces eutrophication in water and deteriorates water quality, the levels of NH_4-N, NO_3-N, NO_2-N and PO_4-P are also required to be measured. NH_4-N, NO_2-N, and NO_3-N and PO_4-P of influent and effluent sampes were quantified using photometric method called Spectroquant® Cell Test (NOVA 60, Merck).

3.2.4. Oxygen uptake rate (OUR) and specific oxygen uptake rate (SOUR)

The oxygen uptake rate (OUR) is referred to as the rate at which microorganims in activated sludge processes use oxygen for consuming substrate. Specific oxygen uptake rate (SOUR) reflects the amount of oxygen using by microorganisms to uitilize one gram of food, which is expressed as the milligram of oxygen consumed per gram of volatile suspended solids (VSS) per hour (Young, 1999).

YSI 5300 biological oxygen monitor was employed in this study with the oxygen electrode. The calibration of OUR and SOUR are shown as follows:

(1) Plotting observed readings (DO concentration %) versus time (minutes) and determine the slope of the line of best fit in Microsoft Excel software;

(2) Calculate OUR in milliliters per oxygen liter per hour as follows

$$\text{OUR (mL } O_2/\text{L·h)} = 6.51\ \mu\text{L } O_2/\text{mL} \times \text{slope} \times 60 \text{ min/h} \times 10^{-3}/10^{-3} \text{ mL/L} \tag{3.1}$$

(3) Calculate SOUR in milligrams per liter per gram per hour as follows:

$$\text{SOUR (mg/g)/h} = 1.308 \text{ g/L} \times \text{OUR (mL } O_2/\text{L·h)} / \text{MLVSS (mg/L)} \times 10^{-3}/10^{-3} \text{ L/g} \tag{3.2}$$

3.2.5. Membrane fouling analysis

A. Filtration characteristics

When completing the filtration experiments, membrane module was disconnected from the suction pump to determine fouling resistance at different fluxes with distilled water. The resistance-in-series model was employed to quantify the filtration characteristics of the membrane in this study (Fig. 3.3), which was described by the following equations (Choo and Lee, 1996):

$$J = \Delta P/\mu R \tag{3.3}$$

$$R_T = R_M + R_C + R_P \tag{3.4}$$

where J is the permeate flux; ΔP is the TMP; μ is the viscosity of the permeate; R_T is total resistance; R_M is the intrinsic membrane resistance, causing by membrane itself and permanent resistance; R_C is the cake resistance, as a result of deposition of foulants on membrane surface; and R_P is the pore blocking resistance, resulting from adsorption of foulants with small size into membrane pores (Shirazi et al., 2010).

At the end of the experiment, the fouled membrane was put into another tank and operated with distilled water. R_T could be obtained by assigning the recorded flux and TMP values to Eq. (3.3). After removing fouling layer from membrane surface with a sponge or soft brush, the filtration test was conducted for the membrane with distilled water to give R_M+R_P. R_C can be calculated from Eq. (3.4) through substrating R_T from R_M+R_P. R_M was computed from results of flux and TMP by distilled water with a cleaned membrane before MBR operation according to Eq. (3.3). R_P was obtained by using Eq. (3.3).

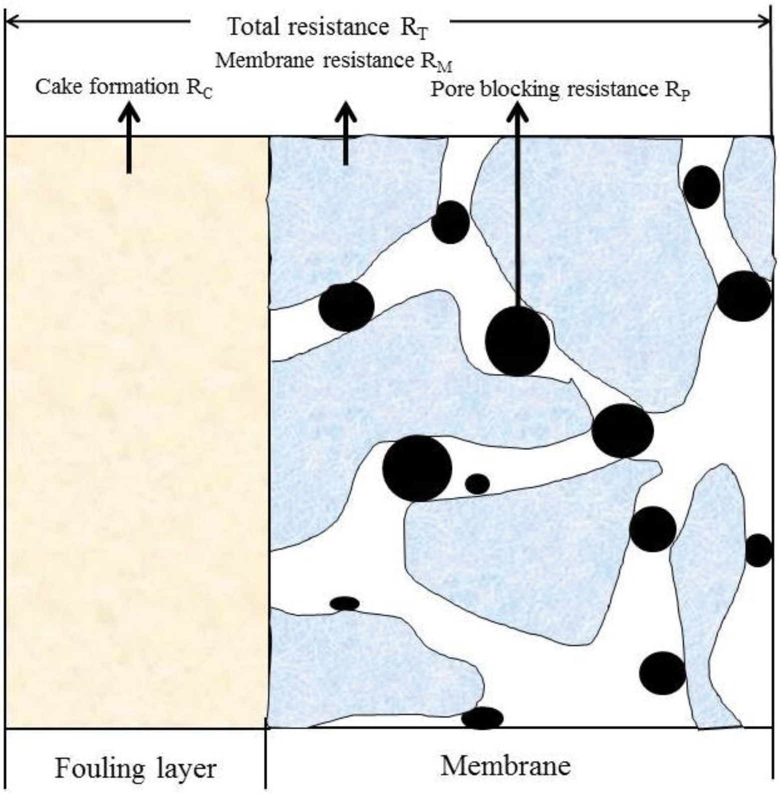


Fig. 3.3. The major resistances of membrane during filtration (modified from Shirazi et al., 2010)

B. Membrane fouling rate

Fouling rate indicating membrane fouling development during MBR operation can be expressed as the following equation:

$$\Delta TMP/\Delta t = (TMP_a - TMP_b)/(a-b) \quad (3.5)$$

where $\Delta TMP/\Delta t$ is fouling rate; TMP is transmembrane pressures; a and b is the measuring time (day or hour).

3.2.6. Biomass concentration and growth rate

Mixed liquor suspended solids (MLSS) measures the concentration of suspended solids in MBR. The volatile solids in mixed liquor sample mainly contain microorganisms and organic matters, corresponding to mixed liquor volatile suspended solids (MLVSS) of the sample. The determination of MLSS and MLVSS concentrations were conducted based on the Standard Methods (APHA, AWWA, WEF, 1998).

Biomass growth rate of mixed lqiuor is calculated as

$\Delta MLSS/\Delta t = (MLSS_c - MLSS_d)/(c-d)$ (3.6)

where $\Delta MLSS/\Delta t$ is biomass growth rate; $MLSS_a$ is MLSS concentration of sludge samples on Day c; MLSSb is MLSS concentration of sludge samples on Day d.

For attached biomass on sponge, sponge was taken out from the reactors, which was then squeezed and washed off using milli-q water to collect almost all biomass from sponge (Ngo et al., 2008). The samples were measured as MLSS and MLVSS concentration.

3.2.7. Characterization of mixed liquor and cake layer

A. Extracellular polymeric substances (EPS), soluble microbial products (SMP) and biopolymer clusters (BPC)

Physical and chemical methods are mainly used for extraction of SMP and EPS, respectively. Centrifugation, as one of physical extraction methods that can be achieved using the external forces, is employed to facilitate SMP to be dissolved in bulk solution. With respect to chemical extraction methods, addition of chemical reagents can help to interrupt the binding interactions between EPS and microbial cells, which promote EPS extraction from sludge samples. In our studies, centrifugation and the cationic exchange resin (CER) method are employed to obtain SMP and EPS. The cation resin is Dowex® Marathon™ C sodium form (Sigma-Aldrich), which shows strong acidity and possesses 20–50 nm mesh. As CER has a high selective ability for Ca^{2+}- and Mg^{2+}-bound EPS, it can remove the multivalent cations involving cross-linking EPS within sludge matrix, resulting in breakup of the flocs and subsequent generation of bound EPS (Park and Novak, 2007; Sheng et al., 2010).

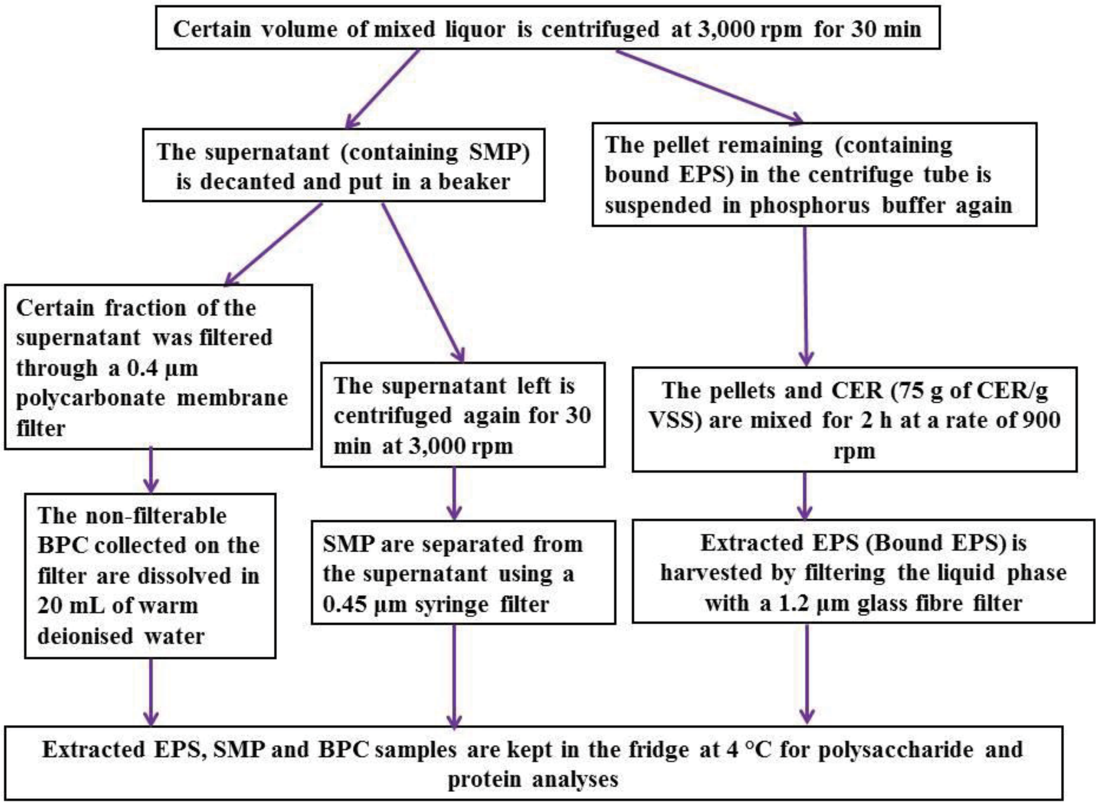


Fig. 3.4. Procedure for EPS, SMP, and BPC extraction

The CER method was modified based on the protocol by Frølund et al. (1996). Fig. 3.4 presents the extraction procedure of EPS, SMP and BPC. Initially, certain volume of mixed liquor (e.g. 30 mL) directly taken from the bioreactor was put in the centrifuge tube, which was subjected to the centrifugation at 3000 rpm for 30 min in the centrifuge (Clement 2000 (Fig. 3.5)). The collected supernatant was then centrifuged for another 30 min at 3000 rpm for further extraction. Subsequently, the organic matters obtained by filtering the supernatant through 0.45 µm Phenex-NY (Nylon) syringe filter was designated as SMP. Besides, after submitting the supernatant to a 0.4 µm polycarbonate membrane filter (Millipore), the non-filtrate substances on the membrane filter were dissolved in 20 mL of distilled water in warm water bath. The obtained organic solution was designed as BPC (Sun et al., 2008). On the other hand, the sludge pellets taken from the bottom of the centrifuge tubes were resuspended with phosphorus buffer solution up to the predetermined volumes (30 mL) as the original mixed liquor samples. Certain amounts of the cation exchange resin were then added into the pellets, which was further exposed to centrifugation for 2 h at 900 rpm. Finally, the liquid phase of the extracted samples in the centrifuge tube was filtered through 1.2 μm Phenex-GF (Glass fiber, Whatman) syringe filter to obtain bound EPS. For the foulants on membrane surface, the cake layer was softly scraped off from the membrane surface, which was suspended in distilled water to 100 mL. The extraction of bound EPS and SMP of cake layer followed the procedure above for those of the mixed liquors.

As the major fraction of bound EPS, SMP and BPC, polysaccharides (EPS_C, SMP_C, BPC_C) and proteins (EPS_P, SMP_P, BPC_P) were measured using Anthrone-sulfuric acid method and the modified Lowry method (Sigma, Australia), respectively (Raunkjer et al., 1994). Determination of polysaccharides is based on the principle of hydrolyses of carbohydrate (or polysaccharides) into simple sugars in hot acid medium (e.g. sulphuric acid), which is further developed to be a green coloured product in the presence of anthrone reagent showing an absorption maximum at 630 nm (Sadasivam et al., 1996). The total protein kit supplied by Sigma is Peterson's modification of the micro Lowry method (Fig. 3.6). Although protein solution can be directly measured through the Lowry reaction, the Lowry procedure may be interfered by other chemicals (e.g. tris, ammonium sulfate, Ethylene diamine tetraacetic acid (EDTA), sucrose, citrate, amino acid and peptide buffers, phenols) in the protein solution. Hence, both deoxycholate (DOC') and trichloroacetic acid (TCA) are employed to cause the precipitation of protein to reduce the interferences. The Lowry Reagent containing sodium dodecylsulfate is further added to the aforementioned precipitants, which promotes the dissolution of relatively insoluble lipoproteins. The subsequent procedure depends on two chemical reactions: (1) the biuret reaction referring to the reaction between the peptide bonds of proteins and alkaline cupuric tartrate reagent complexes; (2) the Folin-Ciocalteau reaction, which reduces Folin & Ciocalteu's phenol reagent by the copper-catalyzed oxidation of aromatic amino acids, resultign in a strong blue or purple color. The adsorbance of the colored solution ranges from 500 to 800 nm (Bensadoun and Weinstein, 1976; Waterborg, 2002).

Fig. 3.5. Photo of centrifuge (Clements 2000)

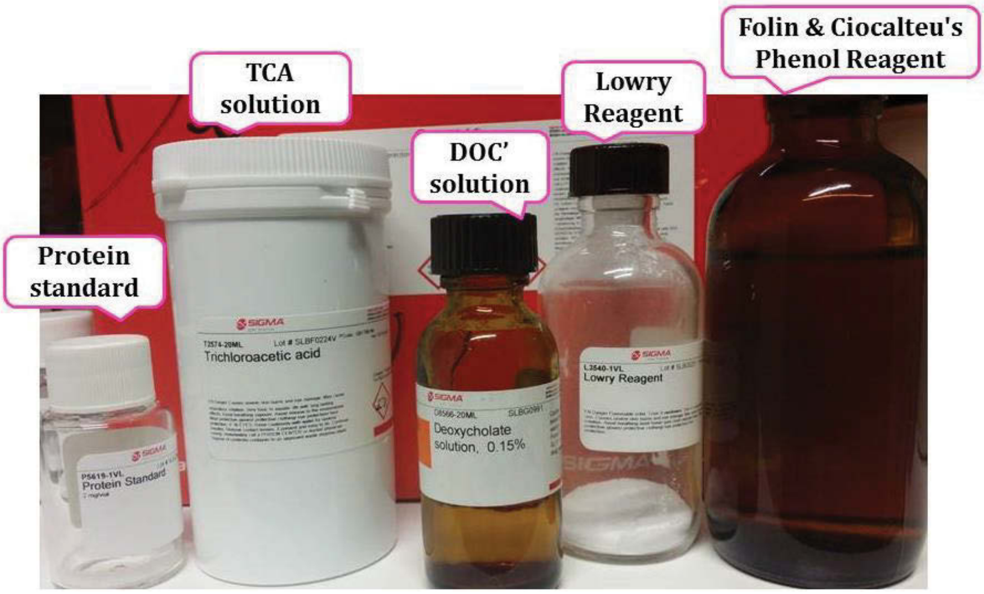


Fig. 3.6. Total protein kit (TP0300 SIGMA, Sigma-Aldrich)
for protein analysis

B. Apparent viscosity, relative hydrophobicity (RH), zeta potential, sludge floc size

The apparent viscosity of activate sludge can be expressed by the ratio of shear stress (τ) to shear rate (γ):

$$\mu = \tau/\gamma \tag{3.7}$$

It implies that the shear stress affect the deformation of the sludge flocs, which indicates the occurrence of the internal and external interactions and forces inside the sludge flocs and the bulk solution (Yang et al., 2009c). In this study, Brookfield Viscometer M/OO-151-E0808 (Brookfield, USA) (Fig. 3.7(a)) was applied for evaluating the apparent viscosity of mixed liquor.

The hydrophobicity and surface charge of sludge flocs could reflect their bioflocculation ability (Liao et al., 2004b). The RH of sludge flocs is referred to as the extent of adherence of sludge flocs to hydrocarbon (n-hexane). According to the method reported by Ji et al. (2010), 50 mL mixed liquor was put in the separatory funnel (Fig. 3.7(b)) and then 50 mL n-hexane was added into the funnel. The mixture was shaken vigorously for 30 min, followed by a 30-min settling. When a clear boundary between two phases (separation line) was observed, the phase at the bottom of the funnel below the boundary was collected in a beaker. The RH was expressed by the ratio of the concentrations of the separated hydrocarbon phase and the initial concentration of the original mixed liquor:

$$RH\ (\%) = (1 - MLSS_e/MLSS_i) \times 100\% \tag{3.8}$$

where $MLSS_e$ is the MLSS concentration in the aqueous phase after emulsification and $MLSS_i$ is the initial MLSS concentration of the mixed liquor sample. The difference between $MLSS_i$ and $MLSS_e$ is hydrocarbon phase and the concentration of sludge flocs adhering to n-hexane, indicating the hydrophobicity of sludge flocs. Generally, sludge flocs show negative charges at neutral pH. The density of surface charge of sludge flocs is reflected by the ionizable groups (e.g. carboxyl, phosphate, and amino groups) in bound EPS and on the surfaces of microorganisms (Liao, 2000). The surface charge of activated sludge in this study was provided by the zeta potential, which was determined using Zetasizer Nano ZS (Malvern Instruments, UK) (Fig. 3.7(c)). As to the sludge floc size, one drop of the mixed liquor was spread on the slide. After that, the image acquisition of sludge flocs were conducted using the Olympus System Microscope Model BX41 (Olympus, Japan) (Fig. 3.7(d)). The Image-Pro Plus software was employed to analyze the microscopic images to measure the floc size.

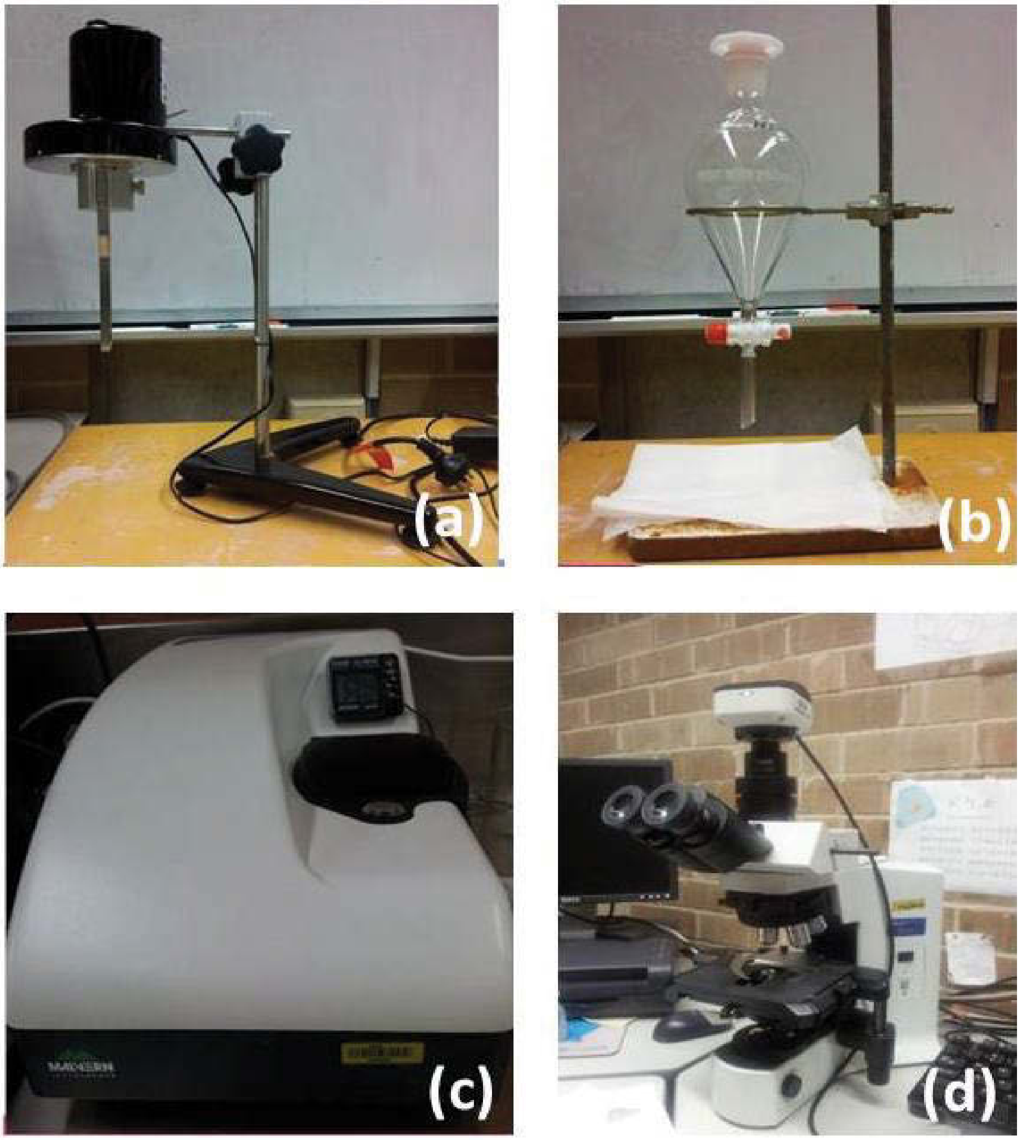


Fig. 3.7. The instruments for analysis of sludge samples: a) Brookfield Viscometer M/OO-151-E0808 for sludge viscosity; b) Separatory funnel for RH; c) Zetasizer Nano ZS for zeta potential; d) Olympus System Microscope Model BX41 for obtaining images of sludge flocs

3.3. Analysis schedule

The sample analyses are conducted at different stages of experiments in the research (Fig. 3.8).

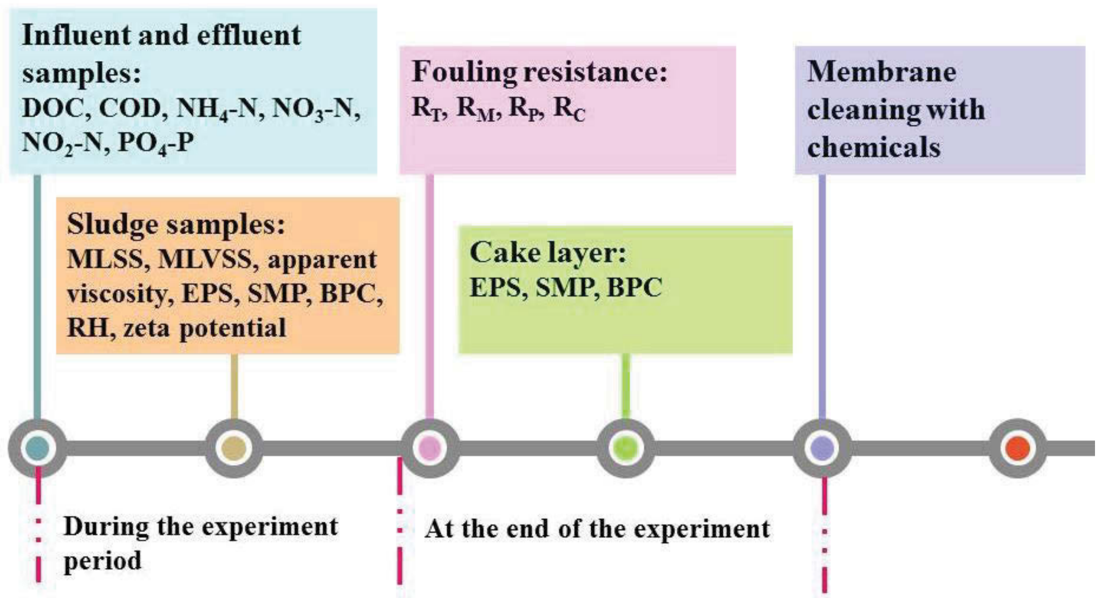


Fig. 3.8. Diagram of analyses in the research

Chapter 4

A Comparison Study on the Performance of a Sponge-Submerged Membrane Bioreactor and a Conventional Membrane Bioreactor

Chapter 4 A Comparison Study on the Performance of a Sponge-Submerged Membrane Bioreactor and a Conventional Membrane Bioreactor

4.1. Introduction

4.1.1. Background

Biofouling is a critical problem during MBR process for water reclamation and reuse. To reduce membrane fouling, a chemical cleaning-in-place (CIP) was investigated by Wei et al. (2011) in a long-term operation of pilot-scale submerged MBR for municipal wastewater treatment. They reported that the chemical CIP, in both transmembrane pressure (TMP) controlling mode and time controlling mode, effectively removed the fouling in terms of membrane pore blockage and gel layer caused by colloids and soluble organic substances. Wu and He (2012) suggested that the low irreversible fouling was found in the cyclic aeration mode, which could be ascribed to the floc destruction and re-flocculation processes. During the short high aeration period, the preservation of the strong strength bonds within activated sludge flocs caused less release of soluble and colloidal material in the supernatant. The weak strength bonds damaged in the high aeration period could be recovered in the re-flocculation process in the low aeration period. Current studies have also explored the feasibility of other methods (e.g. biomass media) for fouling control (Chapter 2, Section 2.5). Using biomass carriers (e.g. plastic media, powdered activated carbon (PAC)) in MBR is an effective and promising method to control membrane fouling by reducing fouling components in mixed liquor and scouring membrane surface (Jin et al., 2013; Ng et al., 2013). The previous studies have suggested that sponge could mitigate membrane fouling as demonstrated by improved sustainable flux, increased permeate flux, and decreased TMP development rate in the sponge-submerged membrane bioreactors (SSMBRs) (Guo et al., 2008a; Nguyen et al., 2012a). However, the effects of sponge on sludge characteristics and membrane fouling have yet to be investigated in MBR systems.

4.1.2. Objectives

A comparison study was conducted to evaluate the performance of a SSMBR and a conventional MBR (CMBR) based on sludge characteristics, such as zeta potential, apparent viscosity, relative hydrophobicity (RH), bound extracellular polymeric substances (EPS) and soluble microbial products (SMP). The cake layer formation on membrane surface was also analysed.

Chapter 4 A Comparison Study on the Performance of a Sponge-Submerged Membrane Bioreactor and a Conventional Membrane Bioreactor

A major part of Chapter 4 has been published as the following paper:

Deng, L., Guo, W.S., Ngo, H.H., Zhang, J., Liang, S., Xia, S., Zhang, Z., Li, J., 2014. A comparison study on membrane fouling in a sponge-submerged membrane bioreactor and a conventional membrane bioreactor. Bioresource Technology 165, 69–74.

4.2. Materials and methods

4.2.1. Experimental setup and operating conditions of the SSMBR and the CMBR

The experimental setup and operating conditions of the SSMBR and the CMBR fed by synthetic wastewater are described in Chapter 3, Section 3.2.1 and 3.2.2. Both MBR reactors had the same effective working volume of 8 L, which were operated at the constant filtration flux of 10 $L/m^2{\cdot}h$ with relaxation mode (59 min on and 1 min off) (Fig. 4.1). The initial MLSS concentrations were 7.03 and 6.98 g/L for the SSMBR and the CMBR, respectively. Table 4.1 summarizes the specifications of the SSMBR and the CMBR.

4.2.2. Sponge specifications

The reticulated porous polyester-polyurethane sponge (PUS) was used in SSMBR system. The PUS has density of 28–45 kg/m^3 and cell count of 45 cells/in (45 cells per 25.4 mm). The dimensions of the sponge cubes are 10 mm, 10 mm, and 10 mm in length, width and thickness, respectively. One of the previous studies (Guo et al., 2008a) investigated impacts of sponge volume fractions (0%, 10%, 20%) on the sustainable flux in SSMBRs. The results indicated that the sponge volume fraction of 10% reduced membrane fouling and improved sustainable flux (about two times) compared to those for the SMBR alone. Therefore, the sponge volume fraction was 10% in the SSMBR in this chapter. Before running the experiments, the sponge cubes were acclimatized to synthetic wastewater for 25 days.

4.2.3. Analysis methods

During acclimatization period, the analysis of attached biomass (including mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS)) of sponge, their oxygen uptake rate (OUR) and specific OUR (SOUR) were performed every two days. For the SSMBR and the CMBR, dissolved organic carbon (DOC), chemical oxygen demand (COD), NH_4-N, NO_3-N, NO_2-N and PO_4-P of influent and effluent samples were measured on different day (i.e. Day 2, 3, 5, 10, 15,…) to determine their treatment performance. Transmembrane pressure (TMP) value was recorded every day. Laboratory analysis was also performed on the sludge samples for the levels of MLSS, MLVSS, extracellular polymeric substances (EPS), soluble microbial products (SMP), biopolymer clusters (BPC), apparent viscosity, zeta potential, relative hydrophobicity (RH), and sludge floc size. At the end of the experiment, cake layer was characterized by EPS, SMP and BPC. Moreover, the filtration characteristics were obtained from various fluxes and the corresponding TMP data.

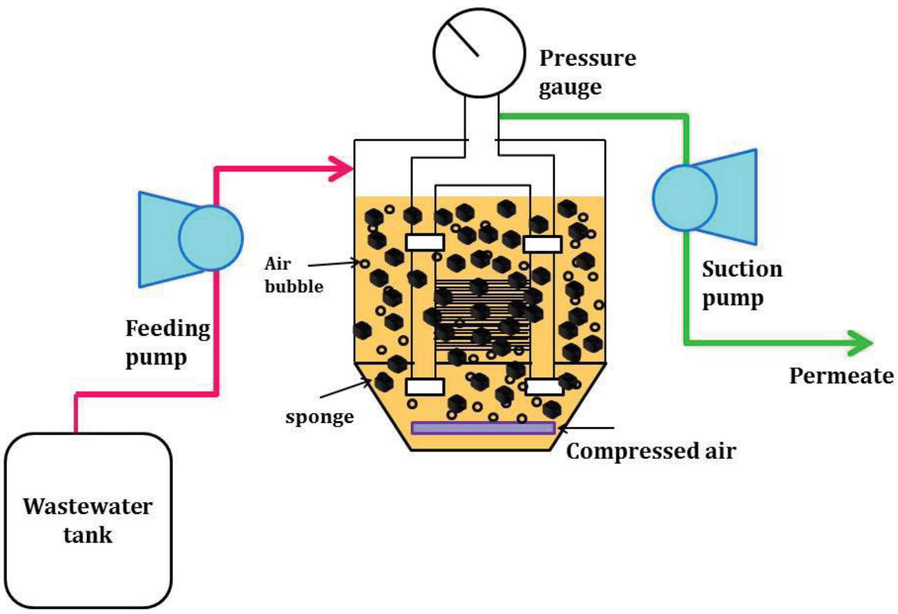


Fig. 4.1. Experimental set-up of the SSMBR

Table 4.1. System descriptions and operating conditions of the SSMBR and the CMBR

Sponge	
Sponge size	10 × 10 × 10 mm
Sponge type	$S_{28\text{-}30}$/45R
Membrane properties	
Membrane configuration: Hollow fiber membrane module	
Material: Polyvinylidene fluoride (PVDF)	
Pore size: 0.2 μm	
Surface area: 0.1 m^2	
Operating conditions	
Constant flux	10 $L/m^2 \cdot h$
Air flow rate	9 L/min
Sponge volume fraction	10%
Working volume	8 L
SRT	no sludge withdrawal
Initial MLSS concentration	7.03 g/L for the SSMBR, 6.98 g/L for the CMBR
Operating mode	59 min ‘‘on’’ and 1 min ‘‘off’’ per hour
Synthetic wastewater	
TOC	100–130 mg/L
COD	330–360 mg/L
NH_4-N	12–15 mg/L
PO_4-P	3.3–3.5 mg/L

4.3. Results and discussion

4.3.1. Attached biomass growth on sponge during acclimatization

Before the experiments, the PUS cubes were put in the aeration tank filling with activated sludge at an initial MLSS concentration of 6 g/L. Fig. 4.2 shows the average concentrations of attached biomass on sponge. After 17 days of acclimatization, MLSS and MLVSS concentrations of attached biomass stabilized at around 19.8 and 18.0 g/L sponge, respectively. As shown in Fig. 4.3, specific oxygen uptake rate (SOUR) reached equilibrium quicker with strong microbial activity for first 10 days, with 99.2% on day 5 and 99.5% on day 7 within 10 minutes, respectively.

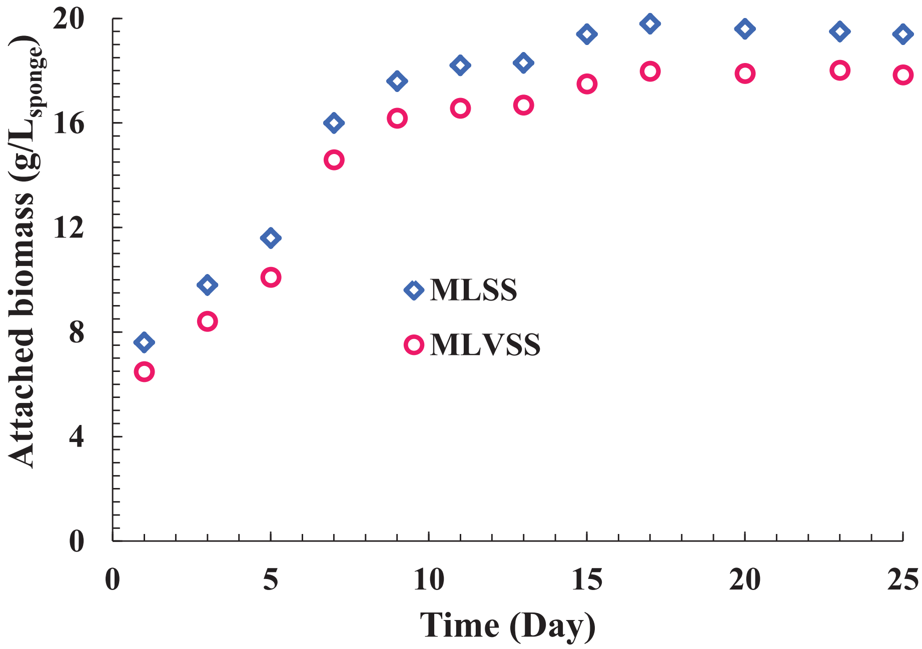


Fig. 4.2. The attached growth on sponge during acclimatization

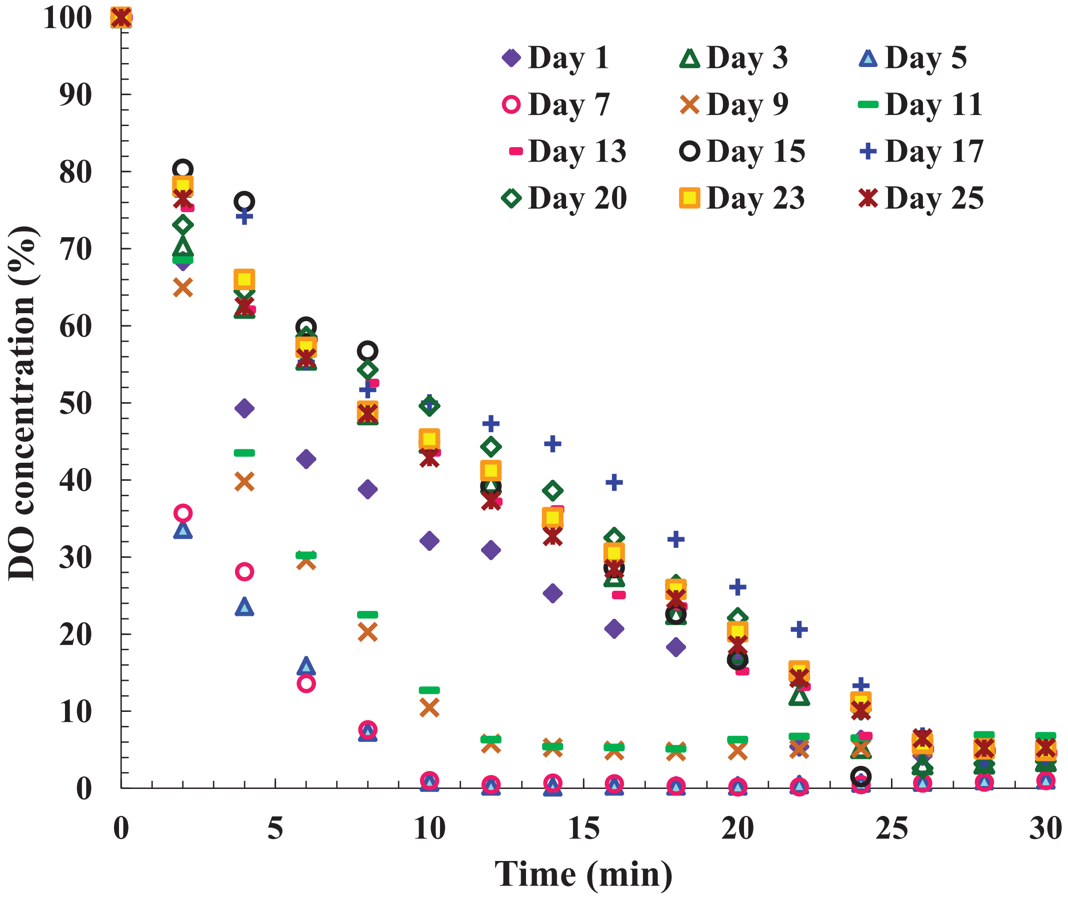


Figure 4.3. SOUR variation of attached growth on sponge during acclimatization

4.3.2. The performance of the SSMBR and the CMBR

A. Treatment performance of the SSMBR and the CMBR

As shown in Table 4.2, more than 90% of organic removal was obtained in both SSMBR and CMBR. The SSMBR showed higher performance for removing NH_4-N (> 70%) and PO_4-P (> 60%), while around 60% of NH_4-N and 30% of PO_4-P were removed in the CMBR. Higher NH_4-N removal in the SSMBR could be attributed to the enhanced population of ammonium oxidation bacteria on the acclimatised sponge during acclimatization period (Nguyen et al., 2012). As sponge could provide the anoxic condition around the surface of the sponge and the anaerobic condition inside the sponge, the SSMBR achieved a higher removal efficiency of PO_4-P (Guo et al., 2008a). As compared to the CMBR (T-N removal < 40%), higher T-N removal (> 50%) was obtained in the SSMBR due to the occurrence of denitrification. Sponge facilitated the formation of the anoxic condition around the surface of sponge or inside the sponge caused by the DO gradient occurring along the inward depth of the sponge (Guo et al., 2008a; Lim et al., 2011).

Table 4.2. Removal efficiencies of DOC, COD, PO_4-P, NH_4-N and T-N in the SSMBR and the CMBR during the operational period

Reactors	DOC (%)	COD (%)	PO_4-P (%)	NH_4-N (%)	T-N (%)
SSMBR	94.74 ± 5.49	93.53 ± 4.46	63.57 ± 5.32	74.35 ± 3.22	53.28 ± 2.16
CMBR	94.17 ± 7.32	91.95 ± 6.53	27.22 ± 6.18	58.14 ± 6.13	37.20 ± 4.58

B. TMP development in the SSMBR and the CMBR

Fig. 4.4 depicts the time course of TMP increase in both SSMBR and CMBR. Both MBRs demonstrated significant difference in TMP profiles. TMP in the SSMBR was maintained at 2.0 kPa up to 90 days. In the CMBR, TMP gradually increased from 5.0 kPa to 7.0 kPa until day 6, followed by a rapid TMP rise. After 35 days, the TMP reached 31.0 kPa, suggesting chemical cleaning should be conducted for the membrane. These results indicated that sponge addition could significantly mitigate membrane fouling, which is further discussed in details in Section 4.3.6.

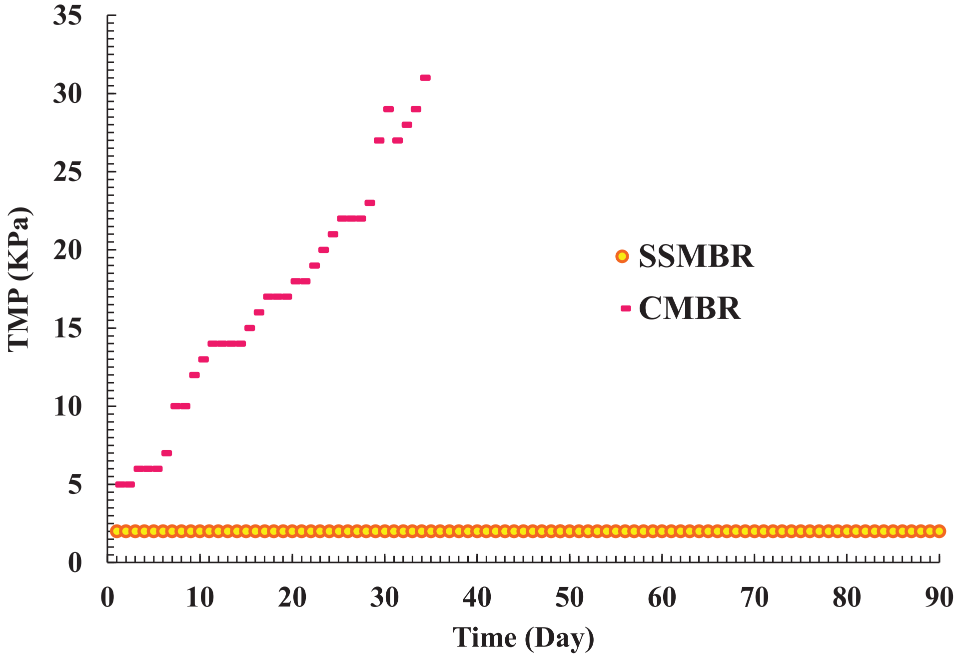
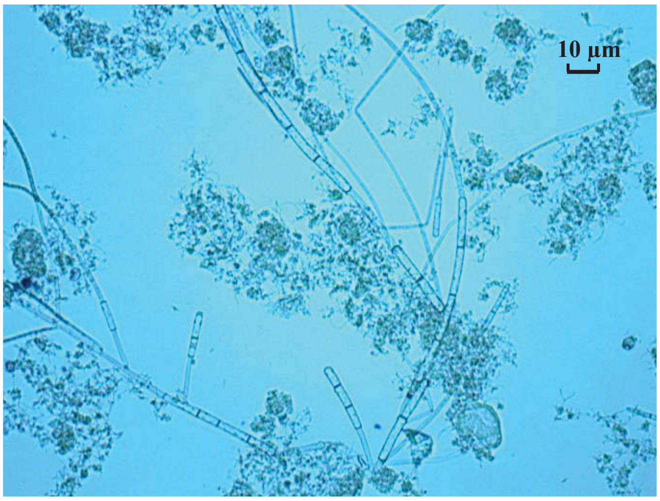


Fig. 4.4. TMP profile for the SSMBR and the CMBR

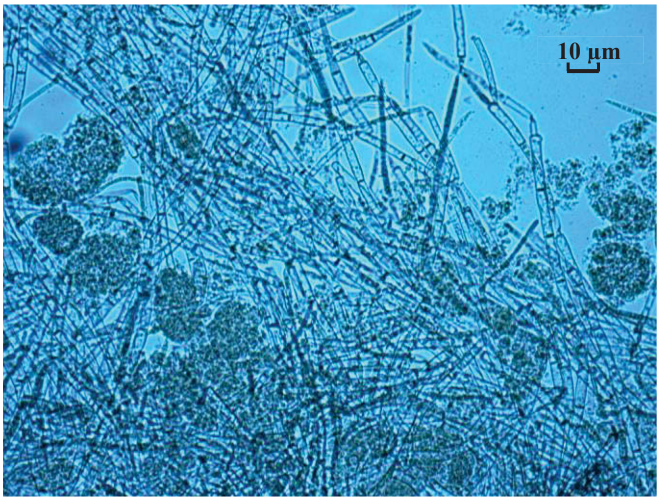
4.3.3. Mixed liquor suspended solids (MLSS) concentration and sludge viscosity

During the experimental period, sludge concentration kept increasing in both MBRs due to no sludge withdrawal. MLSS concentrations were 11.50 ± 4.52 g/L and 9.41 ± 2.38 g/L in the CMBR and the SSMBR after 35 and 90 days of operation, respectively. The lower MLSS concentration in the SSMBR might be attributed to the fact that sponge addition could balance the microorganism growth in suspended activated sludge as well as on and inside the porous sponge cubes (Ngo et al., 2006). Normally, activated sludge contains organic and inorganic matters. As the organic matters in activated sludge indicate the amount of active microorganisms, the sludge activity could be demonstrated by MLVSS/MLSS ratio. In the SSMBR and the CMBR, the average MLVSS/MLSS ratio was 0.86 and 0.82, respectively. It implied that sponge may increase biological components and reduce accumulation of inorganic composition (Fan et al., 2015; Long et al., 2015). Biomass growth rate in terms of ΔMLSS/Δt in both MBRs showed that the biomass growth was 0.053 g/L·d in the SSMBR, which was lower than that without sponge in the CMBR (0.26 g/L·d). In the SSMBR, sponge addition could balance the microorganism growth in suspended activated sludge as well as on and inside the porous sponge cubes through adsorbing biomass, which decreased biomass concentration and lowered their growth in mixed liquors (Ngo et al., 2006).

It was found that there is an exponential relationship between MLSS concentration and sludge viscosity (Reid et al., 2008). In this chapter, sludge viscosity was higher (3.30 ± 0.50 mPa·s) in the CMBR than that (2.60 ± 0.40 mPa·s) in the SSMBR, demonstrating that higher sludge viscosity was attributed to higher MLSS concentration. In addition, it has been reported that the sludge flocs with excess filamentous bacteria showed high viscosity due to presence of high EPS concentration (Meng et al., 2006a). As can be seen in Fig. 4.5, overgrowth of filamentous bacteria was found in the CMBR on day 14, whereas there were less filamentous bacteria in the SSMBR until 83 days, which revealed that higher sludge viscosity in the CMBR was also due to abundance of filamentous bacteria. Similar observations were also recorded by Meng et al. (2007c) who suggested that sludge viscosity was influenced by MLSS concentration, EPS and filamentous bacteria. On the contrary, less filamentous bacteria in the SSMBR may be due to the adsorption effect of sponge, thus lowering sludge viscosity in the SSMBR.



(a)

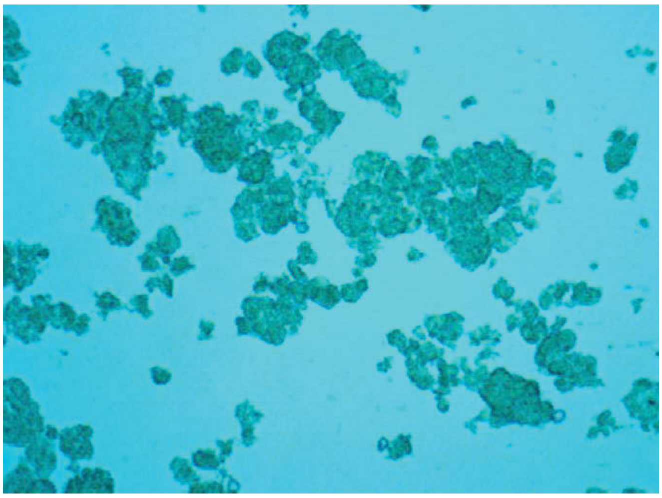


(b)

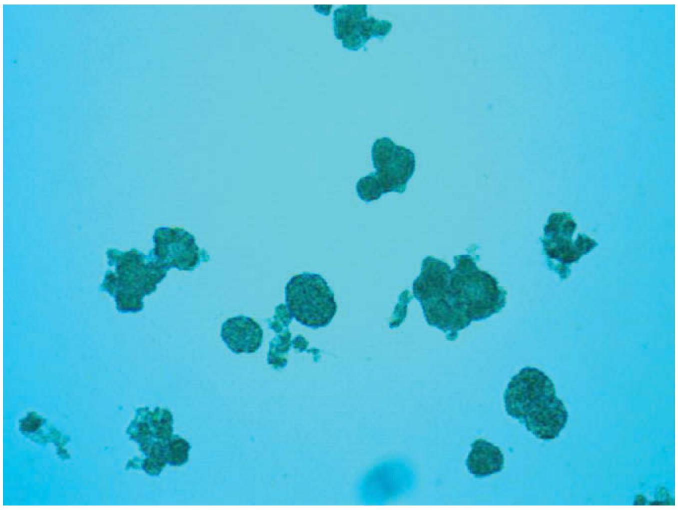
Fig. 4.5. Microscopic images of filamentous bacteria in mixed liquor on (a) day 83 in the SSMBR and (b) day 14 in the CMBR

4.3.4. Zeta potential, relative hydrophobicity (RH) and particle size distribution

The surface charge and hydrophobicity of activated sludge played a key role in their flocculation. The surface charge as indicated by zeta potential affected the extent of agglomeration of sludge flocs. Lower surface charge reduced the electrostatic repulsion but increased interaction energy among sludge flocs, thus enhanced the flocculation of sludge flocs. Moreover, the hydrophobic interaction determined by the hydrophobicity of sludge flocs also contributed to the floc stability and their aggregation ability (Liao et al., 2001; Mikkelsen and Keiding, 2002). In this chapter, activated sludge in the SSMBR had higher zeta potential (- 6.85 ± 3.65 mV) and higher RH (81.00 ± 7.80%) than those in the CMBR (zeta potential of -10.50 ± 4.50 mV, RH of 63.13 ± 13.60%). The results indicated that there might be a positive relationship between surface charge (zeta potential) and hydrophobicity of activated sludge. Moreover, sponge could decrease surface charge and increase hydrophobicity of mixed liquor. It has been suggested that filamentous bacteria promoted the floc formation through forming backbones which facilitated attachment of zoogleal (floc-forming) bacteria to the backbones. However, excess filamentous bacteria could prevent the agglomeration of floc particles by producing a bridge lattice due to the generation of abundant filaments from the flocs into the bulk solution, which reduced settling ability of sludge (Liwarska-Bizukojc et al., 2014; Meng et al., 2006a). Results of particle size distribution in this study showed that the SSMBR contained larger sludge flocs (20–50 μm) than that for the CMBR (10–40 μm) (Fig. 4.6). This suggested that activated sludge had better flocculation ability in the SSMBR, which might be due to higher RH and zeta potential of sludge flocs as well as the presence of less filamentous bacteria.



(a)



(b)

Fig. 4.6. Microscopic images of the sludge flocs in mixed liquor on day 20 in (a) the SSMBR and (b) the CMBR

4.3.5. Bound EPS and SMP in activated sludge

Normally, polysaccharides and proteins are considered as the major fractions of EPS and SMP that contribute to fouling (Guo et al., 2012a). Tables 4.3 and 4.4 exhibit composition of mixed liquor's SMP and bound EPS in the SSMBR and the CMBR. The CMBR demonstrated higher SMP concentrations (around 2–3 times) within 7–day run. The protein concentrations (SMP_P) were similar for both MBRs, while significantly higher polysaccharide concentrations (SMP_C) were observed in the CMBR, suggesting higher fouling propensity of the CMBR. Although activated sludge of both MBRs had similar bound EPS concentrations, slightly higher protein concentrations (EPS_P) but significantly lower polysaccharide concentrations (EPS_C) were obtained in the CMBR. After 7 days of operation, the SMP concentrations (including SMP_P and SMP_C) of both MBRs presented minor difference. On the other hand, bound EPS concentrations (12.3–24.6 mg/L) in the CMBR were higher than those in the SSMBR (12.2–17.3 mg/L), with lower protein concentrations (EPS_P) but significantly higher polysaccharide concentrations (EPS_C). At shorter SRTs, a higher F/M ratio and dissolved oxygen concentration were obtained in MBRs with a more efficient oxygen transfer rate because of decreased fluid resistance at lower sludge concentration and viscosity. Thus, intracellular storage granules and extracellular polymers outside of the cells were formed due to the surplus substrate and energy for microorganisms (Wu et al., 2011a). However, in this chapter, increase of sludge concentration under infinite SRT condition induced the decrease in food to microorganism (F/M) ratio (0.1–0.2 d^{-1}). As a consequence, both MBRs were fed with limited available substrate, which could cause more cell lysis and cell hydrolysis, thereby releasing EPS and SMP in activated sludge (Yigit et al., 2008). Moreover, the excess growth of filamentous bacteria could produce more SMP, resulting in severe fouling (Pan et al., 2010). Therefore, the CMBR exhibited more serious fouling compared with the SSMBR. In the SSMBR, it was obvious that sponge addition could reduce SMP_C concentrations during the first 7-day run and EPS_C afterwards by the means of adsorption onto sponge as well as biodegradation by attached biomass of the sponge.

It has been reported that large quantity of EPS in activated sludge increased floc strength by polymer entanglement, thereby increasing the extent of sludge flocs agglomeration (Mikkelsen and Keiding, 2002). However, in this chapter, lower EPS concentration but larger particles were observed in the SSMBR, pointing out that the flocculation ability of sludge flocs may not only depend on EPS concentration. Lee et al. (2003) found that the ratio of proteins to polysaccharides (PN/PS ratio) in EPS was important in controlling the hydrophobicity and surface charge of sludge flocs. Table 4.4 shows that a significantly higher PN/PS ratio in bound EPS was found in the SSMBR after 7 days operation. Higher RH of activated sludge in the SSMBR proved that higher EPS_P concentration increased the hydrophobicity of sludge flocs by providing amino acids with more hydrophobic side groups, while lower EPS_C concentration contributed to less hydrophilic nature of sludge. Moreover, the amino groups in EPS_P containing positive charges neutralized some of negatively charged activated sludge, thereby inducing higher zeta potential of sludge flocs in the SSMBR (Lee et al., 2003; Liao et al., 2001). Thus, PN/PS ratio in bound EPS could positively influence hydrophobicity and zeta potential of activated sludge, which improved the affinity among sludge flocs, thereby having a positive impact on the agglomeration ability of the flocs and obtaining a stable structure (Yan et al., 2015).

Table 4.3. SMP compositions and total SMP concentrations of mixed liquor in the SSMBR and the CMBR at two different stages (within and after 7 days of operation) during the operational period

Day	Reactor	SMP			
		PN[a] (mg/L)	PS[b] (mg/L)	PN/PS ratio	SMP (mg/L)
Stage I (Day 1-7)	SSMBR	9.9-10.2	7.2-9.4	1.1-1.4	7.4-17.4
	CMBR	10.6-10.8	13.5-14.4	0.7-0.8	24.1-25.2
Stage II (After day 7)	SSMBR	1.0-4.4	1.0-6.9	0.3-2.3	1.5-9.2
	CMBR	0.4-5.7	1.0-5.8	0.1-3.2	1.1-9.8

[a] PN, proteins; [b] PS, polysaccharides

Table 4.4. Bound EPS compositions and total bound EPS concentrations of mixed liquor in the SSMBR and the CMBR at two different stages (within and after 7 days of operation) during the operational period

Day	Reactor	Bound EPS			
		PN[a] (mg/L)	PS[b] (mg/L)	PN/PS ratio	Total EPS (mg/L)
Stage I (Day 1-7)	SSMBR	7.4-9.9	9.4-11.8	0.6-1.1	19.2-19.3
	CMBR	9.3-9.9	1.0-9.4	4.7-9.3	10.3-19.3
Stage II (After Day 7)	SSMBR	9.8-10.6	1.6-7.5	1.3-6.6	12.2-17.3
	CMBR	6.5-10.1	5.8-14.5	0.7-1.4	12.3-24.6

[a] PN, proteins; [b] PS, polysaccharides

4.3.6. Membrane fouling behaviour

A. Fouling resistance distribution

Results of fouling resistance (Fig. 4.7) showed that the CMBR had a higher total resistance (R_T) (5.47×10^{12} m^{-1}) than that of the SSMBR (2.56×10^{12} m^{-1}). The clean membrane resistance (R_M) were the same (1.71×10^{12} m^{-1}) for both MBRs. Higher cake layer resistance (R_C) was found for the CMBR than that for the SSMBR, corresponding to 3.04×10^{12} m^{-1} and 0.85×10^{12} m^{-1}, respectively. Moreover, pore blocking resistance (R_P) for the CMBR was notably higher. R_P of the CMBR accounted for about 20% of R_T, whereas there was no R_P in the SSMBR. These results suggested that cake layer formation was one of the main factors contributing to membrane fouling. Furthermore, sponge could alleviate membrane fouling not only by preventing pore blocking but also by reducing cake layer formation. Some researchers (Jamal Khan et al., 2012b; Yang et al., 2006) have reported the similar findings that R_C was major fraction of R_T and sponge addition could reduce R_C.

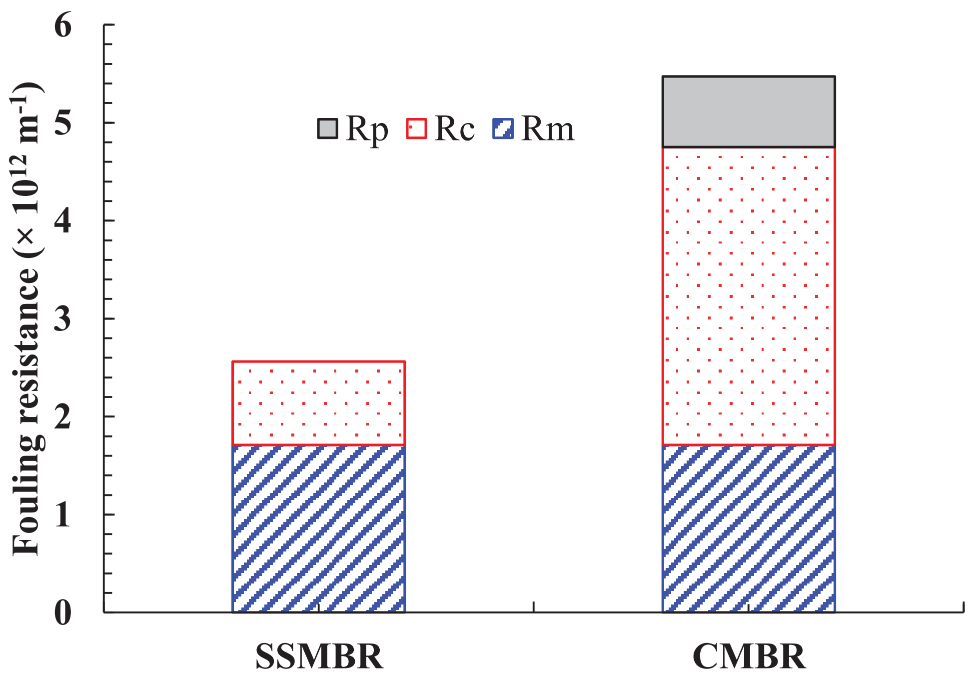


Fig. 4.7. Fouling resistance distribution in the SSMBR and the CMBR

B. Cake layer on membrane surface

According to the aforementioned discussion, activated sludge in both MBRs possessed different properties, which were correlated with membrane fouling potential as well as fouling resistance. When MLSS concentration in mixed liquor was higher, sludge particles, colloids, macromolecular matter and microbial products were accumulated in the bioreactor, which contributed to membrane fouling. The effects of back transport on sludge flocs could be reduced by increased sludge viscosity, leading to increased net force towards the membrane surface. Thus, more particles were deposited on the membrane, thus aggravating membrane fouling (Kornboonraksa and Lee, 2009; Meng et al., 2007c). A sticky cake layer was also formed on membrane surface at higher MLSS concentration due to higher sludge viscosity (Itonaga et al., 2004). Additionally, more easy deposition of the sludge flocs with abundance of filamentous bacteria on membrane surface was achieved owing to their high viscosity. Filamentous bacteria could attach onto the membrane foulants as well as adhere and penetrate between membrane and membrane foulants, leading to the formation of a non-porous cake layer (Meng et al., 2006a). Therefore, it could be noted that higher MLSS concentration, higher sludge viscosity and overgrowth of filamentous bacteria contributed to formation of sticky and non-porous cake layer, giving rise to higher R_C in the CMBR. Being the major fraction of the total fouling resistance, the cake layer was analysed with respect to EPS and SMP (including polysaccharides and proteins). Fig. 4.8 shows the composition of EPS and SMP in the cake layer on membrane surface for both SSMBR and CMBR. Bound EPS concentrations were similar for the SSMBR (15.0 mg/(L·g cake layer)) and the CMBR (13.9 mg/(L·g cake layer)). However, higher concentrations of SMP_C and SMP_P (14.4 and 15.5 mg/(L·g cake layer), respectively) were obtained for the CMBR, while SMP_C and SMP_P of the cake layer were comparatively lower for the SSMBR (9.8 and 7.1 mg/(L·g cake layer), respectively). These results elucidated that higher R_C in the CMBR was mainly caused by SMP (including SMP_C and SMP_P) on membrane surface. At high TMP, more SMP_C and SMP_P could be adsorbed and/or attached onto membrane surface due to the high drag force provided by permeate pump. On contrary, sponge addition effectively reduced SMP_C and SMP_P in cake layer on membrane surface. Apart from adsorption of SMP_C and SMP_P on the sponge and biodegradation by attached microorganisms, reduction of cake layer could be also attributed to physical clearance mechanism of sponge: 1) reducing cake layer through souring effect of sponge on the surface of the submerged membrane; 2) inhibiting the deposition of sludge flocs due to the turbulence of sponge which increased the solute back-transport from the membrane surface; 3) detaching the foulants from the membrane surface by shaking the membrane fibers under the impact of sponge against them (Lee et al., 2006; Yang et al., 2006).

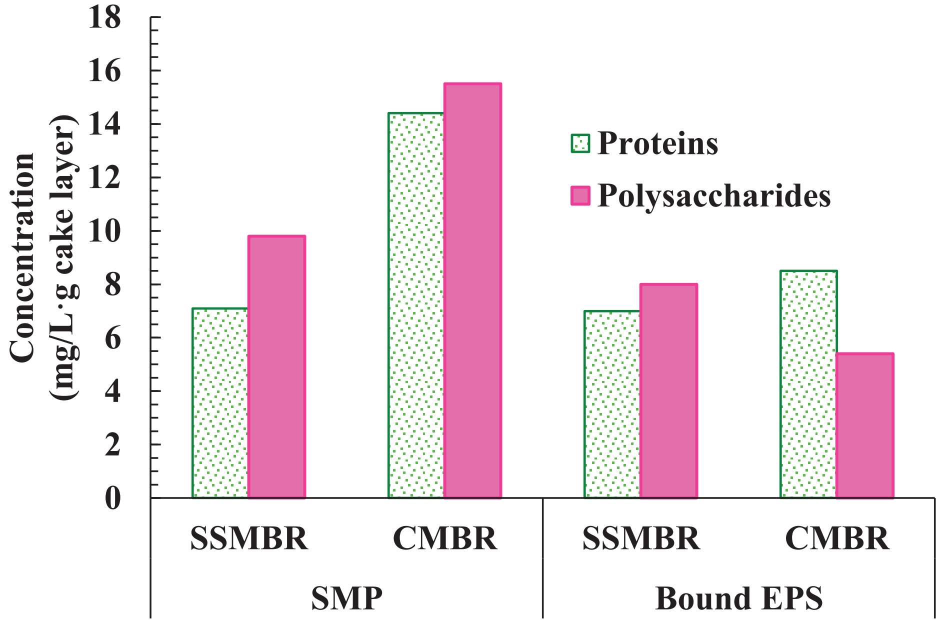


Fig. 4.8. Compositions of bound EPS and SMP in the cake layer in the SSMBR and the CMBR

C. Effects of mixed liquor properties on membrane fouling

Since motions of particles with small floc size was mainly regulated by Brownian diffusion at low shear stress instead of shear-induced diffusion and inertial force, these particles could be readily deposited on the membrane through permeation drag, resulting in severe pore blocking and cake formation on the membrane (Lim and Bai, 2003; Pan et al., 2010; Wang et al., 2011a). The CMBR contained smaller sludge flocs and induced higher TMP increment rate (Fig. 4.4), which illustrated that the presence of smaller sludge flocs contributed to higher R_C and R_P in the CMBR. As larger particles could not easily deposit on membrane surface due to higher shear induced diffusion and inertial lift force, the SSMBR demonstrated significantly lower membrane fouling propensity (Pan et al., 2010).

In addition, SMP in activated sludge appeared as a major contribution to initial membrane fouling. However, in later stage, membrane fouling development was mainly governed by bound EPS in activated sludge. It has been showed that SMP could increase fouling tendency due to the combined effects of pore clogging and adsorption on membrane walls and within membrane pores (Shen et al., 2012). Thus, higher SMP content of the CMBR cake layer led to higher R_P, which was well consistent with the results by Jamal Khan et al. (2012b). Besides, higher concentration of bound EPS in activated sludge could also increase both R_C and R_P in the CMBR. Ng et al. (2006) observed a thick fouling layer on the membrane consisting of microbial cells covered with EPS, which blocked membrane pores. Similar results were also found by Meng et al. (2006b) that the total amount of EPS had a significant positive correlation with the fouling resistance caused by pore blocking and cake formation.

D. A new fouling indicator

Previous studies have reported that PN/PS ratio in EPS or SMP had a significant impact on filtration resistance as well as fouling propensity (Lee et al., 2003; Tian et al., 2011a; Yao et al., 2011). In this chapter, as both SMP and EPS (especially SMP_C and EPS_C) were responsible for membrane fouling in the CMBR, a new fouling indicator ($(SMP_C/SMP_P)/(EPS_C/EPS_P)$) has been developed (Fig. 4.9). There was a strong correlation between fouling rate and fouling indicator ($(SMP_C/SMP_P)/(EPS_C/EPS_P) = 9.6727\ (dTMP/dt) - 8.3431$, $R^2 = 0.9783$). It suggested that fouling rate was remarkably influenced by compositions of SMP (SMP_C/SMP_P) and EPS (EPS_C/EPS_P). Moreover, the fouling rate was lower in the initial stage than that in the later stage as shown in Fig. 4.4, corresponding to relatively lower value of the fouling indicator ($(SMP_C/SMP_P)/(EPS_C/EPS_P)$) during first 7 days of operation. Generally, polysaccharides can penetrate into the cake layer and membrane pores, as well as lead to irreversible fouling due to their partially hydrophilic nature comparing to proteins (Kimura et al., 2004; Meng et al., 2009b; Guo et al., 2012a). Hence, SMP_C can be a greater contribution to irreversible fouling than EPS_C. When activated sludge has higher SMP_C concentration but lower EPS_C concentration, the value of $(SMP_C/SMP_P)/(EPS_C/EPS_P)$ will be higher, indicating more severe membrane fouling and higher fouling rate ($\Delta TMP/\Delta t$), and vice versa.

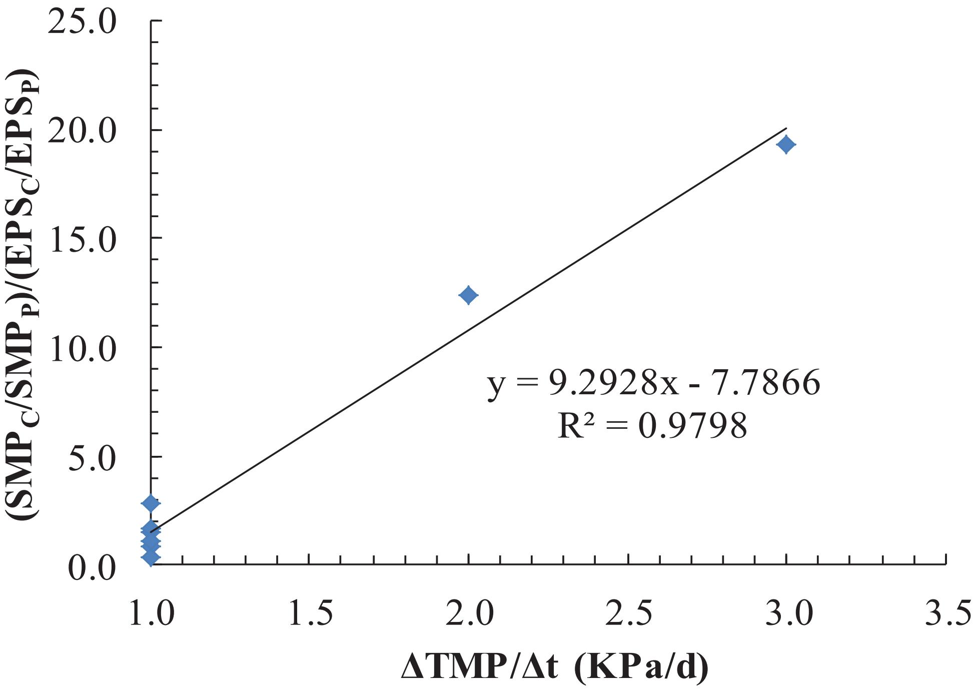


Fig. 4.9. Fouling indicator profile for the CMBR

4.4. Conclusions

An in-depth analysis of membrane fouling behaviour in the SSMBR and the CMBR for synthetic wastewater treatment is presented. The major findings are as follows:

1) In the CMBR, SMP in activated sludge were a major contributor for initial membrane fouling and presented higher concentration in membrane cake layer; Afterwards, membrane fouling was mainly governed by bound EPS in activated sludge, containing lower proteins but significantly higher polysaccharides;
2) Sponge addition could prevent cake formation on membrane surface and pore blocking inside membrane, thereby alleviating membrane fouling;
3) The SSMBR exhibited not only less growth of the biomass and filamentous bacteria, but also lower R_C and R_P due to lower bound EPS concentrations in activated sludge. Less membrane fouling in the SSMBR were also attributed to larger particle size, higher zeta potential and relative hydrophobicity of sludge flocs.

Chapter 5

Enhanced Performance of Submerged Membrane Bioreactor by Bioflocculant Addition

Chapter 5 Enhanced Performance of Submerged Membrane Bioreactor by Bioflocculant Addition

5.1. Introduction

5.1.1. Background

Membrane fouling is a long-lasting and inevitable issue along the development of membrane bioreactor (MBR), which increases the hydraulic resistance to the fluid flow, resulting in less permeability for constant pressure mode or transmembrane pressure (TMP) increment for constant flux mode (Hwang et al., 2012). So far, numerous studies have been devoted to the mechanism and causes of membrane fouling and control strategies. Among six principal fouling mechanisms, biofouling has attracted a significant concern as it is a major cause of fouling in MBRs. Biofouling occurs through deposition and accumulation of undesirable microorganisms and bacterial cells or flocs at membrane surface (Guo et al., 2012a; Meng et al., 2009a). It can lead to cake layer formation, which has been found to be the main contributor to total membrane resistance (Hwang et al., 2012; Meng et al., 2009a; Wang and Li, 2008). For a given MBR, biofouling and membrane filterability as well as cake layer formation are directly associated with sludge characteristics, such as mixed liquor suspended solid (MLSS) concentration, sludge viscosity, floc size, extracellular polymeric substances (EPS), soluble microbial products (SMP) and biopolymer clusters (BPC).

Previous studies have pointed out the advantages of flocculant addition in MBRs for fouling control through improving sludge characteristics (Section 2.5.2). Although these flocculants have their own merits for membrane fouling reduction, the development of a safe biodegradable natural flocculant is essential in order to have less impact on the environment and produce less ‘secondary pollutants’ through wastewater reclamation and reuse processes. Ngo and Guo (2009) developed a new green bioflocculant (GBF) which was modified from a natural starch-based cationic flocculant (HYDRA Ltd., Hungary). It was found that GBF could significantly reduce membrane fouling (TMP increment of 2.5 kPa after 70 days of operation) and energy consumption (less backwash frequency) of a conventional submerged MBR. Based on this research, Gemfloc® was patented by University of Technology, Sydney (UTS). However, membrane fouling behaviour related to sludge properties by Gemfloc® addition has not been well understood and explored yet.

5.1.2. Objectives

In this chapter, the effectiveness of Gemfloc® on fouling reduction in the lab-scale submerged MBR under long-term sustainable operation was evaluated. Organic and nutrient removal were determined in both of the conventional MBR (CMBR) and the MBR with Gemfloc® addition (MBR-G) to assess the impact of Gemfloc® on treatment performance. Membrane fouling behaviour was demonstrated by TMP development and fouling resistance. Furthermore, the fouling reduction through modifying the characteristics of mixed liquor as well as the cake layer were also investigated in terms of SMP, EPS, BPC, zeta potential, apparent viscosity, relative hydrophobicity (RH), and floc size.

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

Deng, L.J., Guo, W.S., Ngo, H.H., Zuthi, M.F.R., Zhang, J., Liang, S., Li, J., Wang, J., Zhang, X., 2015. Membrane fouling reduction and improvement of sludge characteristics by bioflocculant addition in submerged membrane bioreactor. Separation and purification technology 156, 450–458.

5.2. Materials and methods

5.2.1. Experimental setup and operating conditions of the MBR-G and the CMBR

The experiments were carried out in the MBR-G and the CMBR with the same working volume of 8 L (Fig. 5.1(a) and (b)). The MBR-G and the CMBR were set up and operated for synthetic wastewater treatment (Chapter 3, Section 3.2.1. and 3.2.2). The supplementation of Gemfloc® at a dosage of 1 g/day (net weight) was applied into the MBR-G. Initial mixed liquor suspended solids (MLSS) concentrations were 5.04 and 5.00 g/L for the MBR-G and the CMBR. Permeate through the submerged membrane module was withdrawn continuously and maintained at a constant flux of 12 L/m^2·h using a suction pump, corresponding to an hydraulic retention time (HRT) of 6.67 h. During the experiment, the membrane was only backwashed two times/day with duration of 2 minutes/time by pumping a fraction of permeate back through the membrane module at the flow rate of 36 L/m^2·h. More details about operating conditions for the MBR-G and the CMBR were summaried in Table 5.1.

5.2.2. Analysis methods

Chapter 3 (Section 3.2.3–3.2.7) has provided the details of sample analysis methods. The evaluation of fouling resistance was also performed.

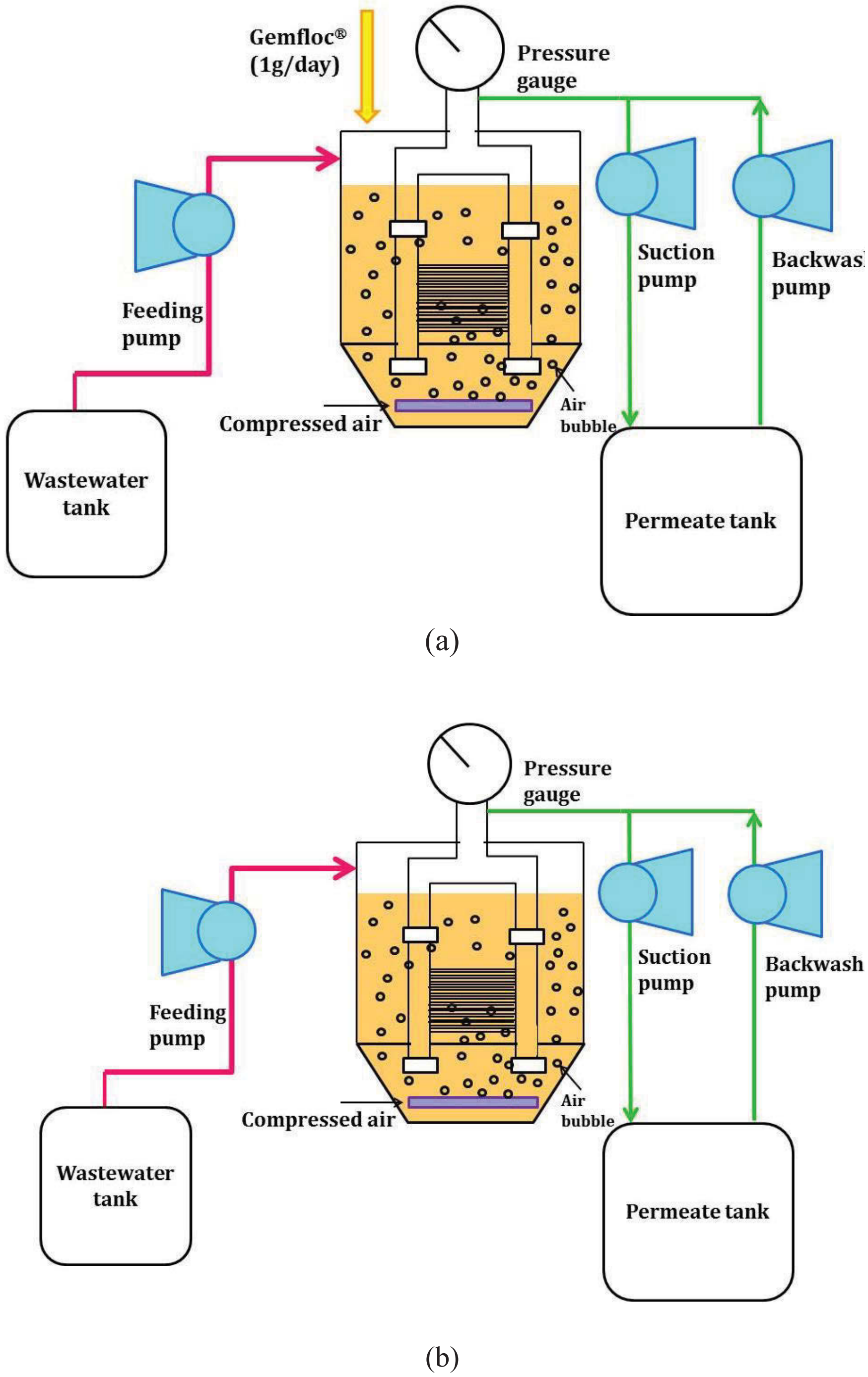


Fig. 5.1. Experimental set-up of the MBR-G (a) and the CMBR (b)

Table 5.1. System descriptions and operating conditions of the MBR-G and the CMBR

Membrane properties		
Membrane configuration: Hollow fiber membrane module		
Material: Polyvinylidene fluoride (PVDF)		
Pore size: 0.2 μm		
Surface area: 0.1 m^2		

Operating conditions		
Constant flux		12 $L/m^2 \cdot h$
Air flow rate		9~10 L/min
Working volume		8 L
SRT		No sludge withdrawal
Initial MLSS concentration		5.04 g/L for the MBR-G 5.00 g/L for the CMBR
Backwash	Times/day	2 times
	Duration per time	2 minutes
	Flux	36 $L/m^2 \cdot h$
Gemfloc®		1 g/day

Synthetic wastewater	
TOC	100–130 mg/L
COD	330–360 mg/L
NH_4-N	12–15 mg/L
PO_4-P	3.3–3.5 mg/L

5.3. Results and discussion

5.3.1. The performance of the MBR-G and the CMBR

A. Treatment performance of the MBR-G and the CMBR

Organic removal efficiencies for the MBR-G and the CMBR averaged 90% or more. More specifically, the MBR-G achieved 96.04 ± 6.33% and 95.36 ± 5.62% of DOC and COD removal, respectively, while lower removal efficiencies of DOC and COD were obtained for the CMBR (94.23 ± 7.69% and 92.76 ± 6.86%, respectively). It indicated that the addition of bioflocculants could improve organic matter removal.

In terms of nutrient removal, the MBR-G showed higher PO_4-P removal (90.12 ± 8.76%) than those of the CMBR (68.75 ± 6.98%) over the entire period of operation, suggesting that PO_4-P removal could be enhanced by enrichment of the activated sludge system with phosphorus accumulating organisms (PAOs) and biomass metabolism due to bioflocculant addition (Ngo and Guo, 2009; Nguyen et al., 2010). Similar NH_4-N removal was found for the MBR-G (87.41 ± 7.52%) and the CMBR (86.41 ± 9.63%), indicating that both MBRs exhibited high degree of biological nitrification as nitrifying bacteria was retained during membrane filtration process at infinite sludge retention time (Chiemchaisri andYamamoto, 2005). Moreover, the MBR-G achieved higher T-N removal of 75.56 ± 6.72%, while only 32.45 ± 8.35% of T-N was reduced in the CMBR during the operation period. It was attributed to the fact that larger flocs obtained in the MBR-G (see Section 5.3.3) promoted the denitrification process inside the biomass due to oxygen gradient existing within the flocs (Nguyen et al., 2010).

B. TMP development in the MBR-G and the CMBR

TMP profile depicting fouling propensity for two MBRs were shown in Fig. 5.2. It was observed that the TMP of the CMBR developed gradually with a sudden jump on day 50 and then reached maximum operation pressure of 35.0 kPa on day 64, implying the requirement of chemical cleaning. Regarding membrane fouling rate, an initial rate of 0.160 kPa/d before 50 days followed by a higher rate of 1.679 kPa/d were obtained. On the other hand, the TMP of the MBR-G only increased from 3.5 to 9.5 kPa within the 75-day filtration period, indicating a fairly low fouling rate of 0.067 kPa/d. The results suggested that Gemfloc® could effectively alleviate membrane fouling in the MBR-G for long-term operation as well as improve filterability. As both MBRs were operated under similar feed characteristics, membrane materials, module configurations and operational conditions, the differences in fouling propensity between the CMBR and the MBR-G were mainly ascribed to the characteristics of mixed liquor and cake layer, which are further discussed in Section 5.3.5.

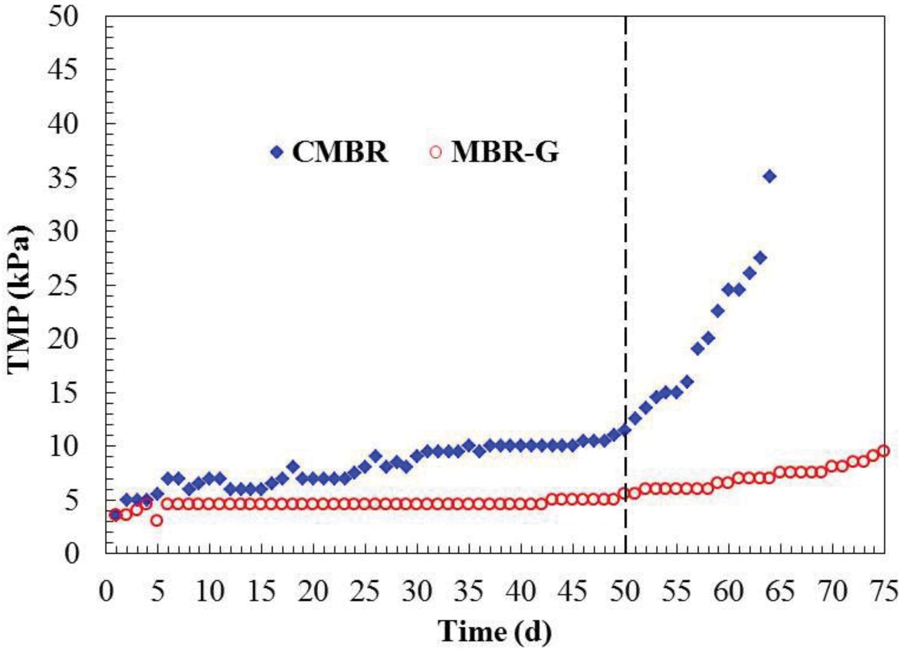


Fig. 5.2. TMP profile for the MBR-G and the CMBR

5.3.2. Mixed liquor suspended solids (MLSS) concentration, mixed liquor volatile suspended solids (MLVSS) and apparent viscosity

In this chapter, the concentrations of initial biomass were 5.00 and 5.04 g/L in the CMBR and the MBR-G, respectively. A consistently increasing trend of MLSS and MLVSS concentrations was observed in the CMBR and the MBR-G without sludge waste during 64 days and 75 days of operation, respectively. At the end of the experimental period, MLSS concentrations increased to 15.22 g/L and 15.12 g/L in the CMBR and the MBR-G, respectively. Moreover, lower MLVSS concentration was obtained for the CMBR than that for the MBR-G, with the values of 12.94 g/L and 13.08 g/L, respectively. As Gemfloc® provided the carbon source for microorganisms, it could improve microbial activity. More activate microorganisms would therefore be accumulated in activated sludge. Consequently, increased MLVSS concentrations were obtained in the MBR-G (Fan et al., 2015; Long et al., 2015). The biomass growth rate in terms of ΔMLSS/Δt was 0.16 g/L·d in the CMBR, which was higher than the rate in the MBR-G (0.13 g/L·d). The CMBR showed higher sludge viscosity (5.5 ± 0.6 mPa·s) than that in the MBR-G (5.4 ± 0.5 mPa·s), pointing out that sludge viscosity was positively correlated with MLSS concentration in both MBRs.

5.3.3. Particle size distribution, zeta potential and relative hydrophobicity (RH)

Results of particle size distribution in this chapter showed different sizes of sludge flocs in two MBRs. The size distribution of biomass particles in the MBR-G was larger (from 80 to 200 μm), whereas the CMBR had smaller sizes of biomass particles, ranging from 20 to 120 μm. These results were confirmed by microscopic images of sludge flocs in mixed liquor in both MBRs at different days of operation (Fig. 5.3). It was observed that the CMBR contained less number of smaller and compacted sludge flocs, while in the MBR-G, the flocs were characterized by more number of larger and looser flocs. The results elucidated that Gemfloc addition could promote the aggregation of sludge flocs in the MBR-G. It also indicated that sludge floc size was related to the floc structure. Small sludge flocs in the CMBR containing dispersed bacteria and small colonies presented higher density, thus increasing the compactness of sludge flocs. On the contrary, the presence of more biopolymer bridging in the larger flocs in the MBR-G decreased their density, which led to the formation of uncompressed sludge flocs (Lin et al., 2009; Massé et al., 2006). The enhanced floc agglomeration was obtained due to Gemfloc® addition, which could be explained by the surface property (i.e. RH) and the surface charge (zeta potential) of sludge flocs (Meng et al., 2006b). Generally, hydrophobicity and surface charge affect flocculation ability of the sludge flocs through hydrophobic interaction and electrostatic repulsion, respectively (Liao et al., 2001; Mikkelsen and Keiding, 2002). In this chapter, higher zeta potential values (–0.86 ± 1.07 mV) of activated sludge were measured in the MBR-G than those in the CMBR (–11.41 ± 5.06 mV), which demonstrated the negative surface charge of the microbial flocs was reduced or neutralized by Gemfloc®. Subsequently, the flocs could attach to each other and promote the production of larger flocs through the charge neutralization mechanism (Lee et al., 2007). Ji et al. (2014) also reported similar results that the charge neutrality was responsible for enhancing flocculation ability of sludge flocs when adding PAM-MGMS into the MBR. In addition, it has been found that the formation of more number of larger permeable sludge flocs is associated with higher RH (Arabi and Nakhla, 2009b). Thus, the increased sludge flocs in the MBR-G demonstrated higher RH (72.19 ± 6.53%) than that for the CMBR (35.64 ± 5.34%), proving that the formation of larger flocs could reduce the retention of water among sludge flocs, resulting in higher hydrophobicity. For the hydrophobic membranes, the sludge flocs with higher hydrophobicity would be more easily deposited on the membranes through the hydrophobic interactions between the sludge and the hydrophobic membrane following the "like dissolved like" rule (Ji and Zhou, 2006). Thus, the hydrophobic substances or foulants could not foul the hydrophilic membrane. In this chapter, higher RH decreased the interaction between the hydrophobic flocs and hydrophilic membrane, thereby improving membrane performance. Overall, increased zeta potential and enhanced hydrophobicity of sludge flocs contributed to better flocculation ability of biomass particles in the MBR-G.

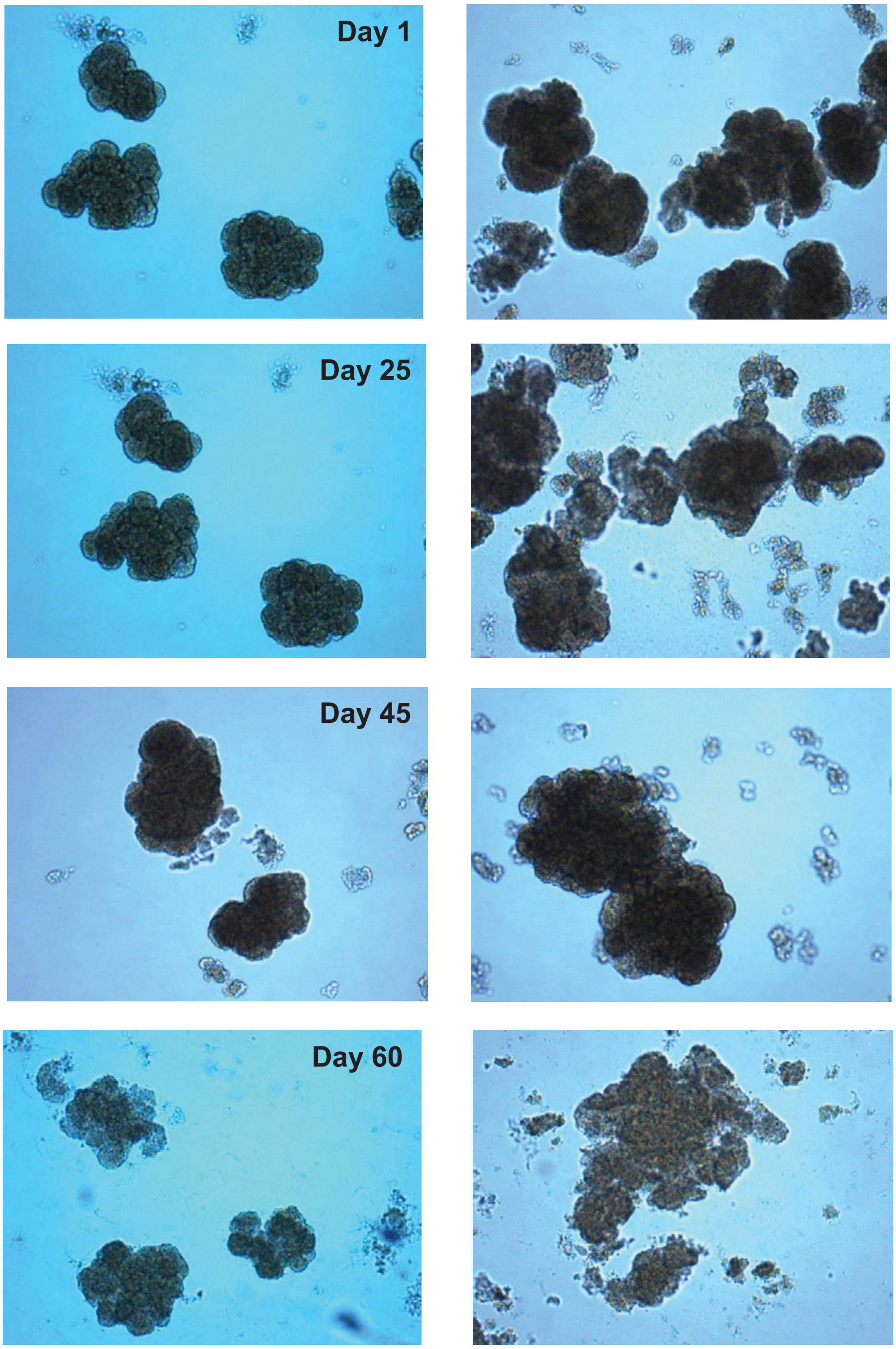


Fig. 5.3. Microscopic images of the sludge flocs in mixed liquor in the MBR-G and the CMBR (100 ×)

5.3.4. EPS and SMP in mixed liquor

As to EPS and SMP, the main compositions were polysaccharides and proteins, with certain amounts of humic acids, nucleic acids, lipids, uronic acids (Lin et al., 2014a). Figs. 5.4 and 5.5 present the profiles of SMP and EPS of the mixed liquor in both MBRs as well as the average values of SMP_P, SMP_C, EPS_P/EPS_C for different operating period. In the CMBR, SMP levels were stable before the fouling period (from day 0 to day 50), with the value of 22.83 ± 9.31 mg/L. The variations of SMP_C and SMP_P were marginal (17.80 ± 5.03 and 5.40 ± 2.11 mg/L, respectively). After 50 days (fouling period with rapid TMP increase), a significant rise in SMP was observed (up to 51.22 ± 14.26 mg/L), while SMP_C and SMP_P increased to 38.45 ± 7.75 and 12.23 ± 2.51 mg/L, respectively. On the other hand, EPS remained steady at 23.02 ± 7.22 mg/L during 60 days operation. During the operation, hydrolysis of bound EPS into small fractions and their subsequent dissolution into bulk solution could lead to more SMP release and EPS reduction (Wu and Lee, 2011). Thus, more serious fouling phenomenon was found in the CMBR. When compared to the CMBR, the MBR-G possessed less SMP (12.70 ± 4.07 mg/L) and higher total bound EPS in activated sludge (43.9 ± 16.2 mg/L), because SMP was adsorbed and/or entrapped onto the flocculated microbial flocs, thereby increasing EPS contents (Dizge et al., 2011a).

Furthermore, floc strength or stability could be enhanced by EPS at high levels due to polymer entanglement, leading to better flocculation ability of sludge flocs and generation of larger flocs (Mikkelsen and Keiding, 2002). The polymeric entanglement can be manifested by physical interactions (e.g. adsorption or Van der Waals forces) or the gel-networks induced by the bridge between EPS and divalent cations. The latter one involves the bridging of EPS containing negatively charged functional groups through divalent cations. It promoted aggregation and stabilization of matrix within biopolymers and microbes, thus facilitating bioflocculation (Mikkelsen and Keiding, 2002; More et al., 2012). In addition, it has been reported that hydrophobicity and surface charge of sludge flocs depend on the ratio of proteins to polysaccharides in EPS (EPS_P/EPS_C) (Lee et al., 2003). As shown in Fig. 5.5, the MBR-G exhibited higher EPS_P/EPS_C ratio (0.55–0.85) than that in the CMBR (0.32–0.52) during the operation period. Normally, amino acids with hydrophobic side groups can be resulted from higher EPS_P, thereby exhibiting higher RH of activated sludge. In addition, declining EPS_C also further reduced hydrophilic nature of sludge. Besides, higher zeta potential of sludge flocs obtained in the MBR-G also indicated that higher EPS_P reduced the surface charge of sludge flocs due to the fact that EPS_P containing amino groups carried positive charge, which neutralized sludge flocs having negative surface charge (Liao et al., 2001; Lee et al., 2003). Overall, the improved aggregation ability of sludge flocs in the MBR-G was ascribed to increased EPS concentrations as well as EPS_P/EPS_C ratio. Hence, although activated sludge presented higher EPS concentrations, the MBR-G exhibited less membrane fouling propensity with lower fouling rate (Deng et al., 2014; Pan et al., 2010).

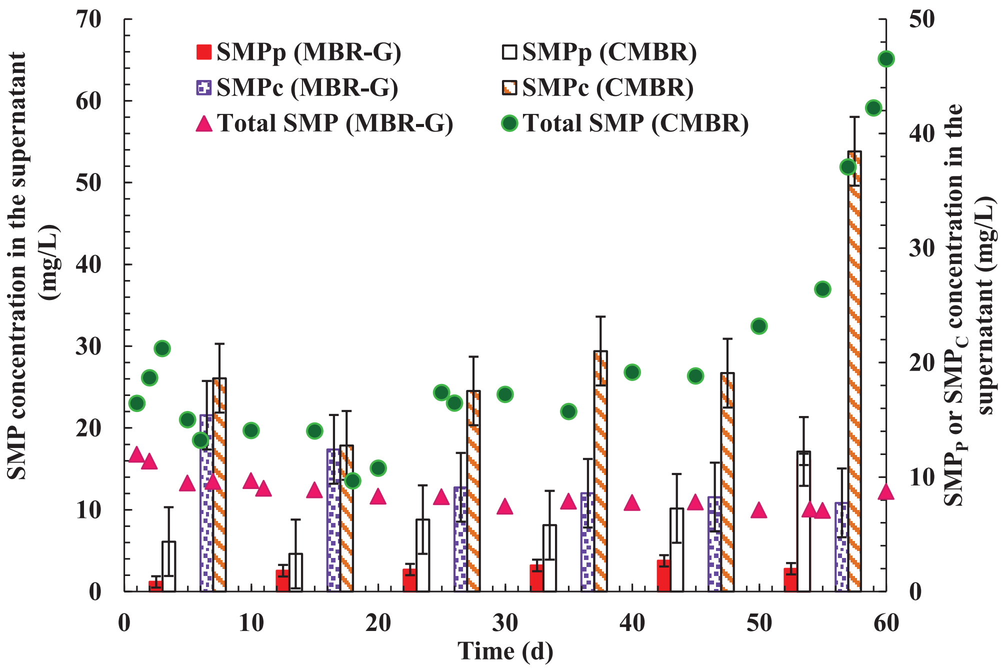


Fig. 5.4. Variations of SMP (including SMP_P and SMP_C) concentrations in the supernatant in the MBR-G and the CMBR

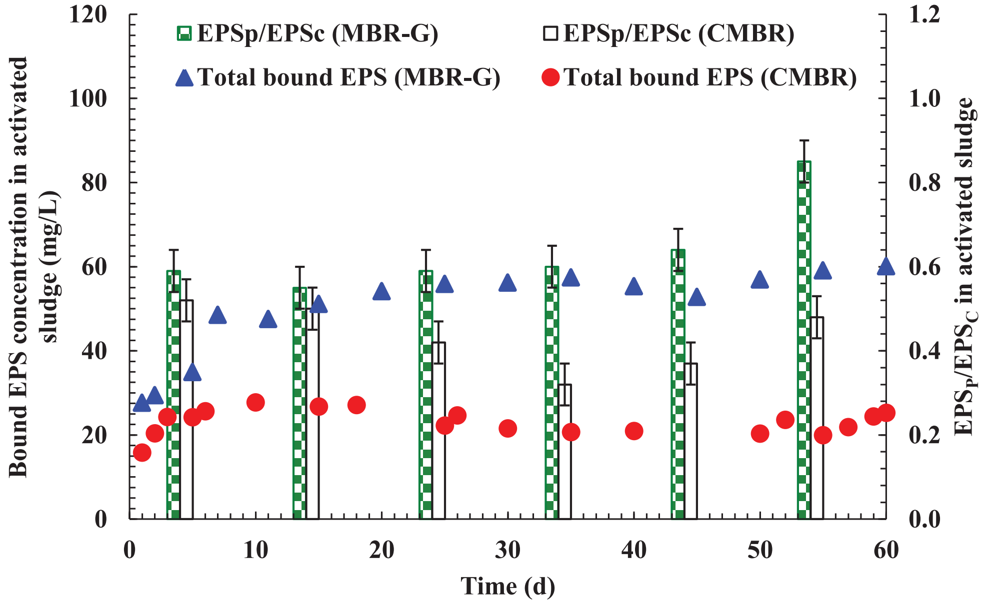


Fig. 5.5. Variations of EPS concentrations and EPS_P/EPS_C ratio in activated sludge in the MBR-G and the CMBR

5.3.5. Membrane fouling behaviour

A. Fouling resistance distribution

Table 5.2 presents the fouling resistance for both MBRs after the experiments. It was observed that the total fouling resistances (R_T) of the CMBR and the MBR-G were 4.05×10^{12} m^{-1} and 3.54×10^{12} m^{-1}, respectively. The CMBR had about 2-time higher pore blocking resistance (R_P of 1.5×10^{11} m^{-1}) than the MBR-G. Cake layer resistance (R_C) of the CMBR (3.00×10^{12} m^{-1}) accounted for 74.1% of R_T, whereas R_C of the MBR-G was 1.94×10^{12} m^{-1}, corresponding to 54.8 % of R_T. These results indicated that cake layer fouling played a significant role in membrane fouling of both MBRs. Additionally, Gemfloc® addition effectively alleviated membrane fouling due to the prevention of cake layer formation and pore blocking. Results reported by Hwang et al. (2007) and Jamal Khan et al. (2012a) with addition of cationic polymer (MPE or MPE50) into MBR systems were consistent with the findings of this chapter. The reduction of R_C made a greater contribution to R_T decrease due to addition of cationic polymer.

Table 5.2. Fouling resistance distribution in the MBR-G and the CMBR

Resistance distribution	MBR-G		CMBR	
	m^{-1}	% of R_T [a]	m^{-1}	% of R_T [a]
Total	3.54×10^{12}		4.05×10^{12}	
Cake layer	1.94×10^{12}	54.8	3.00×10^{12}	74.1
Pore blocking	0.70×10^{11}	19.8	1.50×10^{11}	3.7
Clean membrane	9.00×10^{11}	25.4	9.00×10^{11}	22.2

[a] R_T = total fouling resistance.

B. Cake layer on membrane surface

It has been reported that MLSS affected membrane fouling in two different ways. One is that microbial products and other organic substances (e.g. colloids, solutes) in mixed liquor at higher MLSS concentration could induce membrane pore blocking and/or formation of a fouling layer (gel layer) due to their deposition on membrane surface. Macromolecules and sludge particles could be retained by the gel layer, resulting in more compact layer formation (Wang et al., 2008). The other one involves that higher sludge viscosity at higher MLSS concentration increased net force towards the membrane surface due to less back transport effects on sludge flocs, thus promoting their deposition on the membrane. Since no sludge was withdrawn from MBRs in this chapter, higher MLSS concentration and sludge viscosity induced the deposition of sludge flocs and higher R_C in the CMBR causing by the sticky cake layer also formed on membrane surface (Kornboonraksa and Lee, 2009; Meng et al., 2007a; Watanabe and Kimura, 2006). As the cake layer represented the significant fraction of the total fouling resistance, the compositions of EPS, SMP and BPC in cake layer were further analysed and are shown in Fig. 5.6. EPS_C concentration of the cake layer presented minor difference for both MBRs. However, higher concentration of EPS_P (2.83 mg/g cake layer) was observed in the CMBR than that in the MBR-G (1.42 mg/g cake layer). The cake layer in the CMBR exhibited higher SMP_C and SMP_P concentrations of 0.63 and 0.58 mg/g cake layer, respectively, comparing to lower values for the MBR-G (0.41 and 0.12 mg/g cake layer, respectively). In addition, the CMBR possessed higher concentrations of proteins in BPC (BPC_P) and polysaccharides in BPC (BPC_C) (by 16.94 and 3.13 times, respectively) comparing with the MBR-G. These results implied that EPS_P, SMP (SMP_C and SMP_P) and BPC (BPC_C and BPC_P) on membrane surface contributed to higher R_C in the CMBR. At higher TMP, the drag force from the permeate pump could promote cake layer formation by aggravating the deposition of EPS_P, SMP_C and SMP_P on membrane surface (Jin et al., 2013). More sludge cake on membrane surface could cause endogenous decay or cell lysis at the bottom layer, which led to release of more EPS_P and SMP (Hwang et al., 2008; Yigit et al., 2008). Furthermore, the continuous accumulation of SMP in the sludge layer generated more BPC on membrane surface, which accelerated the formation and attachment of a sticky and impermeable sludge cake on the membrane surface (Sun et al., 2008; Wang and Li, 2008). On contrary, in the MBR-G, Gemfloc® addition not only reduced deposition of EPS_P by lowering TMP development, but also adsorbed SMP_P and SMP_C, resulting in less retention of BPC_P and BPC_C in the sludge cake.

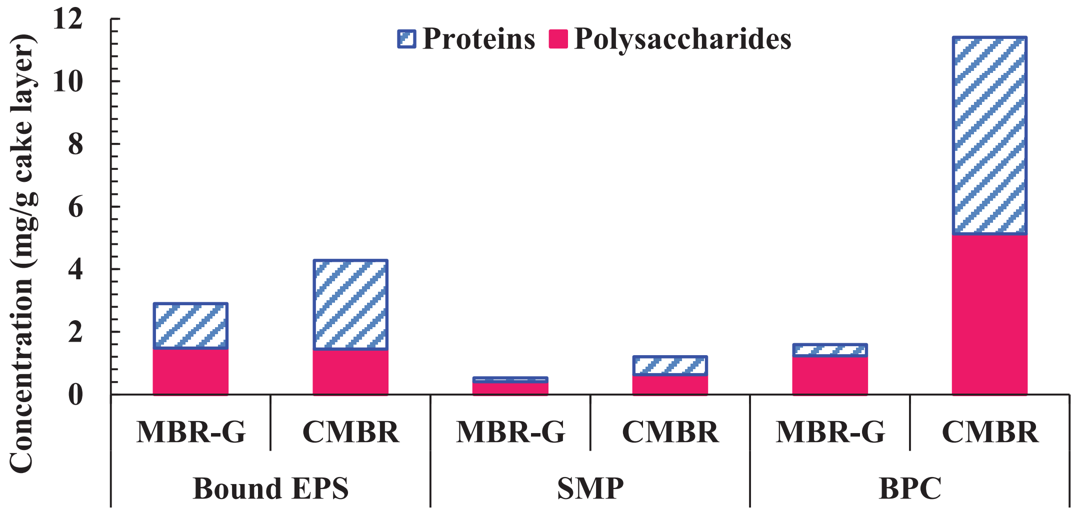


Fig. 5.6. Compositions of bound EPS, SMP and BPC in cake layer in the MBR-G and the CMBR

C. Effects of mixed liquor properties on membrane fouling

It was suggested that smaller biomass particles depended on back transport caused by Brownian diffusion rather than the shear-induced diffusion and inertial lift. Brownian diffusion, which is referred to as the driving force induced by the concentration gradient between the membrane surface and the side of bulk solution, would increase with decreasing particle size of sludge floc. The shear-induced diffusion and inertial lift decreased when sludge floc size was smaller (Bae and Tak, 2005; Pan et al., 2010). Since the CMBR demonstrated faster TMP development (Fig. 5.2) and smaller sludge flocs (Fig. 5.3), the obtained higher R_C and R_P values indicated that the presence of smaller flocs could block membrane pore as well as reduce the porosity of cake layer (Lim and Bai, 2003; Wu and Huang, 2008). On contrary, lower membrane fouling propensity of the MBR-G was due to formation of more porous and permeable cake layer caused by the deposition of larger and looser sludge flocs on membrane surface (Dizge et al., 2011a). Previous studies have suggested that SMP induces internal fouling and decreases filterability since SMP can not only block membrane pores, but also block the pores and spaces between particles in the cake layer (Chang et al., 2001; Lee et al., 2007; Shen et al., 2012; Wu and Huang, 2009). Moreover, SMP_C possessing partially hydrophilic nature could cause irreversible fouling by forming a thin gel layer on membrane compared to SMP_P (Guo et al., 2012a; Kimura et al., 2004; Meng et al., 2009b; Viero et al., 2007). In addition, high portion of SMP_P content in SMP could also cause the cake layer formation due to their stickiness (Gao et al., 2013a). Therefore, SMP played an important role in membrane fouling as well as contributed to the increased R_C and R_P in the CMBR.

5.3.6. Modeling of membrane fouling in the MBR-G and the CMBR

The basic resistance-in-series model can demonstrate the relationship between TMP and flux, identify the main contributor to membrane fouling, and estimate permeate flux during different fouling stages (Shirazi et al., 2010). In this chapter, a modified resistance-in-series model was employed for quantitatively estimating membrane fouling behaviour through the TMP evolution and quantifying the increase of filtration resistance in both MBRs. The model is based on the basic resistance-in-series model as shown in Section 3.2.5 (Chapter 3), which integrates membrane fouling behaviour with sludge properties. It can provide a rational and fundamental framework for understanding membrane fouling process and predicting impacts of sludge characteristics on fouling behaviour. As suggested in Section 5.3.5, SMP and MLSS concentrations were important aspects influencing pore blocking and cake layer formation on membrane surface. Therefore, R_P and R_C can be simulated according to the profiles of SMP and MLSS, respectively.

It can be seen from Fig. 5.7 that the modeled SMP results are reasonably matched up with the experimental results. The simulated data shows a dramatic accumulation of SMP after 50 days in the CMBR, and the CMBR possessed more SMP than the MBR-G during the entire operation period. Regarding pore fouling, Zuthi (2014) has proposed modeling related to pore fouling of the membrane (R_P) in a submerged MBR with membrane pore size of 0.1 μm (Eq. (5.3)). However, in this study, the profile of R_P from trial simulation was not satisfactory even with the significant change of the pore fouling factor n_p and initial porosity. Further reduction or increase of the trial values of the unknowns still could not match the experimental value of R_P for both MBRs. Thus, according to the different membrane pore size (0.2 μm) used in this study, an empirical factor ($(d_{p,used}/0.1)^4$) was assigned to Eq. (5.3). The modified equation of R_P is given in Eq. (5.4). Table 5.3 gives the values of coefficients and constants used for modeling. Plotting of R_P versus the operation time (Fig. 5.8) revealed that the simulation results were in good agreement with the experimental data ($R^2 > 0.9$). R_P in the CMBR showed a gradual increase before 50 days and a significant rise afterward, while the MBR-G presented much lower R_P even after 50 days. These results elucidated that the proposed models can explain the important influence of increase in SMP concentration on pore blocking.

MBR-G: $C_{SMP}(t) = 0.0047t^2 – 0.34t + 16.33$ (5.1)

CMBR: $C_{SMP}(t) = 0.030t^2 – 1.25t + 28.83$ (5.2)

$R_P = \exp(n_p t)\ 8h_m/\ f r_p^2$ (5.3)

$R_P = \exp(n_p t)\ 8h_m/(d_{p,used}/0.1)^4\ f r_p^2$ (5.4)

where C_{SMP} is the time-dependent concentration of soluble particles entering the pores; t is the filtration time; the pore fouling factor n_p is to explain the typically observed exponential rise of TMP due to the pore fouling resistance especially at the final stage of operation of an MBR system; h_m is the membrane's effective thickness; f is the membrane's porosity (variable) and r_p is the membrane pore radius (variable); $d_{p,used}$ is the initial pore diameter of the membrane in μm (0.1μm is the reference membrane pore size). f and r_p are the time-dependent parameters, which are associated with the dynamic change of SMP concentration in the mixed liquor of the bioreactor. These two parameters (f and r_p) can be calculated according to the Eqs. (5.5) and (5.6) proposed by Busch et al. (2007) and Giraldo and LeChevallier (2006).

$\rho_p\ df/dt = –4\eta_f\ J(t)\ C_{SMP}(t)\ m_{d,o}/((m_{d,o})^2-(m_{d,i})^2)$ (5.5)

$dr_p/dt = –\ \alpha_p\ C_{SMP}(t)\ J(t)$ (5.6)

where ρ_P is the density of biomass; the constant related to the porosity (f) is the effective initial porosity of the membrane (60% in this chapter); η_f is the average fraction of soluble particles that accumulate in the pores taken as 0.2 from Busch et al. (2007); $m_{d,o}$ and $m_{d,i}$ are the outer and inner membrane diameter (0.49×10^{-3} m and 0.35×10^{-3} m, respectively, in this chapter); α_p is pore size reduction coefficient (0.000943).

Table 5.3. Values of coefficients and constants used to simulate the model

Coefficient/constant[a]	MBR-G	CMBR
n_p	0.003	0.003
constant for f	0.6	0.6
constant for r_p	1.00×10^{-7}	1.07×10^{-7}
k	0.3	0.1
n_c	0.030	0.042
constant for R_c	75	1000
$(d_{p,used}/0.1)^4$	16	16

[a] f = the membrane's porosity (variable), k = the factor representing the detachment of the cake layer from the membrane surface, n_p = pore fouling factor to explain the typically observed exponential rise of TMP due to the pore fouling resistance especially at the final stage of operation of an MBR system, n_c = cake fouling factor to explain the typically observed exponential rise of TMP due to the cake layer resistance especially at the final stage of operation of an MBR system, r_p = the membrane pore radius (variable).

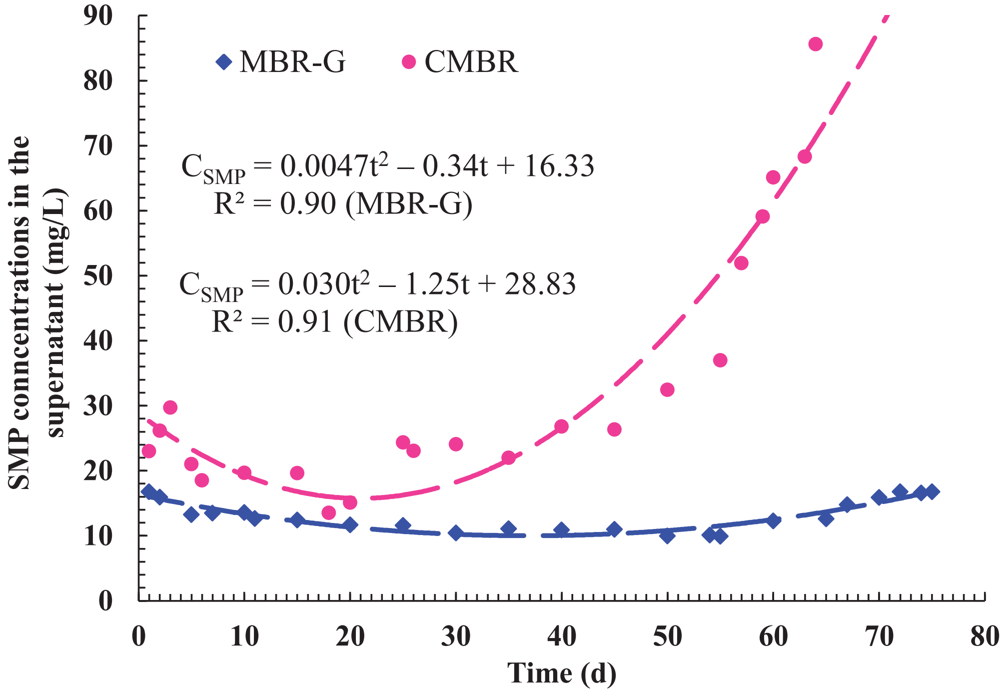


Fig. 5.7. SMP profile of the supernatant in the MBR-G and the CMBR

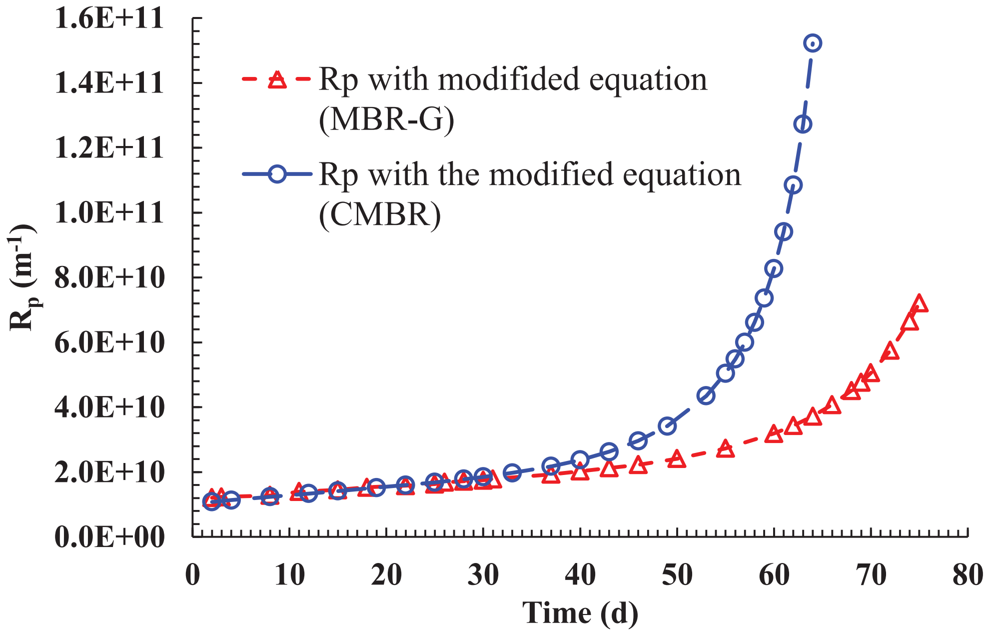


Fig. 5.8. R_P profile drawn for the MBR-G and the CMBR with the modified equation (Eq. (5.4))

Fig. 5.9 displays experimental data vs. simulated results for the variation of MLSS concentration in activated sludge and shows good model fit. It could be referred that sludge concentration increased with filtration time in both MBRs, whereas higher MLSS concentration was obtained in the CMBR. After obtaining the Eqs. (5.7) and (5.8) from Fig. 5.9, R_C could be calculated using the Eqs. (5.9) and (5.10) which were proposed by Zuthi [50] to model the rise of TMP in a submerged MBR.

MBR-G: $C_c(t) = 0.11t + 6.63$ (5.7)

CMBR: $C_c(t) = -0.003t^2 + 0.33t + 4.73$ (5.8)

$R_c = \alpha_c\, h_c(t)\, \rho_c \exp(n_c t)$ (5.9)

$\rho_c\, dh_c/dt = J \cdot (1 - k)\, C_c(t)$ (5.10)

where C_c is concentration of potential cake forming particles in the bulk liquid (e.g. MLSS) which typically varies over time in the MBR-G and the CMBR (Eqs. (5.7) and (5.8), respectively); α_c is specific resistance of the compressible cake layer; h_c is variable depth of the cake layer expressed as a first order differential function in time, which relies on the attachment and detachment of cake layer; ρ_c is density of the cake layer; the factor n_c is to explain the typically observed exponential rise of TMP due to the cake layer resistance especially at the final stage of operation of an MBR system (the MBR-G and the CMBR in this chapter); k is factor representing the detachment of the cake layer from the membrane surface.

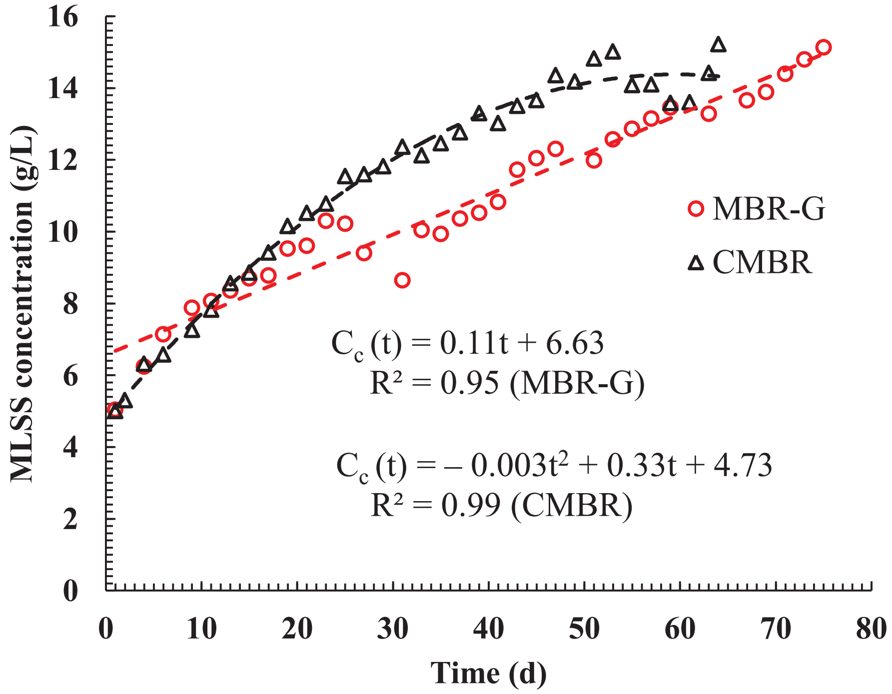


Fig. 5.9. Variation of MLSS concentration in activated sludge in the MBR-G and the CMBR

Based on the calculated values of R_P and R_C as well as Eq. (3.3), the fouling propensity in both MBRs in terms of TMP development can be modeled. Fig. 5.10 shows actual TMP development during experiments and the simulated TMP versus filtration time. It can be observed that the simulated TMP fit well to the experimental TMP profile for both MBRs during the operation period. In addition, the sudden TMP increase recorded in the CMBR was also depicted and predicted by the models. Overall, the results implied that the proposed models in this chapter are capable of predicting the contributions of SMP and MLSS to membrane fouling as well as the TMP development during the operation. In real MBR applications, SMP and MLSS are common parameters and can be easily measured. Thus, by utilizing the profiles of SMP and MLSS, this model can provide an economic way to monitor membrane fouling behavior through simulation of R_P, R_C and TMP.

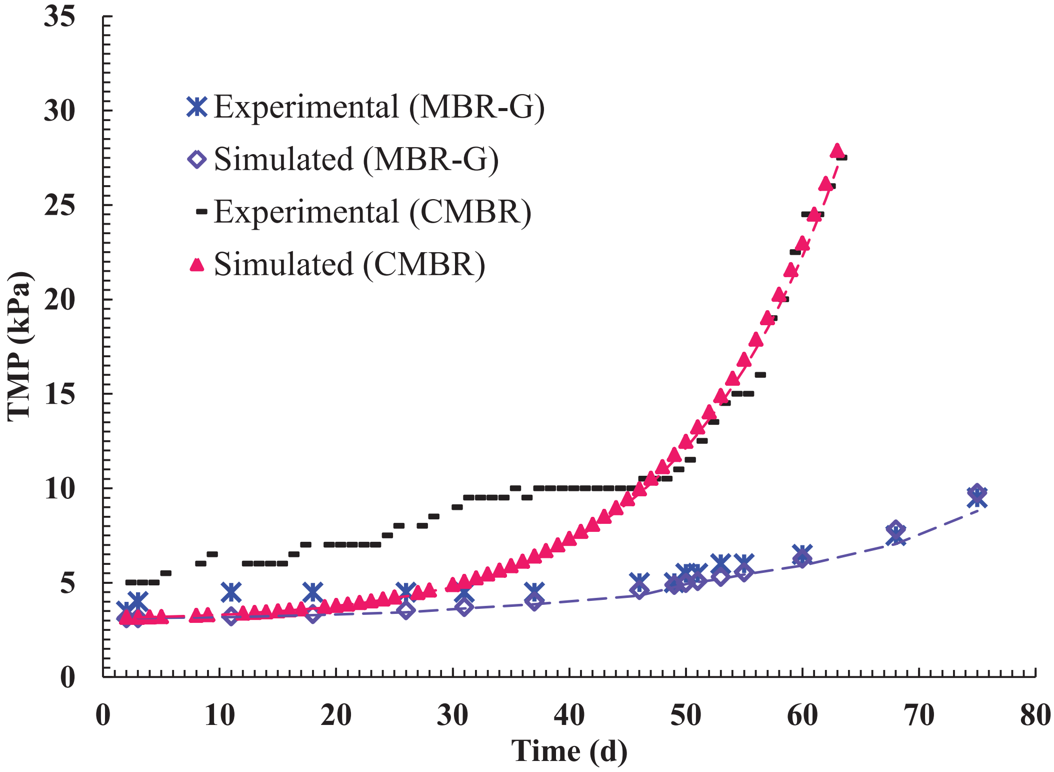


Fig. 5.10. TMP profile after model calibration for the MBR-G and the CMBR

5.4. Conclusions

The effects of Gemfloc® addition on membrane fouling reduction in a SMBR were investigated in this chapter. Specific conclusions could be drawn as follows:

1) Gemfloc® could contribute to membrane fouling alleviation in terms of reduced cake formation, retarded pore blocking and improved membrane filterability;
2) Compared to the CMBR, activated sludge in the MBR-G contained less SMP but more EPS with higher EPS_P/EPS_C ratio, which led to higher RH and zeta potential of activated sludge. The addition of Gemfloc® enhanced aggregation of sludge flocs and increased floc size;
3) Compared to the CMBR, the MBR-G exhibited lower membrane fouling potential as well as lower R_P and R_C due to lower sludge growth rate and viscosity, less deposition of EPS_P and SMP on membrane surface, and less retention of BPC in the cake layer;
4) The proposed mathematical model could successfully simulate R_P, R_C and TMP development based on the profiles of SMP and MLSS during operation. Thus, this

model can help to quantitatively understand the relationship between sludge properties and membrane fouling behaviour in SMBR.

Chapter 6

Effects of Hydraulic Retention Time and Bioflocculant Addition on the Performance of a Sponge-Submerged Membrane Bioreactor

Chapter 6 Effects of Hydraulic Retention Time and Bioflocculant Addition on the Performance of a Sponge-Submerged Membrane Bioreactor

6.1. Introduction

6.1.1. Background

As discussed in Chapter 2 (Section 2.4.2), one of the key operating parameters having a significant impact on membrane fouling (especially biofouling) during membrane bioreactor (MBR) process is hydraulic retention time (HRT), which can alter mixed liquor properties (e.g. extracellular polymeric substances (EPS), soluble microbial substances (SMP) and sludge stability) (Guo et al., 2012a). Current studies have evaluated the performance of conventional MBR (CMBR) when treating various wastewaters at different HRTs. Gao et al. (2012) operated a lab-scale membrane-based process which consisted of an anoxic tank and an aerobic MBR for digested sewage treatment with decreasing HRTs from 8 to 2.5 h. The results suggested that shorter HRT improved NH_4-N and T-N removal, but aggravated membrane fouling, increased filtration resistance and fouling rate. The effects of different HRTs (12, 8 and 4 h) and sludge retention times (sludge retention times (SRTs), 30, 15 and 4 d) on the performance of a submerged MBR were investigated by Aida Isma et al. (2015) for synthetic wastewater treatment. They found that longer HRT prompted PO_4-P removal. At the HRT of 12 h, the MBR with longest SRT of 30 d achieved better removal efficiencies of COD, NH_4-N and PO_4-P, while exhibited less membrane fouling with the slowest transmembrane pressure (TMP) increment. Babatsouli et al. (2014) demonstrated an MBR pilot plant with a short SRT of 20 d for industrial wastewater treatment. At shorter HRT of 19 h, not only lower denitrification and phosphate removal but also higher fouling rate were observed. Although these studies showed HRT plays an important role in MBR performance and membrane fouling, detailed characterisation of sludge and cake layer at different HRTs has not been performed. Another study conducted by Qu et al. (2013) examined the effects of long HRTs (i.e. 0.9 ± 0.1, 1.1 ± 0.1, and 1.7 ± 0.2 d) on the performance of a thermophilic submerged aerobic MBR (TSAMBR) for the treatment of thermomechanical pulping pressate. The results indicated that shorter HRT increased EPS contents in bulk sludge and changed cake layer structure through analysing cake layer morphology, but slightly altered EPS contents of cake sludge. However, the variations of activated sludge and cake layer properties during operation at different HRTs were not identified.

The previous two chapters (Chapters 4 and 5) have convinced that the addition of sponge or bioflocculants (Gemfloc®) in MBR is a promising technology for membrane fouling mitigation. Thanh et al. (2013) also pointed out that lower HRT (higher filtration flux) in sponge MBR led to more severe membrane fouling through monitoring the TMP development. Fouling rate was adversely affected by HRT, following the power equation ($dTMP/dt = 4.2474\ HRT^{-2.225}$). However, systematic analysis and assessment of sludge characteristics and their impacts on fouling in a sponge-submerged MBR (SSMBR) at various HRTs have not been demonstrated yet.

6.1.2. Objectives

This chapter mainly focused on the effects of different HRTs on long-term SSMBR performance (i.e. membrane fouling behaviour, properties of mixed liquor and cake layer) for synthetic wastewater treatment. After determining the optimum HRT, the addition of Gemfloc® for further fouling minimization in a long-term basis was then investigated. For all sponge-submerged MBR (SSMBR) systems, fouling behaviour was evaluated through TMP development, membrane fouling resistance, and characterisation of activated sludge and cake layer including floc size, surface charge, relative hydrophobicity (RH), apparent viscosity, EPS, SMP, and biopolymer clusters (BPC).

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

Deng, L., Guo, W., Ngo, H.H., Du, B., Wei, Q., Tran, N.H., Nguyen, N.C., Chen, S.S., Li, J., 2016. Effects of hydraulic retention time and bioflocculant addition on membrane fouling in a sponge-submerged membrane bioreactor. Bioresource Technology 210, 11−17.

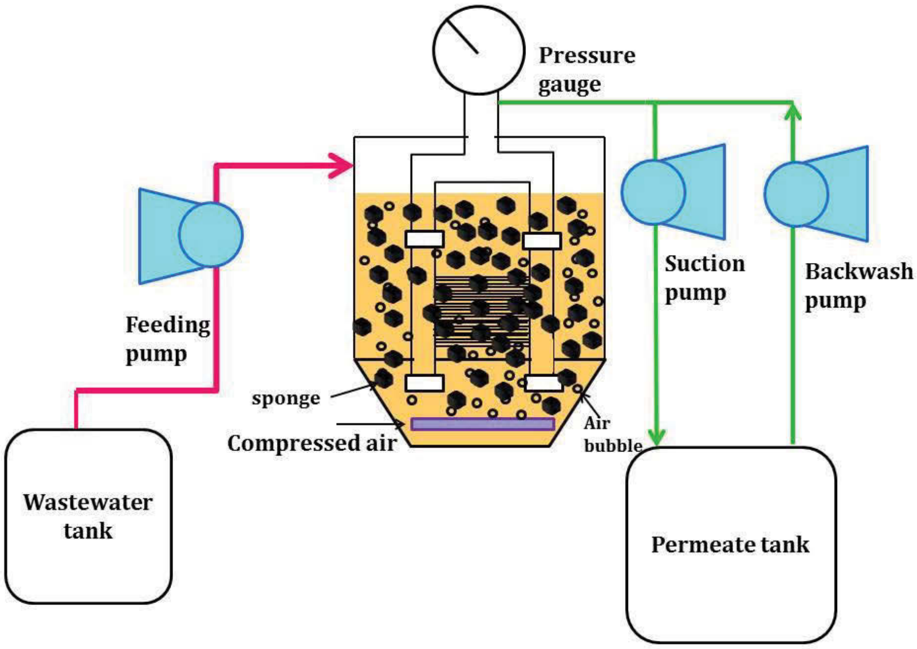
6.2. Materials and methods

6.2.1. Experimental setup and operating conditions of the SSMBR and the SSMBR with bioflocculant addition (SSMBR-G)

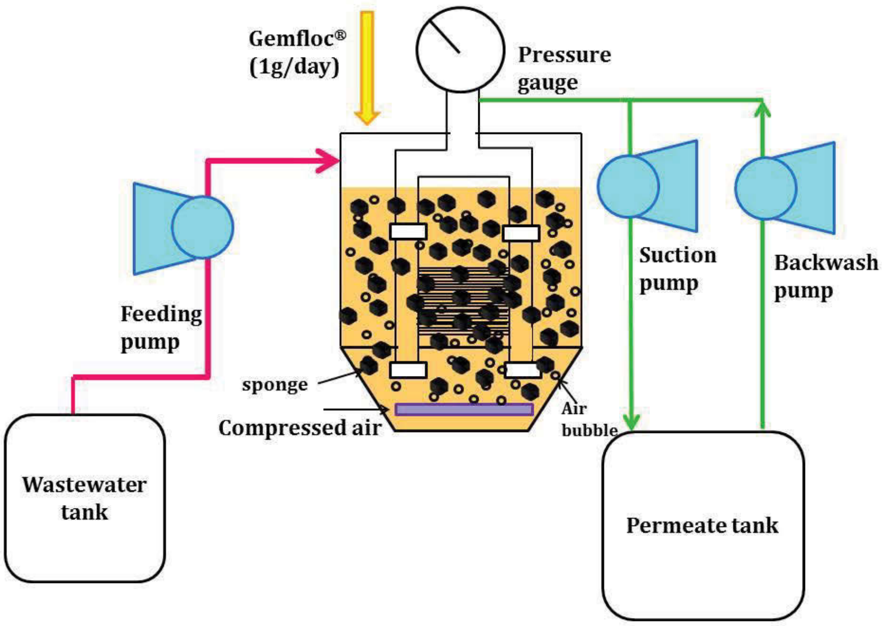
The characteristics of synthetic wastewater, experimental setup and operational conditions of MBR in this chapter could be found in Chapter 3, Section 3.2.1 and 3.2.2. Three experimental trials were conducted by operating three lab-scale SSMBRs (Fig. 6.1a) parallelly at filtration fluxes of 12, 15, and 20 $L/m^2{\cdot}h$, corresponding to HRTs of 6.67, 5.33, and 4.00 h, respectively. Gemfloc® addition was applied to the SSMBR (denoted as SSMBR-G, Fig. 6.1b) with optimum HRT to further mitigate membrane fouling. The same membrane module was immersed in each bioreactor. Each SSMBR was filled with 800 polyester-polyurethane porous sponge cubes (10% of volume fraction) with the dimensions of 10 mm × 10 mm × 10 mm, density of 28–45 kg/m^3 and cell count of 90 cells/in (90 cells per 25.4 mm). Prior to the experiments, sponge cubes were acclimatized to synthetic wastewater for 25 days. For the SSMBR-G, 1 g of Gemfloc® was employed into the bioreactor every day. The initial MLSS concentrations were 5.06, 5.00, and 5.02 g/L in the SSMBRs at HRTs of 6.67, 5.33, and 4.00 h, respectively. MLSS concentration of the SSMBR-G was adjusted to 5.00 g/L before the experiment. During the operational period, membrane fouling was physically controlled by backwashing at 3 times of the corresponding filtration flux and backwash frequency was two times/day with the duration of 2 min/time. The system descriptions and operating conditions of the SSMBRs and the SSMBR-G are displayed in Table 6.1. After the experiments, the membrane should be cleaned by chemical reagents (Chapter 3, Sections 3.2.2).

Table 6.1. System descriptions and operating conditions of the SSMBR and the SSMBR-G

Membrane properties	
Membrane configuration: Hollow fiber membrane module	
Material: Polyvinylidene fluoride (PVDF)	
Pore size: 0.2 μm	
Surface area: 0.1 m^2	
Operating conditions	
Air flow rate	9~10 L/min
Working volume	8 L
SRT	No sludge withdrawal
Initial MLSS concentration	5.06 g/L at 12 $L/m^2 \cdot h$ (HRT = 6.67 h), 5.00 g/L at 15 $L/m^2 \cdot h$ (HRT = 5.33 h), 5.02 g/L at 20 $L/m^2 \cdot h$ (HRT = 4.00 h)
Operating mode	Backwash: 2 times per day with duration of 2 min per time
Gemfloc®	1 g/day
Sponge in SSMBR	
Sponge size	1 × 1 × 1 cm
Sponge type	S_{28-30}/90R
Synthetic wastewater	
TOC	100–130 mg/L
COD	330–360 mg/L
NH_4-N	12–15 mg/L
PO_4-P	3.3–3.5 mg/L



(a)



(b)

Fig. 6.1. Experimental set-up of the SSMBR (a) and the SSMBR-G (b)

6.2.2. Analysis methods

For treatment performance analyses, concentrations of dissolved organic carbon (DOC), chemical oxygen demand (COD), NH_4-N, NO_3-N, NO_2-N and PO_4-P of influent and effluent samples were measured every 5 days. When TMP increased drastically for the consecutively two days, the sludge characterization were conducted, including mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), EPS, SMP, apparent viscosity, zeta potential, RH, floc size. The compositions of EPS, SMP and biopolymer clusters (BPC) of cake layer along with the fouling resistance were analysed at the end of the experiment. The detailed analysis and measurement methods could refer to Chapter 3 (Sections 3.2.3-3.2.7).

6.3. Results and discussion

6.3.1. Effects of HRT on SSMBR performance

A. Treatment performance

The SSMBRs with different HRTs showed good treatment performance in terms of organic removal. The SSMBR at the longest HRT of 6.67 h exhibited 96.53 ± 5.36% of DOC removal, higher than those at HRTs of 5.33 and 4.00 h (95.23 ± 6.89% and 94.56 ± 7.26%, respectively). Shorter HRTs induced a decrease of COD removal efficiencies, with the averages of 95.72 ± 4.35%, 94.29 ± 4.06%, and 93.75 ± 3.95% at HRTs of 6.67, 5.33, and 4.00 h, respectively. These results suggested that decreased HRT reduced organic matter removal owing to that increased organic loading may impair the biological system for the degradation of organic matters (Ng et al., 2014). Nevertheless, at HRTs of 6.67, 5.33 and 4.00 h, the food to microorganism (F/M) ratios were 0.24, 0.28 and 0.33 kg BOD_5/kg MLVSS·d, respectively, which were within the normal F/M range for activated sludge systems (0.2 to 0.5 kg BOD_5/kg MLVSS·d). Hence, although the shortened HRT increased organic loading rate, organic removal did not reduce significantly.

It was observed that nutrient removals in the SSMBRs were obviously different at various HRTs. The highest NH_4-N removal of 91.29 ± 7.43% was achieved at HRT of 6.67 h, while lower removals of 87.93 ± 6.23% and 83.52 ± 5.76% were obtained at HRTs of 5.33 and 4.00 h, respectively. High NH_4-N removal in these reactors was ascribed to retention of nitrifying bacteria by membrane filtration without sludge withdrawal as well as higher population of ammonium oxidizing bacteria (AOB) in the reactor caused by the presence of AOB on the acclimatised sponge during acclimatization period (Chiemchaisri and Yamamoto, 2005; Nguyen et al., 2012b). Three SSMBRs achieved more than 80% of T-N removal, reaching 87.65 ± 4.63%, 83.46 ± 5.69%, and 80.63 ± 6.41% at HRTs of 6.67, 5.33 and 4.00 h, respectively. These results revealed the existence of simultaneous nitrification and denitrification (SND) process, which was ascribed to the anoxic condition around the surface of the sponge and inside the sponge due to distinctive dissolved oxygen (DO) gradient along the sponge inward depth (Lim et al., 2011). PO_4-P removal efficiencies in the SSMBRs at HRTs of 6.67, 5.33, and 4.00 h were 95.23 ± 5.28%, 90.36 ± 6.32 %, and 86.78 ± 4.69%, respectively, suggesting that the presence of anaerobic and anoxic condition at the inner and the outer parts of sponge promoted phosphorus removal (Deng et al., 2014). When decreasing HRT, the higher pollutant loading rates and insufficient retention time depressed nitrification and denitrification, thus obtaining lower NH_4-N and T-N removals. Nitrogen was not effectively eliminated at shorter HRT, as reflected by effluent NO_3-N concentrations of 1.80 ± 0.35, 5.80 ± 0.56 and 8.30 ± 1.40 mg/L at HRTs of 6.67, 5.33 and 4.00 h, respectively. Moreover, since phosphorus release could also be inhibited by high concentration of NO_3-N, PO_4-P removal declined (Wang et al., 2009b; Ng et al., 2014).

B. TMP development

For fouling development, TMP variation at HRT of 6.67 h was characterized by two step fouling phenomenon i.e., a gentle TMP increase from 2.0 to 8.0 kPa before 73 days, and a subsequently faster TMP increment until it reached 18.5 kPa on day 100 (Fig. 6.2). The fouling rate was only 0.175 kPa/d during the entire operational period. At HRTs of 5.33 and 4.00 h, the operating durations of SSMBRs were reduced to 40 and 27 days before membrane module fouled and TMP reached 35.0 kPa, yielding higher fouling rates of 0.825 and 1.204 kPa/d, respectively. These results implied that lower HRT could shorten filtration period, deteriorate filterability, and aggravate membrane fouling. The further interpretation regarding fouling behaviour in SSMBRs is given in Section 6.3.1E.

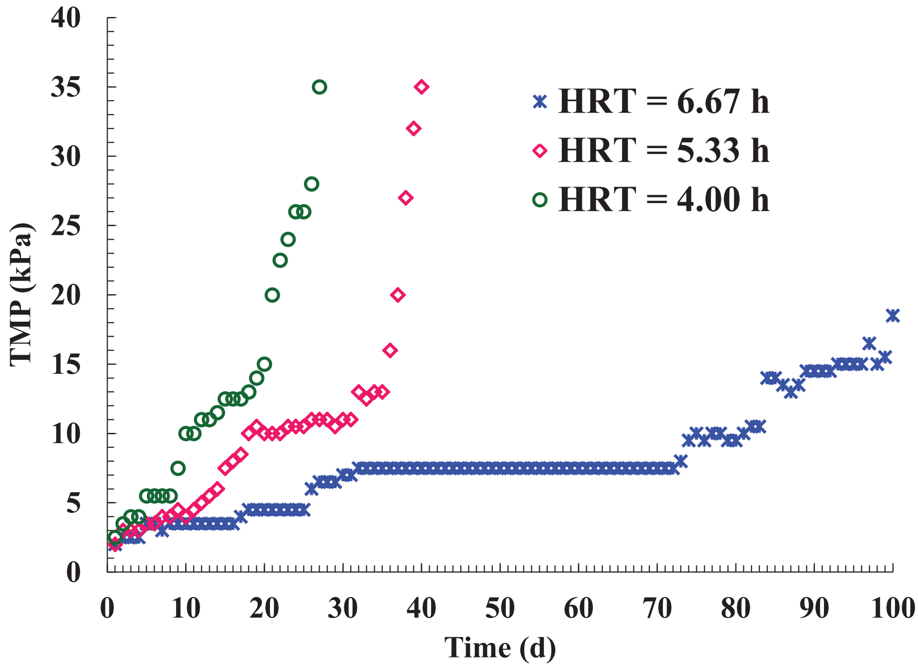


Fig. 6.2. TMP profile for the SSMBRs at different HRTs

C. Mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS) and apparent viscosity

As SRT was infinite, biomass multiplication was found in SSMBRs. The SSMBR at HRT of 6.67 h featured a slower MLSS increment, reaching 13.62 g/L on day 100, corresponding to the lowest biomass growth rate (ΔMLSS/Δt) of 0.09 g/L·d. On the other hand, rapid sludge accumulation was obtained with final MLSS values of 15.40 g/L and 17.24 g/L over the operational period when HRT decreased to 5.33 and 4.00 h, giving higher biomass growth rates of 0.26 and 0.45 g/L·d, respectively. It implied that the reduced HRT increased organic loading rate (OLR), which increased biomass growth and MLSS concentration. The average MLVSS/MLSS ratios increased with decreasing HRTs, corresponding to 0.8600, 0.8623, and 0.8639 at HRTs of 6.67, 5.33, and 4.00 h, respectively. When shortening HRT from 6.67 to 5.33 h, sludge viscosity increased from 3.95 ± 1.75 to 4.45 ± 1.96 mPa·s. A further decrease of HRT to 4.00 h caused a rise of sludge viscosity, obtaining 4.85 ± 1.65 mPa·s. These results suggested that lower HRT induced significantly higher biomass concentration, resulting in higher sludge viscosity.

D. EPS and SMP in mixed liquor

The variations of total EPS concentrations in activated sludge and SMP concentrations in mixed liquor of three SSMBRs responding to different designated TMPs are displayed in Figs. 6.3 and 6.4. Since the development of TMP was less than 20 kPa (18.5 kPa) for HRT of 6.67 h, EPS levels, SMP levels and SMP_P/SMP_C ratios are presented according to 4 designated TMPs. At HRT of 6.67 h, EPS concentrations were in the range of 9.85-13.27 mg/L, while EPS contents varied remarkably at HRTs of 5.33 h (13.77-27.08 mg/L) and 4.00 h (20.10-58.68 mg/L). The elevated EPS levels at shorter HRT could be ascribed to the increased OLR (1.19, 1.49 and 1.98 kg COD/m^3·d for HRTs of 6.67, 5.33 and 4.00 h, respectively), which further accelerated biomass growth. Although the highest concentrations of SMP were observed at HRT of 6.67 h (the maximum value of 7.53 mg/L) compared with those at HRTs of 5.33 h (6.06 mg/L) and 4.00 h (4.26 mg/L), all SMP values in three SSMBRs were considerably low because significantly less SMP in mixed liquor could be found in MBR with sponge addition (Deng et al., 2014). Based on another study of Deng et al. (2015), in a conventional MBR (CMBR) with similar operating conditions (flux of 12 L/m^2, initial biomass of 5 g/L and HRT of 6.67 h), SMP concentration achieved more than 30 mg/L when membrane fouling occurred. It should also be noted that a shortened operation time was recorded for the SSMBRs at low HRTs to achieve the maximum TMP allowed (35.0 kPa), i.e. TMP reached 35.0 kPa after 37 and 21 days of operation at HRTs of 5.33 and 4.00 h, respectively (Fig. 6.2). Hence, higher SMP levels with increased HRT in SSMBR may be due to the accumulation of SMP with evolution of time in the bioreactor. In this case, SMP concentration was not the vital factor to affect membrane fouling.

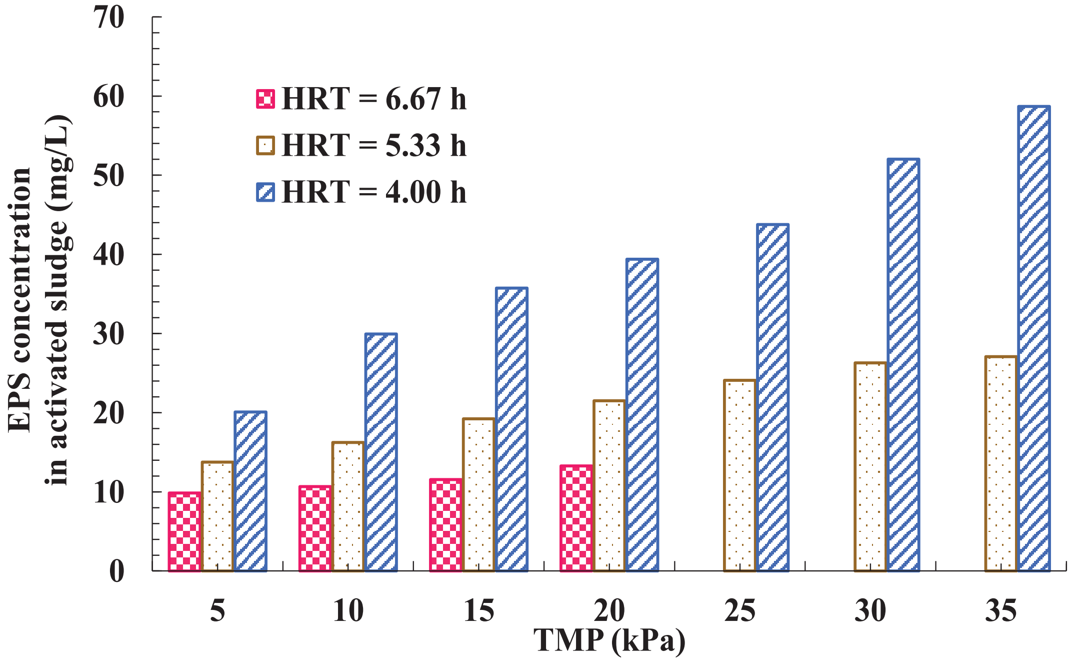


Fig. 6.3. Variations of EPS concentrations in activated sludge in the SSMBRs at different HRTs

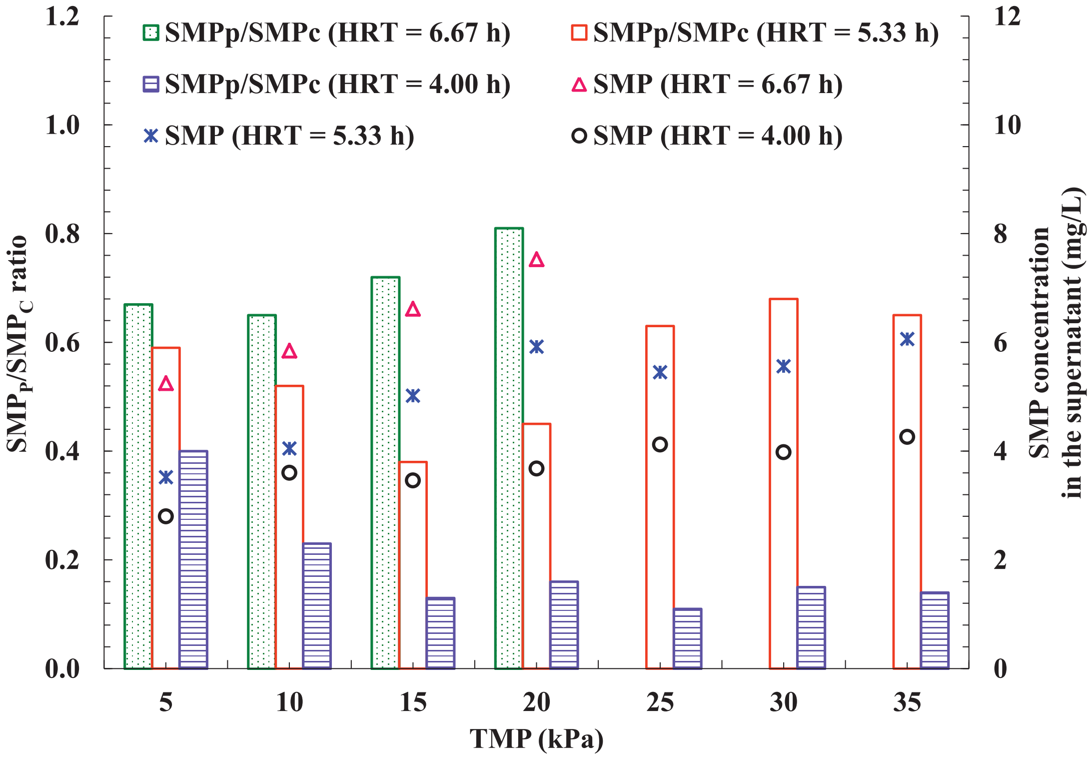


Fig. 6.4. Variations of SMP_P/SMP_C ratio and SMP concentrations in the supernatant in the SSMBRs at different HRTs

E. Membrane fouling analysis

As shown in Fig. 6.5, fouling resistance distributions in three SSMBRs at different HRTs were measured at the end of each experiment. With decreasing HRT from 6.67 to 5.33 h, total fouling resistance (R_T) increased from 2.50×10^{12} to 3.20×10^{12} m^{-1}, which further rose to 4.50×10^{12} m^{-1} at HRT of 4.00 h. Since the same clean membrane resistance (R_M) of 9.00×10^{11} m^{-1} was obtained for all SSMBRs, the dominant filtration resistance was cake layer resistance (R_C) in this chapter. The lowest R_C was 1.49×10^{12} m^{-1} at HRT of 6.67 h, consisting of 59.60% of R_T, while R_C increased to 1.93×10^{12} m^{-1} and 2.85×10^{12} m^{-1} at shorter HRTs of 5.33 and 4.00 h, accounting for 60.31% and 63.33% of R_T, respectively. Pore blocking resistance (R_P) followed the similar patterns as that observed for R_C, with the order of 6.67 h < 5.33 h < 4.00 h (1.06×10^{11}, 3.70×10^{11}, and 7.50×10^{11} m^{-1}, respectively). These results not only highlighted the importance of cake layer on membrane fouling development, but also suggested that deterioration of membrane permeability at shorter HRTs was caused by more cake layer formation and pore blocking.

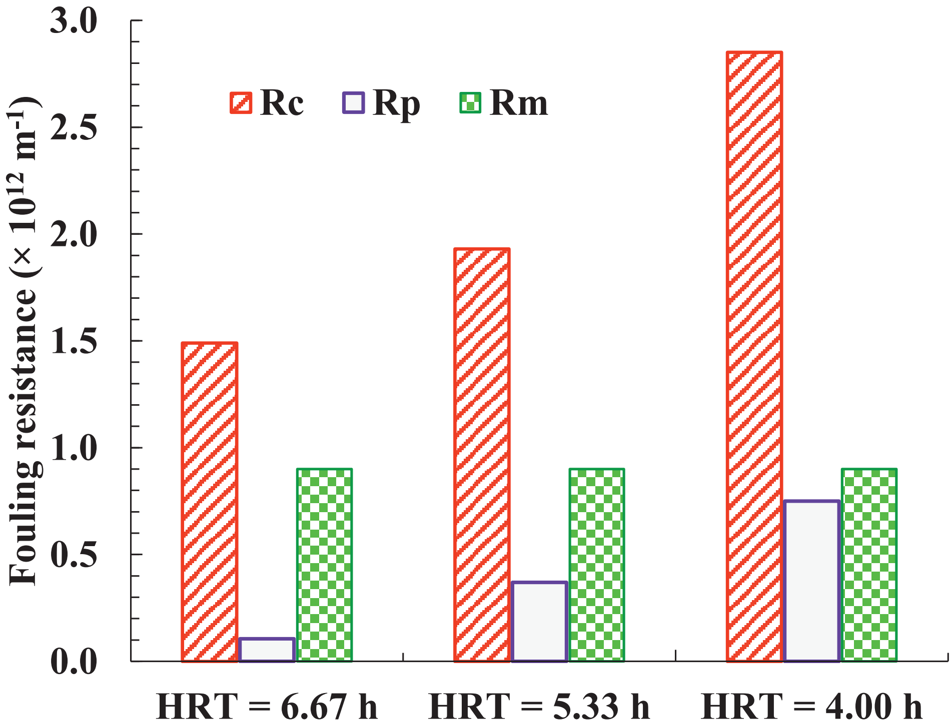


Fig. 6.5. Fouling resistance distribution in the SSMBRs at different HRTs

For biomass particulates with higher MLSS concentration and sludge viscosity, the net force toward membrane surface can be developed on these particulates as a result of reduced back-transport effects. It can lead to the accumulation of more sludge particles and formation of a sticky cake layer on membrane surface, thus increasing R_C at shorter HRT (Kornboonraksa and Lee, 2009; Meng et al., 2007a). In addition, physical clearance effects of sponge on cake layer which was dampened by higher sludge viscosity also gave rise to higher R_C with decreasing HRT (Deng et al., 2014). Cake layer as the predominant portion of R_T was characterized by compositions of EPS, SMP, and BPC (Fig. 6.6). The decline of HRT from 6.67 to 5.33 h induced a pronounced rise of EPS_P and EPS_C from 2.52 to 8.33 mg/g cake layer and 1.48 to 5.28 mg/g cake layer, respectively. Further reduction of HRT from 5.33 to 4.00 h promoted an increase in EPS levels, obtaining EPS_P and EPS_C at 14.40 and 10.10 mg/g cake layer, respectively. Similarly, the contents of BPC extracted from the membrane surface showed significant variations at different HRTs. The cake layer contained the least BPC_P and BPC_C at HRT of 6.67 h at 2.42 and 0.95 mg/g cake layer, respectively. BPC_P

and BPC_C levels at HRT of 5.33 h were 2.1 and 7.3 times comparing with those at HRT of 6.67 h, respectively. A dramatic increase of BPC_P and BPC_C was observed when HRT decreased to 4.00 h, reaching 2.63 and 30.63 mg/g cake layer, respectively. These results indicated that shorter HRTs increased R_C by accumulation of EPS (EPS_P and EPS_C) and BPC (BPC_P and BPC_C) on membrane surface. However, SMP contents of cake layer in three SSMBRs were substantially low and showed slight changes, with the values of 0.58, 1.22 and 1.38 mg/g cake layer at HRTs of 6.67, 5.33 and 4.00 h, respectively. It could be inferred that even the extracted SMP from membrane surface exerted a negligible effect on membrane fouling, which confirmed that SMP were not the key fouling factor under the studied conditions as discussed in Section 6.3.1D.

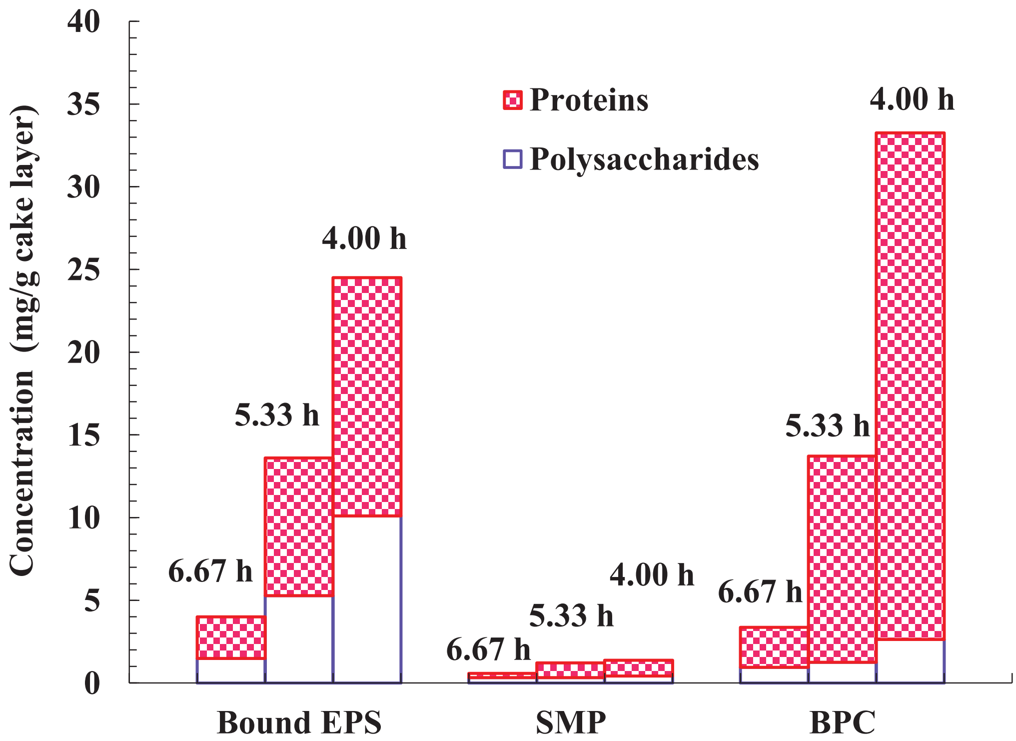


Fig. 6.6. Compositions of bound EPS, SMP and BPC of cake layer in the SSMBRs at different HRTs

At shorter HRTs, the increased suction force at higher TMP and the increased drag force towards the membrane at higher fluxes induced more readily deposition of larger amounts of bound EPS and BPC on membrane surface to form a biofilm/cake layer (Qu et al., 2013). Moreover, cell lysis and the resulting release of bound EPS could take place at the interior section of the bio-cake layer due to the development of an anoxic and endogenous condition (Hwang et al., 2008). As to mixed liquor, EPS enhanced bacterial adhesion and/or attachment onto the membrane as well as promoted formation of a thick fouling layer on the membrane surface, thus clogging membrane pores (Tansel et al., 2006; Ng et al., 2006). Therefore, more EPS in activated sludge led to higher R_C and R_P at shorter HRTs. At the fourth designated TMP, SMP_P/SMP_C ratio declined from 0.81 to 0.45 with decreasing HRT from 6.67 to 5.33 h, which further reduced to 0.16 at the shortest HRT of 4.00 h (Fig. 6.4). As SMP_C presents partially hydrophilic nature as compared to SMP_P (Guo et al., 2012a), lower SMP_P/SMP_C ratio in mixed liquor increased R_C and R_P at shorter HRTs in terms of higher irremovable fouling, and more membrane pore blocking and gel layer formation on membrane surface.

6.3.2. Effects of bioflocculant addition on SSMBR fouling

Based on the above-mentioned results, HRT of 6.67 h could be the optimum HRT for SSMBR. Therefore, SSMBR was further investigated through Gemfloc® addition at HRT of 6.67 h.

A. The performance of the SSMBR-G and the SSMBR

As compared to the SSMBR, the removals of DOC and COD increased to 97.76 ± 4.52% and 96.78 ± 3.25% in the SSMBR-G, respectively, indicating the enhancement of organic removal by Gemfloc® application. Although only slightly higher NH_4-N and PO_4-P removal efficiencies (92.96 ± 5.69% and 97.45 ± 4.68%, respectively) were observed, the SSMBR-G presented the improved T-N removal of 91.69 ± 4.53%, which was consistent with previous findings of the induced denitrification process occurring inside larger sludge flocs (Deng et al., 2015). Comparing to the SSMBR, a lower biomass growth rate of 0.08 g/d was achieved in the SSMBR-G with final MLSS concentration of 13.02 g/L over 100 days of operation, which again proved bioflocculant addition could reduce the growth of biomass in bioreactor (Deng et al., 2015; Ngo and Guo, 2009).

TMP variations for 100 days of operation monitored for the SSMBR and the SSMBR-G are depicted in Fig. 6.7. Higher fouling potential was found in the SSMBR, suggesting that smaller particles aggravated pore blocking and cake layer formation on membrane surface (higher R_P and R_C) (Lim and Bai, 2003). For better comparison of membrane fouling reduction, the TMP development of the CMBR and the CMBR with

Gemfloc® addition (MBR-G) with similar operating conditions (flux of 12 L/m^2, initial biomass of 5 g/L and HRT of 6.67 h) are also presented in the small figure in Fig. 6.7. The SSMBR-G exhibited the least TMP development (4.0 kPa) and fouling rate (0.035 kPa/d) throughout 100-day operation, which were about one-fifth of those in the SSMBR. It suggested that a significant improvement in filterability and membrane fouling reduction in the SSMBR-G were achieved by Gemfloc® addition.

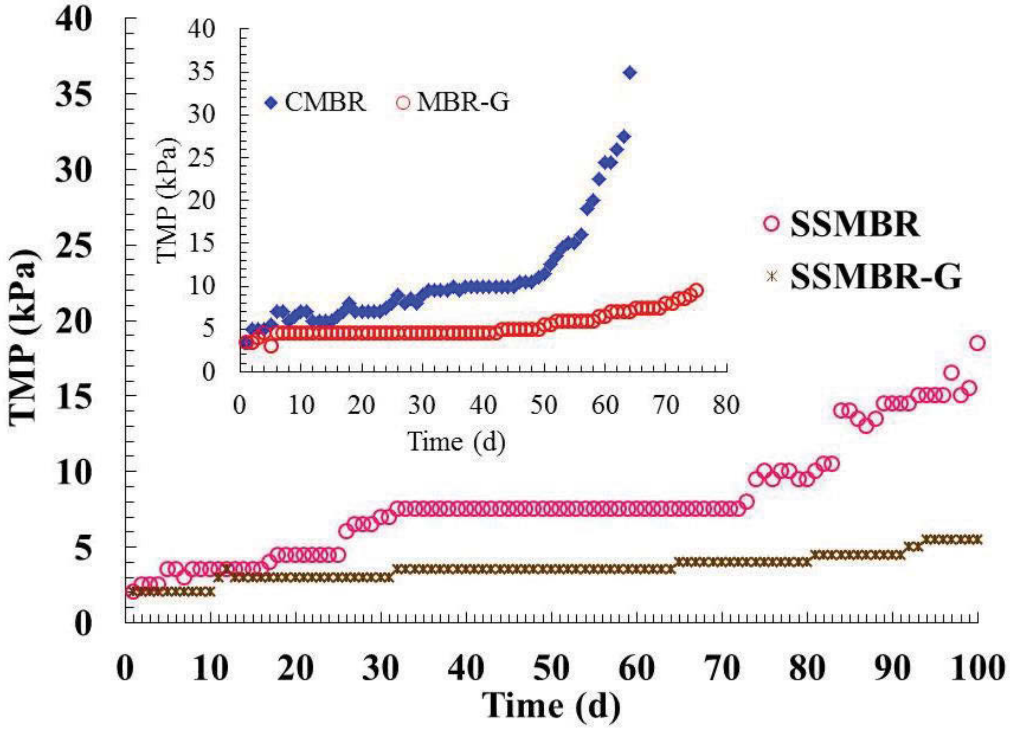


Fig. 6.7. TMP profile for the SSMBR, the SSMBR-G, the CMBR and the MBR-G with similar operating conditions (flux of 12 L/m^2, initial biomass of 5 g/L and HRT of 6.67 h)

B. Membrane fouling analysis

As compared to the SSMBR (2.50 × 10^{12} m^{-1} of R_T and 1.49 × 10^{12} m^{-1} of R_C), the SSMBR-G demonstrated lower R_T and R_C of 2.07 × 10^{12} m^{-1} and 1.10 × 10^{12} m^{-1} (53.14% of R_T), respectively. Moreover, a higher R_P was obtained in the SSMBR (1.06 × 10^{11} m^{-1}), almost 2-fold of that in the SSMBR-G (0.70 × 10^{11} m^{-1}), which accounted for 3.38% of R_T in the SSMBR-G. These results highlighted the capability of Gemfloc® in reducing membrane fouling through eliminating pore blocking and decreasing cake layer fouling which was the major fraction of membrane fouling.

Tables 6.2 and 6.3 display total concentrations of EPS in activated sludge and SMP in the supernatant of mixed liquor as well as average EPS_P/EPS_C and SMP_P/SMP_C ratios, which were divided into three Phases, including Phase I (Day 0-26), Phase II (Day 27-74), and Phase III (Day 75-100). EPS contents in activated sludge of the SSMBR were also lower (8.80–13.27 mg/L) than those in the SSMBR-G ranging from 13.84 to 20.18 mg/L during the operational period. Meanwhile, SMP concentrations in the supernatant of mixed liquor kept relatively stable (4.95–5.29 mg/L) in Phase I, and then increased slightly and maintained in the range between 5.63 and 5.89 mg/L within 74 days before severe fouling occurred (Phase II). Subsequently, a rapid increase of SMP (6.02–7.53 mg/L) was shown in Phase III, corresponding to the sudden TMP jump. Conversely, activated sludge in the SSMBR-G exhibited lower SMP levels, varying from 2.60 to 5.89 mg/L during the entire operational period, which could be ascribed to the entrapment of SMP during flocculation process (Dizge et al., 2011a). Furthermore, lower SMP_P/SMP_C ratios presented for the SSMBR than those for the SSMBR-G (1.40 ± 0.81) also reflected more severe fouling in SSMBR, as higher SMP concentrations with lower SMP_P/SMP_C ratio in activated sludge gave rise to higher R_P and R_C (Deng et al., 2015).

Table 6.2. Total EPS concentrations and EPS compositions (average EPS_P/EPS_C) of activated sludge in the SSMBR and the SSMBR-G at different phases during the operational period

Day		Phase I (Day 0–26)	Phase II (Day 27–74)	Phase III (Day 75–100)
EPS (mg/L)	SSMBR	8.80–10.20	7.70–10.50	10.26–13.27
	SSMBR-G	13.84–20.18	14.01–16.06	13.96–15.42
EPS_P/EPS_C [a]	SSMBR	1.48 ± 0.18	1.35 ± 0.77	1.27 ± 0.65
	SSMBR-G	1.86 ± 0.15	2.80 ± 0.73	2.52 ± 0.64

[a] EPS_P, proteins in EPS; EPS_C, polysaccharides in EPS

Table 6.3. Total SMP concentrations and SMP compositions (average SMP_P/SMP_C) of the supernatant in the SSMBR and the SSMBR-G at different phases during the operational period

Day		Phase I (Day 0–26)	Phase II (Day 27–74)	Phase III (Day 75–100)
SMP (mg/L)	SSMBR	4.95–5.29	5.63–5.89	6.02–7.53
	SSMBR-G	3.01–5.89	2.60–3.95	3.25–3.99
SMP_P/SMP_C[a]	SSMBR	0.72 ± 0.03	0.54 ± 0.11	0.82 ± 0.06
	SSMBR-G	1.26 ± 0.07	1.82 ± 0.39	1.78 ± 0.15

[a] SMP_P, proteins in SMP; SMP_C, polysaccharides in SMP

As shown in Table 6.2, EPS_P/EPS_C ratios were always higher in the SSMBR-G than those in the SSMBR. Generally, EPS_P contains amino acids with hydrophobic side groups and possessed positively charged amino groups, while EPS_C is responsible for hydrophilic nature of activated sludge (Liu et al., 2014). Thus, sludge flocs with higher EPS_P/EPS_C ratio have better flocculation ability and surface properties such as zeta potential and RH. It was found that the sludge flocs in the SSMBR-G exhibited larger particle size distribution were also observed (160–340 μm) than those in the SSMBR (80-160 μm), which was due to the increased EPS in the SSMBR-G which induced polymer entanglement and further promoted aggregation of sludge flocs (Yan et al., 2015). Although activated sludge in both MBRs presented similar RH values (89.43 ± 5.12% and 87.57 ± 9.59% in the SSMBR-G and the SSMBR, respectively), the zeta potentials of sludge flocs in the SSMBR-G (1.43 ± 3.49 mV) were more neutral than those in the SSMBR (– 8.16 ± 4.84 mV), because Gemfloc® addition could improve the surface charge of sludge flocs as well as promote clustering and formation of larger microbial flocs (Lee et al., 2007). Moreover, the reduced zeta potential of sludge flocs associated with larger particle size distribution in the SSMBR-G also indicated the improvement of flocculation ability of the flocs was caused by the decreased repulsive force among the flocs having larger size (Yan et al., 2015).

Through analyzing the cake layer on membrane surface after terminating the experiments, it was found that both MBRs contained similar levels of EPS, with EPS_P and EPS_C of 2.46 and 1.32 mg/g cake layer in the SSMBR-G, respectively. Regarding SMP_P and SMP_C of the cake layer, lower values were attained (0.13 and 0.12 mg/g cake layer, respectively) for the SSMBR-G than those for the SSMBR (0.26 and 0.32 mg/g cake layer, respectively). Bioflocculant addition in the SSMBR-G also reduced BPC_C and BPC_P by 84.8% and 29.34%, reaching 0.60 and 1.71 mg/g cake layer, respectively. Thus, Gemfloc® addition could promote fouling control and permitted a long sustainable operation at low TMP in the SSMBR-G.

6.4. Conclusions

This study evaluated membrane fouling behavior of the SSMBRs at different HRTs and the SSMBR-G at the optimum HRT of 6.67 h. The following specific conclusions were obtained from the study:

1) At shorter HRT, more obvious membrane fouling was caused by exacerbated cake layer formation and aggravated pore blocking. Activated sludge possessed more EPS due to excessive growth of biomass and lower SMP_P/SMP_C ratio;
2) The cake layer resistance was aggravated by increased sludge viscosity and the accumulation of EPS and BPC on membrane surface;
3) SMP showed marginal effect on membrane fouling when SSMBRs operated at all HRTs;
4) The SSMBR with Gemfloc® addition at the optimum HRT of 6.67 h demonstrated superior sludge characteristics such as larger floc size, less SMP in mixed liquor with higher SMP_P/SMP_C ratio, less SMP and BPC in cake layer, thereby further preventing membrane fouling.

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Chapter 7

A New Functional Media for Enhancing Performance of Integrated Moving Bed Biofilm Reactor-Membrane Bioreactor Systems

Chapter 7 A New Functional Media for Enhancing Performance of Integrated Moving Bed Biofilm Reactor-Membrane Bioreactor Systems

7.1. Introduction

It has been widely accepted that biological nutrient removal (BNR) process is a preferable choice for simultaneous organic and nutrient removal during wastewater treatment. Currently, various BNR processes have been developed, including the five-stage Bardenpho process, the anoxic/oxic (A/O), the anaerobic/anoxic/oxic (A^2/O), the University of Cape Town (UCT) process, and attached biofilm reactors (Chen et al., 2011). Among them, the moving bed biofilm reactor (MBBR) is a cost-effective and efficient BNR technology, which can realize high-volume biofilm growth, high specific biomass activity, low headloss, no medium channelling and clogging, and inhibition of the excessive abrasive removal of slow growing microorganisms (Guo et al., 2010b; Guo et al., 2012b; Ødegaard et al., 1999).

During the operation process, characteristics of attached growth media play a key role in MBBR performance. In recent years, different kinds of media have been employed in MBBRs for wastewater treatment, including plastic media (e.g. suspended plastic bio-carreriers, Kaldnes K1, K2, K3 and K5, Kaldnes biofilm Chip M, etc.), polyurethane foam, activated carbon (granular and powdered), natural occurring materials (e.g. sand, zeolite, diatomaceous earth, light expended clay aggregate, etc.), non-woven carriers, ceramic carriers, modified carriers (e.g. BIOCONS carrier, bioplastic-based moving bed biofilm carriers, polyvinyl acochol-gel carrier, biodegradable polymer polycaprolactone carriers, etc.) and wood chips. The most popularly used carrier for MBBR is plastic media. A lab-scale MBBR containing 50% (filling ratio) of the Kaldnes biomedia K1 was operated by Aygun et al. (2008) for synthetic wastewater treatment. It was reported that the increase of the organic loading rate (6-96 g COD/m^2·d) caused the declined organic removal efficiency from 95.1% to 45.2%. Shore et al. (2012) used bench scale MBBRs with 50% fill of BioPortz^{TM} media (high density polyethylene (HDPE)) to treat secondary treated effluent. They found that more than 90% of NH_4-N was eliminated from both synthetic and industrial wastewater at 35 and 40 ºC by the MBBRs. Zhang et al. (2013) used a pilot-scale MBBR with suspended polyethylene (PE) bio-carriers having inclination angle of 60º (50% of working volume fraction) to treat the raw water polluted by NH_4-N at various temperatures (3.7–35.7 °C) and NH_4-N loadings (0.031–0.0473 g NH_4^+-N/m^2·d), achieving average removal of 71.4 ± 26.9%. However, the MBBR systems with plastic media generally do not present high T-N removal due to their limited denitrification capacity. Moreover, under aeration condition in MBBR, the strict anaerobic zone cannot be obtained for effective phosphorus release, which in turn decreases phosphorus removal efficiency (Zhuang et al., 2014). Other constrains include long start-up period required for biofilm growth on the plastic media and stabilizing system performance (Habouzit et al., 2014), as well as easy detachment of biofilm from the plastic media (Rafiei et al., 2014).

As a promising alternative media, sponge has attracted more and more interest, as it is low-cost material and can promote the rapid and stable attachment and growth of microorganisms on the carrier due to its high porosity (Ngo et al., 2008). Some recent studies have highlighted the effectiveness of sponge in MBBRs for organic and nutrient removal. The batch experiments conducted by Lim et al. (2011) showed that high concentrations of 8-mL polyurethane sponge cubes (2 × 2 × 2 cm, 40% (v/v)) induced good T-N removal of 84% in treating low COD/N ratio wastewater. It could enable high capacity of the moving bed sequencing batch reactors (MBRSMBR) for nitrogen removal at low cost. Chu and Wang (2011) reported that the MBBR with sponge (20% filling ratio) at a hydraulic retention time (HRT) of 14 h showed high TOC and NH_4-N removal (90% and 65%, respectively). Feng et al. (2012b) also pointed out that the aerobic MBBR with high polyurethane foam packing rate of 40% could remove average 80% of COD and 96.3% of NH_4-N for artificial sewage treatment at an HRT of 5 h.

This study focused on investigating new functional media (i.e. plastic carrier modified using sponge) developed at UTS for enhancing the treatment performance of MBBR system. The organic, nitrogen and phosphorus removals were elevated and compared between an MBBR with sponge modified plastic carriers (S-MBBR) and an MBBR with plastic carriers only. Both MBBRs were then coupled with membrane bioreactor (MBR) and the performance of two hybrid MBBR-MBR systems were also studied in terms of pollutant removal and membrane fouling.

A major part of Chapter 7 has been published as the following paper:

Deng, L.J., Guo, W., Ngo, H.H., Zhang, X., Wang, X.C., 2016. New functional biocarriers for enhancing the performance of a hybrid moving bed biofilm reactor-membrane bioreactor system. Bioresource Technology 208, 87–93.

7.2. Materials and methods

7.2.1 Wastewater and media specifications

In this study, a synthetic wastewater with COD:N:P ratio of 100:5:1 was used to simulate primarily treated domestic wastewater, which was prepared with glucose, ammonium sulphate, potassium dihydrogen orthophosphate together with trace nutrients by dissolving in tap water. It gives dissolved organic carbon (DOC) of 100-130 mg/L, chemical oxygen demand (COD) of 330-360 mg/L, ammonium nitrogen (NH_4-N) of 12-15 mg/L, and orthophosphate of 3.3-3.5 mg/L. The pH was maintained at 7.0 by adding sodium carbonate or sulphuric acid on a daily basis.

The sponge modified plastic carrier was prepared by combining reticulated porous polyester-polyurethane sponge (Joyce Foam Products, Australia) with plastic carrier (namely Suspended Biological Filter, SBF® from Yixing City Yulong F.P. Co., Ltd., China). Each plastic carrier has the nominal diameter and length of 25 and 9 mm, respectively, with specific density of 950 kg/m^3, specific surface area of 500 m^2/m^3, and void ratio of 95%. The sponge (density of 28-30 kg/m^3, cell count of 90 cells/in (90 cells per 25 mm)) was cut into required size and fixed into alternate holes of the plastic carrier. The average weights of these two kinds of carriers were 1.20 ± 0.04 g per sponge modified plastic carrier and 1.08 ± 0.03 g per plastic carrier.

7.2.2. Experimental setup and operating conditions

Two batch-scale MBBR systems with effective working volume of 12 L were used and both MBBRs were filled with 20% of carriers (working volume fraction). The MBBR with fresh sponge modified plastic carriers (S-MBBR) and the MBBR with fresh plastic carriers (MBBR) were acclimatized for 15 days before operating in continuous mode at the flow rate of 16.7 mL/min, corresponding to a HRT of 12 h. The dissolved oxygen (DO) concentration was controlled in the range of 5.0-6.0 mg/L for both MBBRs. The low air flow rate could promote complete liquid-solid mixing, moderate media up/down motion, and limit the release of biomass from the media.

For the set-up of the hybrid systems, two 10-L submerged MBR units were employed to connect with the S-MBBR and the MBBR, hereafter referred to as S-MBBR-MBR and MBBR-MBR, respectively (Fig. 7.1). For the MBR unit, the membrane module used in this study was hollow fiber membrane which was made of polyethylene (PE) with hydrophilic coating having a surface area of 0.195 m^2 and a pore size of 0.1 μm (Fig. 7.2). Infinite sludge retention time (SRT) was obtained without sludge waste. MBBR effluent was pumped into the MBR unit as the feed through a buffer tank. The membrane permeate was withdrawn from the membrane module by a suction pump at the filtration flux of 10.26 L/m^2·h to maintain the HRT at 5 h. There was a pressure gauge connected with membrane for measuring transmembrane pressure (TMP) value every day. Only two times/day backwash frequency with duration of 2 min/time was employed at flow rate of 30.78 L/m^2·h. Chemical cleaning (1% hydrochloric acid, 2% citric acid, 0.4% sodium hypochlorite plus 4% sodium hydroxide for 6 h soaking, respectively) was conducted when terminating the experiments at TMP of 35.0 kPa.

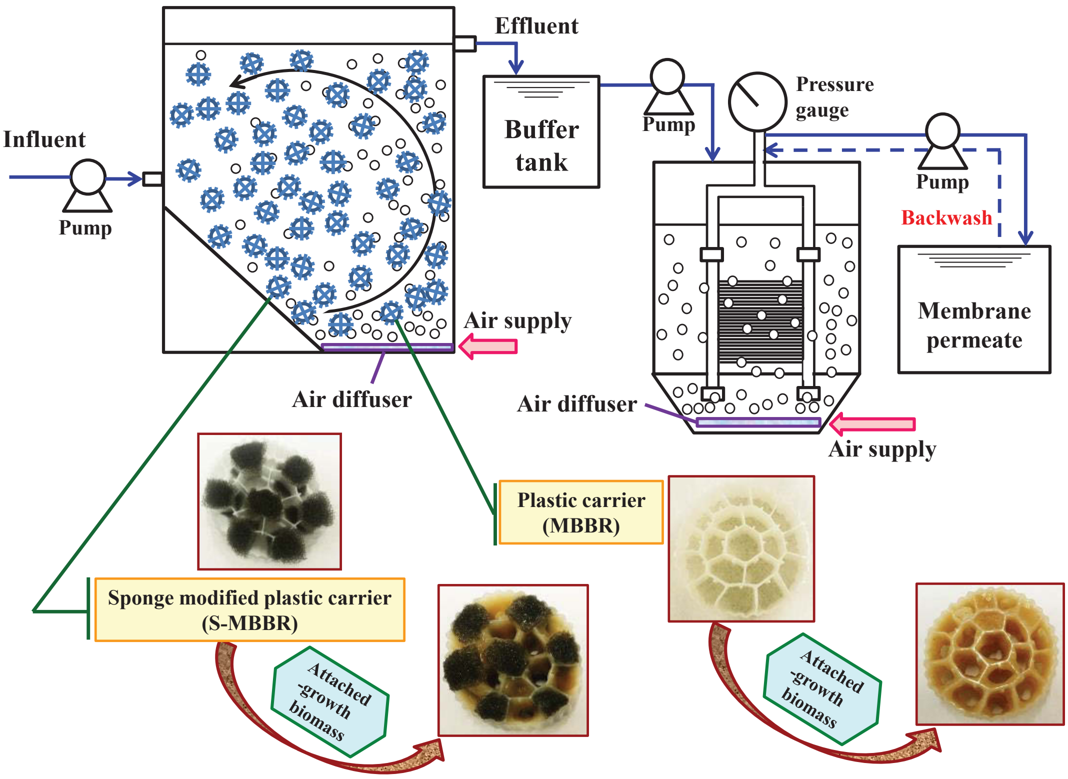

Fig. 7.1. Experimental set-up of the S-MBBR-MBR and the MBBR-MBR

The entire study period consisted of 5 phases according to different operating conditions as displayed in Table 7.1. Phase I (Day 0-15) is the acclimatization period for both MBBRs in batch mode until both systems reached relatively stable treatment performance. In Phase II (Day 16-30), both MBBRs were operated in continuous mode (flow rate of 16.7 mL/min). The stabilization of both MBBR systems was achieved within the first 30-day of operation. During the experimental period, both MBBRs were operated at HRT of 12 h from Day 31 to 60 (Phase III). The HRT was then halved to 6 h from Day 61 to 90 (Phase IV) to match the flow rate requirement of the subsequent MBR unit (33.3 mL/min). Finally, the evaluation of two hybrid systems was conducted at Phase V at the HRTs of 6 h for the MBBR units and 5 h for the MBR units.

Table 7.1. Operating conditions at different phases over the entire experimental period

Phase	Operational day	Systems	HRT (h)	Flow rate (mL/min)
I	0-15 (acclimatization period)	S-MBBR, MBBR	12	16.7
II	16-30 (stabilization period)	S-MBBR, MBBR	12	16.7
III	31-60	S-MBBR, MBBR	12	16.7
IV	61-90	S-MBBR, MBBR	6	33.3
V	91-175	S-MBBR-MBR	6 for MBBR unit, 5 for MBR unit	33.3
	91-122	MBBR-MBR		

S-MBBR: MBBR with sponge modified plastic carriers; MBBR: MBBR with plastic carriers;
S-MBBR-MBR: Hybrid MBBR-MBR system with sponge modified plastic carriers;
MBBR-MBR: Hybrid MBBR-MBR system with plastic carriers

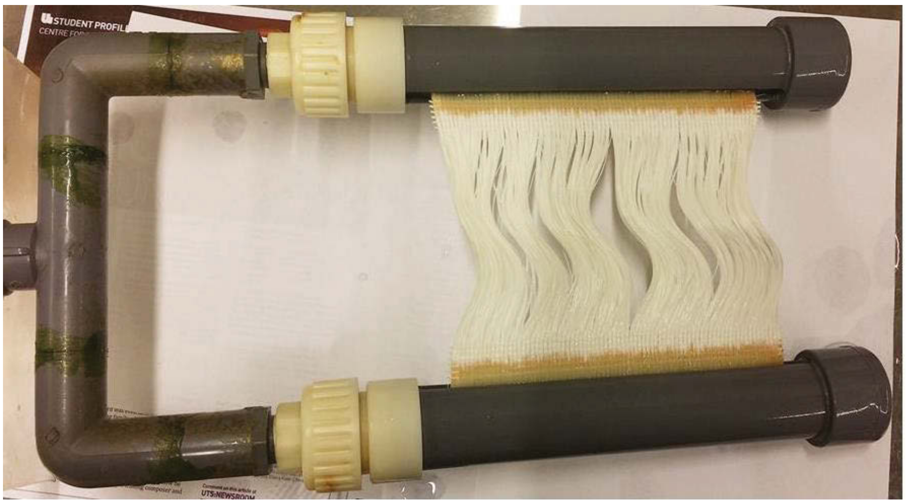

Fig. 7.2. Hollow fiber PE membrane module

7.2.3. Analysis methods

The sample analyses could refer to Chapter 3 as follows:

- Section 3.2.3 for quantification of DOC, NH_4-N, NO_3-N, NO_2-N and PO_4-P;
- Section 3.2.5 for membrane fouling analysis;
- Section 3.2.6 for measurement of attached-biomass and suspended sludge concentrations (including MLSS and mixed liquor volatile suspended solids (MLVSS));
- Section 3.2.7 for extraction and analyses of extracellular polymeric substances (EPS) and soluble microbial products (SMP) in mixed liquor and cake layer in MBR unit.

Besides, the turbidity of MBBR effluent was determined with 2100P Turbidimeter (HACH Company, USA).

7.3. Results and discussion

7.3.1. Treatment performance of the S-MBBR and the MBBR during start-up period

During the first 15-day operation (Phase I, start-up period), organic matter removal was obtained in both MBBRs with small variations. The removal efficiency was 90.03 ± 3.68% for DOC and 89.57 ± 4.62% for COD in the S-MBBR. For the MBBR, DOC and COD removals were 88.90 ± 4.39% and 86.21 ± 5.03%, respectively. Nutrient removal of MBBRs exhibited significant changes with the elapsed time. In the S-MBBR, NH_4-N, T-N, and PO_4-P removals were 60.35 ± 10.21%, 60.32 ± 14.03%, and 63.92 ± 12.87%, respectively, while the MBBR presented less nutrient removals (54.15 ± 11.44%, 51.14 ± 13.46%, and 54.95 ± 13.42%, respectively). As biomass growth was initialized on the carriers during the acclimatization period, nutrient removals were low and unstable in both MBBRs.

From Day 16 to 30, both MBBRs approached steady state (Phase II). Better treatment performance was found in the S-MBBR (94.73 ± 3.85%, 93.26 ± 2.75%, 83.76 ± 4.06%, 75.26 ± 2.17%, and 74.76 ± 3.93% for DOC, COD, NH_4-N, T-N and PO_4-P removals, respectively), compared to the MBBR (94.05 ± 4.76%, 92.03 ± 3.19%, 74.58 ± 5.19%, 59.90 ± 6.34% and 63.28 ± 6.28%, respectively). Additionally, the attached-biomass growth also reached steady state. The carriers in the S-MBBR contained more attached-growth biomass (0.1473 ± 0.0041 g MLSS/g and 0.1341 ± 0.0063 g MVLSS/g sponge modified plastic carrier) than those for the MBBR (0.0677 ±

0.0023 g MLSS/g and 0.0573 ± 0.0016 g MVLSS/g plastic carrier). For the plastic carrier, the biofilm was mainly developed on the outer surface of the carrier. As fresh sponge possesses large amount of pores, microorganisms can be entrapped into the pores and developed on both outer and inner surfaces of sponge (Guo et al., 2010b). Hence, larger amount of biomass was attached onto the sponge modified plastic carrier as compared to that on the plastic carrier. For the suspended growth in the MBBRs, MLSS and MLVSS concentrations of mixed liquor in the S-MBBR remained at 0.251 ± 0.018 and 0.243 ± 0.016 g/L, respectively, which were similar to those in the MBBR (0.262 ± 0.031 and 0.250 ± 0.029 g/L, respectively).

7.3.2. Treatment performance of the S-MBBR and the MBBR during experimental period

After the steady state, the S-MBBR and the MBBR were operated at two HRTs of 12 h (Phase III) and 6 h (Phase IV) and the results are summarized in Table 7.2. At HRT of 12 h, stable DOC and COD removals of 95.63 ± 4.23% and 94.58 ± 5.06% were observed in the S-MBBR, respectively, which were higher than that for the MBBR (93.52 ± 3.25% for DOC removal and 91.27 ± 4.69% for COD removal). It suggested that the MBBR systems demonstrated good performance in organic matter removal. NH_4-N removal in the S-MBBR averaged at 83.46 ± 3.98%, which was approximately 10% higher than the MBBR. Nitrifying microorganisms (including ammonia oxidizing bacteria and nitrite oxidizing bacteria) could be kept by the biofilm on the media, thus giving high NH_4-N removal in the S-MBBR and the MBBR (Shore et al., 2012). Moreover, the results also showed that sponge modified plastic carriers could prevent more nitrifies being washed out with the effluent of the S-MBBR, leading to better NH_4-N removal. Nearly 14% higher T-N elimination achieved in the S-MBBR also implied that simultaneous nitrification and denitrification (SND) process took place, although DO in both MBBRs was maintained at relatively high levels of 5.0-6.0 mg/L in this study. This is due to that the oxic and the anoxic micro-zones could be formed at the outer layer and the inner layer of the biofilm, which was ascribed to DO concentration gradient within the biofilm of media owing to limited oxygen diffusion (Chu and Wang, 2011). Therefore, in the S-MBBR, as the declining DO levels along the inner depth of sponge also favoured the formation of the anoxic zone and permitted more effective denitrification process (Guo et al., 2008a), the sponge modified carriers could enhance the SND process.

Table 7.2. Treatment performance of MBBRs at HRTs of 12 and 6 h during experimental period

Removal efficiency (%)	Phase III		Phase IV	
	S-MBBR	MBBR	S-MBBR	MBBR
DOC	95.63 ± 4.23	93.52 ± 3.25	98.66 ± 1.10	95.89 ± 0.50
COD	94.58 ± 5.06	91.27 ± 4.69	97.52 ± 1.63	93.16 ± 1.45
NH_4-N	83.46 ± 3.98	72.75 ± 5.50	94.17 ± 1.62	81.30 ± 2.03
T-N	74.71 ± 2.06	60.15 ± 6.41	86.66 ± 1.15	71.80 ± 5.01
PO_4-P	70.63 ± 4.15	63.82 ± 6.01	84.52 ± 3.66	70.20 ± 1.89

During the operating period, phosphate can be taken up by phosphorus accumulating organisms in the oxic/anoxic zones (Monclús et al., 2010). As the SND process reduced NO_3-N content in both MBBRs (2.96 ± 0.68 and 3.82 ± 0.84 mg/L in the S-MBBR and the MBBR, respectively), the presence of less NO_3-N in the anoxic zones inside the biofilm resulted in effective PO_4-P release and thus promoted PO_4-P removal (Yuan et al., 2008; Yang et al., 2010a). Furthermore, the sponge modified plastic carrier facilitated PO_4-P elimination by retaining more attached-growth biomass (Guo et al., 2008a). Therefore, the S-MBBR obtained higher PO_4-P removal efficiency (70.63 ± 4.15%) than the MBBR (63.82 ± 6.01%).

At shortened HRT of 6 h, slightly higher DOC and COD removals in the S-MBBR (98.66 ± 1.10% and 97.52 ± 1.63%, respectively) and the MBBR (95.89 ± 0.50% and 93.16 ± 1.45%, respectively) were achieved. Both MBBRs also showed more desirable nutrient removal efficiencies. In addition, better effluent quality in terms of turbidity was observed at HRT of 6 h (17.14 ± 3.12 NTU for the S-MBBR and 56.35 ± 4.72 NTU for the MBBR), compared with higher effluent turbidity values obtained at HRT of 12 h (40.30 ± 3.67 NTU for the S-MBBR and 72.05 ± 4.82 NTU for the MBBR). At HRT of 12 h, the average food to microorganism (F/M) ratios were 0.07 kg BOD_5/kg MLVSS·d for the S-MBBR and 0.17 kg BOD_5/kg MLVSS·d for the MBBR. With decreased HRT of 6 h, F/M ratios increased up to 0.20 and 0.50 kg BOD_5/kg MLVSS·d, respectively. At shorter HRT, F/M ratios in both MBBRs were within the normal range of the activated sludge processes (0.2 to 0.5 kg BOD_5/kg MLVSS·d) (Javid et al., 2013). Thus, adequate substrate could be supplied for the microbial activities (including attached- and suspend-growth), leading to better treatment performance. On the other hand, at longer HRT, lower F/M ratio implied less substrate available for biomass in the reactors, which may cause the risk of sludge bulking and growing filamentous bacteria, thereby deteriorating effluent quality (Javid et al., 2013).

7.3.3. The performance of the integrated MBBR-MBR systems

A. Treatment performance

As shown in Fig. 7.3, the S-MBBR-MBR and the MBBR-MBR showed the excellent DOC removal (98.93 ± 0.89% and 96.64 ± 0.59%, respectively) and COD removal (98.27 ± 0.94% and 94.56 ± 1.06%, respectively). 96.06 ± 1.04% of NH_4-N, 85.60 ± 2.08% of T-N, and 84.08 ± 1.41% of PO_4-P were reduced by the S-MBBR-MBR, while the corresponding pollutant removals in the MBBR-MBR were found to be lower at 82.47 ± 1.88%, 69.59 ± 2.51%, and 68.83 ± 2.36% on average, respectively. It was clear that the MBBR unit could substantially eliminate pollutants in the hybrid systems.

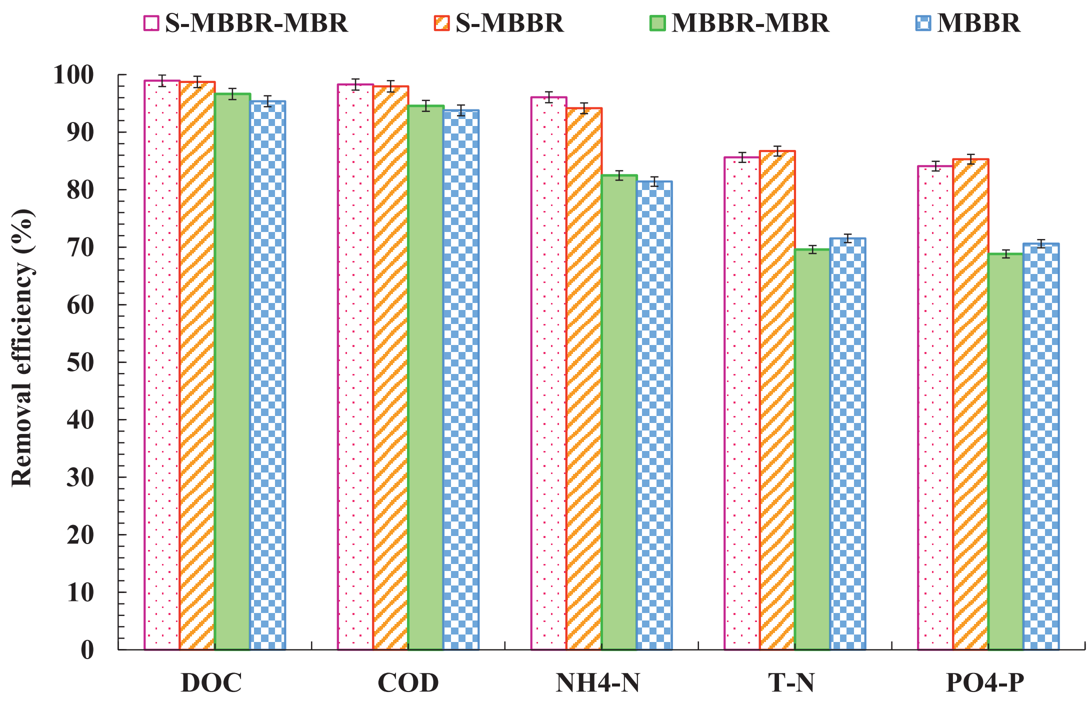

Fig. 7.3. DOC, COD, NH_4-N, T-N and PO_4-P removals in the S-MBBR-MBR, the S-MBBR, the MBBR-MBR, and the MBBR

B. Membrane fouling behaviour

Fig. 7.4 shows the TMP variations for the MBBR-MBR and the S-MBBR-MBR with evolution of time. During the operation, TMP profile of the MBBR-MBR showed a rapid rise until TMP reached 35.0 kPa after 32-day operation, leading to a significantly higher fouling rate of 1.09 kPa/d. On the contrary, a gradual and progressive TMP increment was observed in the S-MBBR-MBR during first 78-day operation with initial TMP of 2.0 kPa and a sudden TMP jump from 20.0 to 35.0 kPa lasting for 7 days thereafter, resutling in a considerably lower fouling rate of 0.39 kPa/d. Thus, more effective membrane fouling mitigation for the S-MBBR-MBR was attributed to extended filtration duration and improved filterability. Moreover, fouling resistance for the fouled membrane in both hybrid systems was measured at the end of the experiment for further fouling analysis. As shown in Table 7.3, total fouling resistances (R_T) were 3.06×10^{12} and 1.42×10^{12} m^{-1} in the MBBR-MBR and the S-MBBR-MBR, respectively. Cake layer in the MBBR-MBR possessed a higher filtration resistance (R_C) of 1.29×10^{12} m^{-1}, while R_C for the S-MBBR-MBR was comparatively lower (0.47×10^{12} m^{-1}), which accounted for 42.16% and 33.10% of R_T, respectively. Pore blocking resistance (R_P) for the MBBR-MBR was higher than 3 times comparing with that for the S-MBBR-MBR, corresponding to 39.54% of R_T and 27.46% of R_T, respectively. The high importance of R_P on R_T in this study may be due to the fact that the MBR unit mainly contained solutes and colloids originating from MBBR effluent, giving rise to serious pore blocking (Defrance et al., 2000; Radjenović et al., 2008). Overall, the S-MBBR-MBR exhibited better membrane permeability by ameliorating pore blocking and cake layer formation.

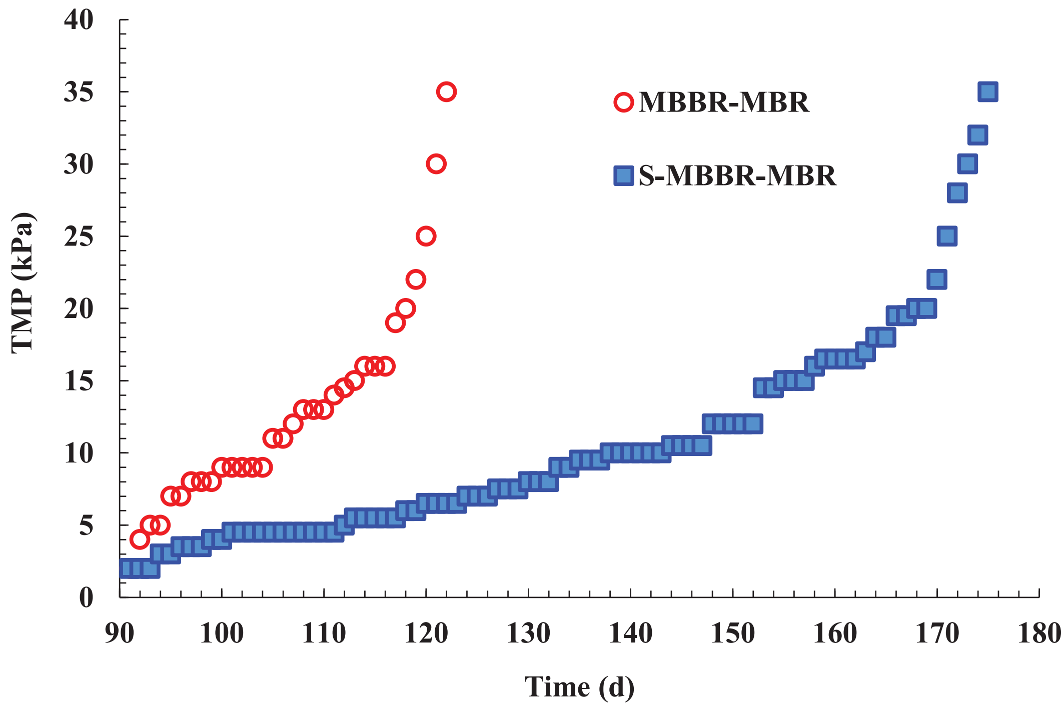

Fig. 7.4. TMP development profile for the MBBR-MBR and the S-MBBR-MBR

Table 7.3. Fouling resistance distribution in the MBBR-MBR and the S-MBBR-MBR

Resistance distribution	MBBR-MBR		S-MBBR-MBR	
	m^{-1}	% of R_T^a	m^{-1}	% of R_T^a
Total	3.06×10^{12}		1.42×10^{12}	
Cake layer	1.29×10^{12}	42.16	0.47×10^{12}	33.10
Pore blocking	1.21×10^{12}	39.54	0.39×10^{12}	27.46
Clean membrane	0.56×10^{12}	18.30	0.56×10^{12}	39.44

R_T = total fouling resistance

It has been reported that EPS facilitated the formation of a cake layer and/or a highly hydrated gel layer containing microbial cells on membrane surface, which further prompted membrane pore blocking (Lin et al., 2014). In addition, SMP encouraged membrane pore blocking, and occupied the space among the particles of cake layer, resulting in a low porosity of cake layer (Domínguez et al., 2012). Figs. 7.5 and 7.6 display the levels of EPS_P and EPS_C of activated sludge, total SMP contents and SMP_P/SMP_C ratio in the supernatant of mixed liquor in the MBR unit at different designated TMP values. Prior to a sudden TMP jump (20 kPa), EPS (EPS_P and EPS_C) of both MBBR-MBRs were at low values and presented slight difference. At TMP of 20 kPa, the notable difference of EPS_P and EPS_C levels between the MBBR-MBR (3.86 and 3.59 mg/L) and the S-MBBR-MBR (2.15 and 1.85 mg/L, respectively) was observed. When TMP reached the highest designated value of 35 kPa, EPS_P and EPS_C contents in the MBBR-MBR reached the highest values of 7.56 and 7.62 mg/L, respectively, which were almost 3 times of the corresponding values for the S-MBBR-MBR (2.31 and 2.85 mg/L, respectively). When TMPs were below 20 kPa, SMP gradually increased from 5.29 to 13.05 mg/L in the MBBR-MBR, while those values maintained at a lower range of 2.99–7.06 mg/L in the S-MBBR-MBR. During the severe membrane fouling period (TMP from 20 to 35 kPa), SMP levels in the MBBR-MBR rose dramatically from 17.58 to 25.86 mg/L. In contrast, the S-MBBR-MBR possessed considerably less SMP and exhibited more stable SMP levels between 7.52 and 9.93 mg/L. The results indicated that total concentrations of SMP were substantially higher than those of EPS in both hybrid systems. Additionally, higher EPS and SMP levels in the MBR unit of the MBBR-MBR were ascribed to higher biomass growth rate in the MBR unit (0.029 g MLVSS/L·d) as compared to that (0.010 g MLVSS/L·d) of the S-MBBR-MBR. Hence, SMP made a greater contribution to membrane fouling development in the MBBR-MBR. Besides, SMP_P/SMP_C ratios in the MBBR-MBR (0.66 ± 0.15) were always lower than those in the S-MBBR-MBR (1.00 ± 0.24) at all the designated TMPs. Since SMP_C could exacerbate irreversible fouling, inducing severe pore blocking and gel layer formation (Jermann et al., 2007), considerably larger amounts of SMP with lower SMP_P/SMP_C ratio of mixed liquor and higher concentrations of EPS of activate sludge were responsible for the elevated R_C and R_P of the MBBR-MBR.

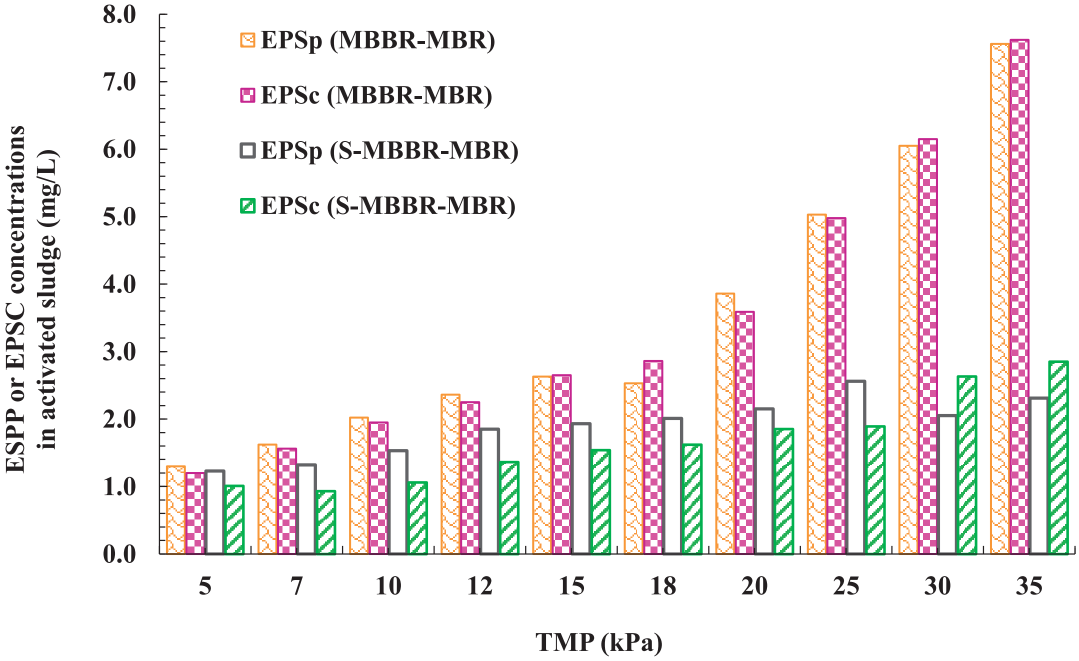

Fig. 7.5. Variations of EPS_P and EPS_C concentrations of activated sludge in the MBR unit at different TMPs

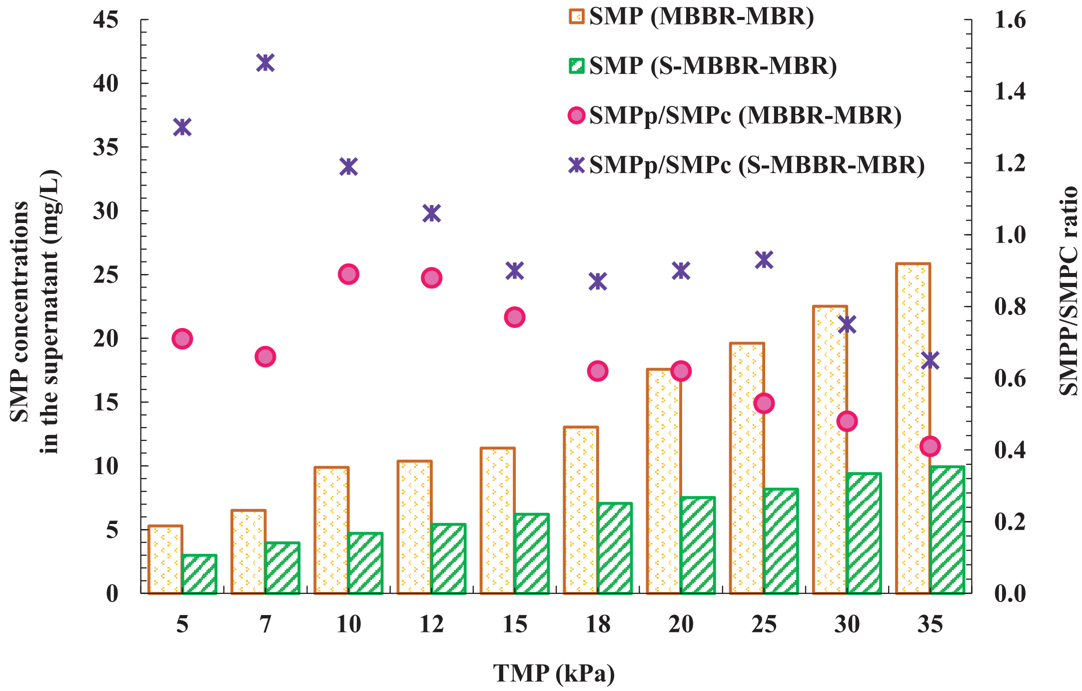

Fig. 7.6. Variations of SMP concentrations and SMP_P/SMP_C ratio of mixed liquor in the MBR unit at different TMPs

The extracted EPS, SMP and BPC from the cake layer were also investigated and characterized by their compositions (including polysaccharides and proteins) (Table 7.4). Both hybrid MBBR-MBR systems had similar EPS levels (including EPS_P and EPS_C) at 4.68 and 3.94 mg/g cake layer for the MBBR-MBR and the S-MBBR-MBR, respectively. Cake layer for the MBBR-MBR was characterized by higher SMP_P and SMP_C than those for the S-MBBR-MBR. BPC_P and BPC_C contents for the MBBR-MBR were 15.27 and 12.16 mg/g cake layer, respectively, whereas those values remarkably decreased for the S-MBBR-MBR, obtaining 8.25 and 5.73 mg/g cake layer, respectively. Hence, cake layer formation for the MBBR-MBR was mainly caused by the accelerated growth of SMP (SMP_P and SMP_C) and BPC (BPC_P and BPC_C) within sludge cake, leading to higher R_C. Moreover, as sponge on the carriers could positively modify the characteristics of suspended biomass through adsorption and biodegradation of attached-biomass of sponge (Deng et al., 2014), it also contributed to the lower SMP and BPC values in the S-MBBR-MBR. In addition, higher drag force due to faster TMP increment in the MBBR-MBR might also enhance the growth of SMP and BPC on membrane surface, which encouraged the development of cake layer, further causing SMP generation by cell lysis and endogenous decay inside the bio-cake layer (Drews et al., 2010). These results again highlighted the significance of SMP on membrane fouling in the MBBR-MBR.

Table 7.4. The compositions of bound EPS, SMP and BPC in membrane cake layer

Reactors	EPS (mg/g cake layer)		SMP (mg/g cake layer)		BPC (mg/g cake layer)	
	EPS_P	EPS_C	SMP_P	SMP_C	BPC_P	BPC_C
S-MBBR-MBR	2.69	1.25	4.13	2.62	8.25	5.73
MBBR-MBR	3.02	1.66	8.63	5.62	15.27	12.16

7.4. Conclusions

This study evaluated the feasibility and performance of sponge modified plastic carriers in both MBBR and MBBR-MBR systems. Compared to MBBR using plastic carriers, sponge modified carriers could not only enhance overall organic and nutrient removal efficiencies, but also prolong the operative time of the hybrid MBBR-MBR system due to efficient fouling reduction. The MBBR-MBR with sponge modified carriers exhibited lower SMP levels in the mixed liquor with higher SMP_P/SMP_C ratio, as well as less pore blocking and cake layer resistances. Therefore, the sponge modified biocarriers could be a promising solution to improve the treatability of the MBBR-MBR system.

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Chapter 8

Conclusions and Recommendations

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8.1. Conclusions

This research mainly aimed to develop the specific integrated membrane bioreactors (MBRs) for membrane fouling control during wastewater treatment for reuse under long-term sustainable operation. The effectiveness of sponge and/or the patented green bioflocculant (Gemfloc®) for membrane fouling alleviation were appraised in submerged MBR (SMBR). The use of integrated moving bed biofilm membrane bioreactor systems with plastic carries and sponge modified plastic carriers for wastewater treatment was also examined. The specific findings from this study are as follows:

- In the conventional MBR (CMBR), soluble microbial products (SMP) and bound extracellular polymeric substances (EPS) in activated sludge have impacts on membrane fouling during the initial stage and later stage, respectively. Comparing to the CMBR, the sponge-submerged MBR (SSMBR) showed less membrane fouling mainly because of the reduction of cake layer formed on membrane surface and the decrease in pore blocking inside the membrane pores. Sponge limited growth of suspended biomass and filamentous bacteria, decreased sludge viscosity, reduced SMP and bound EPS in activated sludge whilst inducing higher zeta potential, greater relative hydrophobicity (RH), and larger particle size of sludge flocs. In addition, less SMP was found on membrane surface in the SSMBR. Hence, the SSMBR possessed lower cake layer resistance (R_C) and pore blocking resistance (R_P).
- Membrane fouling amelioration was accomplished in the MBR-G with regard to less cake formation and pore blocking as well as higher membrane permeability. Gemfloc® positively affected sludge characteristics (including less SMP, higher polysaccharide to protein ratio in EPS (EPS_P/EPS_C), greater RH and zeta potential, larger floc size, and better aggregation ability). The declined R_P and R_C in the MBR-G not only resulted in lower biomass growth rate and sludge viscosity, but also caused less EPS_P, SMP and biopolymer clusters (BPC) within cake layer as compared to the CMBR. According to the profiles of SMP and mixed liquor suspended solids (MLSS), the new mathematical model (the modified resistance-in-series model) was successfully developed to estimated R_C, R_P and transmembrane pressure (TMP). It enables to describe a quantitative understanding on the relationship between sludge properties and membrane fouling behavior in MBR.

- In the SSMBR, more cake layer on membrane surface and pore blocking inside the membrane were increased with decreasing hydraulic retention time (HRT) from 6.67 to 5.33 and 4.00 h, leading to severe membrane fouling. Lower EPS levels in activated sludge and lower polysaccharide to protein ratio in SMP (SMP_P/SMP_C) in mixed liquor obtained at shorter HRT increased R_C and R_P. Higher R_C at the reduced HRT was also ascribed to greater biomass growth and sludge viscosity, together with more EPS and BPC on membrane surface. Membrane fouling in the SSMBRs at different HRTs was not correlated with SMP. At the optimum HRT of 6.67 h, Gemfloc® addition could further mitigate membrane fouling in the SSMBR by enlarging sludge floc size, eliminating SMP of mixed liquor, increasing SMP_P/SMP_C ratio, and reducing SMP and BPC in cake layer.
- The MBBR with sponge modified plastic carriers (S-MBBR) performed better than the MBBR with the plastic carriers (MBBR) in terms of DOC, NH_4-N, T-N and PO_4-P removal. More favorable conditions for nitrogen and phosphorus removals were provided by the sponge modified plastic biocarriers at aerobic/anoxic conditions around the surface of sponge and anoxic/anaerobic conditions inside the sponge. Comparing to the MBBR-MBR with the plastic biocarriers only, the utilization of the sponge modified plastic biocarriers enhanced treatment performance, inhibited membrane fouling, reduced cake layer and prevented membrane pore blocking in the S-MBBR-MBR. Less SMP in mixed liquor, and lower levels of SMP and BPC in cake layer were achieved in the S-MBBR-MBR, resulting in lower R_P and R_C.

8.2. Recommendations

Although this research have evaluated the performance of different integrated MBR systems (including the SSMBR, the MBR-G, the SSMBR with Gemfloc®, the S-MBBR-MBR, or the MBBR-MBR), further studies on these systems to have deeper insight into membrane fouling behavior are necessary. The followings are future research direction:

(1) The identification and characterization of bacteria community are recommended to be added in future studies, as the microbial community was associated with the properties of activated sludge and cake layer. The analysis of molecular weight

distribution should also be performed to determine the molecular fractions of EPS, SMP and BPC, which helps to elaborately explain membrane fouling mechanism;

(2) The clean and fouled membrane surface is also required to be examined by atomic force microscopy (AFM) to have knowledge of the morphology of cake layer;

(3) Influence of temperature or temperature shock on membrane performance in the integrated MBR systems should be examined to simulate temperature fluctuations in their practical application;

(4) High levels of influent cations exerted the adverse influence on MBR performance. Thus, the performance of lab-scale integrated MBR systems needs to be investigated by adding monovalent salt or other cations in synthetic wastewater;

(5) Lab-scale experiments should also be conducted for synthetic wastewater treatment at various COD/N/P ratios to assess the tolerance of the integrated MBR systems for insufficient or excessive nutrients in feed water;

(6) As the presence of micropollutants in the aquatic environment adversely affected aquatic organisms and human health, it is also important to evaluate the effectiveness of the integrated MBR systems for treatment of synthetic wastewater containing micropollutants;

(7) Pilot-scale or full-scale integrated MBR systems for various kinds of wastewater treatment (e.g. industrial, municipal, and domestic wastewater) studies are essential to find more effective ways to control membrane fouling in the large-scale operating conditions.

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Appendix

Appendix

LIST OF PUBLICATIONS

Peer Reviewed Journal Articles

1. **Deng, L.J.**, Guo, W.S., Ngo, H.H., Zhang, J., Liang, S., Xia, S., Zhang, Z., Li, J., 2014. A comparison study on membrane fouling in a sponge-submerged membrane bioreactor and a conventional membrane bioreactor. Bioresource Technology 165, 69–74.
2. **Deng, L.J.**, Guo, W.S., Ngo, H.H., Zuthi, M.F.R., Zhang, J., Liang, S., Li, J., Wang, J., Zhang, X., 2015. Membrane fouling reduction and improvement of sludge characteristics by bioflocculant addition in submerged membrane bioreactor. Separation and purification technology 156, 450–458.
3. **Deng, L.J.**, Guo, W., Ngo, H.H., Du, B., Wei, Q., Tran, N.H., Nguyen, N.C., Chen, S.S., Li, J., 2016. Effects of hydraulic retention time and bioflocculant addition on membrane fouling in a sponge-submerged membrane bioreactor. Bioresource Technology 210, 11−17.
4. **Deng, L.J.**, Guo, W., Ngo, H.H., 2016. New functional biocarriers for enhancing the performance of a hybrid moving bed biofilm reactor-membrane bioreactor system. Bioresource Technology 208, 87−93.

CONTRIBUTIONS TO SCIENTIFIC FORUMS

1. **Deng, L.J.**, Guo, W.S., Ngo, H.H., Zhang, J., Liang, S., Xia, S., Zhang, Z., Li, J., 2014. A comparison study on membrane fouling in a sponge-submerged membrane bioreactor and a conventional membrane bioreactor (Oral presentation). *6th International Conference on Challenges in Environmental Science and Engineering (CESE).* Daegu, Korea, 30 October–2 November, 2013.
2. **Deng, L.**, Guo, W., Ngo, H.H., Du, B., Wei, Q., Tran, N.H., Nguyen, N.C., Chen, S.S., Li, J., 2015. Effects of hydraulic retention time and bioflocculant addition on membrane fouling in a sponge-submerged membrane bioreactor (Oral presentation). *8h International Conference on Challenges in Environmental Science and Engineering (CESE).* Sydney, Australia, 28 September–2 October, 2015.

AWARDS

1. 2014 HDR Students Publication Award from Faculty of Engineering and Information Technology (FEIT), University of Technology, Sydney (UTS) for publishing in high quality journals.
2. 2014 one-off scholarship from Centre for Technology in Water and Wastewater (CTWW), University of Technology, Sydney (UTS).