

**NEW MATHEMATICAL MODELS OF  
BIOMASS VIABILITY AND MEMBRANE  
FOULING IN A MEMBRANE BIOREACTOR**

**By**

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## CERTIFICATE OF ORIGINAL AUTHORSHIP

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

I also certify that the thesis has been written by me. Any help that I have received in my research work and the preparation of the thesis itself has been acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

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## **Abstract**

The optimized performance of a membrane bioreactor (MBR) for wastewater treatment depends not only on the biomass viability but also on the dynamic effects of biomass properties on membrane fouling. This research developed new conceptual mathematical models of biomass viability and fouling using biomass parameters and operational parameters of an MBR. It also presents, as outcomes, new simple and practical models for tracking biomass viability and fouling of an MBR system. The proposed models can be used to track instability in the operation of an MBR, and consequently, measures can be taken to act against instability in the oxygen uptake and for fouling control.

The proposed conceptual models include parameters such as the specific oxygen uptake rate (SOUR) of microorganisms, the soluble or colloidal chemical oxygen demand (COD) of effluent along with the mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) concentrations. The COD parameters of the models represent soluble microbial product (SMP) or bound extra-polymeric substances (bEPS) present within an MBR, offering the possibility of developing practical models with these easily measurable parameters.

The experimental study investigated the effects of biomass parameters on SOUR in a lab-scale sponge submerged MBR (SSMBR) system. Statistical analyses of experimental results indicate that bEPS, SMP, MLSS and MLVSS had significant effects on SOUR and their relative influence on SOUR was  $EPS > bEPS > SMP > MLVSS/MLSS$ . The EPS is considered as a lumped parameter of SMP and bEPS. The progressive change of SMP and bEPS within the bioreactor consistently maintained a negative exponential correlation with SOUR, and two independent models of biomass viability were developed based on

correlations among these parameters. Both the model simulations for biomass viability agreed well with experimental values of the SSMBR system.

The simplified model of membrane fouling considered cake formation on the membrane and its pore blocking as the major processes of fouling. In the model, MLSS is used as a lumped parameter to describe the cake layer formation including the biofilm whereas SMP is assumed as the key contributor to pore fouling. The combined effects of aeration and backwash on detachment of membrane foulants, and new exponential coefficients are included to better describe the exponential increase of transmembrane pressure (TMP). With practical assumptions of these major processes, the new model successfully simulated the fouling phenomena with fairly accurate predictions of the rise of TMP for the operations of two lab-scale submerged MBR systems.

## List of Publications

(06 journal papers published, 02 journal papers submitted, 03 conference presentation)

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## List of Notations and Abbreviations

### A. List of notations

$\Delta P$	Pressure gradient (transmembrane pressure)
$\mu$	Permeate (or effluent) viscosity
$\mu_{20}$	Permeate viscosity at 20 <sup>0</sup> C
$\mu_{\text{aut}}$	Maximum growth rate of autotrophs
$\mu_{\text{het}}$	Maximum growth rate of heterotrophs
$\mu_{\text{SMP}}$	maximum growth rate of SMP
$\mu_{\text{T}}$	Permeate viscosity at T <sup>0</sup> C
$a$	Threshold pore area
$\text{\AA}$	Angstrom
$A_{\text{m}}$ (or $A$ )	Membrane surface area
$A_{\text{t}}$	Total pore area
$B$	First-order endogenous decay rate coefficient
$bE_{\text{i}}/bE_0$	$b\text{EPS}_{\text{i}}/b\text{EPS}_0$
$b_{\text{H}}$	Endogenous respiration rate
$C$	Sludge concentration
$C_0$	Inert COD in the influent
$C_{\text{d}}$	Coefficient of the lifting force of a sludge particle
$C_{\text{m}}$ (or $C_{\text{b}}$ )	Concentration of fouling particles
$C_{\text{m}}^{\text{b}}$	Concentration of particles entering the membrane pore
$\text{COD}_{\text{c,eff}}$	Colloidal COD in the effluent
$\text{COD}_{\text{i}}$	Total inert COD in the influent
$\text{COD}_{\text{perm}}$	COD in the permeate (effluent)
$\text{COD}_{\text{s,eff}}$	Soluble COD in the effluent
$C_{\text{s}}$	Inert COD in the effluent
$C_{\text{SMP}}$	Concentration of soluble particles entering the pores
$d_{\text{f,o}}$ (or $m_{\text{d,o}}$ )	Membrane outer diameter
$d_{\text{i,o}}$ (or $m_{\text{d,i}}$ )	Membrane inner diameter
$d_{\text{p}}$	Sludge particle diameter



$D_s$	Pore area fractal dimension
$E_i/E_o$	EPS <sub>i</sub> /EPS <sub>0</sub>
$f$	Membrane porosity
$f_b$	Fraction of biomass that ends up as microbial products
$f_{bap}$	Fraction of BAP produced during cell lysis
$f_{BAP}$	BAP fraction below critical molecular weight
$f_{EPS}$	growth associated EPS formation coefficient
$f_{EPS,d}$	non-growth associated EPS formation coefficient
$f_s$	fraction of suspended solids produced from EPS hydrolysis/dissolution
$f_{UAP}$	UAP fraction below critical molecular weight
$G$	Geometry factor for fluid flow through a pore
$h_m$	Membrane effective thickness
$I$	Fouling potential index
$J$	Flux (of flow)
$J^*$	Normalized flux
$J_s^*$	Normalized specific flux
$J_{so}$	Specific flux at time zero
$J_t$	Total flux
$K$	constant
$K_1$	UAP formation rate coefficient
$K_2$	BAP formation rate coefficient
$K_{bap}$	Half saturation coefficient for BAP
$K_{eps}$	EPS formation coefficient
$K_{h,bap}$	Hydrolysis rate of BAP
$K_{h,EPS}$	Rate of EPS hydrolysis/dissolution
$k_{hyd}$	First-order hydrolysis rate coefficient
$K_L a_{20}$	Oxygen transfer parameter
$k_{MP}$	Half saturation coefficient for microbial products
$K_{SMP}$	SMP half saturation coefficient for heterotrophs
$k_a$	An empirical parameter
$L_0$	Constant

$L_b$	Biofilm thickness
$M_1/M_2$	MLVSS/MLSS
$MLSS_{\text{sludge}}$	MLSS of sludge
$M_{sc}$	Mass of biomass accumulated on the membrane surface
N	Nitrogen
$N_2O$	Nitrous oxide
NaOCl	Sodium hypochlorite
$n_c$	Exponential coefficient for cake layer resistance
$NH_4\text{-N}$	Ammonia nitrogen
$NO_2\text{-N}$	Nitrite-N
$NO_3\text{-N}$	Nitrate- N
$n_p$	Exponential coefficient for pore fouling resistance
P	Phosphorus
PACl	Poly-aluminium chloride
$PO_4\text{-P}$	Phosphate P
Q	Flow rate
$R^2$	Squared value of correlation coefficient
$R_{\text{biofilm}}$ (or $R_b$ )	Resistance due to biofilm
$r_c$	Specific cake resistance
$R_c(z)$	Time-dependant cake layer resistance
$R_{\text{cake}}$	Resistance due to cake formation
$R_m$	Membrane intrinsic resistance
$R_p$	Pore fouling resistance
$r_p$	Specific resistance of pore fouling
$r_p$	Membrane pore radius
$r_p$	Pearson correlation coefficient
$R_p(z)$	Time-dependant pore blocking resistance
$R_{sc}$	Stable sludge film resistance
$r_{sc}$	Specific resistance of stable sludge film
$R_{sf}$	Dynamic sludge film resistance
$r_{sf}$	Specific resistance of dynamic sludge film

$R_T$	Total resistance
$R_{Tot}$	Total membrane resistance
$S_{BAP}$	BAP (COD units)
$sBOD_5$	Soluble 5-day biological oxygen demand
$sCOD$	Soluble COD
$S_i$	Influent substrate concentration
$SMP_{\text{cake-mem}}$	SMP concentrations in cake layer-membrane interface
$SMP_{\text{permeate}}$	SMP concentrations in the permeate
$SMP_{\text{reactor}}$	SMP concentration within the bioreactor
$S_{ND}$	Soluble biodegradable organic nitrogen
$S_{NH}$	Ammonia or ammonium nitrogen
$SP_i/SP_0$	$SMP_i/SMP_0$
$S_{PO_4}$	Soluble phosphate
$S_S$	Readily biodegradable substrate
$S_{UAP}$	UAP (COD units)
$t$	Filtration period
$t_f$	Elapsed filtration time
$u_b$	Biofilm detachment rate during backwashing
$u_{f,a}$	EPS growth rate due to attachment
$u_{f,d}$	EPS growth rate due to detachment
$U_{Lr}$	Crossflow velocity of tap water
$U_{sr}$	Crossflow velocity of mixed liquor
$V$	Volume of permeate passed through the available membrane area
$V_f$	Water production within the filtration period of the operation cycle
$V_f$	Permeate volume after time $t_f$
$V_s$	Cumulative volume of permeate per membrane surface area
$X$	Biomass concentration
$X_A$	Active autotrophic biomass
$X_{\text{aut}}$	Autotrophic Biomass Concentration
$X_{\text{EPS}}$	EPS concentration
$X_{\text{GLY}}$	Stored glycogen in PAOs
$X_{\text{het}}$	Heterotrophic biomass concentration

$X_{ND}$	Particulate biodegradable organic nitrogen
$X_P$	Particulates from biomass decay
$X_S$	Slowly biodegradable substrate
$Y_{BAP}$	BAP formation constant
$Y_{H2}$	anoxic growth yield coefficient
$Y_{MP}$	Yield coefficient for growth on microbial products
$z_c$	Depth of cake layer
$\alpha$	Stickiness of biomass particles
$\alpha_b$	Specific resistance of biofilm
$\alpha_f$	Membrane porosity reduction coefficient
$\alpha$ -factor	Oxygen transfer rate
$\alpha_{max}$	An empirical parameter
$\alpha_o$	An empirical parameter
$\alpha_p$	An empirical parameter
$\alpha_v$	Air scouring coefficient
$\beta$	Erosion rate coefficient of the dynamic sludge
$\beta$	Soluble Fouling Index (MFI) coefficient
$\gamma$	A coefficient for dynamic sludge compression
$\gamma$	Suspended solids MFI coefficient
$\gamma_{MP,A}$	Autotrophic microbial product formation constant
$\gamma_{MP,H}$	Heterotrophic microbial product formation constant
$\eta$	Viscosity of the permeate
$\eta_f$	Average fraction of soluble particles that accumulate in the pores
$\theta$	Pore tortuosity
$\theta_f$	Filtration period
$v_{air}$	Scouring air surface velocity
$\rho_b$	Biofilm density
$\rho_c$	Density of cake layer
$\rho_p$	Particle density

*B. List of abbreviations*

AOB	Ammonia-oxidizing bacteria
APHA	American public health association
ASMs	Activated sludge models
BAPs	Biomass associated products
BEPR	Biological excess phosphorus removal
bEPS	Bound extracellular polymeric substances
BF-MBR	Hybrid biofilm MBR
bio-P	Biological phosphorus
BNRS	Biological nutrient removal system
BOD	Biological oxygen demand
BPC	Biopolymeric clusters
C/N	Carbon to Nitrogen
C/P	Carbon to Phosphorus
CAS	Conventional activated sludge
CH	Carbohydrate
CIFI	Chemical-irreversible FI
CMBR	Conventional MBR
COD	Chemical oxygen demand
DOC	Dissolved organic carbon
DOM	Dissolved organic matter
EBPR	Enhanced biological phosphorus removal
ED	Electrodialysis
EMBRs	Extractive MBRs
EPS	Extracellular polymeric substances
F/M	Food to microorganisms ratio
FACASM1	Fully Coupled ASM1
FI	Fouling index
FS	Flat sheet
GAOs	Glycogen accumulating organisms
HF	Hollow fibre

List of abbreviations

HIFI	Hydraulic-irreversible FI
HRFI	Reversible FI by hydraulic backwash
HRT	Hydraulic retention time
IC	Inorganic carbon
IUPAC	International union of pure and applied chemistry (IUPAC)
IWA	International water association
LC-OCD	Liquid chromatography- organic carbon detection
MABRs	Membrane-aerated biofilm reactors
MBBR	Moving bed biofilm reactors
MBR	Membrane bioreactor
MF	Microfiltration
MFI <sub>0.45</sub>	Modified fouling index
MFI <sub>MBR</sub>	MFI of MBR
MFI <sub>sol</sub>	MFI of soluble particles
MFI <sub>SS</sub>	MFI of suspended particles
MLSS	Mixed liquor suspended solid
MLVSS	Mixed liquor volatile suspended solids
MT	Mutitube
NF	Nanofiltration
NFFB	Non-oven fabric filter bag
OHs	Ordinary heterotrophic organisms
OLR	Organic loading rate
OUR	Oxygen uptake rate
P	Phosphorus
PAC	Powdered activated carbon
PAOs	Phosphorus accumulating organisms
PFC	Polymeric ferric chloride
PHA	Polyhydroxyalkanoates
PN	Protein
poly-P	Polyphosphate
PS	Polysaccharide

PUS	Polyster-urethane sponge
PVDF	Polyvinylidene fluoride
R	Resistance
RBCOD	Readily biodegradable COD
RO	Reverse osmosis
SBNR	Shortcut biological nitrogen removal
sBOD <sub>5</sub>	Soluble 5-day biological oxygen demand
SCOD	Slowly biodegradable COD
SDI	Silt density index
SEM	Scanning electron micrographs
sEPS	Soluble EPS
SMBR	Submerged MBR
SMBR	Submerged membrane bioreactor
SMP	Soluble microbial products
SOUR	Specific oxygen uptake rate
SRT	Sludge retention time
SS	Suspended solids
SSMBR	Sponge submerged MBR
SSMBR	Sponge submerged MBR
T	Temperature
TC	Total carbon
TEP	Transparent exopolymeric particles
TFI	Total FI
TKN	Total kjeldahl nitrogen
TMP	Transmembrane pressure
TOC	Total organic carbon
TSS	Total suspended solids
TUDP	Technical university of Delf phosphorus
UAPs	Utilization associated products
UCT	University of Cape Town
UCTPHO	UCT phosphorus

List of abbreviations

UF	Ultrafiltration
UMFI	Unified FI
UTS	University of Technology Sydney
UV	Ultraviolet
VSS	Volatile suspended solids





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# **CHAPTER 1**

## **Introduction**

## 1.1 Background

Membrane bioreactor (MBR) has been increasingly used in the wastewater treatment industries around the world because of its reduced physical footprint and less excess sludge production as compared to that of other conventional activated sludge (CAS) systems (Keskes et al., 2012; Li et al., 2008; Lin et al., 2011; Meng et al., 2009; Song et al., 2007; Zhang et al., 2011). However, membrane fouling is still a major hindrance to the widespread commercial application of MBR technology despite its proven advantages over other options of wastewater treatment (Drews et al., 2010; Mannina and Cosenza et al., 2013). Moreover, modification in biomass viability is very likely (Trapani et al., 2011) due to the dynamic changes in biological processes and operational conditions in an MBR subject to the operation of the treatment system with typically higher mixed liquor suspended solid (MLSS) concentrations and medium to high sludge retention time (SRT). Therefore, sustaining a desired rate of oxygen transfer for microbial activity along with maintaining the system's operation with controlled membrane fouling are critically important for sustainable operation of the MBR systems.

As biodegradation potential of microbial culture and filtration performance play critical roles for the sustainable operation of MBR systems (Başaran et al., 2014), assessment of biomass viability and membrane fouling have appeared as major concerns for the researchers working in this field of research. However, only a few researchers (Hasar et al., 2002; Hasar and Kinachi, 2004; Zuthi et al., 2013, 2014) attempted to study biomass viability since complete characterization of biomass culture proved to be troublesome and an expensive procedure as well. Numerous studies have so far been conducted to identify/investigate the foulants (Gao et al., 2010; Lin et al., 2009) and the processes involved with membrane fouling (Zhang et al., 2013; Kim et al 2013), and

hence to devise strategies to control fouling (Meng et al, 2009; Drews et al., 2010; Mannina and Cosenza, 2013) for more efficient operation of the MBR systems. Nevertheless, a reasonably accurate prediction of the fouling behaviour is still an “open challenge” for researchers (Cosenza et al., 2013) pursuing further research in this arena.

The biomass viability, as may be defined within the scope of the research, is the capacity of the living microbial population of the biomass to operate or be sustained under normal operating conditions of the MBR systems. The biomass viability may be affected due to the inhibitory effects of certain microbiological products or processes especially due to few crucial specificities of the MBR system. The microbial culture in the activated sludge systems undergoes changes due to the continuous changes of microbial communities in structure, population and activity with time (Chipasha and Medrzycka, 2008). An assessment of the correlations of the specific oxygen uptake rate (SOUR) of microorganisms to the potential biomass parameters may indicate whether or not the biomass viability is being inhibited by certain biomass product/s that are accumulated within the bioreactor after being rejected by the membrane. However, only a few studies (Başaran et al., 2014; Clouzot et al., 2011; Germain et al., 2007; Han et al., 2005; Hasar et al., 2002; Lee et al., 2003; Malamis et al., 2011; Zuthi et al., 2014) have assessed factors that can affect the oxygen transfer rate and the viability of the microbial culture in an MBR. Among the MBR plant characteristics and operating conditions, the influence of membrane configuration (Clouzot et al., 2011) and sludge retention time (SRT) (Germain et al., 2005; Han et al., 2005) on biomass viability has been investigated. As the specific oxygen uptake rate (SOUR) by microorganisms is associated with substrate utilization rate (Kim et al., 2001), the SOUR was used as an indicator of biomass viability in many studies (Chen et al., 2012; Ngo et al., 2008; Nguyen et al., 2012; Villain and Marrot, 2013). Due to the microbial metabolism within

the MBR, different types of organic polymeric compounds such as bound extracellular polymeric substances (bEPS) and soluble microbial products (SMP) are released to the liquid phase. The roles and effects of these microbial products have also attracted significant scientific attention in this field since these products can also cause membrane fouling. The inhibitory effects of microbial products on microbial activities have been acknowledged in very few studies (Germain et al., 2007; Chuboda 1984; Huang et al., 2000; Lee et al., 2003; Rojas et al., 2005; Zuthi et al., 2014). The solids concentration and carbohydrate fraction of the EPS and the chemical oxygen demand (COD) of the SMP were identified as the factors affecting the oxygen transfer efficiency (Germain et al., 2007). A strong negative correlation between microbial activity and concentration of EPS was observed in a few studies (Lee et al., 2003; Rojas et al., 2005).

To date, only a few researchers (Hasar et al., 2002; Hasar and Kinachi, 2004) attempted to characterize the functional relationships between the factors affecting SOUR and the microbial activity of a submerged MBR (SMBR) system. Hasar and Kinachi (2004) presented an empirical mathematical model of the biomass viability of an SMBR taking into account the mixed liquor suspended solids (MLSS), mixed liquor volatile suspended solids (MLVSS), and inert COD in the influent and effluent. The inert COD was represented in the model mainly as an indicator parameter of SMP in the bioreactor, whereas larger microbial aggregates such as bEPS and colloids were not included in the model. Zuthi et al. (2013) proposed modifications of the above mentioned model in a conceptual model of biomass viability which correlated SOUR with MLVSS/MLSS, and soluble/colloidal inert COD in the effluent as indicator parameters of SMP/bEPS in the bioreactor. The modified model considers the colloidal COD in the effluent as a representative parameter of bEPS in the bioreactor. Nevertheless, soluble/colloidal particles may be retained within the bioreactor by small

membrane pore size leaving no traces of soluble/colloidal COD in the effluent. Therefore, dynamic membrane rejection efficiency of these particles should be included in these models of biomass viability.

Membrane fouling is the undesirable deposition and accumulation of microorganisms, colloids, solutes, and cell debris within/on membranes (Meng et al., 2009) resulting in reduced productivity and the need for frequent cleaning or replacement of the membrane (Kim et al., 2013; Mannina and Cosenza, 2013; Zhang et al., 2014). Suspended particles of comparable sizes to the membrane pore (colloids) and of smaller sizes than the membrane pore (soluble particles) cause internal fouling by pore clogging and pore constriction (Busch et al., 2007). External fouling is ascribed to the cake layer formation associated as well as with the formation of biofilm. In most of the research studies, fouling due to the cake layer formation is considered the major mechanism of fouling (Guo et al., 2012; Johir et al., 2013; Lin et al., 2009; Meng et al., 2009; Wang and Wu, 2009). The cake layer on the membrane is formed by particles larger than the membrane pores and the process is dependent on the concentration of MLSS, membrane flux and the scouring energy induced by the aeration (Giraldo and LeChevallier, 2006). The extra cellular polymeric substances (EPS), especially the SMPs, have also appeared as concerns when found integrated within the cake layer formation and in the pore foulants. Moreover, all of the fouling mechanisms are time-dependent adding further complexity to the phenomena.

Due to the high number of interactions between physio-biochemical conditions within the MBR, a mathematical model-based approach has been adopted to gain a deeper insight into the fouling phenomena. A significant number of modelling studies have been performed on membrane fouling employing a resistance-in-series model in the last decade (e.g. Busch et al., 2007; Li and Wang 2006; Mannina et al., 2011;

Wintgens et al., 2003). Wintgens et al. (2003) presented a semi-empirical model considering the cake layer formation, concentration polarization and irreversible resistance for an SBR system. Reversible and irreversible cake layer, pore blocking and the feed side hydrodynamics were covered in a model by Li and Wang (2006). Giraldo and LeChevallier (2006) provided a set of differential equations to correlate the exponential change of transmembrane pressure (TMP) with cake growth and associated headloss over time, removal of SMP within cakes and in membrane pores, and the associated effects of operational factors on fouling reductions. Nagaoka et al. (1998) developed an approach for the biofilm model of a membrane-based activated sludge system to study the influence of EPS on the membrane surface and the biofouling. Navaratna et al. (2012) provided an explanation of the biofilm production considering the production, accumulation and consolidation of EPS onto the membrane surface as well as the aeration induced effects of shear. However, separate descriptions of the complex effects of different fouling processes and removal of foulants could hardly be integrated to correlate well with basic external measures of fouling such as the practically observed transmembrane pressure (TMP) differences during the operation of MBR systems. None of the above studies has been conducted taking into account the combined effect of aeration and backwashing on membrane fouling. The loss in microbial activity (biomass viability) has not been effectively linked to the membrane fouling phenomena.

## **1.2 Motivations and objectives of this study**

The drawbacks of the bioprocess operations that affect the biomass viability seem to contribute to the membrane fouling process as well. In this context, these two important operational aspects of the MBR are addressed within the scope of the research by experimental studies followed by mathematical modelling of biomass viability and

membrane fouling of a laboratory-scale aerobic SBR system. The major focus of the research is to develop simple but efficient models of biomass viability and membrane fouling which can be applied for better management and operational control of MBR systems. In this context, a summary of the major objectives of this research is as follows:

- i) To conduct an in-depth review of the bioprocesses, filtration and membrane fouling processes of MBR systems. As the fundamental mechanisms of the MBR processes are derived from the conventional activated sludge process, the review is necessarily focused on all aspects of the activated sludge processes followed by the review of the relevant mathematical modelling studies.
- ii) To develop conceptual mathematical models of biomass viability and membrane fouling from an extended literature review and analytical studies based on the findings of previously reported studies.
- iii) To analyse experimental data of several lab-scale SBR systems to assess the operational efficiency/instability linked with potential biomass parameters.
- iv) To develop mathematical models of biomass viability and membrane fouling whilst describing efficient procedures for calibration of the models using experimental results of lab-scale SBR systems.

### **1.3 Organization and major contents of the thesis**

The thesis has been structured into eight main chapters. The research approach of this study is presented in Figure 1.1. The mathematical modelling programs are attached as an Appendix at the end of the thesis.

The first chapter is an introductory chapter where the background, motivations and objectives of the study are stated. This chapter outlines the organization of the thesis with brief information about the contents.

Chapter two presents an extensive review of the fundamentals of the MBR treatment system, its microbiological aspects, membrane fouling processes and other associated factors. The literature review is necessarily elaborated on the bioprocess of the activated sludge process since fundamental treatment mechanisms of the MBR processes are derived from the conventional activated sludge bioprocess only with the sedimentation of conventional systems being replaced in the MBR with a membrane for the solid-liquid separation. The later half of the literature review is about the state-of-the-art in relevant mathematical modelling studies principally focused on the bio-oxidation kinetics of organics and nutrient removal processes. The discussions are extended to factors that may affect the biokinetics and membrane fouling, and how these factors are accounted for in the mathematical models.

Chapter three presents conceptual mathematical models of biomass viability and membrane fouling that are developed from an extended literature review and analytical studies based on the findings of the previously reported studies. The development of conceptual modelling has helped in designing the experimental program of the study and deciding the parameters to be measured in the experimental study of lab-scale SMBR systems.

Chapter four describes the experimental set-up of the lab-scale SMBR system. Also described in this chapter are the methods of measurements of biological and operational parameters of the MBR along with the description of materials and analytical methods used in the study.



Chapter five discusses experimental results of several lab-scale SMBR systems where operational efficiencies or instabilities of the MBR treatment are assessed. The assessment has been done to find correlations to biomass parameters that are reported in the literature as affecting biomass viability and contributing to membrane fouling.

Chapter six and chapter seven respectively describe the development of new mathematical models of biomass viability and membrane fouling. Both the chapters include description of the calibration procedure of the models that have been done using experimental results of a conventional MBR (CMBR) and results of a sponge submerged MBR (SSMBR) system. These chapters also cover validation of the models using additional data from the same experimental MBR systems.

Chapter eight is the final chapter, where a summary of the major contributions of this research is presented. The thesis concludes with recommendations for future work in this area of research.

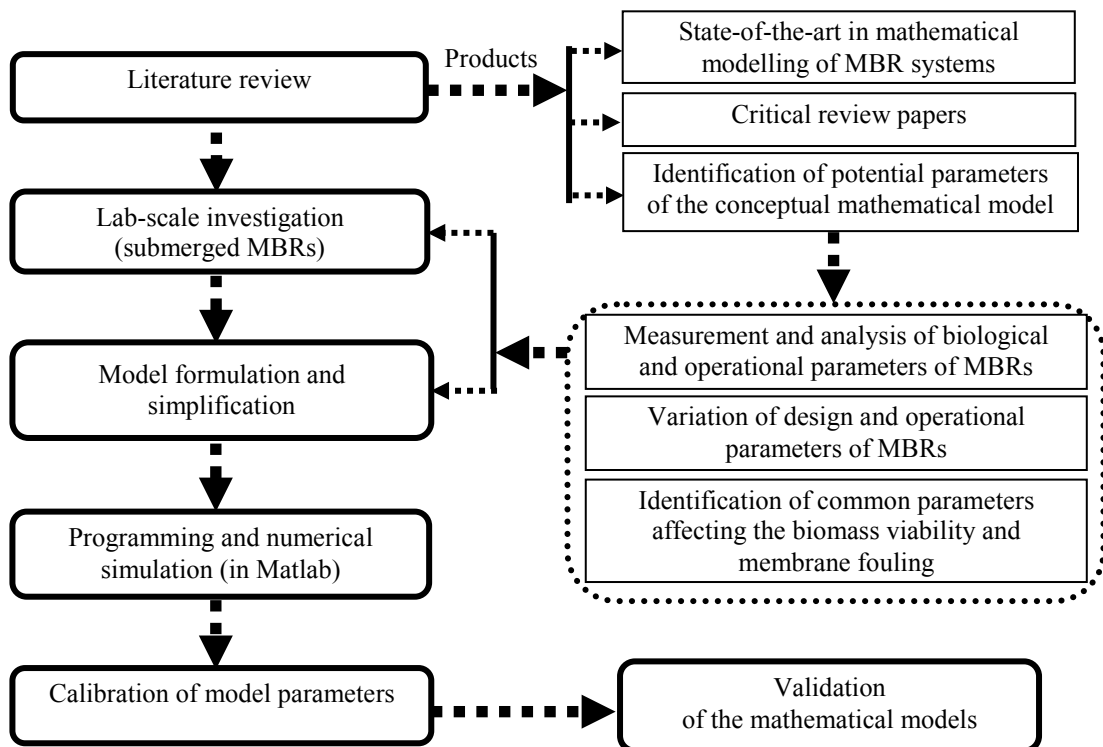


Figure 1.1 Research approach of the study



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## **CHAPTER 2**

### **Literature Review**

## 2.1 Introduction

The efforts for modelling of wastewater treatment systems, so far, have typically been aimed at describing either the activated sludge processes such as how the fractions of biomass/wastewater contribute to the treatment processes or the various aspects of engineering (cost effective design and operation). The development of Activated Sludge Models (ASMs) (Gujer et al., 1999; Henze et al., 1999, 1995, 1987) was an important evolution in the modelling of conventional activated sludge (CAS) processes and their use is now very well established. However, although these ASMs were initially developed to describe CAS processes, they have simply been transferred and applied to MBR processes with little or no modifications. These aspects raise the question as to what extent the ASM framework is applicable to MBR processes (Fenu et al., 2010).

Mathematical modelling studies of MBR systems with major adoption of the ASMs are scattered through the literature. Through a concise and structured overview of the past developments of ASMs and current state-of-the-art in biological modelling of MBR processes, this review mainly explores modelling techniques applied to MBR processes. To cover the whole range of issues relevant to this study, the literature review presented in this chapter is necessarily focused on the fundamentals of membrane processes, MBR definitions and history, microbiological and physical aspects of membrane fouling in an MBR, mathematical modelling of activated sludge treatment and MBR fouling processes followed by the relevant discussions of the models of biomass viability. Particular emphasis is placed on bio-kinetics and SMP or EPS modelling which may be highly linked with the mathematical models of biomass viability and membrane fouling.

## 2.2 Membrane bioreactor for wastewater treatment

### 2.2.1 Membrane processes and applications

The chemical engineering processes involving the use of membranes for phase separation are termed “membrane processes”. According to the International Union of Pure and Applied Chemistry (IUPAC), a membrane is described as “a structure, having lateral dimensions much greater than its thickness, through which mass transfer may occur under a variety of driving forces”. A membrane acts as a barrier to prevent mass movement of selected phases, but allows movement of remaining phases. The phases in an MBR for wastewater treatment include solid phase (suspended solids, dissolved solids, etc.) and liquid phase (water). The driving forces in membrane processes in an MBR are gradients of pressure (transmembrane pressure difference or TMP) (Baker, 2004; Mulder, 1996). Other than driving forces, the nature of a membrane i.e., its pore size and material determines the type of application ranging from the separation of microscopic particles to the separation of molecules of an identical size.

Depending on the mean pore size of the membrane, the membrane processes in MBR include at least five main subcategories for processing water and wastewater (Wang and Menon, 2009). *Microfiltration (MF)* is a pressure-filtration process for the separation of suspended solids (SS) in the particle size-range of about 0.08 to 10  $\mu\text{m}$ . The hydraulic pressure (TMP) applied in MF is about 1 to 2 bars, primarily for overcoming resistance of the cake layer of foulants formed on the membrane. *Ultrafiltration (UF)* is typically used for the separation of macromolecular solids in the particle size range of about 0.01 to 0.1  $\mu\text{m}$  and the TMP variations in the process typically range from 1 to 7 bars (Wang and Menon, 2009). The MF and UF are the major applications of membrane processes in the MBR treatment system to separate suspended solids from the influent stream. However, particles smaller than the mean pore size of the membrane (e.g. dissolved

solids) may also be retained due to filtration effects of the biofilm or sludge cake layer that gradually builds up on the membrane surface. *Nanofiltration (NF)* membranes are multiple-layer thin-film composites of polymer consisting of negatively charged chemical groups that reject movement of molecular solids and multivalent salts equivalent to its pore size in the range of 0.0005 to 0.007  $\mu\text{m}$  (Wang and Menon, 2009). *Reverse osmosis (RO)* membranes are mainly made of cellulose acetate with the pore sizes of about 5 to 20  $\text{\AA}$ , and are used for rejecting salts and organics at osmotic pressure of 20 to 50 bars. *Electrodialysis (ED)* membrane uses voltage or current as the driving force to separate ionic solutes in the range of 0.00025 to 0.08  $\mu\text{m}$ , depending on the pore size of ED membranes.

### **2.2.2 MBR definitions, advantages and history of MBR development**

The principal biological processes used for wastewater treatment can be divided into two main categories: attached growth (or biofilm) and suspended growth process (Tchobanoglous et al., 2003). The principle behind the methods of biological wastewater treatment is to introduce contact with bacteria (cells) to provide its feed on the organic materials in the wastewater, and hence to reduce the biological oxygen demand (BOD) content of the wastewater. Among the attached growth processes, commonly applied treatment processes are the trickling filters, moving bed biofilm reactors (MBBR) and rotating biological contactors. Suspended growth processes are typically aerated systems with activated sludge where biomass is freely suspended in the wastewater and is mixed. Traditionally, the biological treatment system adopts either sedimentation clarification or dissolved air flotation clarification for solids-liquid separation and for microorganisms (sludge) return.

When the quality of influent raw water is good, the membrane process alone should be feasible for treating wastewater. However, during the membrane process operation,

dynamic build-up of a sludge cake layer on the surface of the membrane and adsorption of foulants within the membrane pore structure are the major mechanisms of membrane fouling responsible for membrane flux decline (Wang and Menon, 2009). In the typical treatment facilities of industrial wastewater or domestic sewage, the conventional physical-chemical pretreatment processes are not economically feasible due to the high organic load in the influent streams. As a consequence, the membrane filtration process is preceded by biological treatment in a bioreactor which is a more cost-effective treatment than the physical-chemical processes. When a biological process is used in a reactor in conjunction with a membrane process (either MF or UF), the entire process system becomes a membrane bioreactor (MBR). Wastewater treatment by MBRs involves the biological pretreatment in a suspended growth bioreactor for biochemical reactions such as bio-oxidation, nitrification and denitrification followed by a membrane separator for sequent phase (solid-liquid) separation. The membrane materials used typically in MBRs are organic polymers, e.g., polyethen, polypropylene and polyvinylidene fluoride (PVDF) membranes (Judd, 2006).

Among the many advantages offered by the MBR process over the CAS processes, the smaller footprint and the superior quality of the effluent are generally of critical significance. An MBR effectively displaces three separate process steps in a conventional sewage treatment plant i.e., primary settling, activated sludge system and disinfection; demands only the initial screening stage be upgraded to limit impacts on the membrane. However, MBRs are to some extent constrained by greater process complexity and higher capital equipment and operating costs, as well as other critical issues such as a greater foaming propensity, greater aeration requirements for both the biological and membrane fouling/clogging control, a less readily de-waterable sludge product and generally greater sensitivity to shock loads (Judd, 2006).

The concept of an activated sludge process coupled with UF and MF membrane for biomass separation was first developed and commercialized in the late 1960s by Dorr-Oliver (Bemberis et al., 1971; Smith et al., 1969;), with application to ship-board sewage treatment (Bailey et al., 1971) and other bench-scale MBR systems (Hardt et al., 1970; Smith et al., 1969). The initial MBR systems were all based on side-stream configurations associated with high membrane cost and high energy requirement, as opposed to the now more commercially viable submerged configuration. The submerged MBR was introduced in the late 1980s to reduce the high energy costs (Yamamoto et al., 1989). From the late 1980s to date, rapid commercial developments of MBRs have been taking place around the world. Some pioneer developers of MBR systems around the world include the Thetford Systems of the USA, Japanese company Kubota, Canadian company Zenon Environmental, Kazuo Yamamoto and his co-workers' (Yamamoto et al., 1989) developments of the hollow fibre membrane system.

### ***2.2.3 Classification and configurations of MBRs***

MBRs can be generally classified into three groups: biomass separation MBRs, membrane aeration bioreactors - also called membrane-aerated biofilm reactors (MABRs) and extractive MBRs (EMBRs) (Figure 2.1). Biomass separation MBRs are the most often used MBRs. Their key feature is to use an MF or UF membrane to replace the conventional secondary settling tank in an activated sludge process to separate the biomass from the water phase. The configuration can refer to both the MBR process (and specifically how the membrane is integrated with the bioreactor) or the membrane module. There are two main MBR process configurations (Figure 2.2): submerged or immersed and side-stream.

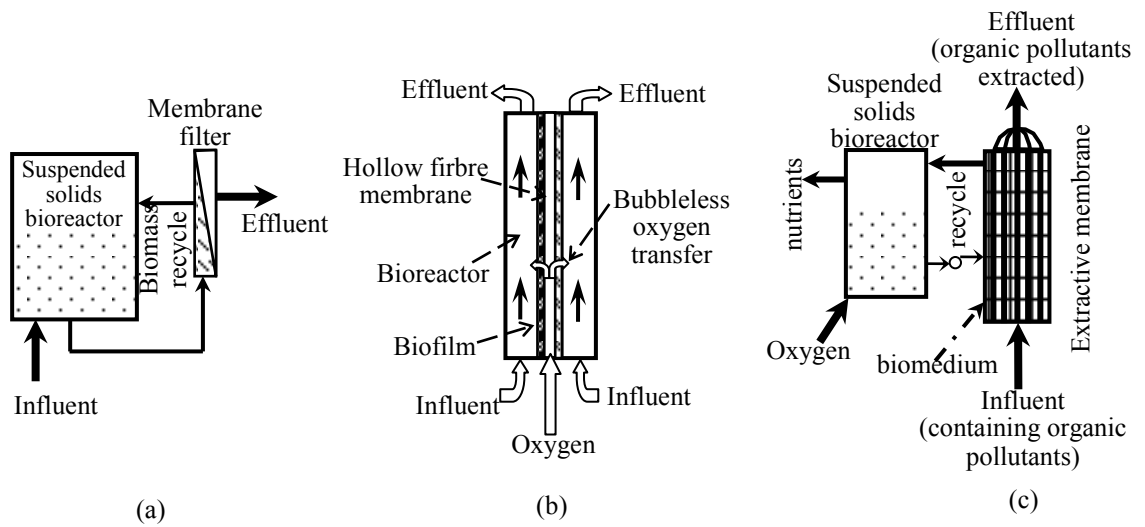


Figure 2.1 Three types of MBR processes: (a) Biomass separation MBRs (b) membrane aeration bioreactors (c) Extraction MBRs (adapted from Stephenson et al., 2009)

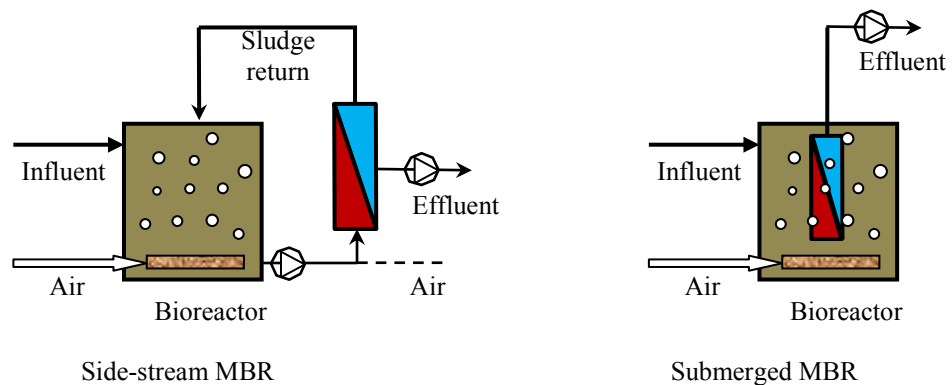


Figure 2.2 Configuration of side stream and submerged MBRs

(adapted from Jiang, 2007)

There are also two modes of hydraulic operation: pumped and air-lift (vacuum pressure), the latter almost exclusively used for submerged systems and the former for side streams. Whilst a number of membrane geometries exist in the membrane market place in general, three predominate in existing commercial MBR technologies, these being flat sheet (FS), hollow fibre (HF) and multitube (MT).

The current trend in MBR design tends to favour submerged over sidestream configurations in the majority of the studies dealing with domestic wastewater treatment (Gander et al., 2000). In side-stream MBRs, the membrane module is separated from the



main bioreactor. The sludge in the bioreactor is pumped into a membrane module, where a permeate stream is generated and a concentrated sludge stream is retained by the membrane and returned to the bioreactor. In the early development of side-stream MBRs, both the transmembrane pressure (TMP) and crossflow velocity were generated by the recirculation pump. However, a few modifications were made to reduce the high energy consumption associated with the side-stream configuration. The biggest advantage of submerged over side-stream configuration is the energy saving by using coarse bubble aeration and lower fluxes (10-30 L/m<sup>2</sup>.h), instead of a high rate recirculation pump and high fluxes (40-100 L/m<sup>2</sup>.h) in side-stream MBRs. The submerged MBR (SMBR) is also easy to build requiring less equipment. The capillary and hollow fibre membranes, used in many submerged MBRs, have very high packing density and low cost which make it feasible to use more membranes and operate at lower fluxes.

#### ***2.2.4 MBR performance and operating factors***

A complete understanding of the interrelationship among the MBR performance and operating factors is yet to be established by research findings. The performance of the MBR, according to its construction, is assessed in two phase operations of the systems namely the biological treatment and the physical membrane filtration process. The factors that determine the MBR operational efficiency in these two phases are briefly described below.

##### *A. Microbiological and MBR design or operational factors for biological treatment*

The MBR technology combines the biochemical and sludge-separation stage as a one step process simultaneously occurring in a bioreactor. There occurs a continuous generation of new sludge with the consumption of feed organic materials, while some

sludge mass is decayed by endogenous respiration (Drews et al., 2010). The maintenance energy is defined by the requirement for the endogenous respiration (Pirt, 1965) which is typically provided with high sludge concentration. However, there should be a balance between the energy available to the microbes (i.e. by the supply of substrate) and the maintenance energy for the optimum biological treatment. The energy supply to the microbes is typically encouraged by increasing the SRT which eventually increases the biomass concentration. On the other hand, external substrate is used only for sustaining the bacterial vital functions with limiting nutrient supply. Moreover, the sludge loading becomes lower with increased biomass concentration (Shahalam and Al-Smadi, 2005); and little or no excess sludge is produced at this condition (Radjenovic et al., 2008; Liu et.al., 2005; Yamamoto, 1989; Yoon et.al., 2002). The retained excess biomass causes the cell dormancy and death affecting the viability of population (Radjenovic et al., 2008; Cicek et. al., 2001; Macomber et. al., 2005), and hence, the operational viability of the MBR is affected as well.

Better organics and nutrient removal in the MBR is achieved especially due to the complete particulate retention by the membrane and avoidance of biomass washout problems commonly encountered in the CAS processes (Daigger et al., 2010; Lesjean et al., 2003; Meng et al., 2012). The ability of microbes to form flocs is vital for the organics and nutrient removal by the MBR biological treatment process since the floc structures enable not only the adsorption of soluble substrates but also the adsorption of the colloidal matter and macromolecules in wastewaters (Radjenovic et al., 2008; Michael and Fikret, 2002; Liwarska-Bizukojc and Bizukojc, 2005). In this regard, the characteristics of sludge morphology (dispersed bacteria, lower amount of large filamentous bacteria, floc densification) certainly play the major role, which in turn, influence the organics and nutrient removal efficiencies. Although a balanced

concentration of the floc-associated EPS (colloidal bEPS) contribute for better nutrient removal of the MBR system, an excess concentration of this microbial product and SMP in the bioreactor are found to pose undesirable effects, for example by inhibiting microbial activity (Germain et al., 2007; Huang et al., 2000; Rojas et al., 2005) and also by potentially contributing to the membrane fouling. The effects of SRT on both membrane performance and fouling potential have recently been investigated by a few researchers (Dereli et al., 2014; Villain and Marrot, 2013; Chen et al., 2012), where correlations between potential biomass parameters (e.g. SMP) and optimum SRT are established indicating that the dynamic biomass properties especially that concerning the EPS have significant effects on both biological and physical (membrane permeability) performance of MBR.

The nitrification capability of the activated sludge is enhanced in MBR systems preferably with long SRTs that retain the nitrifying bacteria within the bioreactor (Radjenovic et al., 2008; Chiemchaisri, 1993; Gao et al., 2004; Muller, 1994). However, the nitrification is generally a rate-limiting step in biological nitrogen removal performance requiring that the net rate of accumulation of biomass in the MBR bioreactor is less than the growth rate of nitrifying bacteria (Radjenovic et al., 2008; Barness and Bliss, 1983). In an aerobic MBR, ammonium is nitrified mostly to nitrate and most phosphates are removed during the aerobic period. Anoxic condition is typically introduced by operating the MBR in an intermittent aeration mode since the denitrification process requires anoxic conditions where the accumulated nitrate is completely denitrified and phosphorus (P) is taken up (Radjenovic et al., 2008).

The enhanced biological phosphorus removal (EBPR) in MBR treatment system is not easily achievable especially with weak sewage and with longer SRT which are common operating conditions in MBR (Lee et al., 2009). The competition between

PAOs and other heterotrophs would limit available carbon and energy for anaerobic P-release in weak wastewater (Lee et al., 2009). Ersu et al. (2010) attributed the decrease of biological phosphorus (bio-P) removal efficiency to the possible increase in lysis of microbial cells at high SRTs along with the low F/M ratio as a result of high suspended solids in the oxic tank. Possible nitrate recycle to the anaerobic zone may also reduce P-release when internal sludge recycle is used. However, the MBR treatment system may achieve significantly better P-removal under conditions that provide suitable environment for the proliferation of PAOs (Silva et al., 2012). Also, the membrane may completely retain the PAOs whose size is typically larger than microfiltration membrane pores (0.2  $\mu\text{m}$ ) (Radjenovic et al., 2008).

There has been much research on the effect of SRT on MBR performance as far as the simultaneous nitrification/denitrification and phosphorus removal are concerned. The long SRTs of MBRs associated with low sludge production and higher MLSS and MLVSS concentration may hinder the luxurious uptake of phosphorus along with the survival of the PAOs due to the limited amount of stored substrate (Rosenberger et al., 2002; Tchobanoglous et al., 2003). A few studies show that EBPR is possible in MBRs operating at sludge ages of up to 26 days (Abegglen et al., 2008; Adam et al., 2002; Fleischer et al., 2005). Yoon et al. (2004) recorded a decreased P-removal at SRT longer than 20 days which was attributed to the possibility that PAOs did undergo competitive conditions with GAOs at longer SRTs (Wang et al., 2001). From an aerobic submerged MBR with anaerobic and anoxic tanks, Ersu et al. (2010) achieved approximately 80% P-removal efficiency at 50 days SRT while the same dropped down at 75 days SRT. The fact behind this was ascribed to an increase in the lysis of microbial cells along with the low F/M ratio in the oxic tank. A full-scale MBR which was not designed for

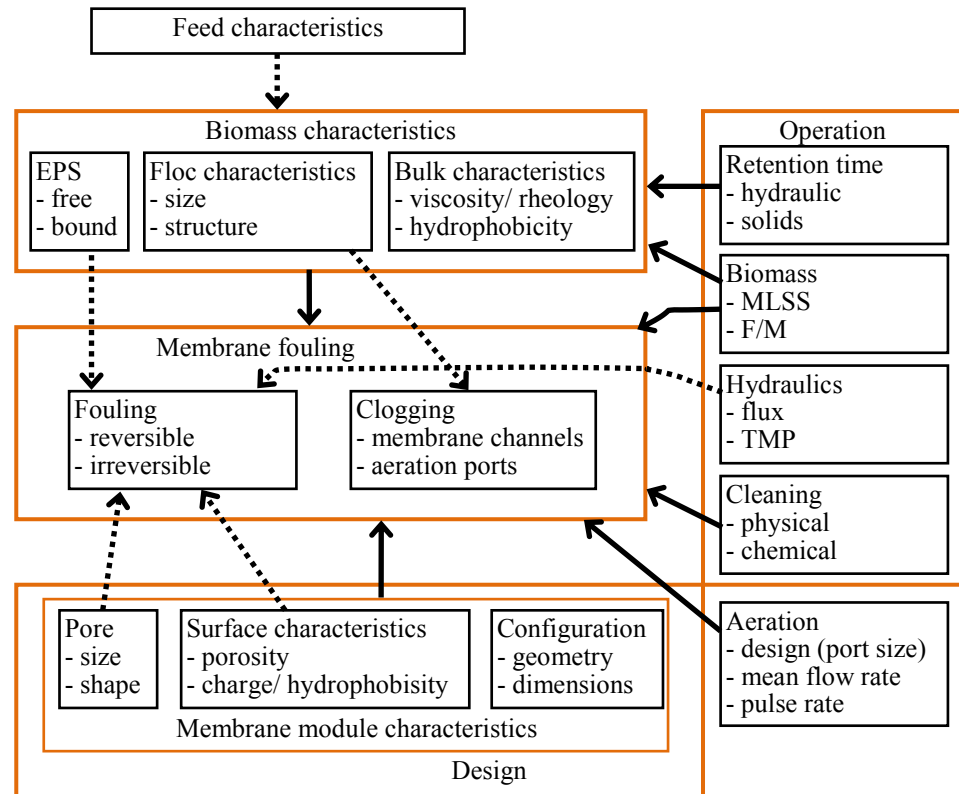
EBPRS, attained high bio-P removal (Silva et al., 2012) perhaps due to the high floc compactness and density of the EPS inducing anaerobic micro-niches.

The external environmental factors may also considerably influence the MBR biological treatment performance. Arévalo et al. (2014) most recently conducted a study on the effect of temperature on MBR performance where a full-scale pre-denitrification MBR system was operated at a constant flow of urban wastewater ( $0.42 \text{ m}^3/\text{h}$ ) at a sufficiently long SRT and HRT. The MBR system was continuously monitored during the period when the mixed liquor temperature varied in the range between 9 and  $31 \text{ }^\circ\text{C}$ . While the variations in temperature did not affect the effluent COD concentration, permeability was reduced both by the increase in membrane flux resistance at temperatures  $<15 \text{ }^\circ\text{C}$  and by viscosity which was attributed to the decrease both in the endogenous respiration rate ( $b_H$ ) and the observable biomass yield coefficient for heterotrophs. The effects led to the increase in the concentration of TSS and the VSS/TSS ratio.

#### *B. Problems of membrane fouling and biological-physical process interactions*

The most critical problem hindering the economic feasibility and sustainable operation of an MBR is the membrane fouling. The factors that affect the membrane fouling may be described into two broad phases. First the membrane resistance is increased due to the pore fouling by particles in the feedwater permanently adsorbing onto or into the membrane pores. Second the resistance at the interfacial region is increased due to the sludge cake deposition on the membrane surface. In fact, the operation of an MBR is defined by complex inter-relationships (Figure 2.3) between the membrane fouling process and the design or operational parameters (flux, TMP, membrane aeration for the SMBR etc.). There are essentially five key elements (Judd et al., 2008) of the SMBR process design and operation, these are:

- 1) the membrane, its design and the sustaining of permeability by cleaning
- 2) feedwater, its characteristics and its pre-treatment
- 3) aeration of both membrane and the bulk biomass
- 4) sludge withdrawal and residence time
- 5) bioactivity and the nature of the biomass.



*Figure 2.3* Inter-relationships between MBR parameters and fouling process variables (adapted from Judd et al., 2008)

The rate at which sludge is withdrawn controls the sludge retention time (SRT) which then determines the concentration of the mixed liquor (or, generally speaking, biomass). The MLSS concentration impacts both upon the biological properties and the physical properties such as the viscosity and oxygen transfer rate. Hence, the operational efficiency of the biological treatment becomes dependent upon biomass viability which is controlled by the mixed liquor concentrations in a more complex manner, while the membrane filtration process (or fouling) is subsequently affected by

the mixed liquor properties. However, the research on biomass viability and how these affect the MBR performance is still at a rudimentary level since it is a much more difficult process to deal with.

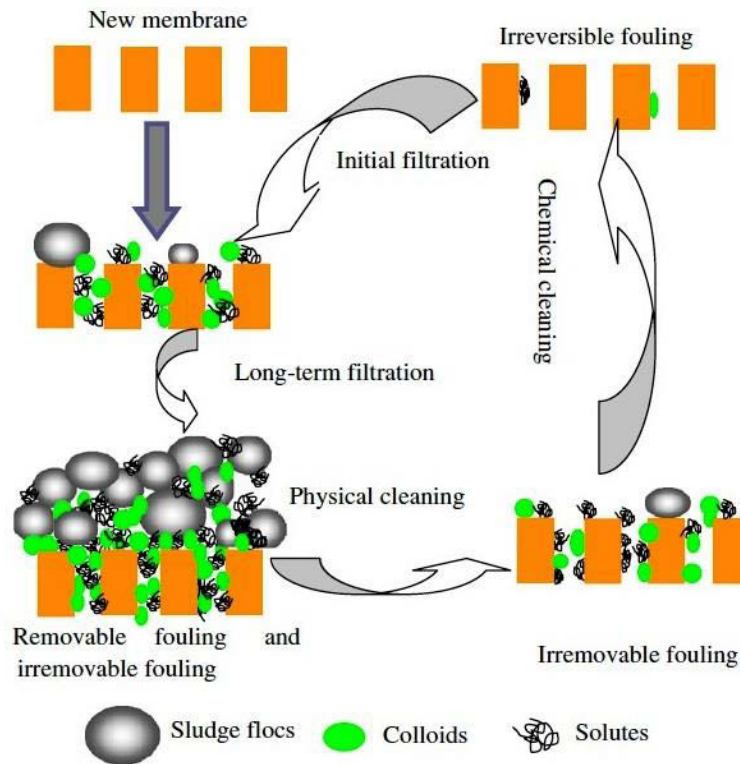
### ***2.2.5 Membrane fouling in MBRs***

The term ‘fouling’ is often used to lump all phenomena that lead to a loss in permeability (Drews, 2010) of a membrane, but in its strict form it is defined as the coverage of the membrane surface by deposits which adsorb or simply accumulate during operation. Fouling results from interaction between the membrane material and the components of the mixed liquor which typically include biological flocs of living or dead microorganisms along with soluble and colloidal microbial cell products. It is the most serious problem affecting the performance of MBRs and as a result, hinders the widespread applications of MBR for wastewater treatment. Fouling leads to a significant increase in resistance to flux, and hence demands frequent replacement or membrane cleaning increasing significantly the system’s operating costs.

#### *A. Types of membrane fouling*

Although there still exists a lack of consensus about the conceptual definitions of different types of membrane fouling, three distinct types of membrane fouling may be defined according to Meng et al. (2009). The removable fouling is generally attributed to the formation of a sludge cake layer by loosely attached foulants which can be easily eliminated by the implementation of physical cleaning (e.g., backwashing). The irremovable fouling, on the other hand, is attributed to pore blocking by permanently attached foulants which needs chemical cleaning to be eliminated. The removable foulants turn back into the mixed liquor due to aeration scouring or backwashing, and

thus, the fouling is termed as reversible (fouling) as well. The irreversible fouling is defined as permanent fouling that cannot be removed by physical or chemical cleaning.



*Figure 2.4* Classification of membrane fouling (adapted from Meng et al., 2009)

When classified according to the components of foulants, the fouling in MBRs can be classified into three major categories: biofouling, organic fouling, and inorganic fouling (Meng et al., 2009). Biofouling is defined as the fouling due to the undesirable microorganisms at a phase transition interface which may occur by deposition, growth and metabolism of bacterial cells or flocs on the membranes (Guo et al., 2012). Biofouling may start with the deposition of an individual cell or cell cluster on the membrane surface, after which the cells multiply and form a biocake (Meng et al., 2009). Many researchers have identified the dominant role of SMP or EPS causing the biofouling in an MBR. Biofouling poses a major problem in an MBR because the foulants are typically larger than the membrane pore size. Organic fouling is basically the deposition of biopolymers (i.e., proteins and polysaccharides) on the membranes which start depositing onto the membranes more readily due to the permeate flow, but

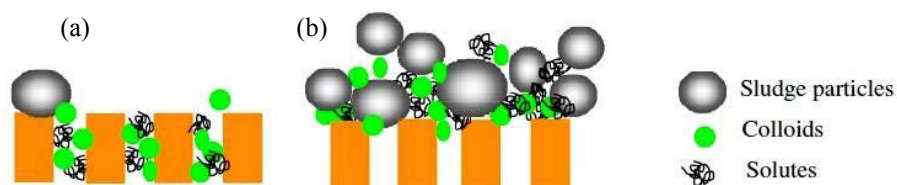


have lower back transport velocity due to lift forces in comparison to large particles (e.g., colloids and sludge flocs). Many studies confirm that SMP or EPS is the origin of organic fouling (Meng et al., 2009).

Although the biofouling and organic fouling mainly govern the overall fouling phenomena of an MBR, the inorganic fouling can also occur in two ways: chemical precipitation and biological precipitation. Chemical precipitation occurs when the concentration of chemical species (e.g.  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Al}^{3+}$ ,  $\text{PO}_4^{3-}$ ,  $\text{OH}^-$  etc.) exceeds the saturation concentrations due to concentration polarisation. The biopolymers may also contain ionic groups such as  $\text{COOH}^-$ ,  $\text{CO}_3^{2-}$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ ,  $\text{OH}^-$  that precipitate leading to inorganic fouling (Meng et al., 2009). The classification of components of foulants may be given in a slightly different way (Guo et al., 2012) into four categories as particulates (inorganic or organic particles/colloids), organic (dissolved components and colloids such as humic and fulvic acids) inorganic (dissolved components such as manganese and silica) and micro-biological organisms (algae and microorganisms such as bacteria).

### *B. Process kinetics of membrane fouling in an MBR*

Membrane fouling in a typical MBR system occurs due to the following general mechanisms (Meng et al., 2009): (i) adsorption of solutes or colloids within/on membranes; (ii) deposition of sludge flocs onto membrane surface; (iii) formation of a cake layer on membrane; (iv) detachment of foulants attributed mainly to shear forces; and (v) the spatial or temporal changes of foulant during the long-term operation.



*Figure 2.5* Illustration of membrane fouling process in MBRs (a) pore blocking (b) cake layer (adapted from Meng et al., 2009)

Experimental evidences suggest that the critical problem of membrane fouling is the dynamic change of bacterial community and biopolymer components in the cake layer particularly due to the high rate of cell lysis of the microbial components of the cake layer. Since the long-term operation of an MBR is typically conducted at a flux lower than the critical flux, the rate of particle convection towards the membrane surface is usually balanced by the rate of back transport and hence the particulate fouling is not a dominant problem to deal with (Drews et al., 2010). This gradually results in more of the membrane pore blocking due to the higher rate of trace foulants' adsorption as opposed to the back-transport of such foulants. These ultimately increase the specific cake resistance, cake compressibility and irreversibility during the long-term operation of an MBR system.

### *C. Effects of MBR operational parameters on membrane fouling*

Since deposits are brought to the membrane mainly by convective transport, the rate of fouling dominantly depends on the velocity orthogonal to the surface - the permeate flux (Drews, 2010). Three main factors that are typically assumed to affect fouling are membrane, sludge characteristics and operation (Chang et al., 2002; Le-Clech et al., 2006). Aeration ports and module dimensions have been added to the original three factors and make up the group of relevant design parameters for the MBR system (Judd, 2006). However, the rate of fouling depends on various other interrelated parameters making the correlation between flux and rate of fouling a dynamic variable. Numerous researches have so far been performed to investigate the influence of the MBR operational parameters on the membrane. Among the different design and operating parameters of an MBR, the organic loading rate (OLR), sludge retention time (SRT), membrane composition and configuration, for example, significantly influence the rate

of membrane fouling. Table 2.1 presents some of the recent findings of the effects of MBR operating parameters on membrane fouling.

*Table 2.1* Recent findings of the effects of MBR operating conditions on membrane fouling

Design and operational factors	Effects on fouling	References
OLR	The fouling potential significantly increases with high OLR which is also highly correlated with bEPS.	Wu et al., (2011); Xia et al. (2010)
Membrane properties and MBR configurations	Hydrophilic membrane with asymmetric, interconnected pore morphology and relatively large pore size (0.3 $\mu\text{m}$ ) and high surface porosity results in good membrane performance with less fouling.	van der Marel et al. (2010); Clouzot et al. (2011)
Substrate	With low ratio C/N or C/P, substrate can significantly influence median particle at steady state leading to lower rate of fouling.	Wu et al. (2012)
SRT	Higher SRT (>10 days) generally contributes to better activated sludge bioflocculation leading to lower rates of fouling.	Broeck et al. (2012)
Aeration intensity	The mixing conditions in low shear aerobic SMBR indirectly results in increased sEPS (or SMP) concentrations and higher fouling potential.	Menniti and Morgenroth (2010)
MLSS	MLSS has a negative correlation with permeate flux and hence with membrane fouling. Activated sludge with higher MLSS (exceeding 10 g/L) may entrap smaller particles (<20 $\mu\text{m}$ ) in the bulk activated sludge which otherwise contribute to increased resistance of the membrane. Activated sludge samples with lower MLSS concentrations generally do not show the capability to entrap particles.	Kornbooraksa and Lee (2009) Schwarz et al. (2006)
Feed characteristics	The colloidal content in feed and mixed liquor plays a dominant role and is more important than the quantity of total suspended solids in controlling fouling.	Gao et al. (2013)
pH	The increase of pH slightly increases the resistance of the virgin membrane and fouled membrane due to the repulsive energy barrier to the foulants approaching the membrane surface. There is a critical pH below which the repulsive energy barrier would disappear.	Zhang et al. (2014)

#### *D. Fouling index for MBR membrane fouling*

A simplified fouling index or mathematical expression may not fully describe the widely variable fouling phenomena that were observed by the researchers under different operating conditions and feed water characteristics of the MBR. de laTorre et

al., (2010) conducted an intensive monitoring campaign of four MBR systems that were operated in Berlin in 2007. The study was intended to characterize the fouling behavior based on mixed liquor properties and understanding their effects on corresponding permeability. A novel parameter named as transparent exopolymer particles (TEP) was chosen as the potential fouling indicator. The study concluded without finding a unique fouling index that could describe the fouling phenomena of MBR systems. Therefore, the search for a universal fouling indicator is typically abandoned in favor of indices that are combination of several parameters and are derived using multivariable analysis.

Among the early initiatives of determining fouling indices, the silt density index (*SDI*) test developed by the DuPont Co. at the end of the seventies is worth mentioning. The *SDI* is measured simply from the rate of membrane plugging of a membrane filter with pores of 0.45  $\mu\text{m}$  at approximately 207 kPa (Alhadidi et al., 2011). This simple fouling index is highly dependent on membrane module properties, and to overcome this limitation a modified fouling index (*MFI*<sub>0.45</sub>) was derived by Schippers and Verdouw (1980) applying the Darcy equation to determine the flow resistance in the case of cake filtration, taking into account the *SDI* as well. Jang et al. (2006) used a modified fouling index (*MFI*<sub>MBR</sub>) of the MBR to predict biofouling potentials dividing the fouling resistances into index (*MFI*<sub>Sol</sub>) of soluble materials and that of suspended solids (*MFI*<sub>SS</sub>), based on the unified theory of Laspidou and Rittman (2002a). Huang et al. (2008) developed a unified modified fouling index (*UMFI*) for low pressure membrane systems with an intention to directly apply it to a low pressure membrane operated at different scales/modes (constant flux versus constant pressure). The fundamental basis of the *UMFI* is a revised *Hermia* model (Hermans and Bredée, 1935; 1936), and the *UMFI* can be representative of total fouling, physically irreversible fouling and chemically irreversible fouling. In the *Hermia* model, the *UMFI* was

mathematically expressed as a function of normalized specific flux and specific volume. Nguyen et al. (2011) developed fouling indices (*FI*) using a resistance-in-series approach applied for a low pressure hollow fibre membrane. The key feature of the *FI* was that the fouling was not attributed to a specific mechanism, but was derived from a series of resistances derived from cake filtration, pore constriction or a combination of both fouling mechanisms, as applicable. The total fouling index (*TFI*) is the sum of the *FI* for reversible fouling by hydraulic backwash (*HRFI*), hydraulic-irreversible *FI* (*HIFI*) and chemical-irreversible *FI* (*CIFI*). All the fouling indices were related with normalized specific flux and specific volume. Mathematical expressions of some of fouling indices, as discussed above, are shown in Table 2.2. The notations for different parameters in the mathematical expressions convey their usual meanings.

Table 2.2 Mathematical expressions of some fouling indices for low pressure MBR systems

Fouling Indices	References
$SDI = \frac{100\%}{t_f} \left(1 - \frac{t_1}{t_f}\right) = \frac{\%P}{t_f}$ where $\%P = 100 * \left(\frac{V_1 - V_f}{V_1}\right)$	Alhadidi (2011); Nahrstedt and Camargo-Schmale (2008)
$MFI = \frac{\mu I}{2 \Delta P A^2}$	Nahrstedt and Camargo-Schmale (2008)
$MFI_{MBR} = MFI_{Sol} + MFI_{SS}$ $MFI_{Sol} = \beta[S(1 - f_s) + BAP(1 - f_{BAP}) + UAP(1 - f_{UAP})]$ $MFI_{SS} = \gamma \frac{QS_i}{V(X_a + EPS)}$	Jang et al. (2006)
$FI = \frac{1}{V} \left(\frac{1}{j^*} - 1\right)$ ; $TFI = \frac{1}{V} \left(\frac{1}{j^*} - 1\right)$ ; $HIFI = \frac{1}{V} \left(\frac{1}{j^*} - 1\right)$ ; and $CIFI = \frac{1}{V} \left(\frac{1}{J^*} - 1\right)$ where $J^* = \frac{(J / \Delta P)_V}{(J / \Delta P)_0}$	Nguyen et al. (2011)
$UMFI_{\text{cake formation}} = \frac{1}{V_s} \left(\frac{1}{J_s^*} - 1\right)$ ; $UMFI_{\text{intermediate blocking}} = \frac{\ln J_s^*}{V_s}$ ; $UMFI_{\text{standard blocking}} = 2V_s (J_s^{1/2} - 1)$ ; $UMFI_{\text{complete blocking}} = -\frac{1}{V_s} (J_s^* - 1)$ where $J_s^* = -\frac{J_s}{J_{s0}}$	Huang et al. (2008)

*E. Fouling control in MBRs*

Since the advent of the MBR technology, a significant number of strategies have been adopted for fouling mitigation which include mechanical, bio-chemical or hydrodynamic means in addition to periodic cleaning of the membrane by backwashing or air-bubbling, pretreatment of feedwater, modification of the mixed liquor properties and innovative design of bioreactors. Many other novel anti-fouling strategies have also been adopted in MBR applications such as the followings:

- Although the rate of aeration is a key parameter in MBR design, beyond an optimum air flow-rate further increase in aeration has no effect on the reduction of fouling. The innovative air-induced cross flow (Cui et al., 2003) in a submerged MBR could better reduce the fouling layer on the membrane surface. The combined use of gas-bubbling and backwashing has also been shown to be more effective in fouling control (Qaisrani and Samhaber, 2011).
- Intermittent permeation, where the filtration is stopped at regular time intervals for a couple of minutes before being resumed. Particles deposited on the membrane surface tend to diffuse back to the reactor; this phenomena being increased by the continuous aeration applied during this resting period.
- In the innovative air backwashing method, membrane modules kept in a pressurized vessel are coupled to a vent system. The pressurized air in the permeate side of the membrane can release a significant pressure within a very short period of time.
- Chemically enhanced backwash or intensive chemical cleaning (e.g. by NaOCl and citric acid)
- Mechanical cleaning (e.g. by abrasive granular material) (Siembida et al., 2010)

- Application of several commercial methods of membrane performance enhancer such as cationic polymer, powdered activated carbon (PAC), polyaluminium chloride (PACl) or ferric salts (Iversen et al., 2007; Koseoglu et al., 2008; Lee et al., 2007; Song et al., 2008; Yoon et al., 2005; Zhang et al., 2004)
- Addition of coagulants to reduce membrane fouling (e.g. Al or Fe coagulant) (Mishima and Nakajima, 2009)
- Feedback control which needs to be suitably adjusted to optimum operating conditions (e.g., Brauns, 2003; Busch and Marquardt, 2006; Drensla et.al., 2010; Li et al., 2003; Smith et al., 2005, 2006)
- Addition of nanomaterials such as Fullerene C60 (both as a coating of ceramic membranes or in the form of colloidal aggregates in suspension) was found to inhibit respiratory activity and attachment of Escherichia coli (Chae et.al., 2009; Fabrega et.al., 2009).
- Systematic optimization of all geometrical and operational parameters which may influence MBR hydrodynamics (tank size, liquid level, riser/downcomer cross-section area, membrane spacing, module height, bottom clearance, aerator dimensions and location, bubble size, aeration rate). (Fane et al., 2005; Hai et al., 2008; Lee et al., 2009; Ndinisa et al., 2006; Phattaranawik et al., 2007).

#### *F. Innovative MBRs for treatability and fouling control*

Researchers around the world are constantly trying innovations in the design of MBRs combining the features of controlled membrane fouling, treatability, sustainability, economy and operational efficiency. However, treatability concerns and the problems of membrane fouling are sometimes found to be inversely correlated. Yang et al. (2011) found that while the addition of polymeric ferric chloride (PFC) could reduce total phosphorus concentration in the effluent, the rate of membrane fouling increased 1.6

times over that of an MBR without PFC addition. Based on further investigation on fouling control in an MBR, Yang et al. (2011) suggested that soluble organic substances (e.g. SMP) and the dose of PFC should be controlled to minimize membrane fouling. In aerobic MBRs, almost complete nitrification can be achieved, while denitrification needs the addition of an anaerobic tank prior to the aeration tank with conventional recycle (Gander et al., 2000). However, the concept of simultaneous phosphorus and nitrogen removal significantly depreciated the most favorable characteristics of long sludge retention time (SRT) control in an MBR. To solve this problem, aerated MBR systems could either be coupled with a chemical treatment process such as coagulation and adsorption (Genz et al., 2004; Yoon et al., 2004), or be associated with a separated anoxic tank for denitrification (Ahn et al., 2003; Hibiya et al., 2003). In the present situation, although these MBR systems have shown an improvement of nitrogen removal, phosphorus has not been removed significantly through these systems.

As a consequence, treatability concerns in relation to sustainable and economic MBR operations have posed significant challenges requiring holistic solution to the problems. Several innovative lab-scale MBR systems have devised impressive mechanisms by which the rate of fouling in an MBR can be significantly reduced while maintaining satisfactory permeate flux. There are few recent innovations in the design of the MBR which deal with different aspects of the above mentioned problems in an integrated manner. Among the different innovative MBRs, the followings are noteworthy innovations claiming milestone achievements in this field of research.

The novel concept of *Single Stage Sponge Submerged MBR (SSMBR)* was developed for alleviating membrane fouling, enhancing permeate flux and improving phosphorus and nitrogen removals simultaneously (Guo et al., 2008; Ngo et al., 2006). The SSMBR includes attached growth bioreactors using specified sponge material in bioreactors to



modify the biological processes. In their innovative design, Guo et al. (2008) and Ngo et al. (2006) introduced sponge as an ideal attached growth media. Experimental investigation of its performance identified that sponge serves the multipurpose objective of the treatment process, for example it acts as a mobile carrier for active biomass and reduces the cake layers formed on the surface of membrane and retains microorganisms by incorporating a hybrid growth system (both their attached and suspended growth) (Chae et al., 2004; Ngo et al., 2006; Pascik, 1990; Psoch and Schiewer, 2006). Sponges may also facilitate metabolic selection via alternating anoxic/aerobic processes within pores increasing the potential of having higher bacterial activities and improved nutrient removal in MBR systems (Liang et al., 2010). Deguchi and Kashiwaya (1994) have reported that the nitrification and denitrification rate coefficients of a sponge suspended biological growth reactor were respectively 1.5 and 1.6 times higher than the same coefficients of the conventional activated sludge reactor.

The performance of the SSMBR system was evaluated (Ngo et al., 2008) using two kinds of polyester–urethane sponges (coarse sponge with higher density S28–30/45R and fine sponge with lower density S16–18/80R) with sponge volume fraction of 10% and bioreactor MLSS of 10 g/L. The results indicated the addition of sponge in the SSMBR could increase sustainable flux (2 times for S28–30/45R and 1.4 times for S16–18/80R) and lower TMP development, thus significantly reducing membrane fouling. S28–30/45R gave a rise in attached growth biomass and the removal efficiencies of DOC, COD and PO<sub>4</sub>-P whilst S16–18/80R had better performance in removing NH<sub>4</sub>-N. Although the SSMBR performed well for most of the trials, superior recycled water quality was achieved when adding S28–30/45R and S16–18/80R together in the SSMBR with the ratio of 2:1 and without any pH adjustment during the operation.

Ren et al. (2010) introduced a new concept of wastewater treatment using a *MBR coupled with Non-oven Fabric Filter Bag* (NFFB). Activated sludge is charged in the nonwoven fabric filter bag and membrane filtration via the fabric is achieved under gravity flow without a suction pump. They found that the biofilm layer formed inside the NFFB achieved 10 mg/L of suspended solids in the permeate within 20 min of initial operation. The dynamic biofilter layer showed good filterability and the specific membrane resistance consisted of  $0.3\text{--}1.9 \times 10^{12}$  m/kg. Due to the low F/M ratio (0.04–0.10 kg BOD<sub>5</sub>/m<sup>3</sup>/d) and the resultant low sludge yield, the reactor was operated without forming excess sludge. Although the reactor provided aerobic conditions, denitrification occurred in the biofilm layer to recover the alkalinity, thereby eliminating the need to supplement the alkalinity. However, there may be some problems of bag failure associated with the design. Bag failure can occur from chemical attack to the fabric. Changes in biomass composition and exhaust material temperatures from industrial processes can greatly affect the bag material. Different weaving patterns increase or decrease the open spaces between the fibres. This will affect both fabric strength and permeability. Fabric permeability affects the amount of air passing through the filter at a specified pressure drop. A tight weave, for instance, has low permeability and is better for the capture of small particles at the cost of increased pressure drop.

Yunxia et al. (2009) based their experimental idea of including packed bed biofilm in the MBR reactor with an aim to achieve stability of nitrite accumulation in the shortcut biological nitrogen removal (SBNR) system. The mechanism in the *MBR Combined with Packed Bed Biofilm Reactor* provides an economic advantage over the conventional biological nitrification-denitrification process. However, maintaining the stable nitrite accumulation in the SBNR posed a challenging task. In the innovative MBR designed by Yunxia et al. (2009), stability of nitrite accumulation is secured by

combining an aerobic SBR and anaerobic packed-bed biofilm reactor. The system was evaluated for treating high strength ammonium bearing wastewater. The MBR was successful in both maintaining nitrite ratio over 0.95 and nitrification efficiency higher than 98% at HRT of 24 h, while PBBR showed satisfactory denitrification efficiency with very low effluent nitrite and nitrate concentration. However, the system required seeding with enriched ammonia-oxidizing bacteria (AOB) which results in the high nitrite accumulation in the MBR.

Phattaranawik and Leiknes (2011) have developed the *Hybrid Biofilm Membrane Bioreactor* (BF-MBR) as an alternative option to the activated sludge MBR. The BF-MBR is a unique design with attachment of a biofilm on a carrier in the biofilm reactor which lowers the suspended solid (SS) environment in the system and also reduces energy consumption from the aeration systems and membrane fouling. The alternative design of the biofilm reactor developed by Phattaranawik and Leiknes (2011) had two vertical chambers filled with small and light plastic carriers for biofilm attachment. Suspended solid (SS) concentration in the BF-MBR is hydrodynamically controlled to be lower than 70 mg/L. The ultraviolet (UV) inactivation unit is integrated with the membrane filtration tank to limit biological activities for biofoulant production and to decelerate the unwanted biofilm formation in the permeate tube. The combinations of membrane relaxation and the UV inactivation in the BF-MBR system have significantly prolonged sustainable operation periods of the membrane filtration.

### ***2.2.6 Correlation between biological process variables and fouling in the MBR***

#### ***A. Effects of dissolved organic matter on fouling***

The biodegradable and non-biodegradable organic matter present in the mixed liquor supernatant (usually referred to as DOM) appear to play a significant role during the

membrane filtration process of the MBR system. A large portion of DOM consisting of soluble and biodegradable organics of microbial origin is frequently referred to the literature as sEPS or SMP. The DOM are adsorbed onto and into the membrane during membrane filtration, block membrane pores and form a partly irreversible gel structure on the membrane surface and into the membrane pores causing fouling. Reviewing the relationship between membrane fouling and DOM concentration Le-Clech et al. (2006) have identified a proportional relationship between the loss of hydraulic performance and DOM. Regarding relevant influence of DOM composition, a direct relationship between the carbohydrate levels in DOM with parameters of fouling propensity has been observed. Rosenberger et al., (2006) have observed a linear relationship between filtration resistance over time and polysaccharide (PS) concentration in sludge supernatant.

The observed correlations of DOM and filtration resistance have also been accounted for in various mathematical modelling efforts. Ishiguro et al. (1994) proposed a simple mathematical expression to describe membrane flux as proportional to the difference of DOM concentration between the mixed liquor and permeate. Liang et al. (2006) developed a mathematical model for both reversible and irreversible fouling rate where permeate flux decline or TMP rise can be determined with model inputs of DOM concentration. Fan et al. (2006) recommended an empirical mathematical expression based on the observed relation between the critical flux and colloidal Total Organic Carbon (TOC). Busch et al. (2007) presented a detailed model of fouling mechanisms for a submerged hollow fibre filtration in which different model variables for different fouling mechanisms literally represents the DOM concentration. Guglielmi et al. (2007) established a subcritical flux fouling model that could predict the time at which a sharp

change in the TMP-time profile occurs considering it inversely proportional to the concentration of the DOM.

The concentration of EPS seems to play an important role in the regulation of DOM concentration assuming these two organic fractions are closely interrelated. Patsios and Karabelas (2010) recently conducted an analysis by Scanning Electron Micrographs (SEM) of suspended biomass aggregates which found bacteria are embedded and mostly immobilized within the slime matrix of EPS. Xuan et al. (2010), after comparing the contribution of granular and flocculent sludge to fouling, concluded that membrane filtration decreases with increasing EPS content in flocculated sludge. However, EPS are found to influence considerably the activated sludge structural characteristics as well as physico-chemical properties (Le-Clech et al., 2006; Meng et al., 2009) which make their impact on the fouling process rather complicated. Based on the observation of the same nature of DOM and EPS in LC-OCD chromatograms, Rosenberger et al. (2006) hypothesized that their relative concentrations are under a dynamic equilibrium which can easily be shifted by changing conditions in the mixed liquor environment. Various processes and/or conditions that result either in the biosorption of DOM by the bioflocs, or the hydrolysis of EPS and the release of DOM in the bulk liquid (Nielsen et al., 1997) affect the EPS - DOM equilibrium.

#### *B. Fractions of mixed liquor solids responsible for membrane fouling*

Unlike the simple mathematical expressions (proposed by Busch et al., 2007; Fan et al., 2006; Guglielmi et al., 2007; Ishiguro et al., 1994; Liang et al., 2006) to describe the relationship between the concentration of Dissolved Organic Matter (DOM) and decline of membrane flux or change of TMP, the MLSS or the Mixed Liquor Volatile Suspended Solids (MLVSS) concentration has a more accurate but complex relation with MBR fouling. Kornboonraksa and Lee (2009) found that MLSS and sludge floc

size are the dominant factors that control the membrane filtration in an MBR. Different researches have proposed different empirical mathematical expressions to describe membrane flux or fouling rate with MLSS/MLVSS included in the expressions. However, contradictory findings about the effect of these parameters on membrane filtration have also been reported (Le-Clech et al., 2006; Meng et al., 2009) perhaps because the MLSS/MLVSS concentration, as a lump parameter, represents different kinds of suspended organic matter with possibly different fouling propensity. However, the major focus with regard to fouling has turned to sticky substances which can be bound to the flocs or freely suspended. These groups of compounds are mostly termed as EPS when they are bound to the flocs or SMP when freely suspended in the supernatant (Drews, 2010). Nevertheless, there still remains disagreement among the researchers regarding the definition of EPS and SMP. Patsios and Karabelas (2010) have defined soluble EPS (sEPS) or SMP as a biodegradable fraction of DOM. The terms biopolymers or biopolymeric clusters (BPC) (Lin et al., 2009; Sun et al, 2011; Wang et al., 2008), neither microbial nor EPS are non-filterable organics, and the transparent exopolymer particles (TEP) (de la Torre et al., 2008, 2010), especially the sticky fraction of EPS, have also come into use.

Table 2.3 gives a few empirical relationships that were based on findings that these fractions of MLSS contribute to membrane fouling. By definition, all these groups of compounds are produced and excreted by microorganisms. However, what is analysed as EPS, SMP, BPC or TEP by commonly agreed methods is not only of microbial origin but can also be terrestrial or man-made (Drews 2010; Judd 2006). A further fractionation of these components of foulants are also reported in the literature identifying that protein, polysaccharide or carbohydrates of the foulants dominate the membrane fouling process (see Table 2.4).

*Table 2.3* Fractions of MLSS and their relationship with membrane fouling (modified after Meng et al., 2010)

Fractions of MLSS	Relation with Membrane Fouling	Reference
EPS	Carbohydrates of EPS $\uparrow$ , $\rightarrow$ (tends to influence) clog membrane	Dvorak et al. (2011)
	EPS, hydrocarbon components and inorganic matters govern membrane fouling layer	Pendashteh et al. (2011)
	$\uparrow$ extracellular polymeric substances (EPS), $\rightarrow$ high membrane resistance indicating severe membrane fouling	Chae et al. (2006)
	$\uparrow$ EPS, fouling rate $\uparrow$	Drews et al. (2006)
	Bound EPS affects cake specific resistance	Cho et al. (2005)
SMP	Hydrophilic fraction (in carbohydrate): major cause for membrane fouling	Pan et al. (2010)
	$\uparrow$ SMP, $\uparrow$ fouling rate and membrane fouling index (MFI)	Arabi and Nakhla (2009)
	Hydrophilic neutrals (carbohydrates) responsible for high fouling potential at short SRTs	Liang et al. (2007)
	SMP influence fouling only under certain conditions such as low sludge age and large pore size	Drews et al. (2008)
	$\uparrow$ MLSS concentrations, $\downarrow$ normalize permeability	Trussell et al. (2007)
	SMP and Polysaccharides influence fouling more than MLSS.	Zhang et al. (2006)
BPC	Concentration $\uparrow$ , fouling $\uparrow$	Sun et al. (2011)
	BPC along with SMP and EPS governs membrane fouling	Lin et al. (2009)
	$\uparrow$ BPC concentration, $\uparrow$ Filtration Resistance	Wang et al. (2007)
TEP	TEP more important for fouling than CH, proteins or total EPS	de la Torre et al. (2010)
	$\uparrow$ TEP, $\rightarrow$ reach the critical flux sooner, $\downarrow$ the mixed liquor filterability	de la Torre et al. (2008)

*Table 2.4* Fractions of microbial products and their effects on membrane fouling

Components of microbial products	Effects on membrane fouling	References
	Proteins of EPS increase membrane resistance.	Ng and Ng (2010)
	PN/PS ratio of both EPS and SMP has significant impact on filtration resistance and fouling propensity.	Lee et al. (2003); Tian et al. (2011)
	The ratio of PN and PS is more important than the total quantity of soluble organic substances in controlling the membrane fouling.	Gao et al. (2013)
Proteins (PN) and polysaccharide (PS)	The contents of polysaccharides in the supernatant and particle size of the bio-flocs increase fouling.	Zhang et al. (2010)
	The higher ratio of PN/PS induces less irreversible fouling and improves the interaction of PN and PS to form cake layer.	Yao et al. (2011)
	In biocake, the EPS polysaccharides are correlated to the filtration resistance (R) and temperature (T) while in mixed liquor, the ratio of PN/PS of SMP is the most correlative factor.	Gao et al. (2013)
	Carbohydrates of both EPS and SMP increase the rate of fouling.	Yigit et al. (2008); Deng et al. (2014)
	The increase in carbohydrate of SMP contributes more to the formation of biofilm than that of protein.	Chu and Li (2005)
polysaccharide (PS) / Carbohydrate	Carbohydrates of EPS tend to clog membrane pores.	Dvorak et al. (2011)
	A low level of biofouling is correlated with the slow rise of the carbohydrates of SMP.	Gao et al. (2013)
	The accumulation of carbohydrates on membrane surfaces significantly increases the TMP.	Khan et al. (2013)

### 2.3 A brief review on mathematical modelling of the MBR

The previous sections of this literature review present a brief overview of the biological processes of the activated sludge wastewater treatment process since all the bioprocess are more or less related to the biomass viability. Individual components of biomass in the mixed liquor exhibit complex bio-oxidation kinetics that is controlled by the oxidation requirements for the removal of particular organics or nutrients. The complexities of the biokinetics of activated sludge processes are significantly increased when the membrane rejection of certain components is retained in the bioreactor.



Consequently, the literature relevant to the effects of membrane fouling on the bioprocesses in an MBR is also described in this review.

The bioprocesses taking place in an activated sludge wastewater treatment system are characterized by great complexity and yet incomplete understanding of some of the process phenomena. The mathematical models of the MBR show inherent deficiencies for its simulation due to additional intrinsic complexities resulting from the interaction between concurrently occurring and dynamic biological processes with membrane filtration, and the straightforward adoption of the Activated Sludge Models' (ASM) frameworks or their modified variations. In this backdrop, this section compiles a brief overview of the previous developments to the current state-of-the-art mathematical modelling approaches of the MBR system. Significant efforts have been applied to identify key variables of mathematical MBR models which can possibly establish an integrated model framework with comprehensive coupling between MBR bioprocesses and membrane fouling. With extended discussions on particular topics such as applications of modified ASMs to MBR modelling, ASM extensions incorporating SMP or EPS concepts, this section also provides a guide for different end-users of mathematical models of MBR systems.

### ***2.3.1 Models of biomass kinetics of the activated sludge process***

#### *A. ASM model family*

The first product of ASM model families, called Activated Sludge Model No1 (ASM1), is the outcome of the work of a task group formed in 1983 by the International Association on Water Pollution Research and Control (now known as the International Water Association - IWA). The model presented in 1987 (Henze et al. 1987) was basically intended to model the biological wastewater treatment process for organic

carbon removal, nitrification and denitrification but excluding the phosphorus removal. Ng and Kim (2007) have done an extensive review on the ASMs model family. The key features of the ASM models as discussed in the following sections have been summarized from the review of the models done by Ng and Kim (2007). Table 2.5 and Figure 2.6 present some important parameters of the ASMs which are also briefly described in the following sections.

Two main concepts have been incorporated into the formulation of *Activated sludge model no. 1 (ASM1)* using Total COD as the suitable parameter for defining the organic matter in the wastewater. Total COD is assumed to consist of biodegradable COD, non-biodegradable (inert) COD and the active biomass. The first concept is that Readily Biodegradable COD (RBCOD) can immediately be used by organisms for synthesis, whereas Slowly Biodegradable COD (SBCOD) must be broken down to be metabolized. The second concept in the model is death-regeneration. When the biomass decays, a portion of the decayed cell material is non-biodegradable. The rest of the decayed material is slowly biodegradable and can be used by active organisms for growth. The inert materials are incorporated into the model as having a zero reaction rate. Many of the model's concepts like the bisubstrate hypothesis, death-regeneration process and Monod type kinetics are still considered state-of-the-art and are widely employed by most of the CAS process models. Nevertheless, a number of simplified assumptions made in the model have limited applications, especially when industrial wastewaters dominate the influents.

The modelling to describe enhanced biological phosphorus removal (EBPR) system has started with the *Activated sludge model no. 2 (ASM2)*, which is the extension of ASM1 incorporating EBPR process and chemical phosphorus (P) removal via precipitation. The model incorporates PAOs as the biomass that is able to accumulate

phosphorus under aerobic conditions and store it in the form of cell internal poly-P and PHA. Growth of PAOs occurs only under aerobic conditions and on cell internal organic material. It is assumed in the ASM2 model that the PAOs, being incapable of any denitrifying activity, can only grow on stored PHA of the cell using energy derived from the hydrolysis of poly-P which eventually leads to the release of soluble phosphates ( $S_{PO_4}$ ). Storage is only possible when fermentation products are available. For the lyses of PAOs, separate process rates are provided in the model. Phosphate precipitation and redissolution are modeled by considering that  $S_{PO_4}$  reacts with metal hydroxides to form a phosphate precipitate. The *ASM2d* model builds on the ASM2 model, adding the denitrifying activity of PAOs so as to allow a better description of the generation and accumulation of phosphate and nitrate. The model additionally assumes that the PAOs can use internal cell storage products for denitrification and thus grow under anoxic conditions leading to the addition of two rate processes to the ASM2 processes: the storage of polyphosphates and growth of PAOs under anoxic conditions.

*Activated sludge model no.3 (ASM3)* (Gujer et al., 1999) was presented by the IWA task group in 1999 where the major changes are the inclusion of internal cell storage compounds in heterotrophs and shifting the focus from hydrolysis to the storage of organic substrates. The growth of the heterotrophic biomass is dependent on internal cell components that are transformed from the readily biodegradable substrates taken up by the heterotrophic biomass. The inclusion of internal cell storage structures has led to the distinction between the decay of biomass and storage products under aerobic and anoxic conditions. The death-regeneration concept of ASM1 is replaced in ASM3 by the growth-endogenous respiration model. The model was developed for domestic wastewater and, therefore, has inherent deficiencies to model industrial wastewater treatment with changed wastewater characteristics.

Table 2.5 Comparison of ASM models with regard to the simulation of MBR bioprocess

Model	ASM1	ASM2 / ASM2d	ASM3
<b>Major components</b>	<ul style="list-style-type: none"> <li>- Active heterotrophic biomass (<math>X_{B,H}</math>)</li> <li>- Active autotrophic biomass (<math>X_{B,A}</math>)</li> <li>- Readily biodegradable substrate (<math>S_S</math>)</li> <li>- Slowly biodegradable substrate (<math>X_S</math>)</li> <li>- Oxygen (negative COD) (<math>S_O</math>)</li> <li>- Ammonia / ammonium nitrogen (<math>S_{NH}</math>)</li> <li>- Nitrate and nitrite nitrogen (<math>S_{NO}</math>)</li> <li>- Particulates from biomass decay (<math>X_P</math>)</li> </ul>	<ul style="list-style-type: none"> <li>- Fermentable substrate / products (<math>S_F / S_A</math>)</li> <li>- Nitrate and nitrite nitrogen (<math>S_{NO3}</math>)</li> <li>- Dissolved oxygen (<math>S_{O2}</math>)</li> <li>- Ammonium and ammonia nitrogen (<math>S_{NH4}</math>)</li> <li>- Inorganic soluble phosphorus (<math>S_{PO4}</math>)</li> <li>- Total suspended solids (<math>X_{TSS}</math>)</li> <li>- Phosphate accumulating organisms (<math>X_{PAO}</math>)</li> <li>- Cell internal storage products of PAO (<math>X_{PHA}</math>)</li> </ul>	<ul style="list-style-type: none"> <li>- Dissolved oxygen (<math>S_O</math>)</li> <li>- Inert particulate organic matter (<math>X_I</math>)</li> <li>- Readily biodegradable organic substrate (<math>S_S</math>)</li> <li>- Active heterotrophic organisms (<math>X_H</math>)</li> <li>- Nitrate and nitrite nitrogen (<math>S_{NOx}</math>)</li> <li>- Cell internal storage product of <math>X_H</math> (<math>X_{STO}</math>)</li> <li>- Ammonium plus ammonia nitrogen (<math>S_{NH4}</math>)</li> <li>- Suspended solids (<math>X_{SS}</math>)</li> </ul>
<b>Major Rate Processes</b>	<ul style="list-style-type: none"> <li>Aerobic growth of <math>X_{B,H}</math></li> <li>Anoxic growth of <math>X_{B,H}</math></li> <li>Aerobic growth of <math>X_{B,A}</math></li> <li>Decay of <math>X_{B,H}</math></li> <li>Decay of <math>X_{B,A}</math></li> </ul>	<ul style="list-style-type: none"> <li>Aerobic / Anaerobic / Anoxic hydrolysis</li> <li>Aerobic growth of <math>X_H</math> on <math>S_F / S_A</math></li> <li>Anoxic growth of <math>X_H</math> on <math>S_F / S_A</math></li> <li>Storage of <math>X_{PHA}</math>, <math>X_{PP}</math></li> <li>Lysis of <math>X_H</math>, <math>X_{PAO}</math>, <math>X_{PP}</math>, <math>X_{PHA}</math></li> <li>Aerobic growth of <math>X_{AUT}</math></li> </ul>	<ul style="list-style-type: none"> <li>Hydrolysis</li> <li>Aerobic/ Anoxic storage of <math>S_S</math></li> <li>Aerobic/ Anoxic growth of <math>X_H</math></li> <li>Aerobic/ Anoxic endogenous respiration of <math>X_H</math></li> <li>Aerobic endogenous respiration of <math>X_{STO}</math></li> <li>Aerobic growth and endogenous respiration of <math>X_A</math></li> </ul>
<b>Major parameters / coefficients in rate processes</b>	different reaction process rates depend on parameters such as maximum growth rates $\mu_H$ , $\mu_A$ ; saturation coefficients $K_S$ , $K_{N,H}$ , $K_{O,H}$ , $K_{O,A}$ , heterotrophic yield coefficient $Y_H$ , decay coefficients $b_A$ , $b_H$	$i_{N,X_S}$ and $i_{P,X_S}$ for N and P contents of $X_S$ , saturation coefficients $K$ for oxygen and ammonium, maximum growth rate of autotrophs $\mu_{AUT}$ , Yield of heterotrophs and PAO, $q_{PHA}$ , $q_{PP}$ rate constants for storage etc.	$Y_{STO,O2}$ and $Y_{STO,NO}$ for aerobic and anoxic storage of $X_{STO}$ , $f_{XI}$ for aerobic and endogenous respiration of $X_I$ , yield coefficient $sY_H$ for $X_H$ and $X_{PAO}$ etc.
<b>Total parameters</b>	13	19/20	13
<b>Total processes</b>	8	19 / 21	12
<b>Bioprocess simulation</b>			
- COD removal	Yes	Yes	Yes
- Nitrification	Yes	Yes	Yes
- Denitrification	Yes	Yes	Yes
- Hydrolysis	Yes	Yes	Yes
Phosphorus removal	No	Yes	No
<b>Key variable for MBR fouling predictions</b>			
- SMP	No	No	No
- EPS	No	No	No

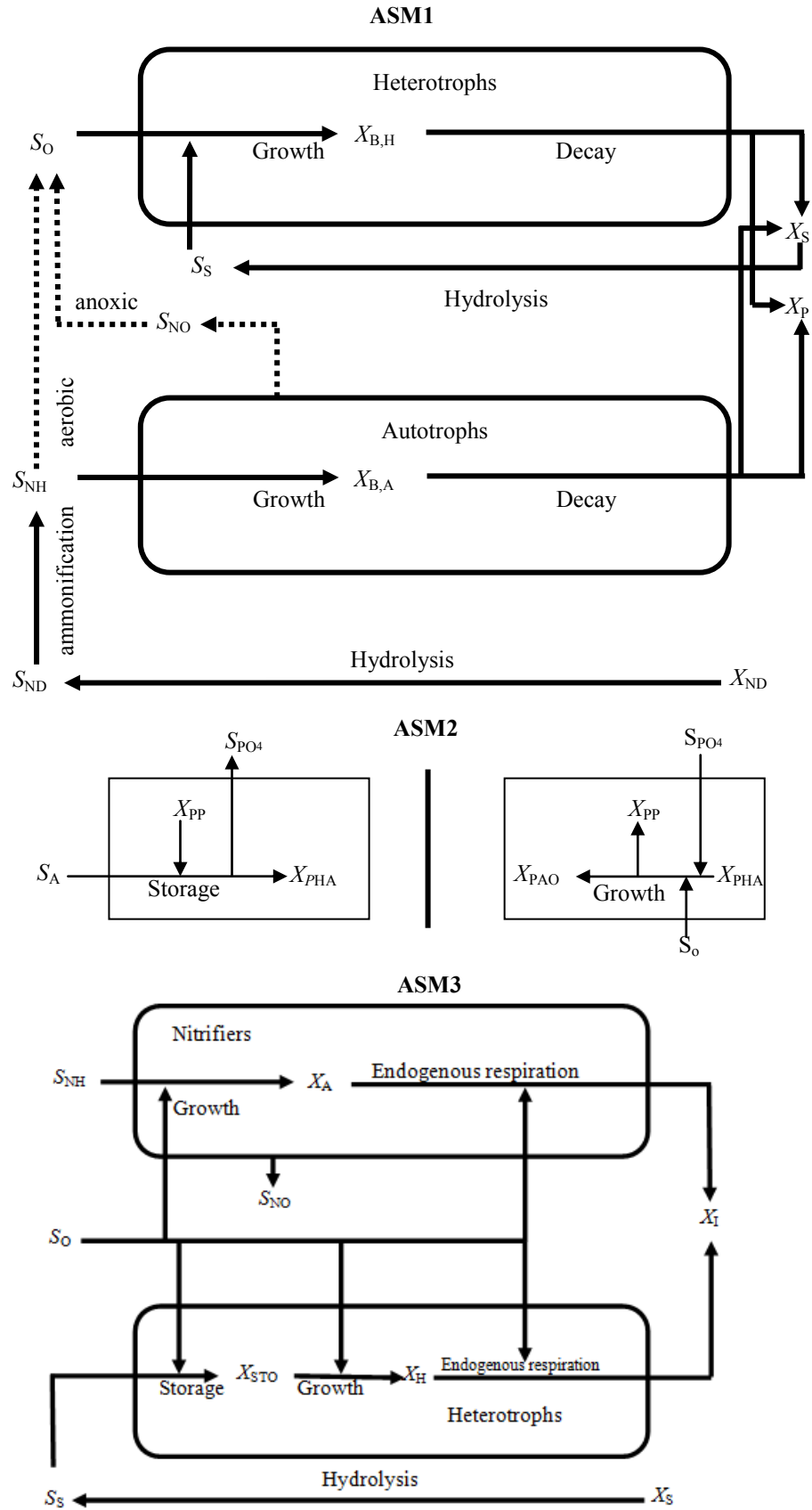


Figure 2.6 Biological parameters and process variables of ASMs

*B. Application of ASMs to MBR modelling: characteristic features and deficiencies*

The application of ASMs are presumably meant for modelling the CAS processes operating conditions of which, depending on the particular treatment process, vary within a typical range e.g. range of SRT 3-15 d, range of HRT 3-5 h and range of MLSS 1.5-4 g/L for completely mixed systems (Tchobanoglous et al., 2003). However, a study on municipal MBRs in Europe has reported the ranges of various operating parameters (Itokawa et al., 2008) that are significantly different from those of the typical CAS system. This obviously raises the question as to what extent the ASM framework is applicable to MBR processes.

As the physicochemical characteristics of the mixed liquor suspension would inevitably affect the filtration performance, models of biomass activity in an MBR should have a dual perspective of adequately describing the complex biological processes as well as accounting for the biomass effects on membrane filtration performance (Patsios and Karabelas, 2010). Previous studies suggest that the applicability of unmodified ASMs for modelling MBR needs to be carefully verified especially to understand the effects of higher SRTs and MLSS concentrations on biomass kinetics (Ng and Kim 2007). The models of biomass kinetics for MBR should at least provide reliable estimates of the EPS concentration in the activated sludge flocs, the elevated concentration of DOM and the typically higher MLSS concentrations in the bioreactor particularly taking into account the existence of SMP.

An early investigation (Chaize and Huyard, 1991) of the MBR using the unmodified ASM1 has found that a non-calibrated ASM1 is able to give a reasonable estimate of the MBR effluent COD and TKN while it fails to predict fairly accurate solids concentration particularly at very low HRT and very high SRT. Recent efforts have emphasized systematic calibration of ASMs' key parameters along with biomass

fractionations and/or influent wastewater characterization into various ASM based fractions (Delrue et al., 2008; Fenu et al., 2010; Spèrandio and Espinosa, 2008). However, there still exists dispute among researchers about the chemical, physical, biological or trial-and-error procedures to adopt for wastewater characterization (Fenu et al., 2010; Jiang et al., 2005, Spèrandio and Espinosa, 2008). Different researchers have tried adjusting values of ASMs' kinetic and stoichiometric parameters for matching model prediction with experimental data which eventually results in a range of values for these model parameters suggested in the literature (e.g. Hocaoglu et al., 2011; Parco et al., 2007). This may be attributed to the fact that certain factors of the MBR system have dominant effects on process kinetic parameters such as the sludge suspended solids ( $X_{TSS}$ ) impacting the excess sludge production and the oxygen transfer rate ( $\alpha$ -factor), the removed and residual nitrogen species ( $S_{NH}$ ,  $S_{NO}$ ), the residual phosphorus concentration ( $S_{PO_4}$ ) and oxygen consumption rate (OUR, and  $S_O$ ) (Fenu et al., 2010). Because of this lack of coherence, it is not appropriate to suggest best set of ASMs' (unmodified) model parameters for MBR modelling studies.

### *C. MBR modelling studies with modified ASMs*

Large fractions of flocs, bacteria, biopolymers such as polysaccharides, proteins and organic colloids are mostly retained in the bioreactor of an MBR which may significantly affect biodegradation kinetics within the bioreactor. The ASM models do not specifically consider the impacts of these retained particles on the biokinetics although in MBR systems with typically low organic loads, the retained molecules may have a significant impact on the metabolic path allowing further use of carbon based metabolites (Fenu et al., 2010; Furumai and Rittmann, 1992). Furthermore, ignoring SMP and EPS formation may lead to a general overestimation of true cellular growth

rates and this would severely underpredict the COD effluent (Fenu et al., 2010; Jiang et al., 2008).

Mathematical modelling of MBR systems to incorporate the above mentioned specificities have been usually attempted through modifying ASMs with SMP/EPS concepts. Some models have been developed as stand-alone descriptions of the concepts of production and degradation of both SMP and EPS while others have focused on integrating only the SMP concepts into the ASM type of models (Baek et al., 2009; Lu et al., 2001). To model biomass accurately without calibration using only experimental data is one of the specific advantages of standalone SMP model over ASMs.

*Standalone SMP/EPS models:* Researchers have so far encountered great difficulties in the determination of individual fractions of SMP. Substrate utilization, biomass decay and EPS hydrolysis are the major processes responsible for the SMPs' formation. The utilization associated products of the SMPs i.e. the UAPs are generally classified as compounds produced during substrate metabolism at a rate proportional to the rate of substrate degradation (Aquino and Stucky, 2008; Laspidou and Rittmann 2002a; Namkung and Rittmann, 1986) (see Figure 2.7 and Table 2.5). Unlike UAPs, there exist ambiguous concepts for describing the production mechanism of the BAPs. It was assumed in earlier studies that BAPs are produced from the decay of active biomass (Furumai and Rittman, 1992; Namkung and Rittmann, 1986; de Silva and Rittman, 2000). Laspidou and Rittmann (2002a, b) clearly differentiated the active biomass into active cells and bound (floc-associated) EPS that were hydrolysed to form BAP. In contrast, Ramesh et al. (2006) compared the physicochemical characteristics of hydrolysed EPS and BAPs but found them non-identical in all properties. However, the researchers have come to a general consensus that both the UAP and BAP are biodegradable and thus cycle back to become substrate cells.



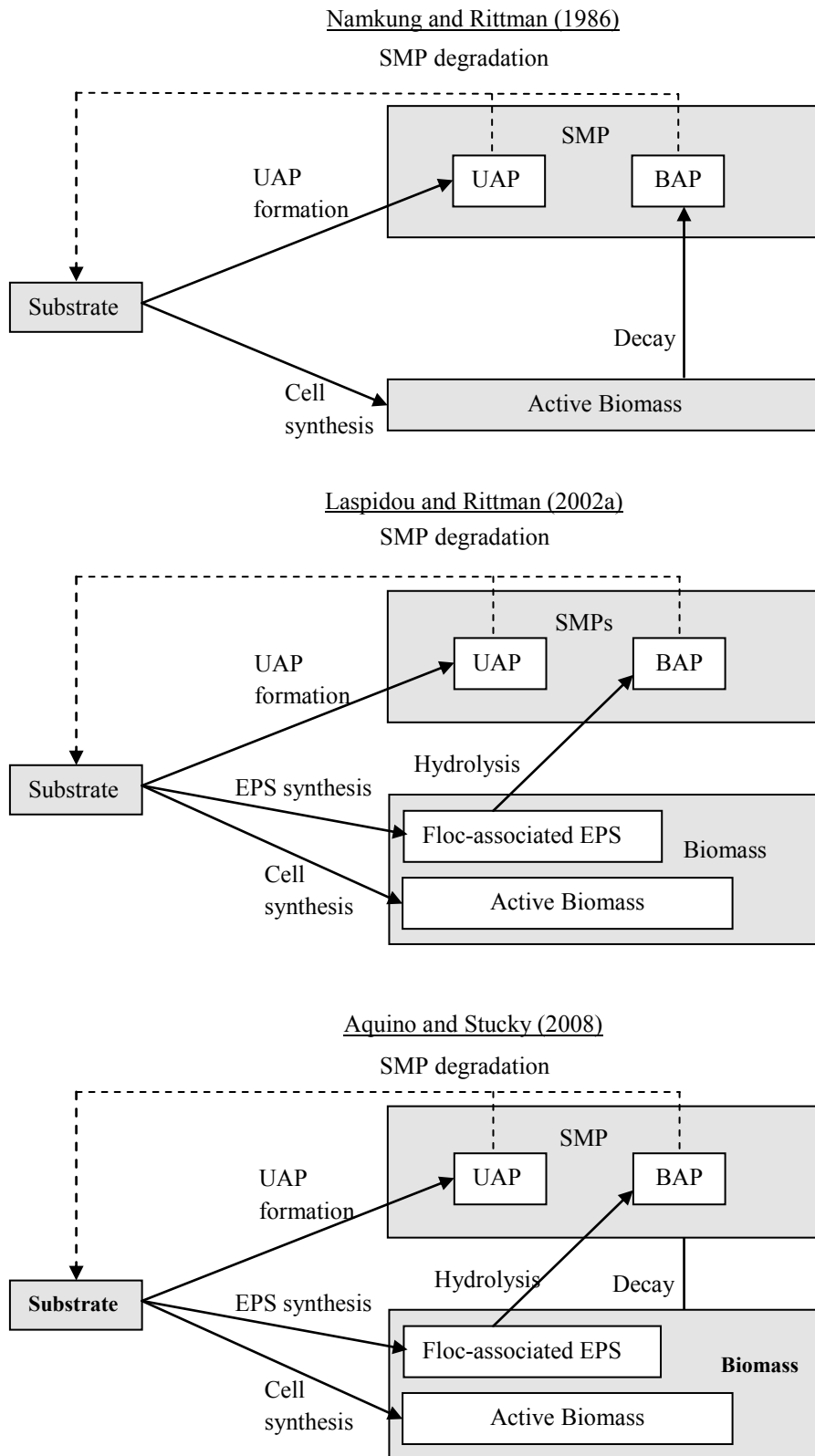


Figure 2.7 Different concepts of the formation and degradation of SMPs used in typical modelling studies (modified after Menniti and Morgenroth, 2010)

Despite the observation that there still remains disagreement about exact kinetics of SMP, it is crucial to include SMPs in the modelling of CASPs/MBRs. However, the correlation of SMP fractions with other parameters makes the MBR modelling a quite complex task resulting in a large number of variables in the model formulation. In a recent review, Fenu et al. (2010) mentioned that neglecting one of the two SMP fractions could lead to a simplification of the mathematical model at least by reducing the number of state variables. Based on the subjective judgement of the operating and environmental conditions of the treatment system for example, the SRT of the MBR system may determine the predominant fraction case by case for the different process specific conditions. Table 2.6 presents some of the empirical expressions that have been proposed by different researcher from their respective findings about the UAP and BAP formation and degradation kinetics.

*Table 2.6* Biokinetics of formation and degradation of SMPs (after Fenu et al., 2010)

Parameters	Equations	References
	$r_{UAP} = \mu_{het} X_{het}$ or $r_{UAP} = \mu_{aut} X_{aut}^*$ or	Lu et al. (2001)
UAP Formation rate	$r_{UAP} = \left( k_1 \cdot q_{uap} S / (K_s + S) \right) X_{bm}$	Laspidou and Rittman (2002b)
	$r_{UAP} = (Y_{uap}/Y) \cdot \mu \cdot X$	Janus and Ulanicki (2010)
UAP degradation rate	$r_{UAP} = -\mu_{SMP} \left( S_{SMP} / (K_{SMP} + S_{SMP}) \right) X_{het}^*$	Lu et al. (2001)
	$r_{UAP} = \left( -\cdot q_{uap} UAP / (K_{uap} + UAP) \right) X_{bm}$	Laspidou and Rittman (2002b)
BAP formation Rate	$r_{BAP} = K_{hyd} EPS$	Laspidou and Rittman (2002b)
	$r_{BAP} = K_2 X + K_{hyd} EPS$	Aquino and Stuckey (2008)
	$r_{BAP} = f_{bap} \cdot b \cdot X + (1 - f_s) \cdot K_{h,EPS} \cdot EPS \cdot Y_{BAP}$	Janus and Ulanicki (2010)
BAP degradation rate	$r_{BAP} = -K_{h,bap} S_{bap} X_H$	Jiang et al. (2008)
	$r_{BAP} = \left( -q_{uap} BAP / (K_{bap} + BAP) \right) X_{bm}$	Laspidou and Rittman (2002b)

Although the importance of EPS to cell aggregation has long been recognized, there has been limited modelling study on EPS formation. Luedeking and Piret (1959) proposed the first model (Eq. 2.1) to characterize microbial products' formation from the fermentation of lactic acid including only the consistent EPS formation as a growth and a non-growth associated product. This very simple model did not include the mechanism of EPS dissolution.

$$r_{\text{EPS}} = k_1\mu X + k_2X \quad \text{Eq. (2.1)}$$

The first term of the equation accounts for EPS formation associated with a first-order growth and the second term represents EPS formation associated with a non-growth term. An improved kinetic expression (Eq. 2.2) for EPS formation was introduced by Hsieh et al. (1994) where an EPS loss term is introduced by the third term of Eq. (2.2).

$$r_{\text{EPS}} = K_1\mu X_a + fK_2X_a - \mu_{\text{diss}}K_3\text{EPS} \quad \text{Eq. (2.2)}$$

Lapidou and Rittmann, (2002 a,b) studied the EPS mass balance (Eq. 2.3) in a continuous flow reactor which was latter applied in a submerged membrane bioreactor (Jang et al., 2006). The second term in Eq. (2.3) quantifies the rate of EPS loss due to hydrolysis, using a first order relationship.

$$r_{\text{EPS}} = K_{\text{eps}}r_sX_a - K_{\text{hyd}}\text{EPS} \quad \text{Eq. (2.3)}$$

The modified model has been based on the hypothesis that the formation of bound EPS is only growth associated, and is in direct proportion to substrate utilization. Aquino and Stucky (2008) proposed to model the formation of EPS as a non-growth associated product in anaerobic condition considering that both the soluble EPS and cell lysis products are the sources of BAP.

*Modified ASMs incorporating SMP/EPS concepts:* Ohron et al. (1989) first attempted to integrate of the formation and degradation kinetics of SMP into the ASM1 when

modelling a sequencing batch reactor. The model includes the so called soluble residual products SRP (equivalent to non-degradable BAP) that is assumed to contribute only to the soluble COD of the mixed liquor. Later Artan et al. (1990) further developed the model to include UAP. However, strong parameter correlations due to the model's combined concepts of formation and degradation of UAP and BAP affect their correct determination. Lu and coworkers, on the contrary, incorporated a very complex SMP model into ASM1 (Lu et al., 2001) (Figure 2.8a) and ASM3 (Lu et al., 2002) in MBR studies. Most of the original parameters of ASM1 were used, but the denitrification correction factor was enhanced to account for higher sludge concentrations in the system. The model contains eight SMP related parameters which can be determined by trial and error or can be approximated from references in literature. Besides significant underestimation of MLVSS concentration, the ASM1-SMP model predictions have been found to be close to the experimental observations for an intermittent aerobic MBR system. Oliveira-Esquerre et al. (2006) proposed modification of ASM3 by introducing five new SMP kinetic parameters ( $\gamma_{MP,H}$ ,  $\gamma_{MP,A}$ ,  $k_{MP}$ ,  $f_b$ ,  $Y_{mp}$  with values adopted from Lu et al., (2001), and two new processes. UAP and BAP are lumped into a general term MP in the modified ASM3 (Oliveira-Esquerre et al., 2006). Evaluation of both model predictions for a submerged MBR system has showed that the carbonaceous materials were more accurately estimated by the modified ASM3, while the model of Lu et al. (2001) performed slightly better in the estimation of nitrate. Jiang et al. (2008) has extended the existing ASM2d to ASM2dSMP by introducing kinetics for formation and degradation of SMP by hydrolysis steps, creating three new processes and imposing variations in thirteen other processes. The study has revealed the SRT as the key parameter controlling the SMP concentration.

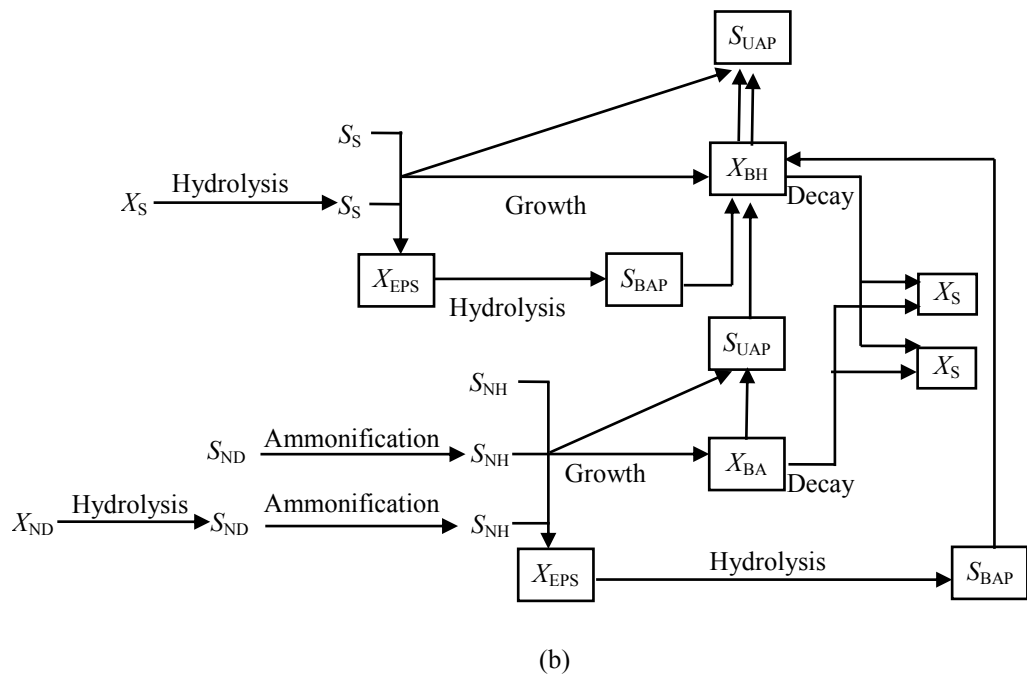
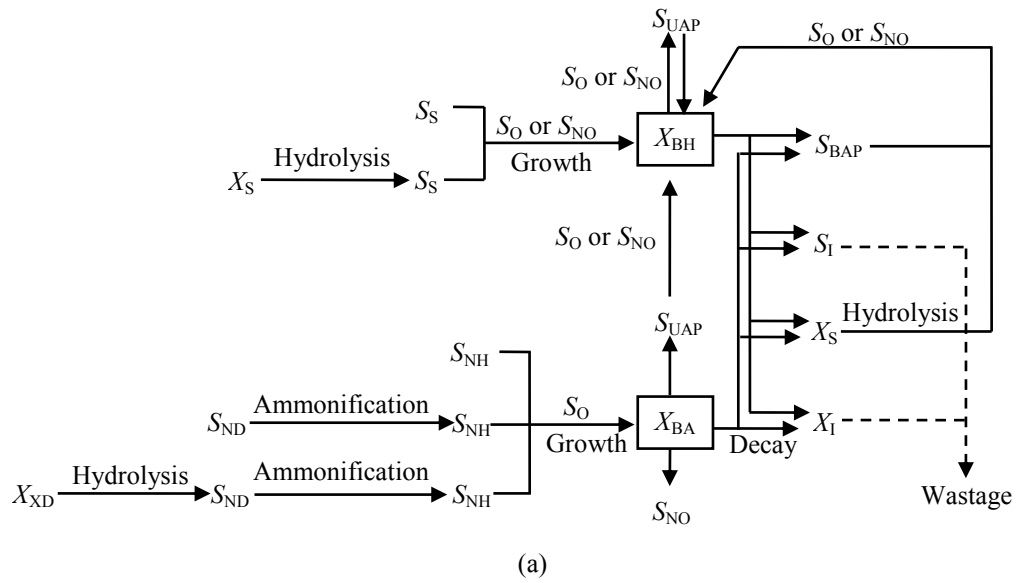


Figure 2.8 Schematic of the (a) ASM1-SMP hybrid model (adapted from Ng and Kim, 2007) (b) ASM1-SMP-EPS hybrid model (modified from Ahn et al., 2006)

Recently a new model, the CES-ASM3 model, has been proposed by Janus and Ulanicki (2010) in order to predict SMP formation and EPS production in an activated sludge system based on the Luedeking-Piret (1959) hypothesis. Janus and Ulanicki (2010) reformulated non-growth associated term with an additional reaction for EPS hydrolysis;

$$r_{EPS} = f_{EPS}\mu X + f_{EPS,d}bX - K_{h,EPS}EPS \quad \text{Eq. (2.4)}$$

where, UAP production is associated with biomass growth and substrate utilization while BAP formation is associated with biomass decay and hydrolysis or dissolution of EPS.

#### *D. Activated sludge/ MBR modelling including biological phosphorus removal*

The biological phosphorus (bio-P) removal process is not included in the common ASM modelling with ASM1 or ASM3. The bio-P removal or enhanced biological phosphorus removal (EBPR) in activated sludge systems has been typically described by two basic types of mathematical models, namely the metabolic models and the ASM2/2d. Considering the relevance of a particular model including kinetics of bio-P removal, the following models are discussed briefly regardless of their classification under the metabolic or ASM2 model or the combination of both. Table 2.7 give comparison of some of the basic mathematical model parameters, their default stoichiometric and kinetic values respectively which are typically used for the modelling of bio-P-removal in activated sludge process (ASP) and MBR.

The research group at the Delft University of Technology associated with IWQW task group presented *the TUDP* (Brdjanovic et al., 2000; Meijer, 2004; van Veldhuizen et al., 1999) model to describe EBPR of the ASPs. The TUDP model uses the maintenance concept instead of the decay concept of the ASM2/ASM2d model, and it is assumed that the bio-P-organisms always have internal substrate  $X_{PHA}$  available to satisfy the requirement for the maintenance of cell structure (van Veldhuizen et al., 1999).

Table 2.7 Comparison of different mathematical models for bio-P-removal (updated from Garnaey et al., 2004)

Models	ASM2	ASM2d	ASM3-bio-P	TUDP	UCTPHO+
Major processes involved with the phosphorus accumulating organisms	Storage of $X_{PHA}$	Storage of $X_{PHA}$	Storage of $X_{PHA}$	Aerobic storage of $X_{PP}$	Growth of $X_{PAO}$ on $X_{PHA}$ with $S_{NH4}$
	Storage of $X_{PP}$	Aerobic storage of $X_{PP}$	Aerobic storage of $X_{PP}$	Anoxic storage of $X_{PP}$	Growth of $X_{PAO}$ on $X_{PHA}$ with $S_{NO3}$
	Lysis of $X_{PAO}$	Anoxic Storage of $X_{PP}$	Anoxic storage of $X_{PP}$	Anoxic storage of $S_A$	Heterotrophic decay but $S_{PO4}$ limited
	Lysis of $X_{PP}$	Anoxic growth on $X_{PP}$	Aerobic lysis of $X_{PP}$	Aerobic $X_{PHA}$ consumption	Conversion of $S_f$ to $S_A$ but $S_{PO4}$ limited
	Lysis of $X_{PHA}$	Lysis of $X_{PAO}$	Anoxic lysis of $X_{PP}$	Aerobic $X_{GLY}$ formation	Anoxic growth of $X_{PAO}$ on $X_{PHA}$ with $S_{NH4}$
	Aerobic growth of $X_{PAO}$ on $X_{PHA}$	Lysis of $X_{PP}$	Anoxic growth on $X_{PHA}$	Anoxic $X_{GLY}$ formation	Aerobic growth of $X_{PAO}$ on $X_{PHA}$ with $S_{NO3}$
	Precipitation of $S_{PO4}$	Lysis of $X_{PHA}$	Anoxic respiration of $X_{PHA}$	Anaerobic Storage of $S_A$	Aerobic decay $X_{pp}$ lysis on anaerobic decay
Redissolution of $S_{PO4}$	Precipitation of $S_{PO4}$	Aerobic growth of $X_{PAO}$ on $X_{PHA}$	Anoxic $X_{PHA}$ consumption	$X_{PHA}$ lysis on anaerobic decay	
		Redissolution of $S_{PO4}$	Aerobic respiration of $X_{PHA}$	$X_{pp}$ cleavage for anoxic maintenance	
		Aerobic growth of $X_{PAO}$ on $X_{PHA}$	Aerobic endogenous respiration of $X_{PAO}$	$X_{pp}$ cleavage for anaerobic maintenance	
			Eendogenous respiration of $X_{PAO}$	Sequestration of $S_A$ by $X_{PAO}$	
EBPR	Yes	Yes	Yes	Yes	Yes
Chemical P-removal	Yes	Yes	No	No	No
Fermentation	Yes	Yes	No	Yes	Yes
Reactions	19	21	23	22	35
State variables	19	19	17	17	16
Full-scale application	CAS Yes	Yes	Yes	Yes	BNRS
	MBR No	Yes	No	No	No

The integrated metabolic model (TUDP model) was tested on full-scale WWTP's Haarlem Waarderpolder (Brdajanovic et al., 2000) and WWTP Hardenberg (Meijer et al., 2001), but the shortcomings of the model kinetics specifically to simulate the kinetics of glycogen were observed. Meijer et al. (2004) modified the model to solve the kinetic problems of the model, and concluded that operational conditions greatly influenced the WWTP operation. They also indicated that steady state conditions were not suitable to calibrate model kinetics since the growth of PAO's was mainly determined by the glycogen formation rate.

In order to model the biological behaviour for carbonaceous material removal, nitrification, denitrification and biological excess phosphorus removal (BEPR) for an activated sludge system with external nitrification, Hu et al. (2007a) developed a kinetic model called UCTPHO<sup>+</sup>. This model is a combination of metabolic and ASM2/2d models which has been derived from the UCTPHO model (Wentzel et al., 1992) but with modifications to address some of the deficiencies of the model ASM2/2d and Barker and Dold model (Barker and Dold, 1997). The basic UCTPHO model represented kinetics for ordinary heterotrophic organisms (OHOs) and nitrifiers as well as for the PAOs. The modified UCTPHO model, namely UCTPHO<sup>+</sup> model, was comprised of anoxic growth of PAO with associated anoxic uptake/denitrification/death/maintenance of PAO, provision for a separate reduced anoxic growth yield coefficient ( $Y_{H2}$ ) for OHO growth, and the linkage of the organic N and P fractions/transformation to the corresponding COD fractions/transformation (Henze et al., 1995). The model has been evaluated against a large number of experimental data sets under anaerobic, anoxic, and aerobic conditions and has been successfully used to simulate a wide variety of conventional BNRAS systems (Hu et al., 2007b). Simulation results have demonstrated that the model is capable of predicting



COD removal, nitrification and denitrification as well as aerobic and anoxic/aerobic P-uptake in EBPR with appropriately calibrated parameters. However, the model considers the hydrolysis process simultaneously with growth but without taking into account the anaerobic hydrolysis which may cause limitation in its usage as it is especially important for bio-P models to make substrate available for storage. Besides, like the above-mentioned models discussed, as denitrification and nitrification were modeled as one-step and the same decay rate under all electron acceptor conditions is not consistent with the experimental observations, the model is also not suitable to predict nitrite accumulation or  $N_2O$  production (Hauduc et al., 2013).

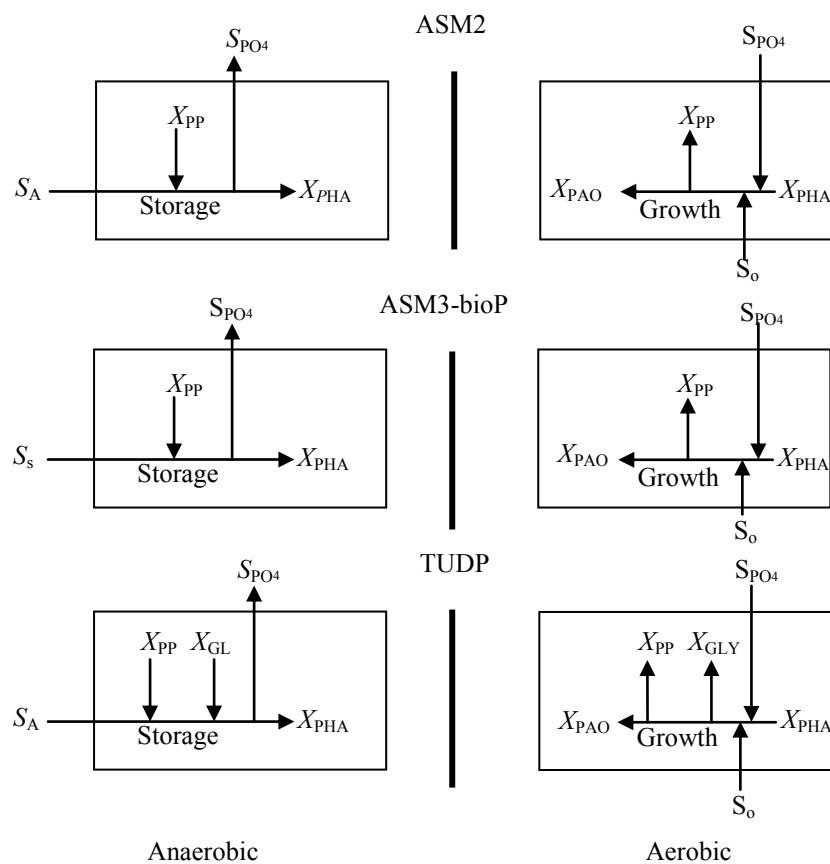


Figure 2.9 Flow diagram of anaerobic storage and aerobic growth of PAOs in ASM2 and ASM3-bio-P model (ASM2 adapted from Ng and Kim, 2007; TUDP model adapted from van Loosdrecht et al., 2008)

The ASM3-bioP model (Rieger et al., 2001) integrated the bio-P-removal to ASM3 (Gujer et al., 1999) including both the EBPR by the PAOs and the P-uptake during the growth of organisms. The model has four specific state variables ( $S_{PO_4}$ ,  $X_{PAO}$ ,  $X_{PHA}$ ,  $X_{PP}$ ) identical to ASM2d as well as 13 components of ASM3. The main limitation of the ASM3-bioP model is that no reliable characterization methods are suggested for some important parameters such as poly-P and glycogen. The model cannot be validated for a low resolution of COD, N and P and it also has limitation to accurately describe P-removal in all growth phases. The model does neither consider the decreasing phenomena of storing and response of PHA under anoxic condition nor does it include the anaerobic decay and chemical precipitation. In addition, fermentation is not considered in the ASM3-bio-P model and hydrolysis is considered as a rate-limiting step. Thus, this can be a major limitation of the model especially in cases where hydrolysis is no longer the rate limiting step (Hauduc et al., 2013). Sun and Song (2009) proposed an advanced model based on the ASM3-bioP model considering the effects of competition among microorganisms for organic carbon, nitrate and ammonia. In the so called Fully Coupled Activated Sludge Model No. 1 (FCASM1), Sun and Song (2009) added two equations into the kinetic expression to show the restraint on the nitrifier growth and the storage of  $X_{PHA}$ . However, the representation of the interaction mechanism among the microorganisms cannot adequately describe the competition among them for oxygen, ammonia and nitrogen.

It is evident from the above discussion that there are significant differences among the assumptions and kinetics involved in the modified ASMs models some of which were modified to account for the factors of a particular nutrient removal process, such as the TUDP, ASM3-bioP models for EBPR. In addition, the models mentioned are also based on the crude assumption that all processes including N and P-removal are

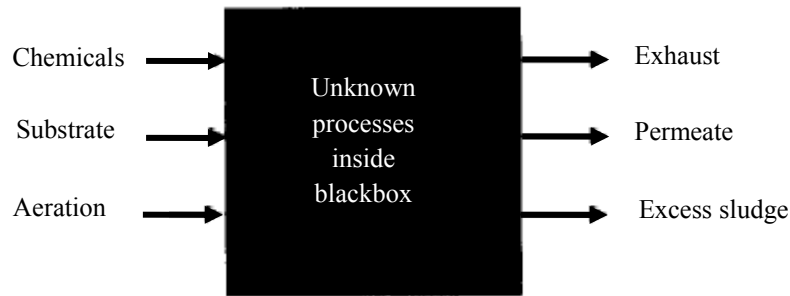
independent, thereby allowing no interactions among those processes. Therefore, due to the complexity of interaction (the coexistence of PAOs, autotrophic and heterotrophic organism) together with the asymmetry of aeration and hydraulics (no absolute area of anaerobic, aerobic and anoxic reaction) in the activated sludge system, all the models discussed in the review can only partially reflect the real processes and the application of the models are limited by factors such as temperature, toxicity and alkalinity. However, the modifications of the basic ASM models considerably increase the model parameters, and hence the complexity of model simulations and calibrations are also significantly increased. In this backdrop, some simplified mathematical modelling of activated sludge processes are also reported in the literature that describe limited aspects of the activated sludge treatment processes of an MBR. A few of the simplified mathematical models are described below considering their relevance to the study.

#### *E. Blackbox and shortcut models for modelling MBR biological processes*

A black box model generally applies a mass or mole balancing of input and output streams without considering the processes in between. Distribution coefficients are derived from experimental data in order to calculate output streams and concentrations from input streams and composition. Figure 2.10 depicts the typical conceptual formation of a black box model (Gehlert and Hapke, 2002) for a continuous aerobic MBR, detailed development of which is described in (Gehlert et al., 2001).

After the mass or mole balancing of the input and output streams using the blackbox model, Gehlert and Hapke (2002) developed two successive shortcut models of different complexities. In contrast to black box models, a shortcut model considers processes within the MBR system, such as biological growth or biochemical reactions. Its difference to rigorous models is that the whole MBR system is modeled as one functional group or one volume element. The subdivision into volume elements or

functional groups is the subject of rigorous models. This leads potentially to the advantage of subdivision of shortcut models of different complexities to form rigorous models.



*Figure 2.10* Blackbox model for continuous aerobic MBR process (adapted from Gehlert and Hapke, 2002)

The basic shortcut model developed by Gehlert and Hapke (2002) predicted TOC degradation as well as sludge production by applying the Monod kinetics for biological growth whereas the enhanced shortcut model additionally considered an inhibition of biological growth due to insufficient oxygen supply. The enhanced shortcut modelling may be applied as an indirect indicator of the biomass viability since the enhancement resulted in an additional inhibition term in the growth kinetics as well as a mass transport expression for oxygen passed from gas into liquid.

#### *F. Mathematical modelling of biomass viability in an activated sludge system*

Although a significant number of modelling studies were conducted on the biokinetics of the activated sludge, only a few modelling studies were aimed at identifying the biomass viability in connection to potential biomass parameters. The operators at the field level frequently need to devise operational control to maintain stable performance of a treatment which is still done by following some rules of thumb such as controlling an optimum SRT, HRT etc. In this regard, the mathematical models of biomass viability are important to track the stability in the performance of a biological treatment system.

Hasar et al. (2002) first proposed mathematical models of biomass viability by developing empirical correlations of SOUR with the inert fractions of the biomass. Assuming a fraction of the inert compounds might pass through the membrane, Hasar and Kinachi (2004) confirmed the accumulation and the production of inert compounds within the bioreactor by tracing increasing fractions of those in the effluent. The change of inert COD in the effluent was ascribed to the inert compounds that were produced due to microbial activity within the bioreactor. They proposed empirical mathematical expressions (Eqs. 2.5 and 2.6) representing the exponential decay or loss of microbial activity (viability) with its correlation established with the increasing ratio of inert chemical oxygen demand (COD) in the effluent to that in the influent ( $C_s/C_0$ ), and also with the ratio of volatile suspended solids to the mixed liquor suspended solid's concentration (MLVSS/MLSS). However, they took into account the production and accumulation of total inert compounds within the system without any fractionation of its constituent compounds whatsoever.

Power function:

$$\text{SOUR} = 1.811 * (C_s/C_0)^{-3.12} * (\text{MLVSS}/\text{MLSS})^{-1.389} \quad \text{Eq. (2.5)}$$

Exponential function:

$$\text{SOUR} = 1.33 + \exp(7.245 - 4.832 * (C_s/C_0) - 2.526 * (\text{MLVSS}/\text{MLSS})) \quad \text{Eq. (2.6)}$$

With the growing interest in identifying membrane foulants, it is important to identify the constituents/fractions of inert compounds such as SMP/EPS or colloidal compounds which may have a direct or indirect impact on biomass viability and membrane fouling as well. Germain et al. (2007) has reviewed the biomass effects on oxygen transfer in MBR where useful information can be found for better mathematical modelling of biomass viability including the effects of potential biomass parameters

such as SMP/EPS or their further fractionation into components such as proteins, polysaccharides and carbohydrates.

### ***2.3.2 Membrane fouling models***

The capacity of the membrane of an MBR system declines over time due to several fouling resistances such as pore blocking, porosity reduction, cake layer formation, biofilm formation, concentration polarization and a few other resistances. In this section, the stand-alone mathematical models of physical membrane fouling are discussed that account for the combined effects of the different individual fouling resistances to the physical fouling phenomenon. The modelling of membrane resistance to flux is fundamentally derived from Darcy's law that relates pressure head differences to time during constant flow operation of an MBR system.

#### *A. Empirical hydrodynamic model*

The crossflow velocity is an important parameter affecting the formation of the sludge cake layer on the membrane surface. The empirical hydrodynamic model investigates the influence of hydrodynamic conditions on the mixed liquor crossflow velocity and the membrane fouling rate in an MBR. In the hydrodynamic model proposed by Liu et al. (2003), the following equation was empirically derived from experimental investigations for the mixed liquor crossflow velocity:

$$U_{sr} = 1.311U_{Lr}^{1.226}e^{-0.0105X} \quad \text{Eq. (2.7)}$$

where  $U_{Lr}$  (m/s) is the crossflow velocity of tap water and  $X$  is the suspended solids concentration. The TMP was used for the calculation of membrane fouling rate as the rate of increasing membrane filtration resistance. The filtration resistance ( $R$ ,  $\text{m}^{-1}$ ) was computed from the following equation:  $10^9$

$$R = 3.6 \times 10^9 \frac{\Delta P}{\mu J} \quad \text{Eq. (2.8)}$$

where  $\mu$  (mPa-s) is the viscosity of the permeate and the factor,  $3.6 \times 10^9$ , stems from using the units given in parentheses for each of the variables.

Although the model is easy to use due to the straight-forward adoption of the empirical hydrodynamic equation, it cannot capture the contribution of the individual fouling resistances to the overall development of fouling resistance. The typically observed phenomena of reversible and irreversible membrane fouling are not accounted for in the model development. The model may be useful for illustrating hydrodynamic effects on membrane fouling, but it may not be suitable for the purposes of operation and design of MBR systems (Ng and Kim, 2007).

### *B. Fractal permeation model*

Meng et al. (2005) developed a fractal permeation model based on Darcy's law. They hypothesized that the microfiltration of activated sludge formed a disordered and complicated sludge cake layer on the membrane which could not be modeled by the application of traditional geometry. In order to evaluate the permeability of such an irregular cake layer they applied the fractal geometric theory to determine the pore area fractal dimension,  $D_s$ , of a cake layer in terms of its average self-similar properties. The permeability of the porous cake was described according to the following equation:

$$\kappa = \frac{\mu L_0 Q}{\Delta P A_t} = \frac{G}{A} C_0 \frac{1}{A_t} \frac{2 - D_s}{3 - D_s} a_{\max}^{3 - D_s} \quad \text{Eq. (2.9)}$$

where  $\mu$  is the permeate viscosity,  $L_0$  is a constant,  $\Delta P$  is the pressure gradient,  $A_t$  is the total pore area,  $Q$  is the flow rate,  $G$  is the geometry factor for fluid flow through a pore,  $C_0$  is a constant and  $a$  is a threshold pore area. Since the model is composed of relatively few parameters, it is easy to calibrate the model parameters. However, the validation of the model based on true experimental observation is complex. The model

does not illustrate how transient operational parameters and conditions affect cake layer formation on the membrane.

### C. Sectional resistance model

In a submerged MBR system, coarse bubbles from aeration provide a cleaning mechanism for the immersed membrane modules by the application of shear force and thus scouring the surface of the foulant layer. The variable effective shear forces over the section of the membrane gives rise to uneven cake formation and detachment at different sections of the membrane. Li and Wang (2006) proposed a sectional resistance model in order to model such uneven cake formation and degradation due to varying shear distribution over the length of the membrane. The membrane surface is divided into equal fractional areas,  $\Delta\varepsilon$ , and the total resistance ( $R_T$ ) is calculated from the separate resistances determined for each section;

$$R_T = R_m + R_p + R_{sf} + R_{sc} \quad \text{Eq. (2.10)}$$

$$R_p = r_p \sum J \theta_f ; \quad R_{sf} = r_{sf} M_{sf}; \quad R_{sc} = r_{sc} M_{sc} \quad \text{Eq. (2.11)}$$

where  $R_m$ ,  $R_p$ ,  $R_{sf}$ ,  $R_{sc}$  denote inherent membrane resistance, pore fouling resistance, dynamic sludge film resistance and the resistance due to stable sludge film respectively;  $r_p$ ,  $r_{sf}$ ,  $r_{sc}$  are the specific resistance of pore fouling, dynamic sludge film and sludge cake layer respectively;  $J$  is the permeate flux;  $\theta_f$  is the filtration period of an operational cycle;  $M_{sf}$  and  $M_{sc}$  respectively are the mass of the dynamic sludge film and biomass accumulated on the membrane surface. The mass of the sludge in the dynamic film was determined from the following equation:

$$\frac{dM_{sf}}{dt} = \frac{24CJ^2}{24J + C_D D_p G} - \frac{\beta(1-\alpha)GM_{sf}^2}{\gamma V_f t + M_{sf}} \quad \text{Eq. (2.12)}$$

where  $C$  is the sludge concentration,  $J$  is the local permeate flux in the membrane section,  $C_d$  is the coefficient of the lifting force of a sludge particle of diameter  $d_p$ , and



$G$  is the shear intensity on the section of the membrane surface,  $\beta$  is the erosion rate coefficient of the dynamic sludge,  $\alpha$  is the stickiness of biomass particles,  $\gamma$  is the compression coefficient for dynamic sludge,  $V_f$  is water production within the filtration period of the operation cycle and  $t$  is the filtration time. The first and second terms of Eq. (2.12) represent the rate of attachment and detachment of the sludge cake respectively.

The advantages of this transient model are that it accounts for cleaning cycles and characterizes fouling development over time with varying sludge concentrations, filtration fluxes, and aeration intensities. However, the model parameters are not easy to determine and calibration of the model for the total fouling resistances may lead to further complexity due to the many parameters involved in the model.

#### *D. Combined mechanistic model*

Kim et al. (2013) proposed a combined mechanistic model by combining four constant-flow rate blocking mechanisms. The model was developed to account for two effects on the fouling process: (1) Loss of available membrane area by the membrane blocking and (2) increase of hydraulic resistance by cake formation. Once the membrane is blocked by the combined effect of complete or partial blocking mechanisms, the volume of permeate passed through the available membrane area is expressed as follows:

$$V = \frac{\exp(k_i J_0 t) - 1}{k_i} - \frac{J_0}{k_b} \ln(1 - k_b t) \quad \text{Eq. (2.13)}$$

The increase in hydraulic resistance to flux was assumed by the combined effect of cake formation and standard blocking mechanisms, and was expressed by the following equation:

$$\frac{R}{R_0} = (1 + k J_0 V) + \left(1 - \frac{K_s V}{2}\right)^{-2} \quad \text{Eq. (2.14)}$$

where  $k$ ,  $k_s$  and  $V$  are the parameters for cake filtration, standard blocking and volume of permeate respectively. The model simulation identified that the membrane fouling was extensively progressed from the intermediate blocking to cake filtration.

#### *E. Dynamic mathematical model of membrane fouling*

The dynamic mathematical model proposed by Giraldo and LeChevallier (2006) was intended to obtain a description of cake formation and growth, removal of substrate due to cake-membrane behavior, change in membrane permeability over time, increase in cake head-loss over time, removal of SMPs by the cake and the associated change of TMP over time. The model differentiates between the internal fouling of membranes due to SMP during subcritical flux operation and supercritical flux fouling due to cake formation and compression. Additionally, it allows for changing the operational conditions of the membrane compartment of the bioreactor as a function of time such as modifications of permeate fluxes e.g. membrane relaxation, modification of aeration rates, backflushing and changes in water quality variables during one run while capturing the filtration effect generated by the cake. In case of internal fouling, the following expression was used to correlate membrane permeability, pore size and membrane resistance:

$$R_p = \frac{8\theta h_m}{f r_p^2} \quad (\text{Eq. 2.15})$$

where  $\theta$ = pore tortuosity,  $h_m$ = membrane effective thickness,  $f$ = membrane porosity,  $r_p$ = effective pore radius. As the adsorption is a time-dependent phenomenon,  $f$  and the  $r_p$  have been considered as time-dependent variables. The differential equation to account for the effect of porosity/pore size reduction due to the adsorption of soluble particles within the pores is given in equations (2.16) and (2.17).

$$\frac{df}{dt} = -\alpha_f C'_m J \quad \text{Eq. (2.16)}$$

$$\frac{dr_p}{dt} = -\alpha_p C'_m J \quad \text{Eq. (2.17)}$$

where  $\alpha_f$  is the membrane porosity reduction coefficient,  $\alpha_p$  is the membrane pore reduction coefficient,  $C'_m$  is the concentration of fouling particles at the membrane surface e.g. SMPs and  $J$  is the permeate flux.

The external membrane fouling was considered to be caused by the irreversible deposition of cake layer, and the cake filtration effects accounting for the cake compressibility is included in the mathematical expressions for cake resistance ( $R_c$ ) as shown in equation (2.18).

$$R_c = \widehat{R}_c z_c \quad \text{Eq. (2.18)}$$

where  $\widehat{R}_c$  = specific resistance of the compressible cake layer,  $z_c$  = depth of cake layer to be determined according to the following mass balance equation around the membrane surface:

$$\rho_c \frac{dz_c}{dt} = J \cdot C_b - \alpha_v \cdot v_{\text{air}}^\beta \quad \text{Eq. (2.19)}$$

where  $\alpha_v$  is the air scouring coefficient,  $C_b$  is the concentration of potential cake forming particles in the bulk liquid,  $\rho_c$  is the density of the cake layer,  $v_{\text{air}}$  is the scouring air surface velocity and  $\beta$  is the air scouring dependence exponent.

Using the dynamic model, Giraldo and LeChevallier (2006) could simulate some of the complex but commonly observed effects such as: exponential increase in TMP due to high MLSS, reduced fouling rates at increased aeration intensities, subcritical operation fouling and effect of increased particle size on the filterability of the microbial suspension. The results from the membrane and cake resistance were combined into Eq. (2.20) to obtain the change of TMP and cake pressure differential as a function of time:

$$J = \frac{\Delta p}{\mu(R_m + R_{\text{biofilm}} + R_{\text{cake}} + \dots)} \quad \text{Eq. (2.20)}$$

The modelling approach, however, does not allow modelling of the variation in colloidal particles that might take place during dynamic influent flow conditions to the bioreactor. There may be a change in the backward transport rate of cake-forming particles as well due to differences in particle size. These effects should be added in the model if increased prediction ability of the dynamic model is required.

Busch et al. (2007) presented a model covering the geometry of the system, the hydrodynamics of the feed and of the permeate flow, and the filtration resistance. The filtration resistance model considers membrane resistance, pore blocking, cake layer formation, poly-dispersed particles, biofilm formation and concentration polarization. The mathematical expression for the total membrane resistance ( $R_T$ ) is as follows in Eq. (2.21)

$$R_T(z) = R_m + R_c(z) + R_p(z) + R_b \quad \text{Eq. (2.21)}$$

where,  $R_m$  is the intrinsic membrane resistance;  $R_c(z)$ ,  $R_p(z)$  and  $R_b$  are the time-dependant resistances due to cake layer, pore blocking and biofilm formation respectively. The time-dependent pore blocking and cake formation was fundamentally based on the model developed by Broeckmann et al. (2006) with one extension accounting for irreversible pore blocking. The concept was extended to the case of backwashing, when the filtration flux is negative. The increasing porosity,  $\varepsilon$  is then described according to the following Eq. (2.22)

$$\rho_p \frac{d\varepsilon(z)}{dt} = \begin{cases} -4J(z)C_m^b \frac{d_{f,o}}{(d_{f,o})^2 - (d_{f,i})^2} & \text{if } \varepsilon < \varepsilon_{\max}(z) \\ 0 & \text{if } \varepsilon = \varepsilon_{\max}(z) \end{cases} \quad \text{Eq. (2.22)}$$

where,  $\rho_p$  is the particle density,  $d_{f,o}$  and  $d_{f,i}$  are the outer and inner membrane diameter  $C_m^b$  is the concentration of particles entering the pore.

It is assumed in the model (Busch et al., 2007) that the cake layer is completely reversible while biofilm accumulating between the cake layer and membrane surface is

at least partially resistant to backwashing. The biofilm resistance  $R_b$  was described as the product of its thickness  $L_b$ , its density  $\rho_b$  and its specific resistance  $\alpha_b$  according to the following equation:

$$R_b = \alpha_b L_b \rho_b \quad \text{Eq. (2.23)}$$

The specific resistance  $\alpha_b$  was estimated according to Nagaoka et al. (1989) as follows

$$\frac{d\alpha_b}{dt} = k_\alpha (\alpha_{\max} - \alpha_b) \quad \text{Eq. (2.24)}$$

$$\alpha_{\max} = \alpha_0 + \alpha_p \Delta p \quad \text{Eq. (2.25)}$$

where  $k_\alpha$ ,  $\alpha_{\max}$  and  $\alpha_{\max}$  are empirical parameters and  $\Delta p$  is the transmembrane pressure difference.

The thickness of  $L_b$  depends on the attachment and detachment of EPS around the membrane, and the dynamic variation of it was obtained as follows:

$$\frac{dL_b}{dt} = \begin{cases} u_f, & J(z) > 0 \\ -u_b, & J(z) < 0 \end{cases} \quad \text{Eq. (2.26)}$$

$$u_f = u_{f,a} - u_{f,d} \quad \text{Eq. (2.27)}$$

where  $u_{f,a}$  and  $u_{f,d}$  are growth rates due to attachment and detachment during filtration while  $u_b$  is the detachment rate during backwashing.

The dynamic fouling models proposed by Giraldo and LeChevallier (2006), and Busch et al. (2007) include a comprehensive description of the different fouling resistances. However, the models are too complicated to be used for practical design and modelling purposes, especially the determination of reliable model parameters for the rate of cake layer detachment due to air scouring (Giraldo and LeChevallier, 2006) or backwashing (Busch et al., 2007) needs complicated calibration of the models. The combined effects of aeration and backwashing on the cake layer detachment need to be

integrated in the fouling model by more simple assumptions about the phenomenon of the cake layer detachment.

### ***2.3.3 Integrated and hybrid MBR models***

An integrated MBR model refers to a model integrating the biological process with a membrane filtration, i.e., a model that accounts for the reciprocal impact of MBR biology on membrane fouling. The formulation of an integrated model needs a few sub-models, i.e., a biological model preferably an activated sludge model, a hydrodynamic model and a filtration (resistance) model. The key integration is to select suitable variables to link these sub-models. There are no truly mechanistic integrated MBR models developed so far due to a lack of complete understanding of the true interactions among different sub-models. The following sections present a brief review of a few efforts for integrated model development that are reported in the literature.

Various by-products of the metabolic activity of bacterial cultures, specially the SMPs and EPSs, have been found to be correlated with floc strength and resistance to shear and to influence various properties of activated sludge (Janus and Ulanicki, 2010). For this reason, researchers adopted different ASM models for improved bioprocess modelling and then coupled those biological models with sub-models of membrane fouling (e.g. Di Bella et al., 2008; Lee et al., 2002; Mannina et al., 2011; Zarragoitia-Gonzalez 2008). The choice of ASM models for bioprocess modelling also varied depending on the type and purposes of different MBR systems. However, early efforts of integrated MBR model development ignored the incorporation of formation and degradation kinetics of SMPs/EPSs. Early efforts of integrated MBR model development ignored the incorporation of formation and degradation kinetics of SMPs/EPSs into the bioprocess models. Lee et al. (2002) presented an MBR model based on the model of Lu et al. (2001) coupled with a resistance-in-series filtration

model for simulating fouling phenomena. The concentration of SMP in the model was assumed negligible compared to the TSS. Wintgens et al. (2003) introduced an integrated model employing ASM3 with a resistance-in-series model to describe the filtration performance of a submerged capillary hollow fibre module in an MBR. The simulation results for the evolution of permeability over time matched well the data from the pilot plant except for a major deviation at the end of the considered period.

Lee et al. (2002) developed a model for SMBR by coupling an ASM1-SMP hybrid model with a conventional resistance-in-series model. Four additional processes are presented in the model for describing the fate of SMP while the process rates and stoichiometry for the carbon and nitrogen are kept the same as ASM1. All the UAP but only a portion of BAP is considered to be biodegradable in the model. The membrane fouling in the model is captured by the conventional resistance-in-series model which has its components derived from the bioprocess sub-model. Although the contribution of the SMP is considered for its influence on the specific resistance, its contribution is ignored in the total cake mass deposited on the membrane.

Jang et al. (2006) developed a model based on the unified theory (Laspidou and Rittmann 2002a) for the production and degradation of EPS and SMP. The model was extended with several additional equations using the modified fouling index ( $MFI_{MBR}$ ) of the MBR aimed at predicting the biofouling potentials caused by soluble and suspended solids (Eq. 2.28). The  $MFI_{MBR}$  in the model was divided into soluble materials ( $MFI_{Sol}$ ) and suspended solids ( $MFI_{SS}$ ) and were measured by Eq. (2.29) and Eq. (2.30) respectively.

$$MFI_{Sol} = \beta[S(1 - f_S) + BAP(1 - f_{BAP}) + UAP(1 - f_{UAP})] \quad \text{Eq. (2.29)}$$

$$MFI_{SS} = \gamma \frac{QS_i}{V(X_a + EPS)} \quad \text{Eq. (2.30)}$$

The simulation results showed that the  $MFI_{Sol}$  contributed more to biofouling of the MBR than the  $MFI_{SS}$  except for a SRT less than 5 days. The model is over parameterized and it has yet to be validated by the observed results of the full-scale MBR plants. Janus and Ulanicki (2010) implemented the theory of production and degradation of SMP and EPS as proposed by Laspidou and Rittmann (2002a, b) in an ASM3-SMP/EPS hybrid model. The model simulations indicated an increased production of SMP and EPS at higher MLSS, lower temperatures and lower SRT. The model also predicted a slight increase in SMP and EPS with increased DO concentration.

Zarragoitia-Gonzalez et al. (2008) presented a hybrid model linking part of an ASM1-SMP hybrid model (Lu et al., 2001) and a membrane fouling model. Only heterotrophs were considered in the model. Although biomass decay was considered for the BAP production, the SMP concentration was used to calculate bound EPS as  $S_{UAP} + S_{BAP} / 0.8 X_{TSS}$ . Considering the usual operating condition of an MBR, the use of the model was limited to aerobic condition only. As the model was based on Lu's model, the model predicted an incomplete and incorrect COD balance. Although the model contains a high number of SMP parameters, it neglects some important physical mechanisms and phenomena, such as the dynamic deep-bed filtration of the cake layer and their possible influence on the removal of organics (Mannina et al., 2011).

Another adoption of the biological mechanism of Lu's model was by Di Bella et al. (2008) and it was connected to the physical mechanism of the membrane for the removal of organics. The formation of the cake layer on the membrane was described according to the Li and Wang (2006) model. In addition, COD removal by the cake layer and the physical membrane were quantified in the model. The model was calibrated well and could predict the COD better than the previous models of its kind.



However, the SMP concentration was missing in the work. The complexity of the model calibration was reduced by the calibration using the most sensitive parameters only. di Bella et al. (2008) carried out about 10,000 Monte Carlo simulations for the calibration of the model followed by the calibration by trial and error in order to define the values of the most sensitive parameters of the model. It was found in the sensitivity test that the  $Y_{SMP}$  and  $\gamma_{UAP,A}$  had strong influence on the majority of output variables while  $b_{BAP,h}$  and  $\mu_{SMP}$  had mainly affected the parameters  $NH_4$  and  $NO_3$ . According to Mannina et al. (2011), the major limitations of the model were that the different effects of aeration on the cake deposition were not taken into account in the model, and the filtration was considered uniformly distributed on the entire surface of the membrane which is impractical especially in the case of a hollow fibre membrane in submerged configuration.

In order to overcome the above limitations of the above model (di Bella et al., 2008), a few modifications were proposed by Mannina et al. (2011) e.g. the kinetics of the SMP formation and degradation (according to Jiang et al., 2008), the dynamic phenomena of the attachment and detachment of the cake layer on the membrane and their influence of the development of fouling, the variation of the transmembrane pressure (TMP) and the variation of the membrane resistance due to fouling. An innovative calibration protocol based on a step-wise approach followed by preliminary global sensitivity analysis was employed to calibrate the model. In the new model, the biological process was based on a modified version of the modelling approach proposed by Jiang et al. (2008) which was linked with the sectional resistance-in-series model for modelling the physical mechanisms of the membrane. From the results of the sensitivity tests, it was reported that  $Y_a$ ,  $f_{Si}$  and  $f_{BAP}$  influenced COD in the reactor while the  $i_{x,p}$  is influential on the  $NH_4$  concentration in the permeate. Like the earlier models, SMP

concentration was also missing in the work but the model predicted well the MLSS,  $\text{COD}_{\text{perm}}$ ,  $\text{NO}_3$  and  $R_{\text{tot}}$  during the long-term behaviour of the treatment system. The simulations showed a discrepancy in some cases which was ascribed to the poor experimental investigation during the first days of the start-up period and also to the initial inoculum biomass acclimatization that was not modeled.

In a study to find out the influences of start-up strategies on MBR performance and fouling development of membrane fouling, Mannina and Di Bella (2012) applied the model developed by Jiang et al. (2008) in order to assess different fouling rates. Mannina and Di Bella (2012) monitored an MBR pilot plant for two experimental periods, each of 65 days, and within this time the plant was started up with (period 1) and without (period 2) inoculation of biomass. Higher net SMP production during the start-up period with sludge inoculation was observed compared to the period without sludge inoculation. The finding was attributed to the increased biological activity in the experimental period 2 because of the different SRT. However the stickiness of the biomass particles in period 1 was slightly higher than that in period 2 which was assumed due to the high SMP concentration in the initial phase of start-up without inoculation. Also assuming that the SMPs can be changed by the biological and physical actions which can be retained at a different proportion inside the cake layer, three different SMP concentrations were calculated at the different MBR sections: reactor ( $\text{SMP}_{\text{reactor}}$ ), cake layer-membrane interface ( $\text{SMP}_{\text{cake-mem}}$ ) and permeate ( $\text{SMP}_{\text{permeate}}$ ). The results of the model simulation (Mannina and Di Bella, 2012) confirmed the complex relationship between MLSS and SMP formation-degradation especially during the start-up phase. Lower concentration of SMPs was found in period 1 due to higher SRT. However, higher specific SMP concentration was observed during

the first 25 days of plant operation in period 1 which might be due to the variations in operational conditions i.e. F/M and MLSS.

Various by-products of the metabolic activity of bacterial cultures, specially the SMPs and EPSs, have been found to be correlated with floc strength and resistance to shear and to influence various properties of activated sludge (Janus and Ulanicki, 2010). However, early efforts of integrated MBR model development ignored the incorporation of formation and degradation kinetics of SMPs/EPSs. There also exist differences among different empirical expressions that were independently developed for a particular MBR system having system dependent relations between the hydrodynamic variables and membrane fouling. As such, there is no general consensus about the correlation of hydrodynamic variables with membrane fouling independent of the MBR systems. This associated with the lack of coherence among different methods of influent wastewater characterization makes the task of calibration of ASMs for MBR bioprocess modelling a very complicated one. Some of the process variables introduced into the hybrid models are impossible to be determined experimentally in full-scale MBR systems (e.g. UAP and BAP) and, thus, serious identifiability issues are raised (Patsios and Karabelas, 2010).

Most of the integrated models developed have been evaluated for a bio-system with rather simple substrate inputs that were considered merely soluble and readily biodegradable. In contrast, the organic matter in influent wastewater of a real treatment plant is very complex and consists of both soluble and particulate fractions with different biodegradability rates. Moreover, most of the ASM-SMP/EPS hybrid integrated models developed so far are generally too complicated and over-parameterized. In this backdrop, it is very difficult to formulate a complete but easily implementable integrated mathematical model framework for an MBR system. Zuthi et

al. (2013) proposed some ideas (Figure 2.11) that can be integrated into the conventional integrated MBR model framework, i.e. incorporation of empirical expressions in order to derive fractions of SMP/EPS from other measurable parameters, shortcut sub-models with variables determined using the black box model. These, along with other simplifications, may lead to reformulation of MBR bioprocess modelling using ASM-SMP/EPS hybrid models.

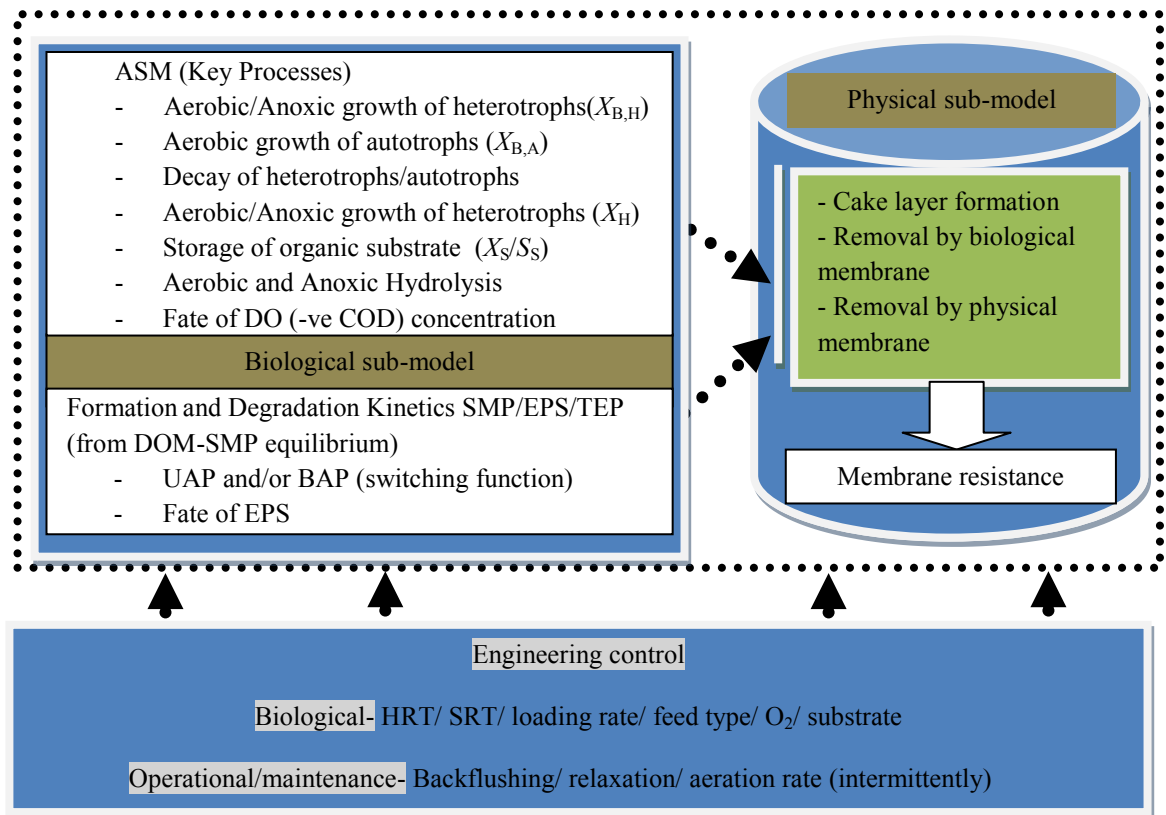


Figure 2.11 Conceptual diagram of integrated model framework for MBR system (Zuthi et al., 2013)

## 2.4 Concluding remarks

The conventional mathematical model, by construct, is used to describe observed phenomena of something that can be interpreted with scientific arguments and correlations. While the mathematical modelling of CAS and/or MBR systems helps scientists understand the mechanics of the bioprocesses of the activated sludge

treatment processes and the processes' interdependencies, the end-users of the mathematical model may be additionally interested in tracking the health of the treatment system and devising operational control strategies by the application of mathematical models such as how optimum treatability of the system can be sustained and how the membrane fouling can be controlled by the change of parameters such as MLSS, SMP and bEPS. The focus of this PhD research is on the later aspect of the mathematical modelling of biomass viability and membrane fouling aimed at operational control of MBR systems.

The bacterial population in biomass exhibits heterogeneous oxidation kinetics. The review of the MBR biokinetics, membrane fouling, and state-of-the-art CAS and MBR mathematical models revealed that the individual bacterial component play specific roles in biological treatment of water for certain organics or nutrients while simultaneously contributing to dynamic oxidation kinetics of the mixed liquor. The oxidation biokinetics of a biomass component are also affected by several other biological and/or environmental parameters or process variables. Consequently, a complete mathematical model of biomass viability would inevitably include all the parameters that affect to a lesser or greater extent the biological treatment processes. However, the mathematical modelling of biomass viability in relation to all the components of biomass would obviously become highly complicated due to the need to include many parameters in the model.

The biomass properties in different activated sludge treatment processes may be characterized by analysing the morphological properties of biomass (e.g. floc size), the physical parameters such as dynamic viscosity and potential bio-chemical components such as SMP/EPS (Ji et al., 2010). The daily measure of biomass viability is important for tracking the health or efficiency of any wastewater treatment system, and the

conventional practice is to monitor the morphological properties of biomass such as floc size and structure, the filamentous problem in addition to the oxygen uptake rate (SOUR) of the microbes. However, the monitoring or tracking of biomass viability in terms of gross biomass parameters (e.g. MLSS/MLVSS) may help prevent critical upsets of the treatment processes by controlling mixed liquor properties such as changing the mixed liquor concentration which subsequently would affect the dominant membrane-fouling components (e.g. SMP/bEPS). In this regard, the following chapters are dedicated to present a new mathematical model of biomass viability including the gross biomass parameters such MLSS or MLVSS as well as the dominant membrane-fouling components such as SMP/EPS. This would help in tracking the health of the MBR treatment system and in devising operation control strategies against critical upsets of treatability and fouling.



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## **CHAPTER 3**

# **New Conceptual Mathematical Models for Biomass Viability and Membrane Fouling of a Membrane Bioreactor**

### **3.1 Introduction**

With the growing interest in identifying membrane foulants, researchers have identified that certain constituents of inert compounds of wastewater such as soluble or colloidal microbial products contribute potentially to the development of membrane fouling. These soluble or colloidal inert compounds also have direct or indirect effects on the oxygen transfer for microbial activities and hence on the biomass viability. Basically, the optimal efficiency for removal of organics or nutrients by MBR systems while enabling control of membrane fouling depends on the bio-oxidation kinetics facilitated by a suitable environment with minimum inhibition by any product, processes or environmental disturbances.

However, a direct correlation among these potential biomass parameters concerning the biomass viability and membrane fouling has yet to be developed. Consequently, this chapter aims at proposing new conceptual models of biomass viability considering the potential effects of the soluble and colloidal inert particles which also have strong correlation with the model of membrane fouling. The ultimate objective is to develop new conceptual mathematical models of biomass viability and membrane fouling which can be used in an integrated way for the operational control of an MBR for treatability and fouling control.

### **3.2 Methods of the development of conceptual models**

#### *3.2.1 Background and state-of-the-art*

The biochemical functions of the microbial communities within the MBR evolves by a continuous generation of new sludge soon after the organic feed enters the bioreactor and contacts the biomass (Hasar and Kinachi, 2004; Navaratna et al., 2012). There have been a lot of studies evaluating the efficacy of the treatment and fouling control of



different MBR systems credited to the particular configuration of the treatment system. However, only a few studies (Chen et al., 2012; Clouzot et al., 2011; Hasar et al. 2002; Hasar and Kinachi, 2004) were intended to reveal the biomass viability, which is particularly important for the operational control of an MBR for better treatability.

Generally, the viability of microbial activities in the mixed liquor suspended solids (MLSS) can be conveniently monitored by the oxygen transfer efficiency within the treatment system. However, controlling the biomass (MLSS) properties that affect the oxygen transfer for the microbial activities is rather complex as biomass is a heterogeneous mixture of particles, microorganisms, colloids, organic polymers and cations, of widely varying shapes, sizes and densities (Germain and Stephenson, 2005). There is also a lack of general consensus about the methods of characterization of the components of MLSS and thus, no single component/s has yet been identified exclusively influencing the oxygen transfer for microbial activities.

The decrease of specific oxygen uptake rate (SOUR) generally corresponds well with the loss of microbial activity associated with the lower substrate utilization rate (Kim et al., 2001). As a consequence, this results in higher chemical oxygen demand (COD) concentration in the reactor with an increased fraction of colloids present in the bioreactor along with soluble microbial products (SMP) or extracellular polymeric substances (EPS). Therefore, the biomass viability of the activated sludge processes is conventionally monitored by the SOUR of the sludge supernatant of the bioreactor while evaluating its correlation to some other biomass parameters traced at the effluent flow. Hasar et al. (2002), for example, investigated the relationship between the inert COD in the influent and that in the effluent of a submerged MBR (SMBR) system. They reported that the inert COD in the effluent was higher than that in the influent

wastewater which was ascribed to the production of inert compounds by the microorganisms and their progressive accumulation within the bioreactor.

The mixed liquor volatile suspended solids (MLVSS) and the SOUR have been conventionally used as simple indicators of the biomass viability as the accumulation of inert (non-biodegradable) matter within the bioreactor was commonly observed to increase the MLVSS with time resulting in a reduction of the oxygen uptake rate within the system (Hasar et al., 2002; Hasar and Kinachi, 2004). Hasar and Kinachi (2004) proposed an empirical model of the viability of microbial activity in an SBR where the accumulation of inert compounds within the bioreactor was identified affecting the biomass viability with operation time. They proposed empirical mathematical expressions (Eqs. 3.1 and 3.2) to represent the exponential decay or loss of microbial activity (viability). Hasar and Kinachi (2004) also established important correlations between the biomass viability and the increasing ratio of inert COD in the effluent ( $C_s$ ) to that in the influent ( $C_0$ ), and also the ratio of volatile suspended solids to the mixed liquor suspended solid's concentration (MLVSS/MLSS).

Power function:

$$\text{SOUR} = 1.811 * (C_s/C_0)^{-3.12} * (\text{MLVSS}/\text{MLSS})^{-1.389} \quad \text{Eq. (3.1)}$$

Exponential function:

$$\text{SOUR} = 1.33 + \exp(7.245 - 4.832 * (C_s/C_0) - 2.526 * (\text{MLVSS}/\text{MLSS})) \quad \text{Eq. (3.2)}$$

The empirical model, however, did not consider the relative contributions of the individual fractions of inert compounds (e.g. soluble microbial products, colloidal components) to the biomass viability. The parameters of the model cannot be linked directly to the physical sub-model of the membrane fouling. Although the amount of SMP is negligible compared to the total suspended solids in the mixed liquor, research findings suggest that the SMP attached to the suspended solids has a great influence on

specific resistance (Lee et al., 2002). The colloidal content in the feed and mixed liquor plays a dominant role in controlling the membrane fouling (Gao et al., 2013).

Many researchers acknowledged the inhibitory effect of on microbial activity (Table 3.1), and the SMP has been identified as one of the major foulants responsible for irreversible membrane fouling. However, there is no unambiguous method of measurement of SMP/EPS to characterize the biomass and the available methods are too complex to be adopted in general practice. Hence, the trend in many scientific studies is to investigate DOC (dissolved organic carbon), COD or TOC (total organic carbon) in the mixed liquor supernatant to represent SMP and EPS concentrations.

Soluble COD in the supernatant ( $COD_{ss}$ ) which was likely to be SMP had significant impact on the cake formation rate, and the significant difference between  $COD_{ss}$  and  $COD_{s,eff}$  (soluble COD in the effluent) suggested that the membrane retained some of the high molecular weight SMP (Lin et al., 2010). The authors concluded that the SMP in the  $COD_{ss}$  might have served as the binding sites for sludge cake formation. Wu et al. (2012) modelled specific cake resistance as the cake consolidation process due to the entrapment of colloidal material within the cake layers which could explain the acceleration of cake resistance and the increase of transmembrane pressure (TMP) in the MBR. The cake layer was assumed to be formatted by MLSS and consolidated by the entrapment of colloidal components, resulting in a decrease in cake porosity and subsequent increase in specific cake resistance.

*Table 3.1* Studies on the effect of microbial products on microbial activity

Reference	System studied	Findings
Zhang and Yamamoto (1996)	Wastewater reuse system of an existing building was investigated using membrane separation activated sludge process.	The accumulated microbial products might be one of the limiting factors to bacterial activity and viability.
Huang et al. (2000)	A submerged membrane bioreactor treating synthetic wastewater was studied during long term operation	The accumulation of SMP in the supernatant of the bioreactor proved to be inhibitory towards the metabolic activity of the activated sludge, as well as contributing to the poor membrane permeability of the mixed liquor
Shin and Kang (2003)	Experiments were performed in lab-scale SMBR fed with synthetic wastewater.	Microbial inhibition was not observed by the accumulation of SMP during operation time
Chipasha and Medrzycka (2004)	A modified UCT process was studied where activated sludge was used from a local municipal wastewater treatment plant. Variations in pH, dissolved oxygen concentration, soluble biological and chemical oxygen demands (sBOD5 and sCOD) as a measure of microbial activity in synthetic wastewater were monitored.	Biomass developed in the presence of SMP degraded the substrates irregularly, suggesting that some microbes were dependent on the metabolic products of those that could utilize the feed components.
Li et al.(2006)	Crossflow dynamic membrane bioreactor (CDMBR) kinetics was investigated by treating caprolactam wastewater over a period of 180 days.	The sludge activity was possibly inhibited by the accumulated SMP or affected by the pump shear stress
Germain et al. (2007)	Both municipal and industrial pilot and full scale SMBRs with mixed liquor suspended solids concentrations (MLSS) ranging from 7.2 to 30.2 g L <sup>-1</sup> were studied	The presence of SMP <sub>COD</sub> had a negative effect on oxygen transfer suggesting that SMP affected microbial activity.
Chipasha and Medrzycka (2008)	A modified UCT process was studied where activated sludge was used from a local municipal wastewater treatment plant	Results suggested that accumulation of SMP was one of the intrinsic regulatory mechanisms that control viability and dormancy of microbial communities in activated sludge

In the following section, the new proposed conceptual model of biomass viability presents effective correlations of SOUR with MLSS/MLVSS and total/soluble COD in the bioreactor supernatant and effluent stream. The model of biomass viability may be

subsequently linked to the predicted rejection efficiency of the membrane at the actual fouling state using the dynamic changes of parameters such as MLSS and SMP/EPS concentration in the supernatant and TMP.

### *3.2.2 New conceptual model of biomass viability*

The literature review in the previous section suggests that the production and accumulation of colloidal and soluble products from microbial metabolism may develop progressive effects on biomass viability and membrane fouling as well. As already discussed in the review, the SMPs were commonly observed to inhibit microbial growth in the activated sludge processes. The proposed model, consequently, includes SMP in the bioreactor as the main parameter affecting the biomass viability. The model also considers that the accumulation of colloidal inert compounds in the mixed liquor also affects the biomass viability. Both of these fractions of inert compounds are considered in the proposed conceptual model of biomass viability including as well the common indicators such as the ratio of MLVSS/MLSS and the SOUR. The proposed models consider soluble COD in the effluent ( $COD_{s,eff}$ ) as a reflective parameter of SMP (within the bioreactor) part of which is retained on the membrane surface contributing to the membrane fouling. The colloidal COD in the effluent ( $COD_{c,eff}$ ) is assumed to be reflective of the concentration of inert colloidal microbial products in the bioreactor which are known to be major foulants and thus may be given as a function of the rejection efficiency of the membrane at the actual fouling state (Galinha et al., 2012).

The new proposed model of biomass viability is based upon two major assumptions, a fraction of the inert COD present in the bioreactor will pass the membrane unchanged and the biodegradable COD in the effluent is negligible. Following the basic structure of the empirical model given by Hasar and Kinachi (2004), the new conceptual model of biomass viability is given in Eqs. (3.3) and (3.4).

Exponential function:

$$\text{SOUR} = a_1 + \exp[a_2 + b_1 * (\text{COD}_{s,\text{eff}} / \text{COD}_i) + b_2 * (\text{COD}_{\text{cp}} / \text{COD}_i) + b_3 * (\text{MLVSS} / \text{MLSS})] \quad \text{Eq. (3.3)}$$

(or) Power function:

$$\text{SOUR} = a_1 * (\text{COD}_{s,\text{eff}} / \text{COD}_i)^{b_1} * (\text{COD}_{c,\text{eff}} / \text{COD}_i)^{b_2} * (\text{MLVSS} / \text{MLSS})^{b_3} \quad \text{Eq. (3.4)}$$

where  $\text{COD}_i$  is the total inert COD in the influent,  $a_1$ ,  $b_1$ ,  $b_2$  and  $b_3$  are constants to be estimated during model calibration.

In summary, the model considers that the following criteria are influential for the biomass viability:

- i) The progressive change in the concentration of SMP is assumed to cause changes in the composition of the microbial communities (Chipasa and Medrzycka, 2008) which mainly inhibit the microorganism activity for other nutrient treatment/removal. The SMP/EPS concentration in the supernatant of the bioreactor is the controlling parameter for biomass viability and also for the progressive built-up of irreversible membrane fouling. The progressive development of SMP/EPS within the bioreactor can be traced by monitoring the soluble COD concentration in the effluent which is accounted for in the model by a parameter of its ratio to the total inert COD of the influent.
- ii) The effects of colloidal microbial products is perhaps more significant for membrane fouling. The fraction of this can also be traced in the effluent and for this reason, it is introduced into the model of biomass viability by the ratio of the colloidal COD of effluent to the total inert COD of the influent.

### 3.2.3 Conceptual mathematical model of membrane fouling

Membrane fouling is not a discrete physical phenomenon, rather it is highly linked with the dynamic biochemical process occurring within the bioreactor. The recent trend of mathematical modelling of MBR is therefore to develop a bioprocess model of MBR integrated with the model of membrane fouling. Hence, a conceptual model of the membrane fouling is given in this section which incorporates some of the bioprocess parameters of the model of biomass viability proposed in the previous section. The proposed model of the membrane resistance at the actual fouling state is based upon the basic resistance-in-series model. The total membrane resistance given in Eq. (3.5) is made up of the membrane's intrinsic resistance, pore fouling resistance and cake layer resistance.

$$R_t = R_m + R_c + R_p \quad \text{Eq. (3.5)}$$

The resistance  $R_m$  is the non-varying intrinsic resistance of the membrane typically determined by Darcy's law.  $R_p$  is the pore fouling resistance caused by solute deposition inside the membrane pores, and this can be calculated by the Eq. (3.6) as followed by Li and Wang (2006).  $R_c$  (cake layer resistance) can be calculated according to the Eq. (3.7) proposed by Lee et al. (2002) where  $r_c$  implies for specific cake resistance,  $V_p$  is the total volume filtered,  $A_m$  is the membrane filtration area,  $X_{TSS}$  is the total suspended solids, and  $k$  is a coefficient value of which ranges from 0 to 1. Cho et al. (2005) found a relationship among specific cake resistance, MLSS, bound EPS (bEPS), TMP, and expressed the specific cake resistance ( $r_c$ ) as a function of bEPS, TMP (or  $\Delta p$ ), MLVSS and viscosity ( $\mu$ ) (Eq. 3.8).

$$R_p = r_p V_p \quad \text{Eq. (3.6)}$$

$$R_p = \frac{r_c k V_p X_{TSS}}{A_m} \quad \text{Eq. (3.7)}$$

$$r_c = \frac{\Delta p}{\mu^2} \left( 9.3 * 10^{12} + 1.803 * 10^4 (1 - \exp(-115.2 (\frac{EPS}{MLVSS})))^{36.66} \right) \quad \text{Eq. (3.8)}$$

Later, Zarragoitia-Gonzalez et al. (2008) modified the equation assuming bound EPS (bEPS) is associated with SMP and expressed the bEPS in terms of SMP concentration in the bioreactor ( $S_{SMP}$ ) with an appropriate coefficient, and the modified equation is as follows:

$$r_c = \frac{TMP}{\mu^2} \left( a + b(1 - \exp(-c(\frac{S_{SMP}}{0.8X_{TSS}})))^d \right) \quad \text{Eq. (3.9)}$$

To maintain the link with the proposed mathematical model of biomass viability through the parameters of  $COD_{s,eff}$  and  $COD_{c,eff}$ , the proposed equation for the specific cake resistance is given as follows:

$$r_c = \frac{TMP}{\mu^2} \left( a + b(1 - \exp(-c(\frac{COD_{s,eff} + COD_{c,eff}}{d MLVSS})))^e \right) \quad \text{Eq. (3.10)}$$

where, TMP is the transmembrane pressure, and  $a$ ,  $b$ ,  $c$ ,  $d$  and  $e$  are the constants.

According to the proposed modification of the equation for specific cake resistance (Eq. 3.9), it is implied that the accumulation of both the soluble and colloidal inert compounds within the bioreactor contributes to the cake resistance. By the integration of these easily measurable parameters in the equation, the researchers could find a way to measure membrane resistance avoiding the measurement of controversial SMP parameters. Finally, all the parameters could be integrated in the basic Darcy's law for estimating the rejection efficiency of the membrane at the actual fouling state, and the equation is as follows:

$$TMP = \mu * j_t * R_t, \text{ where } j_t \text{ is the total flux} \quad \text{Eq. (3.11)}$$



### 3.3 Conclusion

The proposed new conceptual models presented in this chapter consist of parameters to represent the soluble and colloidal (inert) microbial products of wastewater that potentially affect the biomass viability and membrane resistance of an MBR as well. The measurements of soluble and colloidal COD of the effluent can be effectively used to establish empirical correlations to estimate the SMP/EPS concentration of the supernatant. Easily measurable parameters of the proposed model may also serve to estimate SMP/EPS concentration in the bioreactor of the MBR avoiding their tedious and expensive measurement. Because of the common parameters in both the models, the proposed model of biomass viability can be effectively linked to the model of membrane fouling. The development of such empirical correlation would help integrate models of biomass viability and membrane fouling for better operational control of MBR systems.

The models, however, do not incorporate the dynamic membrane rejection efficiency of soluble/colloidal particles and rate of transport of fractions of these particles back into the bioreactor. Depending on the pore size of the membrane, traces of colloidal COD may not be found at the effluent and hence, the effects of colloidal (inert) microbial products on biomass viability will be excluded from the model. These limitations of the models warrants further improved models of biomass viability and fouling based on the true contributions of inhibitory products such as SMPs/bEPSs on the biomass viability and fouling of the MBR systems.



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## **CHAPTER 4**

# **Experimental Investigations**

## 4.1 Introduction

The experimental program of the research was designed to investigate the biomass viability and membrane fouling in several lab-scale aerobic submerged MBR (SMBR) systems. With only slight changes of bioreactor dimensions, the experimental set-up of all the MBR systems was necessarily the same. A novel sponge submerged MBR (SSMBR) system was also operated under the experimental conditions where sponges were additionally introduced in the bioreactor to facilitate for the attached growth of biomass.

During the MBR systems' operation, periodic measurements of several biological and (operational) indicator parameters were taken and later analysed to investigate for the biomass viability and fouling in the MBR systems. This chapter presents detailed descriptions of the experimental set-up of the SSMBR system, materials and methods used for the measurements of biological and operational parameters along with the detailed description of materials and analytical methods used in the study.

## 4.2 Materials and methods

### 4.2.1 *Experimental set-up*

One of the experiments was performed on a continuously aerated lab-scale SSMBR system. The membrane module was submerged within the bioreactor. Specifications of the membrane module, operating conditions of the experiment and the system performance are mentioned in Table 4.1, and the diagram of the experimental set-up is shown in Figure 4.1. Both the influent and effluent flow rates were controlled by a two channel pump, and a separate pump was used for backwashing. The SSMBR was filled with the sludge collected from a local wastewater treatment plant and acclimatized with the synthetic wastewater to be treated. A pressure gauge was used to measure the trans-

membrane pressure (TMP). A soaker hose air diffuser was used to provide air flow and an airflow meter was used to maintain a constant air flow rate of 7L/min. The experiment was conducted at room temperature which was about 25<sup>0</sup>C.

*Table 4.1* Design parameters, operating conditions and system performance of the SSMBR

<u>Membrane details:</u>	
Membrane material	Polyethylene with hydrophilic coating (Figure 4.3)
Manufacturer	Mitsubishi-Rayon, Tokyo, Japan
Pore size	0.1 $\mu$ m
Outer diameter, $m_{d,o}$	0.41 mm
Inner diameter, $m_{d,i}$	0.27 mm
Effective thickness, $h_m = m_{d,o} - m_{d,i}$	0.14 mm
Surface area	0.195 m <sup>2</sup>
<u>Sponge details:</u>	
Name	S28-30/90R; Joyce Foam Products, Australia
Density	28-30 kg/m <sup>3</sup> with 90 cells per 25 mm
Size	1 cm $\times$ 1 cm $\times$ 1 cm
Volume fraction of bioreactor	10%
<u>Operating conditions:</u>	
Flux, $J$ (L/m <sup>2</sup> .h)	12
Reactor volume (L)	10
MLSS (g/L)	5-18
Temperature ( <sup>0</sup> C)	21-24.5
Aeration rate (L/m <sup>2</sup> .h)	2.2
HRT (h)	4.3
DO (mg/L)	7.5-8.5
Operation period (d)	50
Physical cleaning frequency	1 min after every 1 hour of filtration
<u>Influent characteristics:</u>	
COD (mg/L)	350-380
PO <sub>4</sub> -P (mg/L)	3.1-4.0
NH <sub>4</sub> -N (mg/L)	9-15
<u>Removal efficiency (%):</u>	
COD	95-98
PO <sub>4</sub> -P	85-100
NH <sub>4</sub> -N	70-90

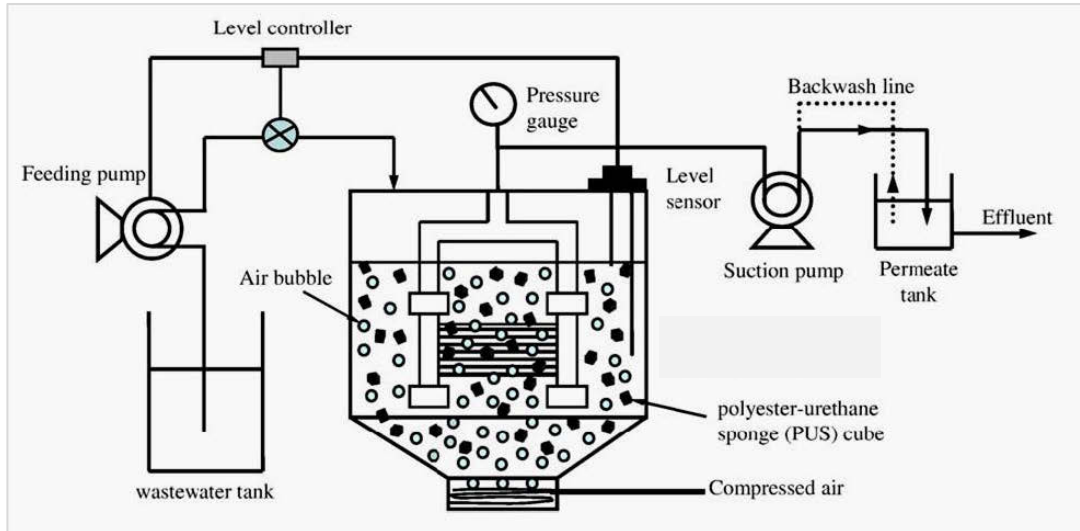
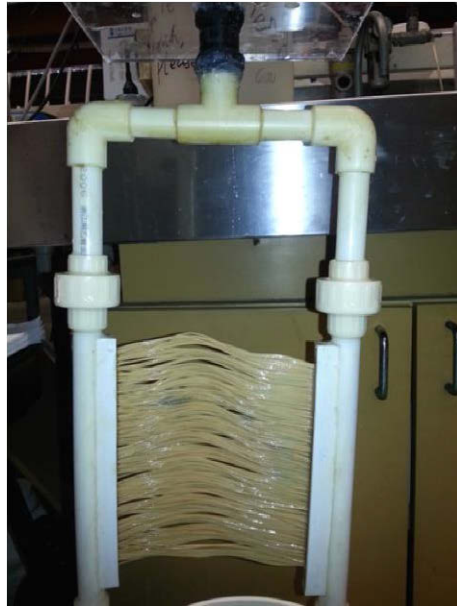


Figure 4.1 Schematic diagram of the SSMBR experimental system



Figure 4.2 The SSMBR experimental system



*Figure 4.3* The membrane module used for the SSMBR

Germain et al. (2007) conducted a study on the oxygen transfer efficiency for different MLSS concentrations with varying volumetric airflow rates. While an increase in volumetric airflow rate naturally led to an increase in oxygen transfer parameter ( $k_{L}a_{20}$ ), an increase in MLSS resulted in an exponential decrease in  $k_{L}a_{20}$ . The maximum value of  $k_{L}a_{20}$  ( $\approx 28h^{-1}$ ) was found at an MLSS concentration of 9.3 g/L but with high volumetric air flow rate ( $6 \text{ m}^3/\text{m}^3 \cdot \text{h}$ ). However, they observed adequate oxygen transfer efficiency ( $k_{L}a_{20} \approx 20h^{-1}$ ) at a lower MLSS concentration of 7.2 g/L when the volumetric air flow rate was maintained approximately at a rate of  $4.4 \text{ m}^3/\text{m}^3 \cdot \text{h}$ . With a reasonably high air flow rate ( $2.2 \text{ L}/\text{m}^2(\text{membrane surface})/\text{h}$ ), the initial MLSS concentration for the SSMBR was set approximately at 5g/L in this study.

#### ***4.2.2 Compositions of the substrate and sponge specifications***

Synthetic wastewater containing glucose, ammonium sulphate, potassium dihydrogen orthophosphate phosphate and trace nutrients (Table 4.2 [Lee et al., 2003]) was used as substrate in the experiment. The synthetic wastewater had COD concentration of 340-

390 mg/L, NH<sub>4</sub>-N of 15-20 mg/L and PO<sub>4</sub>-P of 3.5-4.0 mg/L. NaHCO<sub>3</sub> or H<sub>2</sub>SO<sub>4</sub> was used to adjust the pH to 7.

*Table 4.2* Compositions of the substrate used for the SSMBR

Compounds	Molecular weight (g/mol)	Concentration (mg/L)
<u>Organics and nutrients</u>		
Glucose (C <sub>6</sub> H <sub>12</sub> O <sub>6</sub> )	180.0	280
Ammonium sulphate ((NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> )	132.1	72
Potassium phosphate (KH <sub>2</sub> PO <sub>4</sub> )	136.1	13.2
<u>Trace nutrients:</u>		
Calcium chloride (CaCl <sub>2</sub> ·2H <sub>2</sub> O)	147.0	0.368
Magnesium sulphate (MgSO <sub>4</sub> ·7H <sub>2</sub> O)	246.5	5.07
Magnesium sulphate (MgSO <sub>4</sub> ·7H <sub>2</sub> O)	197.9	0.275
Zinc sulphate (ZnSO <sub>4</sub> ·7H <sub>2</sub> O)	287.5	0.44
Ferric chloride anhydrous (FeCl <sub>3</sub> )	162.2	1.45
Cupric sulphate (CuSO <sub>4</sub> ·5H <sub>2</sub> O)	249.7	0.391
Cobalt chloride (CoCl <sub>2</sub> ·6H <sub>2</sub> O)	237.9	0.42
Sodium molybdate dihydrate (Na <sub>2</sub> MoO <sub>4</sub> ·2H <sub>2</sub> O)	242.0	1.26
Yeast extract		30

The sponges used in the SSMBR were acclimatized with the synthetic wastewater to be treated for at least 25 days before commencing the experiment. The sponge was poured within the bioreactor when the sludge MLSS was about 5g/L. The sponge specification was reticulated porous polyester-urethane sponge (PUS) named S28-30/90R (density of 28-30 kg/m<sup>3</sup> with 90 cells per 25 mm; Joyce Foam Products, Australia). Sponge volume fraction of 10% (of bioreactor volume) with size of 1cm×1cm×1cm was used in the study, which was determined according to previous critical flux experiments (Guo et al., 2008).

### 4.2.3 Analysis

#### *A. Mixed liquor suspended solids (MLSS) and Mixed liquor volatile suspended solids (MLVSS)*

The MLSS and MLVSS of the sludge samples were analysed daily according to standard methods (APHA, 1998). About 2-5 ml of the sample was filtered through a

Whatman filter paper of 1.2  $\mu\text{m}$  size, and then the filter paper was dried for at least 2 hour at  $105^{\circ}\text{C}$  for MLSS analysis. The filter paper was then incinerated for 20 mins at  $550^{\circ}\text{C}$  in a furnace for the analysis of the MLVSS of the sample.

#### *B. Specific oxygen uptake rate (SOUR)*

YSI 5300 biological oxygen monitor was used to measure the oxygen consumption through the use of oxygen electrode with oxygen permeable Teflon membrane. The oxygen uptake rate (OUR) was calculated by a linear regression of the dissolved oxygen versus time plot. This quick test has many advantages such as rapid measures of influent organic load and biodegradability, indication of the presence of toxic or inhibitory wastes, degree of stability and condition of a sample (Nguyen et al., 2012). The specific oxygen uptake rate (SOUR) was then calculated in  $\text{mgO}_2/\text{gVSS}/\text{h}$  as follows;

$$\text{SOUR} = \text{OUR}/\text{MLVSS} \quad \text{Eq. (4.1)}$$



*Figure 4.4* YSI 5300 biological oxygen monitor

#### *C. Analysis of extra cellular polymeric substances*

Both the SMP and bEPS within the bioreactor were analysed according to modified method of Le-clech et al. (2006) and Menniti and Morgenroth (2010). The fresh 50 mL of mixed liquor sample was centrifuged at the rate of 3500 rpm for 30 mins. The supernatant was then decanted carefully and filtered using glass fibre filter (Whatman



934-AH) with a nominal pore size of 1.2  $\mu\text{m}$ . The filtrate was further filtered using a 0.2 $\mu\text{m}$  cellulose acetate filter for the SMP analysis. The bEPS was extracted by filling the centrifuge tube (up to 50 mL mark) with distilled water and then kept in ultrasonic water bath for 10 mins. After heating at 80<sup>0</sup> F for 10 minutes, it was further centrifuged for 10 mins. The supernatant was then decanted and filtered through 1.2 $\mu\text{m}$  syringe filter. Bothe the bEPS and SMP were quantified as COD of the sample.



Figure 4.5 Ultrasonic water bath used for the EPS extraction

#### D. COD analysis

The COD analysis was done by COD reagent of the *Hanna Instruments* following their prescribed procedure. Two ml of sample (for low range COD, 0-150mg/L) or 0.2 ml (for high range COD, 0-15000mg/L) of sample was added into the COD reagent tube, and after mixing in the tube properly the sample was digested at 150<sup>0</sup>C for 2 hours. After digestion, the tube was kept for 20 mins in the heater (*Hanna reactor*) to cool to about 120<sup>0</sup>C, and then was kept in the rack to cool to room temperature after inverting the tube several times while it was still warm. The COD of the sample was then quantified in the photometer using the specific wavelength (*Hanna spectrophotometer*).

### *E. Dissolved organic carbon (DOC) analysis*

The DOC was measured by using Multi N/C 3100 DOC analyser in this study. The sample was filtered through 0.45 $\mu$ m syringe filter prior to the analysis. The instrument measures the value of total carbon (TC) and inorganic carbon (IC). DOC was then calculated by subtracting IC from TC.

### *F. Nutrient analysis*

The nutrients were measured by photometric method using Spectroquant<sup>®</sup> Cell test (Figure 4.6, NOVA 60- Merck). Sample cell test was used to analyse ammonium nitrogen (NH<sub>4</sub>-N), nitrite-nitrogen (NO<sub>2</sub>-N), nitrate-nitrogen (NO<sub>3</sub>-N) and phosphate phosphorus (PO<sub>4</sub>-P).



Figure 4.6 Spectroquant<sup>®</sup> Cell photometer (NOVA 60- Merck)

### *G. Measurements of resistances*

Before commencing the experiment, the clean membrane resistance was measured in distilled water at various fluxes (5 L/m<sup>2</sup>.h, 10 L/m<sup>2</sup>.h, 15 L/m<sup>2</sup>.h, 20 L/m<sup>2</sup>.h, 25 L/m<sup>2</sup>.h, 35 L/m<sup>2</sup>.h). After every 1 hour (h) flux step, 1-min backwash was provided at a backwash rate of 30 L/m<sup>2</sup>.h. The clean membrane resistance was then calculated

according to Darcy's law at the operational flux through linear regression from the plot of TMP versus flux (Figure 4.7).

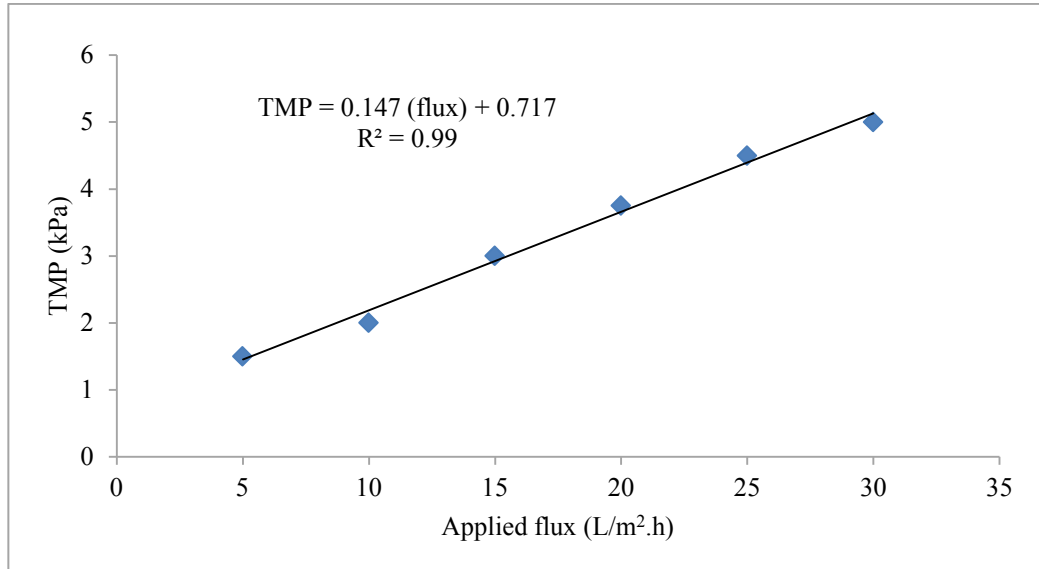


Figure 4.7 TMP versus flux plot

At the end of experiment, the membrane module was taken out of the bioreactor and submerged in distilled water to measure for the total resistance ( $R_T = R_m + R_p + R_c$ ) of the fouled membrane. Darcy's law was applied to calculate total membrane resistances ( $R_T$ ) according to in Eq. (4.2):

$$J = \frac{\text{TMP}}{\mu R_T} \quad \text{Eq. (4.2)}$$

where:  $J$  is the permeate flux; TMP is the transmembrane pressure,  $\mu$  is the viscosity of the permeate at 20°C;  $R_T$  is the total resistance which is the combination of the membrane's intrinsic resistance ( $R_m$ ), pore fouling resistance ( $R_p$ ) and cake (sludge) layer resistance ( $R_c$ ).

The membrane was cleaned by gently shaking with distilled water to remove the deposited sludge cake layer from the membrane surface, and then submerged again in the distilled water for the measurement of  $R_p$  and  $R_m$ . Finally, the membrane was chemically cleaned to measure the intrinsic membrane resistance ( $R_m$ ). For the

calculation of daily total resistances from the measurements of TMP, the value of  $\mu$  was corrected for temperature as follows (Delrue et al., 2011);

$$\mu_T = \mu_{20} e^{-0.0239(T-20)} \quad \text{Eq. (4-3)}$$

where  $T$  is the temperature of mixed liquor temperature in  $^{\circ}\text{C}$ .

#### *H. Chemical cleaning of membrane*

The membrane was cleaned following two step cleaning procedure to remove its internal pore foulants. To remove the organic foulants, the membrane was soaked in 2% citric acid [ $\text{C}(\text{OH})(\text{COOH})(\text{CH}_2.\text{COOH})_2 \text{H}_2\text{O}$  210.14] for 6 hours, and then the module was soaked next day in 0.4% NaOCL and 4% NaOH for 6 hrs to remove organic foulants from the pore of the module. During the soaking period, the module was stirred very carefully every 1-2 hr. After the two-step cleaning, the module was kept in distilled water for 1 day before measuring for the  $R_m$  again on the following day.

#### *I. Statistical analysis*

Statistical analysis was performed to identify the potential biomass parameters affecting the SOUR by bivariate correlation analysis. The relationships between the biomass characteristics and SOUR were examined by computing a Pearson product-moment correlation coefficient ( $r_p$ ) matrix between each pair of parameters. The Pearson correlation coefficient,  $r_p$ , ranges from -1 to + 1 where  $r_p = -1$  or +1 indicates a perfect negative or positive correlation respectively, and  $r_p = 0$  represents a lack of correlation. The analysis was carried out using the statistical package for the social science, SPSS V21.0 produced by IBM, USA.

#### *J. Mathematical model simulations and parameters estimation*

The mathematical model equations were solved in Matlab 2014a based on the measured data of SOUR, MLSS, MLVSS, SMP and bEPS concentrations of the SMBR systems.

The algorithm used in the solution by *fitnlm* function of Matlab 2014a was of a nonlinear regression analysis by the iterative reweighted least squares method. The process was run with multiple initial values of parameters to ensure a maximum and acceptable value of  $R^2$  (squared value of the correlation coefficient,  $R$ ).



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## **CHAPTER 5**

# **Performance Evaluation of the Submerged Membrane Bioreactors for Wastewater Treatment**

## 5.1 Introduction

The experiments were done on two lab-scale aerobic submerged MBR (SMBR) systems to evaluate their performance for treatability and fouling behaviour. An enhanced system of SMBR was investigated under the experimental plan where sponge was additionally introduced in the bioreactor to facilitate the attached growth of biomass. The improved system of the aerobic SMBR was named as sponge submerged MBR (SSMBR). In the SSMBR system, cube sized sponges were introduced as an ideal attached growth medium. Several biological and physical parameters were analyzed during the course of operation of the MBR systems. The novel SSMBR system was previously evaluated by a series of studies (Guo et al., 2009, 2008; Ngo et al., 2008; Nguyen et al., 2012 ) at the UTS to assess conditions (design or operational requirements) for effective and stable performance. The performance of the SSMBR system was assessed for its operation with new and acclimatized sponges, and the effects of different biomass parameters were investigated for the removal of organics and nutrients from wastewater at different suspended solid ( $MLSS_{sludge}$ ) concentrations of the sludge.

This chapter discussed the experimental results of the lab-scale SMBR systems by analysing the results for treatment efficiency with particular reference to the indicator parameters of biomass viability. The discussions in this chapter are done in two main sections. Firstly, the performance of the SSMBR system was evaluated for the effects of different gross biomass parameters on the efficiency of organics removal. The measured results of the nutrient removal efficiencies were also discussed in the first section. The discussions in the second part were on the assessment of the effects of potential biomass parameters on the microbial viability in a conventional aerobic SMBR system (without

sponges), and the analyses of the test data were done especially to investigate the correlations between the potential parameters of biomass viability as are indicated in the conceptual mathematical models (Chapter 3). Table 5.1 shows the system description and the operating conditions of the SMBR systems. The specifications of the sponges used in the bioreactor of the SSMBR are already mentioned in the detail description of the SSMBR system in the previous chapter.

*Table 5.1* System descriptions and operating conditions of the SMBR systems

MBR systems		
	SSMBR	CMBR
<u>Membrane details:</u>		
Membrane material	Polyethylene with hydrophilic coating	
Manufacturer	Mitsubishi-Rayon, Tokyo, Japan	
Pore size	0.1 $\mu\text{m}$	0.2 $\mu\text{m}$
Surface area	0.05 $\text{m}^2$	0.1 $\text{m}^2$
<u>Operating conditions:</u>		
Flux, J ( $\text{L}/\text{m}^2/\text{hr}$ )	10	10
Reactor volume (L)	06	08
Initial MLSS (g/L)	10, 15	10
Aeration rate ( $\text{L}/\text{min}$ )	9.0	9.0
Physical cleaning mode and frequency	Backwash twice a day for 01 min	Relaxation for 01 min in after every cycle (01 h) of operation

## 5.2 Evaluation of the performance of the SSMBR

There have been a lot of studies on the operational factors such as the concentrations of MLSS (Ferreira et al., 2010), food to microorganism (F/M) ratio (Wu et al., 2012), sludge retention time or SRT (Villain and Marrot, 2013), hydraulic retention time or HRT (Hong et al., 2012) and other factors which may affect the optimum performance of an MBR. A



few researchers (inter alia Delrue et al., 2011; Ren et al., 2005) also investigated the mathematical relationships among the operational conditions and efficiency of the MBR. Among other factors, an effective support medium can greatly accelerate the attached growth process which eventually results in efficient oxygen transfer, and higher removal efficiency of the organic pollutants and nutrients (Nguyen et al., 2010; Odegaard, 2000; Tavares et al., 1994). In the SSMBR system, cube sized sponges were introduced as an ideal attached growth medium which could act as a mobile carrier for active biomass (Ngo et al., 2008) in the continuously aerated system. The addition of sponges to the SSMBR could not only remove over 96% of the DOC and nutrients but could also significantly reduce membrane fouling, and consequently enhanced sustainable flux (Ngo et al., 2008). However, the operational conditions may greatly affect the performance of any MBR.

In this context, this section specifically aimed at discussing the effects of sponges on the performance of an SSMBR for the removal of DOC and nutrients. The sponge specification was reticulated porous polyester-urethane sponge (PUS) named S28-30/90R (density of 28-30 kg/m<sup>3</sup> with 90 cells per 25 mm; Joyce Foam Products, Australia). Sponge volume fraction of 10% (of bioreactor volume) with size of 1cm×1cm×1cm was used in the study, which was determined according to critical flux experiments (Guo et al., 2008). The sludge used in the study was taken from a local wastewater treatment plant and was acclimatized with synthetic wastewater to be treated.

Depending on the subjective judgment on the bioreactor volume, aeration rate, backwashing frequency and wastewater constituents, two different (initial) MLSS concentrations of the sludge were employed in the bioreactors of the SSMBR. The performance of the SSMBR was evaluated for the treatment efficiency by measuring the concentrations of dissolved organic carbon (DOC), NH<sub>4</sub>-N and NO<sub>3</sub>-N in the influent and

effluent stream. The analytical methods of measurements of the biomass parameters, organics and the nutrients are explained in Chapter 4. The experiments were conducted with two different conditions of sponges, new sponge and acclimatized sponge (acclimatized with activated sludge in the laboratory for at least 25 days before commencing the experiments).

### 5.2.1 DOC removal efficiency of the SSMBR system

As the new sponges would eventually take time to be acclimatized with the mixed liquor ( $MLSS_{sludge}$ ) of the bioreactor, it was expected that the acclimatized sponges would perform better for the organics removal at least during the first few days of operation of the SSMBR. Table 5.2 shows the measured DOC concentrations in the influent and effluent for different MLSS concentrations of the sludge used in the SSMBR system. The DOC removal efficiencies of the SSMBR system was consistently over 96% regardless of the variations of sponge conditions (new or acclimatized) used in the SSMBR system. Figures 5.1 and 5.2 respectively compare the DOC removal efficiencies of the SSMBR at different initial  $MLSS_{sludge}$  concentrations of approximately 10 and 15 g/L, both with the new and acclimatized sponges.

*Table 5.2* Dissolved Organic Carbon (DOC) concentrations in the influent and effluent at different MLSS concentrations in the SSMBR system

Sponge Condition	MLSS concentration (mg/L)	DOC concentration (mg/L)		DOC removal efficiency (%)
		Influent	Effluent	
New Sponge	5	105.8 ± 3.5	3.3 ± 1.0	96.9
	10	102.1 ± 3.9	1.90 ± 1.2	98.1
	15	92.2 ± 11.1	1.12 ± 0.5	98.8
Acclimatized Sponge	5	126 ± 18.0	2.29 ± 2.4	98.2
	10	113.7 ± 11.7	2.0 ± 1.1	98.2
	15	119.1 ± 8.7	3.0 ± 2.1	97.5

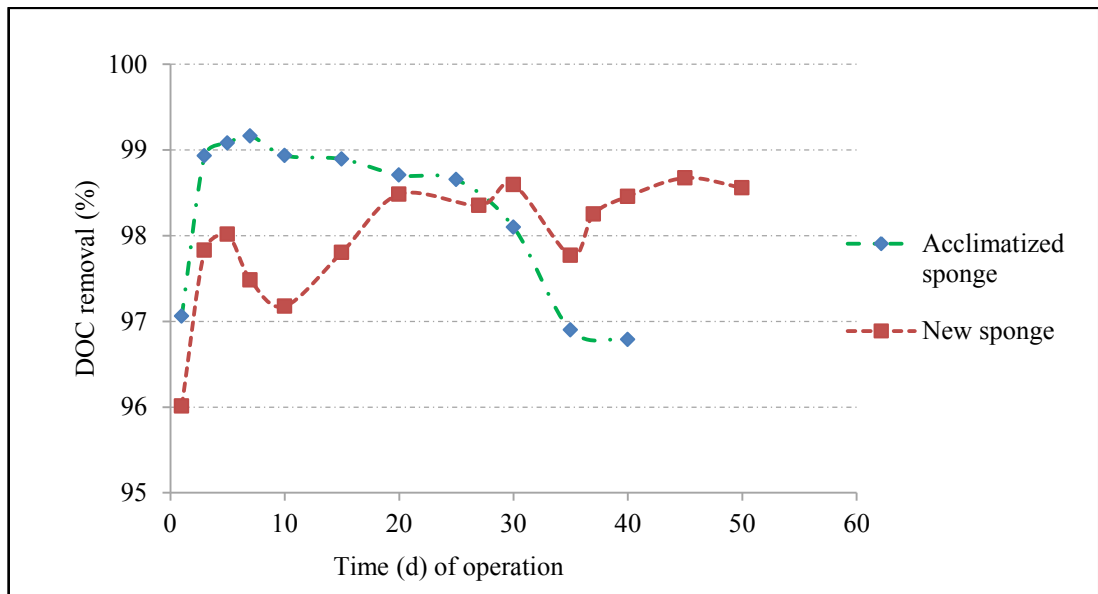


Figure 5.1 DOC removal efficiency (%) of SSMBR @ initial  $MLSS_{sludge} \approx 10$  g/L

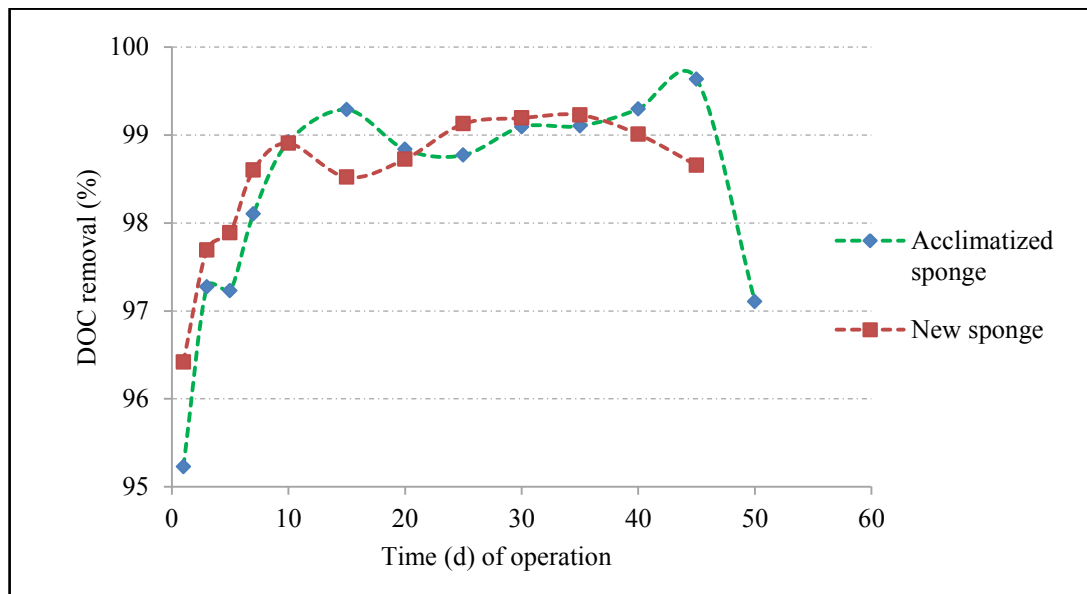


Figure 5.2 DOC removal efficiency (%) of SSMBR @ initial  $MLSS_{sludge} \approx 15$  g/L

It is evident from the comparisons shown in Figures 5.1 and 5.2 that the performance of the acclimatized sponges for the removal of DOC was generally better than the new sponges in the bioreactor. Although the DOC removal efficiency was generally good at initial  $MLSS_{sludge}$  concentration of approximately 15 g/L with acclimatized sponges, the

performance of the SSMBR system seemed to be unstable at different instances during the operation time. At the initial  $MLSS_{sludge}$  concentration of 15 g/L, the DOC removal efficiencies peaked twice but with drops in between the peaks (Figure 5.2) which was indicative of the instability in the system's operation. This may be due to the problems of acclimatization of the sponges in the bioreactor. On the other hand, the SSMBR system with acclimatized sponges quickly achieved the maximum DOC removal efficiency (>99%) followed by a slow and steady drop in the removal efficiency when the initial  $MLSS_{sludge}$  concentration was approximately 10 g/L. The desired DOC removal efficiency, therefore, could be sustained for longer time.

With the acclimatized sponges and with initial  $MLSS_{sludge} \approx 10$  g/L in the SSMBR, the efficiency of the SSMBR for the removal of  $PO_4\text{-P}$  was in the range of 97.1 to 99.5% while its average efficiency for the removal of  $NH_4\text{-N}$  was also relatively high (85.6 to 87.9%) during the first 25 days of operation of the system. For the operation of the SSMBR with the acclimatized sponges and with initial  $MLSS_{sludge} \approx 15$  g/L, the efficiency of the SSMBR for the removal of  $PO_4\text{-P}$  varied between 98.6 and 99.7% while its efficiency for the removal of  $NH_4\text{-N}$  was in the range between 79.2 and 90.1% during the first 25 days of the operation of the SSMBR. Moreover, the membrane required more frequent cleaning to avoid the rise of TMP when the SSMBR operated with initial  $MLSS_{sludge}$  15 g/L.

It appears that the better performance of the SSMBR with the acclimatized sponges is not only due to the optimum initial  $MLSS_{sludge}$  (of approximately 10 g/L) concentration in the bioreactor but also due to the better acclimatization of the sponges with biomass in the bioreactor at initial  $MLSS_{sludge}$  concentrations of 10 g/L. As a consequence, the following section presents further analysis to identify the effects of biomass for the removal of organics from the wastewater.

### 5.2.2 Effects of biomass on the DOC removal

The better efficiencies of the SSMBR with the acclimatized sponges may be attributed to the positive effects of acclimatized sponges for the organics and nutrients removal from wastewater. The effects of sponges on the DOC removal may be compared among the different experiments by comparing the DOC removal efficiency (%) with the ratio of the  $MLSS_{\text{sponge}}$  or  $MLVSS_{\text{sponge}}$  to the corresponding  $MLSS_{\text{sludge}}$  concentration. Figures 5.3 and 5.4 compare the DOC removal efficiencies for different normalized biomass parameters of acclimatized sponges in the  $MLSS_{\text{sludge}}$  concentrations of 10 g/L and 15 g/L respectively.

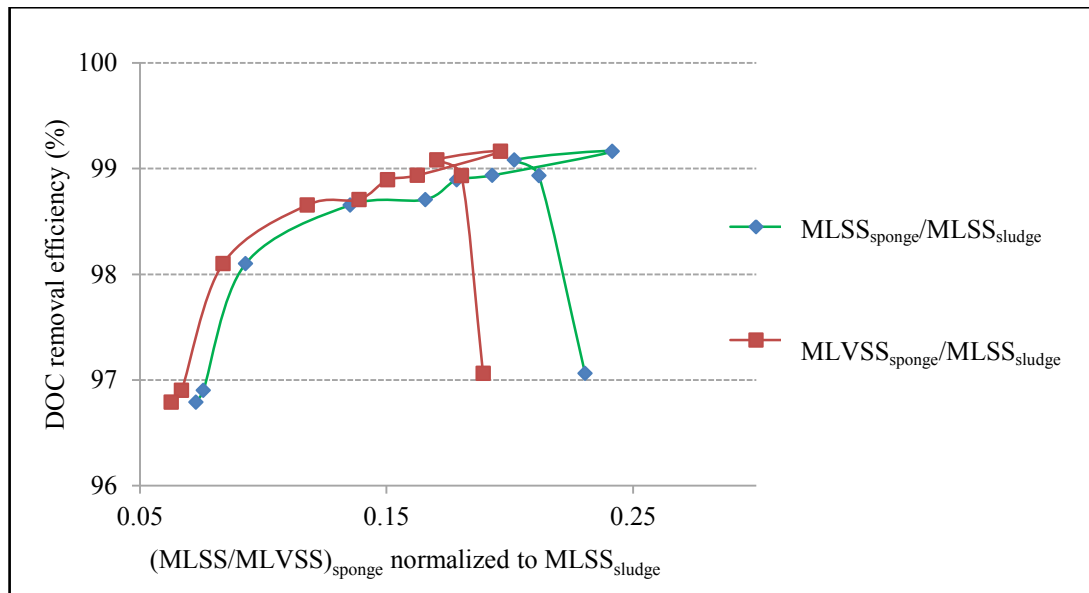


Figure 5.3 DOC removal at various  $(MLSS/MLVSS)_{\text{sponge}}/MLSS_{\text{sludge}}$  (for the acclimatized sponge and initial  $MLSS_{\text{sludge}} \approx 10$  g/L)

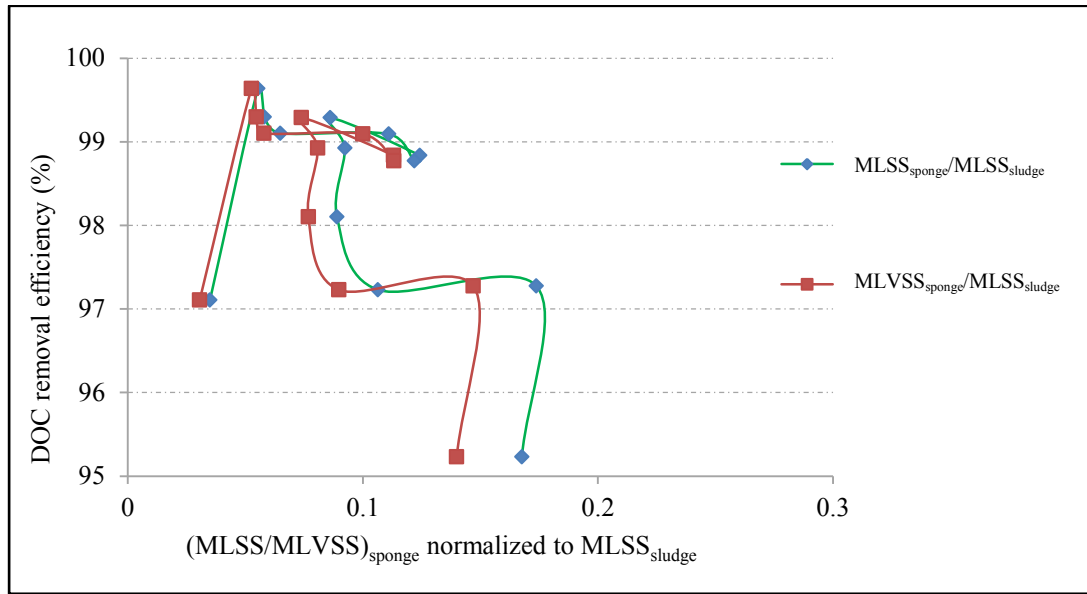


Figure 5.4 DOC removal vs.  $(\text{MLSS}/\text{MLVSS})_{\text{sponge}}/\text{MLSS}_{\text{sludge}}$   
(for the acclimatized sponge and initial  $\text{MLSS}_{\text{sludge}}$  of 15 g/L)

From the comparison shown in Figures 5.3 and 5.4, it appears that the effect of acclimatized sponges on the removal of DOC was more stable when the initial  $\text{MLSS}_{\text{sludge}}$  was approximately 10 g/L. The SSMBR (with initial  $\text{MLSS}_{\text{sludge}} \approx 10$  g/L) had the maximum DOC removal efficiency achieved at or near the point where the normalized  $\text{MLSS}_{\text{sponge}}/\text{MLSS}_{\text{sludge}}$  was approximately 0.24. The  $\text{MLSS}_{\text{sponge}}/\text{MLSS}_{\text{sludge}}$  at the first day of the operation of the SSMBR was about 0.23 which was close to 0.24. As a result, the effects of sponges might have been quickly stabilized to the system's operation and a steady operational efficiency was maintained by the system for longer time. The similar trend line of  $\text{MLVSS}_{\text{sponge}}/\text{MLSS}_{\text{sludge}}$  similar to that of  $\text{MLSS}_{\text{sponge}}/\text{MLSS}_{\text{sludge}}$  (Figure 5.3) indicates that the sponges might also have a positive influence on the biomass viability by accumulating volatile suspended solids within their pores or on their surface. On the other hand, the system behaved in an unstable manner when the SSMBR was operated with initial  $\text{MLSS}_{\text{sludge}}$  of 15 g/L (Figure 5.4). The maximum removal of DOC occurred at

$MLSS_{\text{sponge}}/MLSS_{\text{sludge}} \approx 0.05$  whereas the system started operating on the first day with  $MLSS_{\text{sponge}}/MLSS_{\text{sludge}} \approx 0.15$ .

The above analyses of test results suggest that the instability in the SSMBR operation might be strongly influenced by the gross biomass parameters of sludge and that of acclimatized sponges as well. The operation of SSMBR systems, from these results, should be maintained with the optimum concentrations of these biomass parameters to achieve the optimum treatment efficiency. For example, the operation of the lab-scale SSMBR system is suggested to be maintained with initial  $MLSS_{\text{sludge}} \approx 10$  g/L. When acclimatizing sponges with activated sludge, a balanced concentration of  $MLSS_{\text{sponge}}/MLSS_{\text{sludge}}$  at or around the value of 0.20 seems to be appropriate for sustaining stable operation of the lab-scale SSMBR for the removal of organics and nutrients from the wastewater.

### ***5.2.3 Mathematical functions for the effects of biomass on the DOC removal***

It was discussed in the previous section that the SSMBR performed best for the DOC removal when the SSMBR was operated at the initial  $MLSS_{\text{sludge}}$  concentration of approximately 10 g/L using acclimatized sponges as the attached growth medium. From further analyses of experimental results, a general mathematical function for the correlation of different characteristic biomass parameters with the DOC removal efficiency is developed in this section. The general mathematical expression is shown in Eq. (5.1).

$$D_i = a_i S_i^2 + b_i S_i + \Delta d_i \quad \text{Eq. (5.1)}$$

where  $D_i$  is the DOC removal (%) and  $S_i$  represents different characteristic biomass parameters of the sponge and the sludge that could be correlated well with the DOC removal efficiencies.  $a_i$  and  $b_i$  in Eq. (5.1) are the coefficients values which are shown in

Table 5.3, and in Figures 5.5 (*a, b, c, d*) for each of the representative cases of  $S_i$ .  $\Delta d_i$  is a constant which has a value even when the value of  $S_i$  is zero.

The following are the main biomass parameters ( $S_i$ ) that were found affecting significantly the DOC removal of the SSMBR:

- i)  $MLSS_{\text{sponge}}/MLSS_{\text{sludge}}$  (for the effects of sponge)
- ii)  $MLVSS_{\text{sponge}}/MLSS_{\text{sludge}}$  (for the effects of sponge)
- iii)  $Biomass_{\text{sponge}}$  (biomass on sponge, g Biomass/g sponge)
- iv)  $MLSS_{\text{sludge}}$  (MLSS concentration of the sludge)

All the analyses show a nonlinear relationship (Figures 5.5) between the DOC removal (%) and the characteristic biomass parameters ( $S_i$ ). Neglecting the initial values of the experimental results, a 2<sup>nd</sup> order polynomial in Eq. (5.1) describes reasonably well the effects of sponge on the DOC removal. In a similar study but without normalization of biomass parameters, Ren et al. (2005) found 4<sup>th</sup> order relationship between the MLSS and chemical oxygen demand (COD) removal efficiency of an SBR. This indicates that the normalization of biomass parameters may lead to the development of more simple but practical relationships among the parameters.

*Table 5.3* Mathematical functions for the effects of different biomass parameters on the DOC removal

$S_i$ (Parameters)	Mathematical equation of $D_i$ (%)	$\Delta d_i$ (%)	Coefficients		$R^2$
			$a_i$	$b_i$	
$S_1$ ( $MLSS_{\text{sponge}}/MLSS_{\text{sludge}}$ )	$D_1 = -a_1 S_1^2 + b_1 S_1 + \Delta d_1$	94.27	-108.34	45.63	0.93
$S_2$ ( $MLVSS_{\text{sponge}}/MLSS_{\text{sludge}}$ )	$D_2 = -a_2 S_2^2 + b_2 S_2 + \Delta d_2$	93.82	-174.52	60.50	0.95
$S_3$ ( $Biomass_{\text{sponge}}$ )	$D_3 = -a_3 S_3^2 + b_3 S_3 + \Delta d_3$	90.09	-10.44	19.08	0.92
$S_4$ ( $MLSS_{\text{sludge}}$ )	$D_4 = -a_4 S_4^2 + b_4 S_4 + \Delta d_4$	84.21	-0.11	2.57	0.94



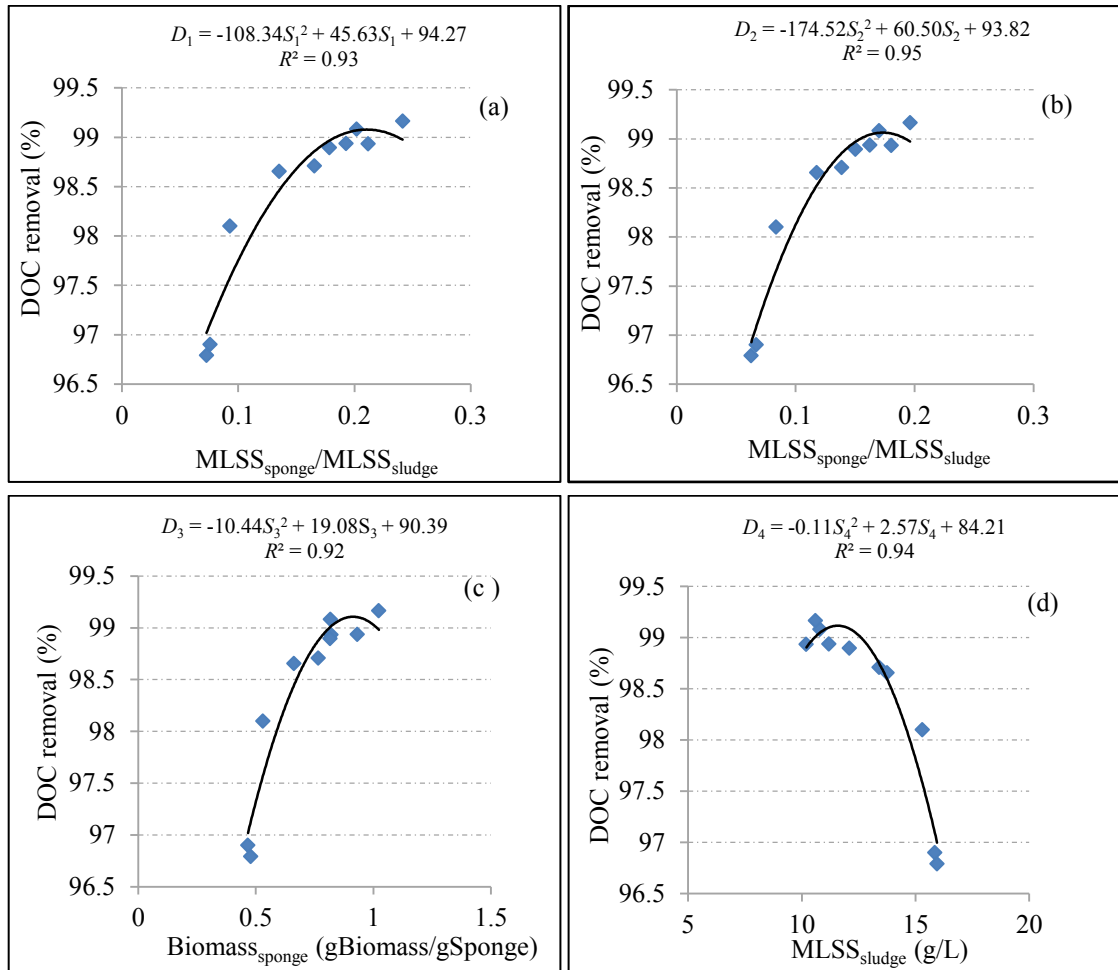


Figure 5.5 Effects of different biomass parametrs on DOC removal: (a)  $MLSS_{sponge}$  and (b)  $MLVSS_{sponge}$  (normalized to  $MLSS_{sludge} \approx 10\ g/L$ ) (c) biomass of sponge (d)  $MLSS$  concentration of the sludge

### 5.3 Assessment of biomass viability in SMBR

In the compact configuration of the MBR system, modification in biomass activity is more likely (Trapani et al., 2011). Besides mixed liquor properties, different types of organic compounds are released during microbial metabolism (SMPs or bEPS) that may pose inhibitory impacts on the biomass activity in an MBR (Hasar et al., 2002). As a consequence, the performance of the treatment system may be hampered due to factors such as the reduced rate of oxygen transfer to sustain functions of the microorganisms.

Maintaining a desired rate of oxygen transfer for microbial activity is, therefore, critically important for the optimum performance of an MBR system.

It is evident from the discussions in the previous section that the efficiency of SBR for the removal of organics and nutrients is affected by gross biomass parameters such as the concentrations of MLSS or MLVSS of the sludge and that of the sponges as well in the SSMBR system. The variable treatment efficiencies of the SSMBR were possibly due to the effects of these biomass parameters on the oxygen uptake of microorganisms. However, the correlations of these parameters to the rate of microbial activity is rather complex. Among other parameters the specific oxygen uptake (SOUR) rate is typically used as a true indicator of microbial activity (Chen et al., 2012; Clouzot et al., 2011; Hasar and Kinachi, 2004; Liang et al., 2010). In this context, the main objective of the extended analyses done in this section is to determine additionally the effects of the microbial products such as the SMPs or bEPS on the biomass viability through establishing their correlations with the SOUR. It is acknowledged in many studies that there are no unambiguous methods of determination of SMPs or bEPS. Therefore, soluble or colloidal chemical oxygen demand in the effluent stream could be measured instead as representative parameters for SMP or bEPS of the bioreactor respectively (Galinha et al., 2012).

The experimental SBR that was used for the investigation started operating with the initial MLSS concentrations of 10g/L which varied up to 11.72 g/L during the operational period of the MBR (20 days). The procedure of analyses of SOUR and the biomass parameters was discussed in Chapter 4.

### 5.3.1 Relationships between specific oxygen uptake rate and mixed liquor properties

According to the experimental results of the SBR, an exponential relation (Figure 5.9) between the MLVSS and SOUR was found with a reasonably good correlation coefficient ( $R^2= 0.66$ ). The SOUR exponentially decreased (3.5 to 0.2 mg/gVSS/hr) with the increase in MLVSS (8.9-11.2 g/L) and the relationship between these parameters can be expressed by the following equation:

$$\text{SOUR} = 13770 \exp (-0.99 \cdot \text{MLVSS}) \quad \text{Eq. (5.2)}$$

The normalized parameter MLVSS/MLSS gives indication of the relative change of volatile solids in the bioreactor. The SOUR plotted against the MLVSS/MLSS also shows a similar exponential relationship as that of SOUR vs. MLVSS (Figure 5.6).

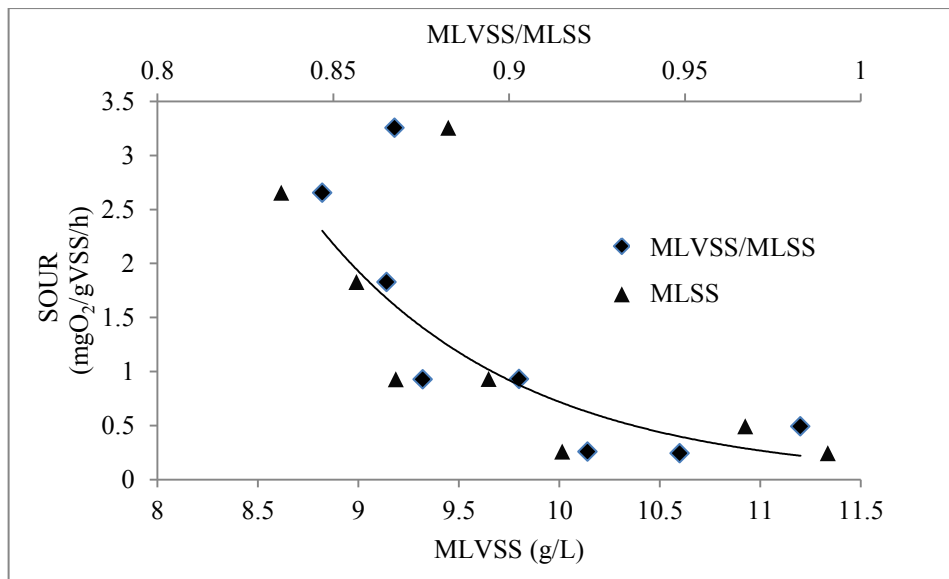


Figure 5.6 Relationships of SOUR with MLVSS and MLVSS/MLSS

### 5.3.2 Relationships between specific oxygen uptake rate and SMP indicator parameter

Soluble COD in the effluent ( $\text{COD}_{s,\text{eff}}$ ) is considered one of the important indicator parameters of the biological treatment performance. In Figure 5.7, the SOUR is plotted

against soluble COD in the effluent ( $\text{COD}_{s,\text{eff}}$ ). An exponential correlation between these two parameters was observed in the experimental study, and the correlation can be expressed by the following equation:

$$\text{SOUR} = 11.32 * \exp(-0.20 \text{COD}_{s,\text{eff}}); R^2 = 0.77 \quad \text{Eq. (5.3)}$$

As the specific oxygen uptake decreased, the soluble COD in the effluent increased exponentially.

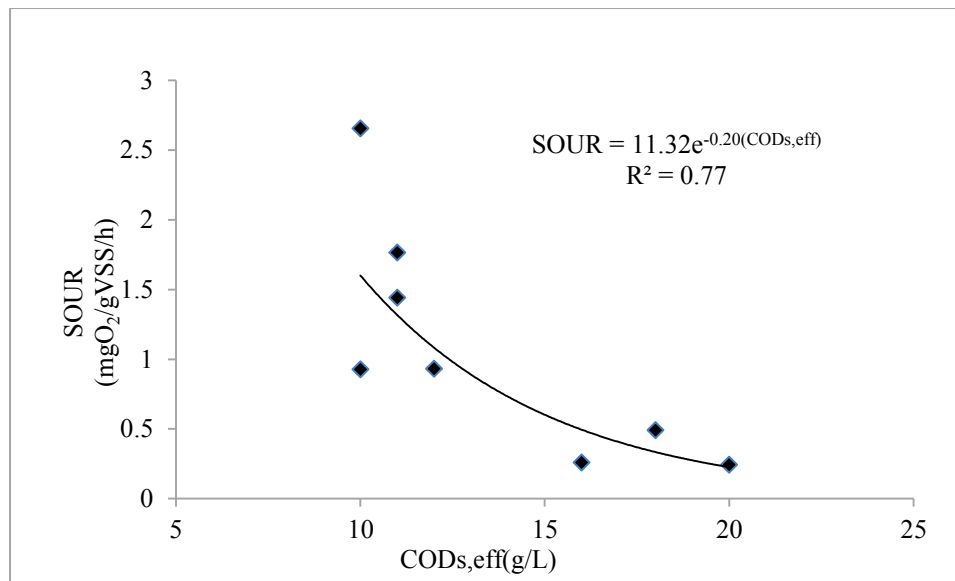


Figure 5.7 Relationship of SOUR with  $\text{COD}_{s,\text{eff}}$

It is assumed that biodegradable COD is negligible in the effluent, and almost all the COD present in the effluent is the fraction of non-biodegradable (inert) of the COD within bioreactor that can pass through the membrane. This soluble residual COD of effluent is reasonably assumed to be a fraction of the total soluble inert COD initially present in the influent and inert soluble microbial products (e.g. SMP) generated during the course of biochemical reactions (Hocaglu and Orhon 2010). The experimental results showed that the ratio of  $\text{COD}_{s,\text{eff}}$  was not increasing with time but was showing a fluctuating trend which was contrary to the relevant findings of Hasar et al. (2004). The less soluble inert COD in

the effluent as compared to that found in the earlier days may be due to the rejection of the membrane module. However, these observations need to be further investigated based on experimental results of SMBRs operated for longer periods of time.

Existence of soluble microbial products due to microbial metabolism within an MBR has already been acknowledged by many researchers, and it has been suggested to incorporate this fraction of microbial products in the case of membrane fouling and sCOD prediction studies (Fenu et al., 2010). Although very few researchers conducted studies on the effects of microbial products on the microbial viability, a negative relation was generally found between microbial activity and microbial products (SMPs/bEPS). From the analysis of test results of the SMBR, it was found that SOUR is exponentially correlated with the concentration of SMP in the bioreactor (Figure 5.8).

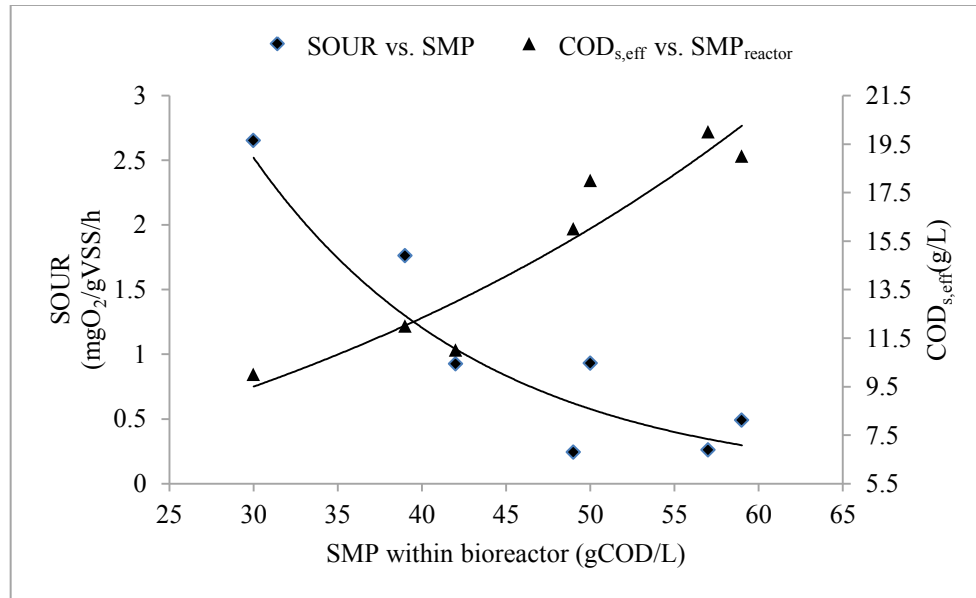


Figure 5.8 Relationships of SMP with SOUR and COD<sub>s,eff</sub>

Soluble COD in the effluent is a fraction of SMP produced within the bioreactor (Galinah et al., 2012). Based on the results of the experiment, an exponential relationship

between  $COD_{s,eff}$  and  $SMP_{reactor}$  is found with a high correlation coefficient ( $R^2= 0.90$ ). The relationship can be expressed by the following equation;

$$COD_{s,eff} = 4.34 * \exp(0.026SMP_{reactor}) \quad \text{Eq. (5.4)}$$

It was observed that there was an exponential increase of  $COD_{s,eff}$  with the increased accumulation of SMP within the bioreactor ( $SMP_{reactor}$ ). With such established correlation as expressed in Eq. (5.4),  $SMP_{reactor}$  can be known from the easily measurable  $COD_{s,eff}$  avoiding the tedious and controversial experimental measurement of SMP.

#### **5.4 Further discussions and future perspectives**

This chapter presents different experimental results to evaluate the performance of two lab-scale SMBR systems for wastewater treatment. In the first section, the analyses of the results are done to evaluate the performance of sponges for the removal of organic pollutants and nutrients in a novel SSMBR system. As compared to new sponge, the acclimatized sponge was found to perform better as an attached growth support medium and as a mobile carrier of biomass. However, the optimum DOC removal by the SSMBR system could be achieved when the MLSS concentration of the acclimatized sponge is at an optimum with respect to the MLSS concentration of the sludge.

The treatability of a conventional lab-scale SMBR system was also assessed using normalized biomass parameters that are well correlated with true measures of microbial viability in the SMBR such as the SOUR. The concentration of MLVSS and SMP in the bioreactor showed inverse exponential correlation with SOUR, and were found to have affected the microbial activity within the bioreactor. The findings of the study suggest that soluble COD measured in effluent ( $COD_{s,eff}$ ) may be used as a representative parameter of

SMP in the bioreactor, and therefore,  $COD_{s,eff}$  may be used in the mathematical models of microbial viability in the MBR avoiding tedious measurement of SMP in the bioreactor.

The discussions and correlations established in this chapter indicate the possibility that the potential parameters of biomass viability can be correlated to the parameters affecting the membrane fouling in an MBR system. More simple but practical mathematical models could be developed taking into account the combined effects of these parameters on both the biomass viability and fouling. These aspects will be explored further in the following chapters that are presented with discussions of simplified mathematical modelling of biomass viability and membrane fouling in the SMBR systems using these potential biomass parameters in the models.



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## **CHAPTER 6**

# **Biomass Viability: Identification of Influencing Factors and Mathematical Modelling for a Membrane Bioreactor**



## 6.1 Introduction

Maintaining a desired rate of oxygen transfer for microbial activity is critically important for the better management and operational control of any biological wastewater treatment system. However, modification in biomass viability is more likely (Trapani et al., 2011) in the MBR treatment systems especially because of the compact configuration with typically higher suspended solids' (MLSS or MLVSS) concentration in the bioreactor. Therefore, assessment of biomass viability has always been a concern for the operational control of MBR systems for treatability since the microbial activity or biodegradation potential of microbial culture play critical roles for the sustainable operation of MBR (Başaran et al., 2014).

It was discussed in the previous chapters that the physical indicator parameter of biomass viability (SOUR) has correlations with various biomass parameters. A few mathematical expressions have been developed based on the correlations among the potential biomass parameters as measured during the short term operation of an aerobic SSMBR (Chapter 5). The potential biomass parameters that are found to be indicators of biomass viability are MLVSS, MLSS and soluble inert COD in the effluent as indicator parameter of SMP in the bioreactor. In the conceptual models proposed in Chapter 3, the colloidal COD in the effluent is also included as an indicator parameter of the concentration of bEPS in the bioreactor since the bEPS affects the microbial activity as well. However, colloidal particles of typically larger sizes than the membrane pores may be retained within the bioreactor leaving no trace of colloidal COD in the effluent.

Consequently, improvements of the mathematical model of biomass viability are proposed in this chapter. The experimental measurements of the concentrations of SMP and bEPS along with the MLSS or MLVSS in the bioreactor of a lab-scale SSMBR are correlated with the SOUR. This chapter extends with systematic formulation and

validation of the model of biomass viability based on the experimental data of the SSMBR for its operation of about 50 days.

## **6.2 Materials and methods**

### ***6.2.1 Experimental set-up and operational parameters***

The experiment was performed on a continuously aerated lab-scale SSMBR system. The specifications of the membrane module and sponges used in the SSMBR, its operating conditions and the system performance are mentioned in Table 4.1 of Chapter 4. A schematic diagram of the experimental set-up is also shown in Chapter 4 (Figure 4.1). The experimental arrangements for controlling the influent and effluent flow rate, back-washing and aeration are the same as those used in the previously described lab-scale MBR systems. The sponges used in the SSMBR were acclimatized with the synthetic wastewater to be treated for at least 25 days before commencing the experiment. Sponge volume fraction of 10% (of bioreactor volume) with size of 1cm×1cm×1cm was used in the study, which was determined according to previous critical flux experiments (Guo et al., 2008). With a reasonably high volumetric air flow rate (7 L/min to provide 2.2 L/m<sup>2</sup> (membrane surface area)/h), the initial MLSS concentration for the SSMBR was set approximately at 5g/L in this study. The characteristics of the sludge and the synthetic wastewater are the same as were used in the previously mentioned experiments.

### ***6.2.2 Methods of analysis of biological parameters***

The MLSS and MLVSS of the sludge samples were analysed daily according to standard methods (APHA, 1998). Oxygen uptake rate (OUR) was calculated by measuring dissolved oxygen concentration using the YSI 5300 biological oxygen monitor. The SOUR was then calculated in mgO<sub>2</sub>/gVSS/h as follows;

$$\text{SOUR} = \text{OUR}/\text{MLVSS} \quad \text{Eq. (6.1)}$$

The concentration of the SMP and bEPS within the bioreactor were analysed according to the methods of Le-clech et al. (2006) and Menniti and Morgenroth (2010), respectively. Fresh mixed liquor sample (50 mL) was centrifuged @ 3500 rpm for 30 mins. The supernatant was then decanted carefully and filtered using fiber-glass filter (Whatman 934-AH) with a nominal pore size of 1.2  $\mu\text{m}$ . The filtrate was further filtered using a 0.2  $\mu\text{m}$  cellulose acetate filter for SMP analysis. bEPS was extracted by filling the centrifuge tube (up to 50 mL mark) with distilled water and then kept in ultrasonic water bath for 10 mins. After heating at 80  $^{\circ}\text{C}$  for 10 mins it was further centrifuged @ 3500 rpm for 10 mins. The supernatant was then decanted and filtered through 1.2  $\mu\text{m}$  syringe filter. Both the bEPS and SMP were quantified as COD of the sample. COD analysis was done using COD reagent (*Hanna Instruments*) following their prescribed procedure.

### **6.2.3 Statistical analysis**

Statistical analysis was performed to identify the biomass parameters affecting SOUR by bivariate correlation analysis. The relationships between the biomass characteristics and SOUR were examined by computing a Pearson product-moment correlation coefficient ( $r_p$ ) matrix between each pair of parameters. The Pearson correlation coefficient,  $r_p$ , ranges from -1 to + 1 where  $r_p = -1$  or +1 indicates a perfect negative or positive correlation respectively, and  $r_p = 0$  represents a lack of correlation. The analysis was carried out using the statistical package for the social science, SPSS V21.0 produced by IBM, USA.

#### **6.2.4 Parameter estimation**

The model equation was solved in Matlab 2014a based on the measured SOUR, MLSS, MLVSS, SMP and bEPS concentrations of the SSMBR for 32 and 49 days. The algorithm used in the solution process was of a nonlinear regression analysis using *fitnlm* function. The process was run with multiple initial values of parameters to ensure a maximum and acceptable  $R^2$  value. The Matlab program files for the analyses are attached in Appendix 1. During the evaluation of the proposed models, a different set of experimental data was used rather than that used in the calibration of the model.

### **6.3 Results and discussion**

#### **6.3.1 MLSS and SOUR profile with operation time**

The daily measure of biomass viability is important for tracking the health or efficiency of any wastewater treatment system, and the conventional practice is to monitor few physical parameters such as SOUR of the microbes in addition to the morphological properties of biomass such as floc size and structure, filamentous problem etc. However, these properties are controlled by the dynamic change of mixed liquor properties (generally speaking, the biomass properties or concentrations) (Germain et al., 2007; Hasar et al., 2002; Ji et al., 2010). The trace of any instability in the bioprocess operation can be detected based on experienced judgment of the mixed liquor properties e.g. by detecting whether the MLSS or MLVSS concentration are changing at a steady manner as usually expected. This may help prevent critical upsets of the treatment processes by controlling mixed liquor properties such as changing the mixed liquor concentration which subsequently would affect the dominant inhibitory or membrane-fouling components (e.g. SMP or bEPS).

Figure 6.1 shows MLSS and SOUR profile for the operational period of the SSMBR. With the initial MLSS concentration of the sludge at about 5 g/L, the SSMBR was operated for about 50 days. The MLSS concentration steadily increased up to a maximum of 18.84 g/L within 32 days of operation, after which some sludge was withdrawn (MLSS 10 g/L) to avoid rapid increase of the transmembrane pressure (TMP). It was shown in the figure that up to 32 days of operation of the MBR system, the increase of the concentration of MLSS was more or less steady while the SOUR also decreased nearly exponentially as was expected. These indicate that the system behaved in a stable manner up to 32 days after which time intervention was required to avoid the rapid rise of TMP.

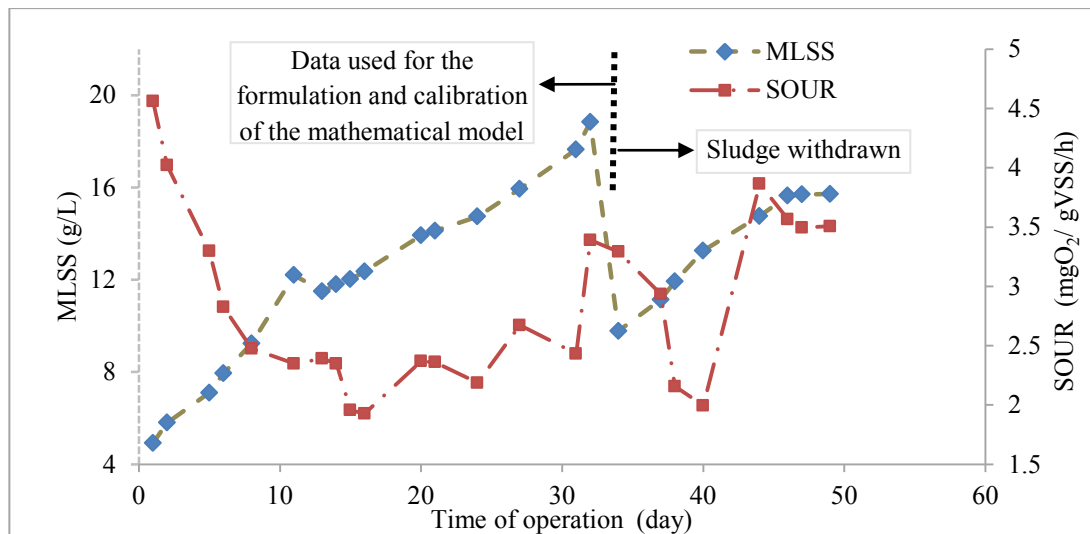


Figure 6.1 Variation of MLSS and SOUR as a function of time (SSMBR)

### 6.3.2 Correlation among biomass parameters and SOUR

The biomass parameters are often intricately related to each other, and therefore, the effects of many variables may be mixed or confounded in the practical treatment processes (Germain et al., 2007). Amongst other parameters, many researchers consider SOUR as the most reliable parameter indicating the microbial activity (Ngo et al., 2008; Nguyen et al., 2012; Villain and Marrot, 2013). The choice of the other variables of the

mathematical model of biomass viability, however, has been a challenging task for the researchers. It is difficult to isolate one parameter to represent a group of similar biomass parameters in the mathematical model.

Table 6.1 shows correlations between SOUR and several biomass parameters (i.e., MLSS, MLVSS, bEPS, and SMP). When EPS was considered as a lumped parameter of SMP and bEPS, it provided the best correlation to SOUR ( $r_p = -0.939$ ) supporting the findings of Lee et al. (2003) in a previous study. However, the individual correlations of either bEPS or SMP to the SOUR were also significant with the  $r_p$  for these parameters found to be -0.910 and -0.863 respectively. Although the correlations between SOUR and the mixed liquor concentrations were weak (for MLSS,  $r_p = -0.257$ ; for MLVSS,  $r_p = -0.325$ ), the correlation of SOUR to MLVSS/MLSS was strong ( $r_p = -0.846$ ). This is, however, contradictory to the findings of Germain et al. (2007), who found MLSS as the most important parameter affecting the oxygen transfer efficiency. The negative sign of the correlations indicate that these biomass parameters have negative effects on the SOUR. Hence, the relative influence of biomass parameters on SOUR of the microbes was in the order of EPS > bEPS > SMP > MLVSS/MLSS.

Table 6.1 Pearson- $r_p$  correlation matrix of the biomass parameters to SOUR

	SOUR					
MLSS	-.257	MLSS				
MLVSS	-.325	.997**	MLVSS			
bEPS	-.910**	.268	.325	bEPS		
SMP	-.863**	.098	.160	.794**	SMP	
EPS	-.939**	.219	.280	.974**	.910**	EPS
MLVSS/MLSS	-.846**	.003	.083	.729**	.763**	.778**

\*\* Correlation is significant at the 0.01 level (2-tailed); No of observation (N) = 24

The MLSS, as a lumped parameter of the microorganisms, colloids and particles, provided the weakest correlation to SOUR which is intuitively reasonable. Similarly, the MLVSS also does not have significant correlation to SOUR since only a small

percentage of the organisms in the activated sludge are viable while the dead mass contained in the MLVSS does not contribute to the biological treatment. The state-of-the-art mathematical models of biomass viability (Hasar and Kinachi, 2004) include the parameter MLVSS/MLSS, instead of the MLVSS or MLSS (Hasar et al., 2002). The normalized parameter of MLVSS to MLSS may be an indicator of microbial viability since this largely represents the viability of active biomass for biological treatment.

### 6.3.3 SOUR profile with the progressive change of microbial products

The SOUR typically follows an exponential decay over the days of operation of a biological treatment system. The correlations between the SOUR and several variables were also better determined based on exponential functions. Figure 6.2 shows the individual correlations of the SOUR with several potential biomass parameters.

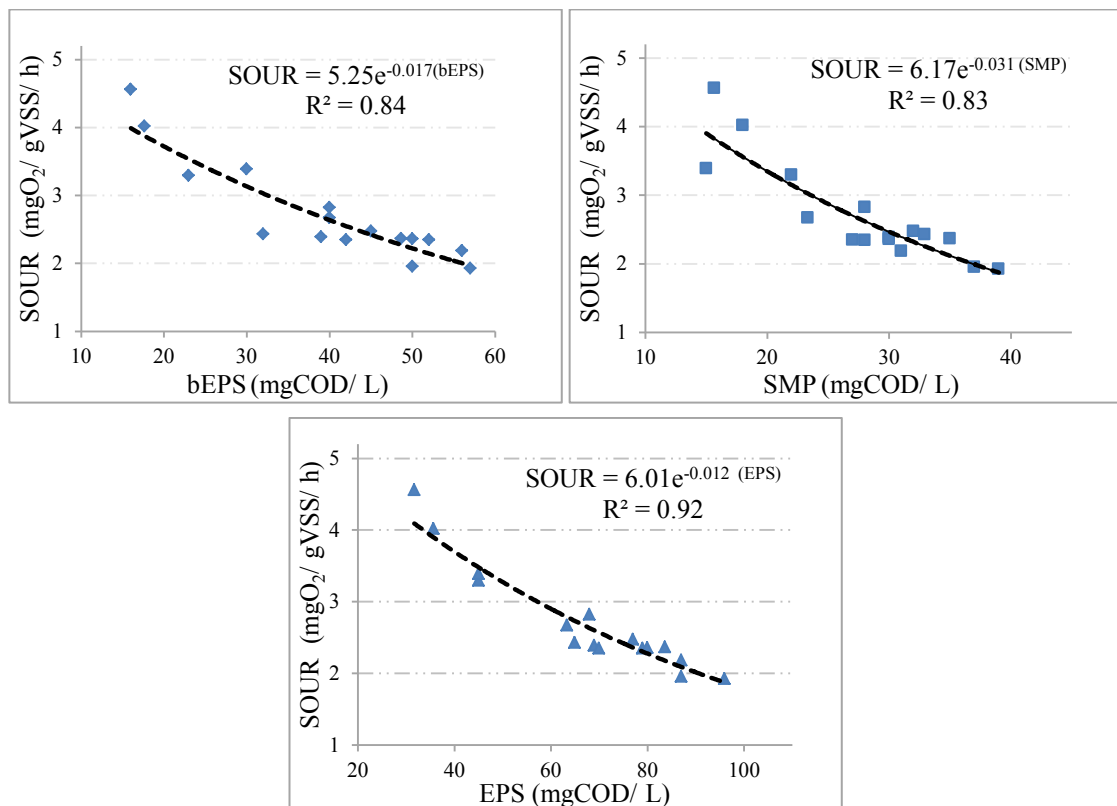


Figure 6.2 Relationship between SOUR and biomass parameters (up to 32 days of SSMBR operation)

The increase or decrease of several biomass parameters was typically maintaining inverse correlation through exponential best fit curve. The bEPS and SMP were individually related to the SOUR by exponential functions, and even better exponential correlation of SOUR to the EPS could be found where EPS was taken as the sum of bEPS and SMP. In an earlier study of the authors (Zuthi et al., 2014), the SOUR was found to be related with the bEPS by an exponential best fit function, whereas the variations of the SMP with SOUR did not follow any specific trend although the SOUR generally decreased with the increase of SMP in the bioreactor.

#### **6.4 Mathematical modelling of biomass viability and validation of the model**

The development of new mathematical model of biomass viability is aimed at achieving significant advancement over the currently available models of biomass viability. The most significant advancement is that the new model can be used to derive quantitative data about the biomass components e.g. the concentrations of SMP or bEPS in the bioreactor that directly or indirectly inhibit the biomass viability. The quantitative data of biomass components will help devise operation control strategies for treatability and fouling control by changing the mixed liquor properties (e.g. MLSS) which would obviously affect the concentration of SMP or bEPS. The currently available models (Hasar and Kinachi, 2004) including the conceptual model (Zuthi et al., 2014) proposed previously by the authors, however, can be used to explain rather qualitatively that the SMP or bEPS are correlated with the biomass viability based on the trace of soluble or colloidal components (soluble or colloidal COD) in the effluent stream.

The individual correlations between SOUR and several biomass parameters, as shown in Figure 6.2, are dependent on wastewater and sludge characteristics (e.g., MLSS). These individual mathematical expressions of SOUR will change with the change of wastewater or sludge characteristics. Therefore, normalization of these



parameters with respect to relevant biomass parameters is required to make a mathematical model independent of the effects of change of wastewater or sludge characteristics. Normalized biomass parameters such as  $bEPS_i/bEPS_0$ ,  $SMP_i/SMP_0$ ,  $EPS_i/EPS_0$  may be included in the mathematical model of biomass viability along with  $MLVSS/MLSS$ . The subscripts  $i$  and  $0$  for any biomass parameter indicate, respectively, the concentration of that parameter measured at the  $i^{\text{th}}$  day and that measured at the initial stage (at  $0^{\text{th}}$  day). As the wastewater used in the study was synthetic wastewater, the normalization of the biomass parameters was done with respect to the initial concentration of a biomass parameter in the supernatant of the bioreactor. As mentioned earlier, the normalized parameter  $EPS_i/EPS_0$  denotes a lumped parameter of  $bEPS$  and  $SMP$  given as  $(bEPS+SMP)_i/(bEPS+SMP)_0$ . Figure 6.3 shows the correlations of the measured values of  $SOUR$  to these normalized parameters for the operational period of the MBR up to 32 days.

Significant correlations of  $SOUR$  to these normalized biomass parameters indicate that these are the potential biomass parameters affecting biomass viability, and hence need to be included in the mathematical expression of biomass viability. Following the basic form of the empirical model proposed by Hasar and Kinachi (2004) and based on the findings of this study, two potential mathematical models of biomass viability have been formulated as shown by Eq. (6.2) and Eq. (6.3). From the correlation study,  $EPS$  was found most influential on  $SOUR$  as compared to the individual effects by  $bEPS$  or  $SMP$ . Therefore, two variations of the mathematical models of biomass viability are proposed, one including  $bEPS$  and  $SMP$  along with  $MLVSS$  and  $MLSS$  (model 1) while in the other model (model 2) the lumped parameter  $EPS$  replaces  $bEPS$  and  $SMP$ .

Model 1:

$$\text{SOUR} = a_1 + \exp[a_2 + b_1 \cdot (M_1/M_2) + b_2 \cdot (bE_i/bE_0) + b_3 \cdot (SP_i/SP_0)] \quad \text{Eq. (6.2)}$$

Model 2:

$$\text{SOUR} = a_1 + \exp[a_2 + b_1 \cdot (M_1/M_2) + b_2 \cdot (E_i/E_0)] \quad \text{Eq. (6.3)}$$

where  $a_1$ ,  $a_2$ ,  $b_1$ ,  $b_2$  and  $b_3$  are constants,  $M_1/M_2$  is the ratio of MLVSS and MLSS,  $bE_i/bE_0$  is  $bEPS_i/bEPS_0$ ,  $SP_i/SP_0$  is  $SMP_i/SMP_0$  and  $E_i/E_0$  is  $EPS_i/EPS_0$ .

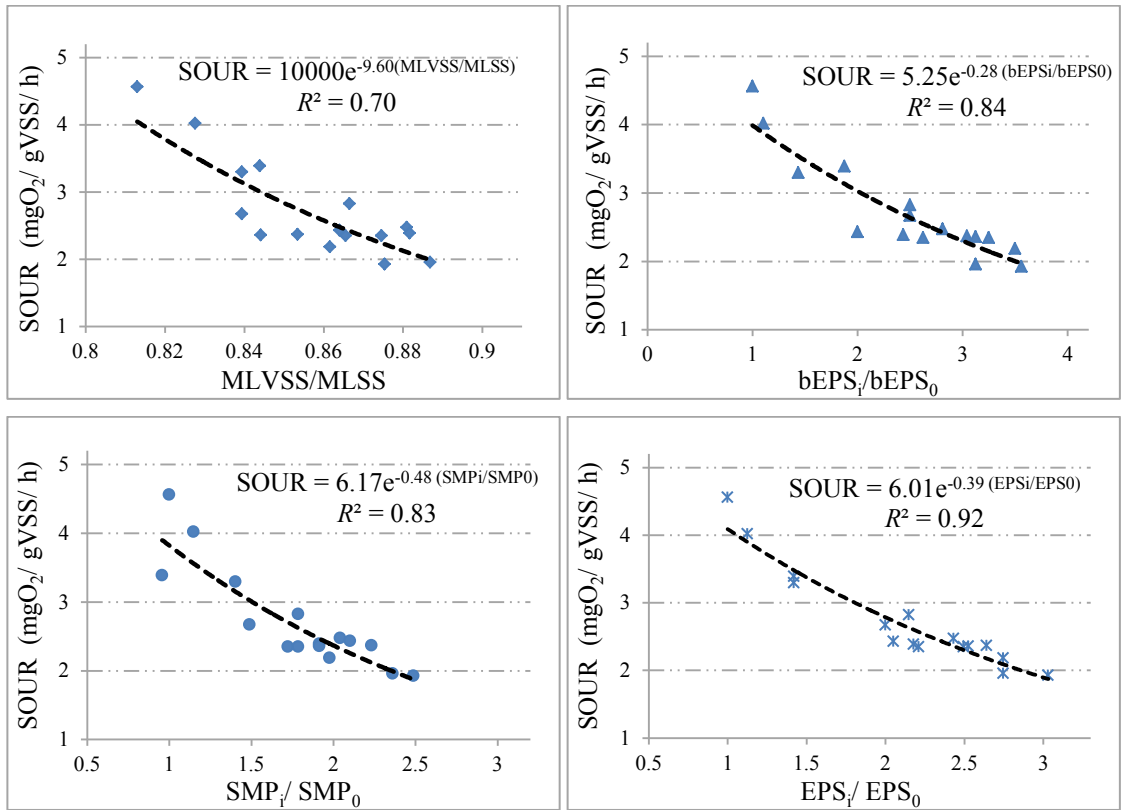


Figure 6.3 Relationship between SOUR and normalized biomass parameters

Both models were solved by nonlinear regression analysis using the *fitnlm* function of Matlab 2014a. With the coefficients determined by regression analysis, the models of biomass viability are determined as shown in Eq. (6.4) and Eq. (6.5):

Model 1:

$$\text{SOUR} = 1.31 + \exp[5.63 - 4.65 \cdot (M_1/M_2) - 0.22 \cdot (bE_i/bE_0) - 0.46 \cdot (SP_i/SP_0)]; R^2 = 0.974 \quad \text{Eq. (6.4)}$$

Model 2:

$$\text{SOUR} = 1.20 + \exp[6.026 - 5.30 \cdot (M_1/M_2) - 0.53 \cdot (E_i/E_0)]; R^2 = 0.972 \quad \text{Eq. (6.5)}$$

The models are then simulated for the total investigated period of the SSMBR whereas data from the first 32 days of operation have been used for the calibration of the models and estimation of the coefficients. As some sludge was withdrawn at 32 days of operation of the MBR system, there were abrupt changes in the MLSS, MLVSS, BEPS and SMP concentrations in the mixed liquor after 32 days of operation of the SSMBR (Figure 6.1). Figure 6.4 shows that the model simulation results for both the variations of the model are almost identical and compare well with the experimental results for the total operational period (49 days) of the SSMBR system.

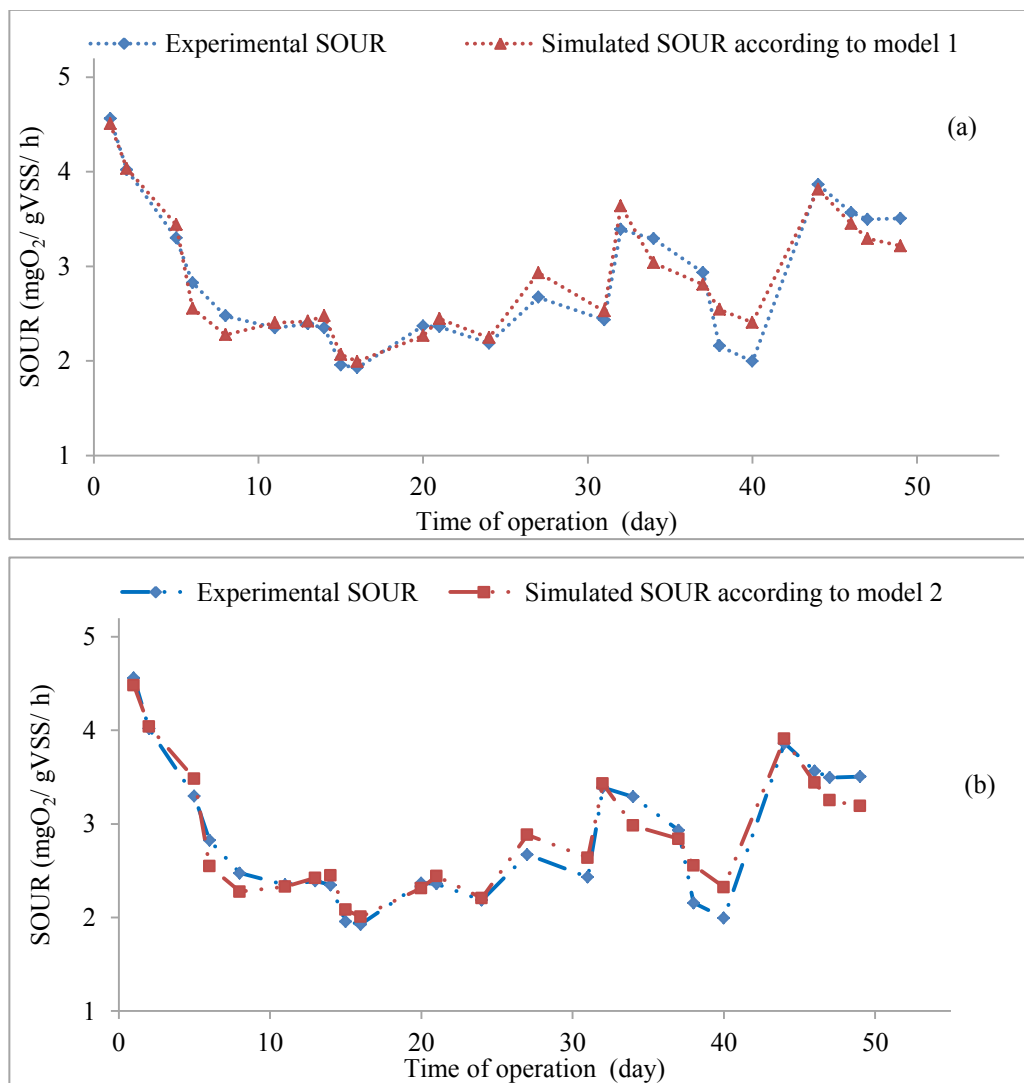


Figure 6.4 Comparison of experimental and simulated SOUR profile: (a) simulation of model 1; (b) simulation of model 2

The model simulations of the SOUR agree well with the experimental SOUR data for both variations of the model. The model's predictions of SOUR for the period after the sludge withdrawal (32 days) are also found reasonably well when compared to the respective experimental data of the SOUR. The EPS represented as a lumped parameter in the model (model 2) gives slightly better simulation results by the mathematical model of biomass viability. Hence, the calibrated models of biomass viability are also validated against the experimental data from the SSMBR for its total operational period. However, the models need to be validated further with a wide range of data from MBR systems of different operating conditions.

## 6.5 Conclusion

The effects of biomass on microbial activity of a lab-scale SSMBR were investigated in the study. Based on the statistical analysis of test results and trends of SOUR affected by biomass characteristics, two independent mathematical models were formulated and subsequently validated by the experimental data obtained from the SSMBR. The following specific conclusions were obtained from the study:

- bEPS, SMP, MLSS and MLVSS significantly affected the SOUR and their relative influence on SOUR was  $EPS > bEPS > SMP > MLVSS/MLSS$ .
- the progressive change of SMP and bEPS within the bioreactor consistently maintained a negative exponential correlation with SOUR.
- Microbial products such as bEPS, SMP or EPS should be considered in the mathematical model of biomass viability. Based on correlation study, two models of microbial viability have been formulated as a function of MLSS, MLVSS, bEPS and SMP (model 1); and MLSS, MLVSS and EPS (model 2).

- The simulated results of both models agree well with the experimental results. Model 2 performed better than model 1 as the EPS, as a lumped parameter of bEPS and SMP, showed the highest correlation with SOUR.



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## **CHAPTER 7**

# **New and Practical Mathematical Model of Membrane Fouling in an Aerobic Submerged Membrane Bioreactor**

## 7.1 Introduction

Membrane fouling in an MBR occurs due to the dynamic development of resistances by several fouling components dominant among which are the fouling due to the cake (sludge) layer formation and the pore blocking of the membrane. This chapter presents systematic development and calibration of a semi-empirical mathematical model of membrane fouling successfully using these major dynamic fouling components to describe the fouling phenomena. The experimental data used for the model calibration are that of the continuously aerated SSMBR system (described in Chapter 4) which was operated at constant flux for about 50 days.

Due to the high number of interactions between physio-biochemical conditions within MBR, mathematical model-based approach has been adopted to gain a deeper insight of the fouling phenomena. In this context, this chapter describes development of a simple mathematical model of membrane resistance combining three separate fouling resistances considering as well the effects of aeration and backwashing on the reduction of fouling. As the overall fouling process is a dynamic one, the mathematical expressions for the membrane fouling processes proposed here necessarily include differential equations with time-dependent variables and constants. The model calibration is performed using experimental data of the SSMBR for the first 32 days of its operation. The progressive increase of TMP is considered as a direct indicator of the dynamic membrane fouling phenomena, and the internal fouling resistances calculated by the mathematical model are used to derive comparable prediction of TMP with the experimental TMP data. The biomass parameters that have been used in the model of biomass viability are also used in the model of membrane fouling, which will facilitate integrated use of the models for devising efficient operational control and management of MBR systems for better treatability and fouling control.

## 7.2 Methods of measurement of fouling resistances and analysis procedure

### 7.2.1 Measurements of fouling resistances

Before commencing the experiment, the clean membrane resistance was measured in distilled water at various fluxes from 5 to 30 L/m<sup>2</sup>.h flux at the increment of 5 of L/m<sup>2</sup>.h. At the end of experiment, the membrane module was taken out of the bioreactor and submerged in distilled water to measure the total resistance ( $R_T = R_m + R_p + R_c$ ) of the fouled membrane. Darcy's law was applied to calculate total membrane resistances ( $R_T$ ) following Eq. (7.1):

$$J = \frac{\text{TMP}}{\mu R_T} \quad \text{Eq. (7.1)}$$

where  $J$  is the permeate flux; TMP is the transmembrane pressure,  $\mu$  is the viscosity of the permeate at 20°C;  $R_T$  is the total resistance which is the combination of membrane's intrinsic resistance ( $R_m$ ), pore fouling resistance ( $R_p$ ) and cake layer resistance ( $R_c$ ).

The membrane was cleaned with distilled water by gently shaking to remove the deposited sludge cake layer from the membrane surface, and then submerged again in distilled water for the measurement of  $R_p$  and  $R_m$ . Finally, the membrane was chemically cleaned with 2% citric acid for 6 hours to remove internal pore fouling, and then cleaned with 0.4% NaOCl and 4% NaOH for 6 hours to determine the intrinsic membrane resistance ( $R_m$ ). For the calculation of daily total resistances from the measurements of TMP, the value of  $\mu$  was corrected for temperature according to Eq. (7.2) (Delrue et al., 2011);

$$\mu_T = \mu_{20} e^{-0.0239(T-20)} \quad \text{Eq. (7.2)}$$

where  $T$  is the mixed liquor temperature in °C.

It was expected that some of the measured data of variables would be inconsistent which might occur due to the environmental disturbances (e.g., temperature



fluctuations) or simply because of the system fluctuations. Filtration to screen out those data was done carefully, which might otherwise affect the correlation and subsequently the mathematical model of membrane fouling.

### **7.2.2 Estimation of parameters of the mathematical model**

The mathematical model equations were solved in Matlab 2014a based on the measured TMP, fouling resistances, MLSS and SMP concentrations in the bioreactor of the SSMBR. The algorithm used in the solution process was that of a nonlinear regression analysis using *fitnlm* function. The process was run with different initial values of parameters to ensure a maximum and acceptable value of  $R^2$  (squared value of the coefficient of variance). However, a different set of experimental data was used during the verification of the proposed models rather than that used in the calibration of the model.

## **7.3 Model development**

In the simplified approach of mathematical modelling, the development of fouling of the membrane is better linked with biological indicator parameters such as the concentrations of SMP and MLSS in the bioreactor. In this regard, the dynamic membrane fouling is considered occurring in two major phases.

- The internal pore fouling of the membrane is assumed to occur by the adsorption of soluble particles within/onto the pores (e.g. SMP).
- The external cake layer resistance to flux is assumed to occur as the main fouling resistance integrated in which is the resistance due to biofilm.

### **7.3.1 Resistance due to pore blocking**

A fraction of soluble products are adsorbed within the pores, and therefore reduces the effective pore sizes as well as the surface porosity of the membrane causing the internal

pore fouling of the membrane. The soluble particles that are considered to cause pore blocking of the membrane are the SMPs (soluble EPS of the microbial products). The mathematical formulation of internal pore fouling is typically expressed by relationships of pore blocking resistance with progressive reduction of the effective pore radius ( $r_p$ ) and effective porosity ( $f$ ) of the membrane as shown by Eq. (7.3).

$$R_p = e^{(n_p t)} \frac{8h_m}{f r_p^2} \quad \text{Eq. (7.3)}$$

where  $h_m$  = membrane's effective thickness. The expression for  $R_p$  was first proposed by Wiesner and Aptel (1996) and later modified by Bowen et al. (1997). An extension of the mathematical expression for  $R_p$  is proposed by introducing an exponential term with a factor  $n_p$  to better explain the typically observed exponential rise of TMP especially after the initial stages of operation of an MBR system. The mass balance equation for particles around the membrane causing the reduction of porosity is calculated following Busch et al. (2007).

$$\rho_p \frac{df}{dt} = -4\eta_f J(t) c_{SMP}(t) \frac{m_{d,o}}{(m_{d,o})^2 - (m_{d,i})^2} \quad \text{Eq. (7.4)}$$

where  $\rho_p$  is the density of biomass,  $c_{SMP}$  is the concentration of soluble particles entering the pores which may be taken as  $c_{SMP}$  in the bioreactor,  $\eta_f$  is the average fraction of soluble particles that accumulate in the pores. Equation 7.4 can be rewritten as shown in Eq. (7.5) following the basic equation proposed by Giraldo and LeChevallier (2006).

$$\frac{df}{dt} = -\alpha_f c_{SMP}(t) J(t) \quad \text{Eq. (7.5)}$$

where  $\alpha_f$  is the membrane porosity reduction coefficient to be determined from Eq. (7.6)

$$\alpha_f = \frac{1}{\rho_p} \cdot \frac{4\eta_f m_{d,o}}{(m_{d,o})^2 - (m_{d,i})^2} \quad \text{Eq. (7.6)}$$

The differential equation to account for the effect of pore size reduction due to the adsorption of soluble particles within the pores is given in equation 7.7 (Giraldo and LeChevallier, 2006).

$$\frac{dr_p}{dt} = -\alpha_p c_{SMP}(t)J(t) \quad \text{Eq. (7.7)}$$

where,  $\alpha_p$  = pore size reduction coefficient. The concentration of SMP in the bioreactor ( $c_{SMP}$ ) is a time dependent variable which depends on the design and operation of an MBR system, particularly depending on the initial concentrations of MLSS.

### ***7.3.2 Resistance due to cake layer formation***

External membrane fouling is caused by the deposition of cake layer over the membrane surface which gradually grows in size and thickness over time. It was found in the earlier studies that the membrane fouling generally increases with the increase in the MLSS concentrations (Kornboonraksa and Lee, 2009) that mainly contribute to the progressive formation of cake layer on the membrane surface. The formation of the cake layer on the membrane surface integrates in it the formation of biofilm. However, the cake layer resistance due to the formation of biofilm is a complex process in the mathematical modelling, especially due to the hardly understood process of the detachment of biofilm (Busch et al., 2007). As the formation of biofilm is inevitable in an MBR system and is acknowledged as one of major causes of membrane fouling (Gao et al., 2013), fouling prediction would not be complete without taking it into consideration. Consequently, the formation and detachment of the biofilm layer is not separately treated in the mathematical modelling, but is assumed to be integrated in the process of the formation and detachment of the cake layer. The concentration of MLSS in the bioreactor is taken as a gross parameter affecting the cake layer formation on the

membrane while the dynamic effects of the formation of biofilm and cake layer is still accounted for by taking the changes of MLSS concentrations in the model formulations.

Due to the continuous aeration and periodic backwashing in the MBR system, part of the cake layer is detached from the membrane surface back into the mixed liquor suspension. An average rate of detachment of the cake layer ( $k$ ) is assumed to represent this phenomenon which is accounted in the mass balance equation of the formation of cake layer over the membrane surface. In this simplified mathematical model, the variation of concentrations of MLSS (lumped parameter including SMP and bEPS and other microorganisms) in the bioreactor is assumed to be linked with the net rate of the attachment of cake layer (including biofilm) on the membrane surface. The cake filtration effects accounting the cake compressibility is included in the mathematical expressions of cake resistance ( $R_c$ ) as shown in Eq. (7.8).

$$R_c = \alpha_c h_c(t) \rho_c e^{(n_c t)} \quad \text{Eq. (7.8)}$$

where  $\alpha_c$  = specific resistance of the compressible cake layer,  $h_c$  = variable depth of the cake layer expressed as a first order differential function in time. Considering the mass balance of the cake forming particles (e.g. MLSS) over the membrane surface, the  $h_c$  can be expressed in the following differential equation:

$$\rho_c \frac{dh_c}{dt} = J \cdot (1 - k) c_c(t) \quad \text{Eq. (7.9)}$$

where  $c_c$  = concentration of potential cake forming particles in the bulk liquid (e.g. MLSS) which typically varies over time,  $\rho_c$  = density of the cake layer. The factor  $k$  accounts for the detachment of the cake layer from the membrane surface a reasonable value for which may be determined from the model calibration. The depth of the cake layer ( $h_c$ ) is calculated from the solution of Eq. (7.9) and is replaced in Eq. (7.8) to calculate the value of  $R_c$ . Finally, the total resistance is calculated from the equation of the resistance-in-series model as follows:

$$R_T = R_m + R_p + R_c \quad \text{Eq. (7.10)}$$

Here the intrinsic membrane resistance ( $R_m$ ) is a static component in the mathematical expression which can be determined experimentally, but the total membrane resistance ( $R_T$ ) becomes a dynamic variable as it includes  $R_p$  and  $R_c$ . The external physical parameter indicative of the membrane fouling at any stage of the operation of an MBR system is the TMP (or  $\Delta P$ ). The state of fouling of the membrane at any instance of time ( $t$ ) can be obtained from the current TMP( $t$ ) the mathematical expression for which are related to the respective measured data of flux ( $J$ ) and total internal resistance ( $R_t$ ) to flux according to Darcy's law as shown in Eq. (7.1).

## 7.4 Results and discussion

### 7.4.1 Variation of MLSS and SMP with operation time

The operation of full-scale MBR systems is typically conducted with MLSS concentrations in the range of 8 to 18 g/L (Drews, 2010). Among the two common practices in the operation of the MBR systems, one is to keep the MLSS concentration fixed more or less around a certain value which, however, needs frequent removal of excess sludge or activated sludge from the mixed liquor to avoid any instability in the operation of treatment such as to avoid the rapid rise of TMP. In the continuously operated MBR systems without sludge removal, the concentration of MLSS often increases steadily in most of the MBR systems depending on the feed characteristics and microbes present in the sludge (Hernandez et al., 2014). From the operational point of view, the latter practice of the MBR operation may offer advantages, for example it may promote more nitrification process due to the development of nitrifying bacterial community in the increased MLSS concentration (Kornboonraksa and Lee, 2009). However, the excess activated sludge may still need to be withdrawn in the

continuously operated MBR systems to maintain its operation for longer term or to avoid any sudden instability in its operation. In a study of an MBR system for treating domestic wastewater, Hasar et al. (2002) withdrew sludge in two stages to sustain stability in the operation of the system as the MLSS steadily increased to much higher value resulting in rapid rise of TMP.

In this study, the SSMBR system was operated up to 49 days and started with the initial MLSS concentration of 5 g/L. The MLSS concentration steadily increased to 18.8 g/L after 32 days of operation of the system when the rapid rise of TMP was first observed. Consequently, some sludge was withdrawn (to reduce the MLSS concentration around 10 g/L) after 32 days to avoid the rapid rise in TMP. The MLSS again steadily increased from 10 g/L to 17 g/L after 49 days when the operation of the system was terminated since the TMP increased to about 50 kPa.

The effects of the withdrawal of sludge at 32 days were also evident in the concentrations SMP in the bioreactor of the SSMBR. The concentration of SMP in the bioreactor of MBR depends on the microbial activity, membrane rejection efficiency and many other factors. During the first 32 days of operation of the SSMBR, the SMP concentration was found in the range of 15 to 39 mg COD/L but relatively lower SMP concentrations (15 to 22 mg COD/L) were found in the latter stage of the operation period. Menniti and Morgenroth (2010) studied an MBR system under different operating conditions created by high shear and low shear aerations and with different membrane size. In the high shear MBR, the SMP concentration in the bioreactor was found to be around 50 to 100 mg COD/L, while the SMP concentration in the low shear MBR varied within the range between 50 and 350 mg COD/L.

## 7.4.2 Model analysis and application

### A. Inputs for model calibration

In the calibration of the model, the MLSS and SMP concentrations in the bioreactor are input as the best representative mathematical functions of time for easier solution of Eq. (7.7) and Eq. (7.9). The data chosen to derive the functions are the representative data measured during the first 30 days of the operation of the SSMBR system. The variation of the concentrations of MLSS (in g/L) up to 32 days of operation of the system can be best represented by a linear function with a reasonably good correlation coefficient ( $R^2=0.95$ ), as shown in Eq. (7.11) and in Figure 7.1.

$$\text{MLSS} = 0.41 t + 5.6 \quad \text{Eq. (7.11)}$$

where,  $t$  represents the days of operation of the SSMBR. In any continuously operated MBR systems with no sludge withdrawal, the MLSS concentrations mostly increase at a steady rate (Hernandez et al., 2014; Kornboonraksa and Lee, 2009). Therefore, this type of best approximated function may be developed for the typical MBR system's operation.

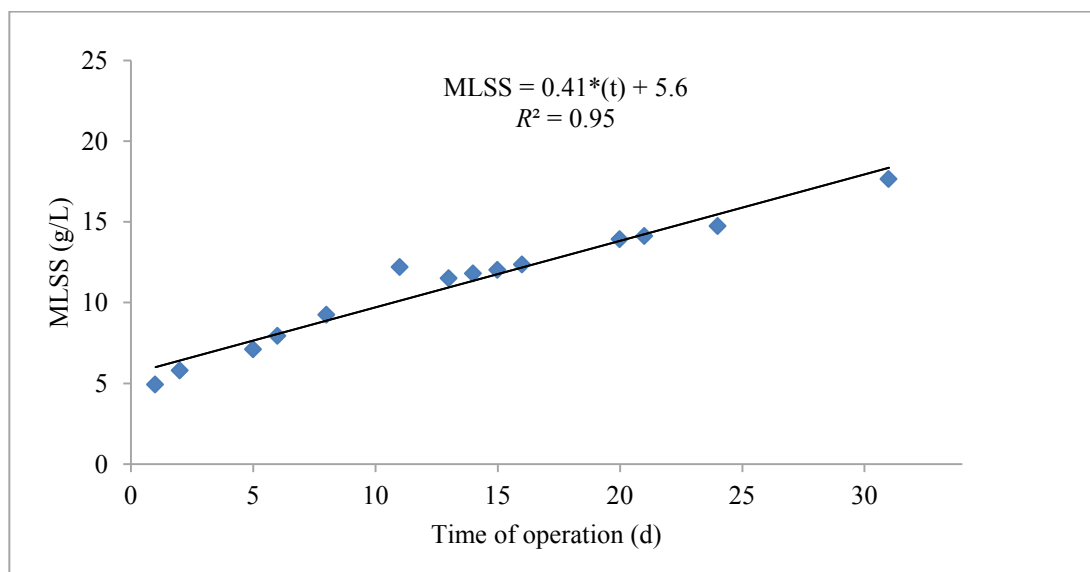


Figure 7.1 Variation of MLSS in bioreactor during the first 32 days of SSMBR operation

The variations of the concentrations of SMP in the bioreactor for the first 32 days of operation of the MBR can be better represented by a power function ( $R^2$ : 0.79), as shown in Eq. (7.12) and in Figure 7.2.

$$c_{\text{SMP}} = 16.3*(t)^{0.24} \quad \text{Eq. (7.12)}$$

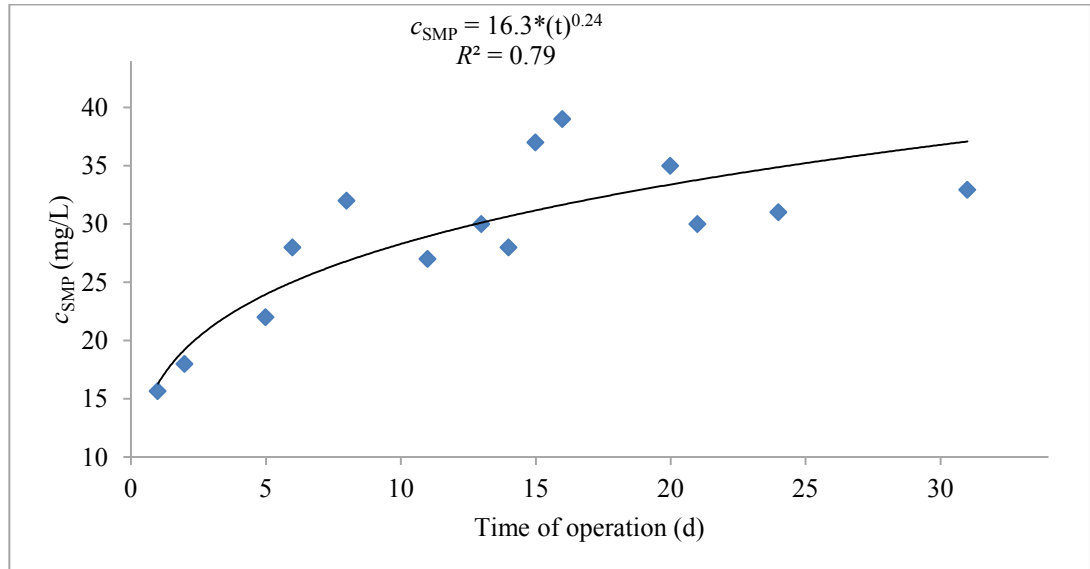


Figure 7.2 Variation of SMP in bioreactor during first 32 days of the SSMBR operation

The numerical model simulation is easy if the rate of change of MLSS or SMP concentrations is given as best approximated functions. The best approximated functions, as that are found in this study, may not be found in other MBR systems although with continuous operations of the system. In fact, the dynamic change of SMP concentration within bioreactor depends on the SMP growth rate, rate of degradation, the membrane rejection efficiency and many other factors. Menniti and Morgenroth (2010) found fluctuating (no trend) concentration of SMP in a high shear MBR while the SMP concentration steadily increased in a low shear MBR. However, the proposed model's simulation can also be done by the inputs of discrete value of these parameters.

With specific operational and design parameters (Table 4.1, Chapter 4), the value of the membrane porosity reduction coefficient ( $\alpha_f$ ) was determined to be  $3.25 \text{ m}^2/\text{Kg}$  according to Eq. (7.6) while the value for the membrane pore size reduction coefficient



( $\alpha_p$ ) is reasonably assumed to be  $0.000943 \text{ m}^3/\text{Kg}$  ( $=1/\rho_p$ ). An average value for the specific cake resistance  $\alpha_c = 1 \times 10^{14} \text{ m/Kg}$  was adopted which fell between the upper and lower bound value as reported in the literature (Li and Wang, 2006). With other design and operational parameters (e.g. measured values of  $J$ ,  $\mu$ ,  $R_m$ ) and the expressions for  $R_p$  and  $R_c$  are derived from Eq. (7.3) and Eq. (7.8) respectively, TMP was calculated according to Darcy's law (Eq. (7.1)).

### B. Model calibration and reliability analysis

The unknowns involved in the solution of the mathematical expressions have some characteristic features by definition. The coefficients  $n_p$  and  $n_c$  should preferably have positive values, the value of effective initial porosity of the membrane should be in the range between 5% and 34% (Yoon et al., 2006), and the coefficient for the rate of cake layer detachment ( $k$ ) should have a value between 0 and 1. With the input values of variable TMP (experimental) for the first 32 days of operation of the MBR, the resulting equation for TMP (Eq. (7.1)) could be solved by non-linear regression analysis to find unknowns and hence to predict reasonably accurately the TMP (Figure 7.3). The Matlab program file and output for the simulation is attached in Appendix 2(a).

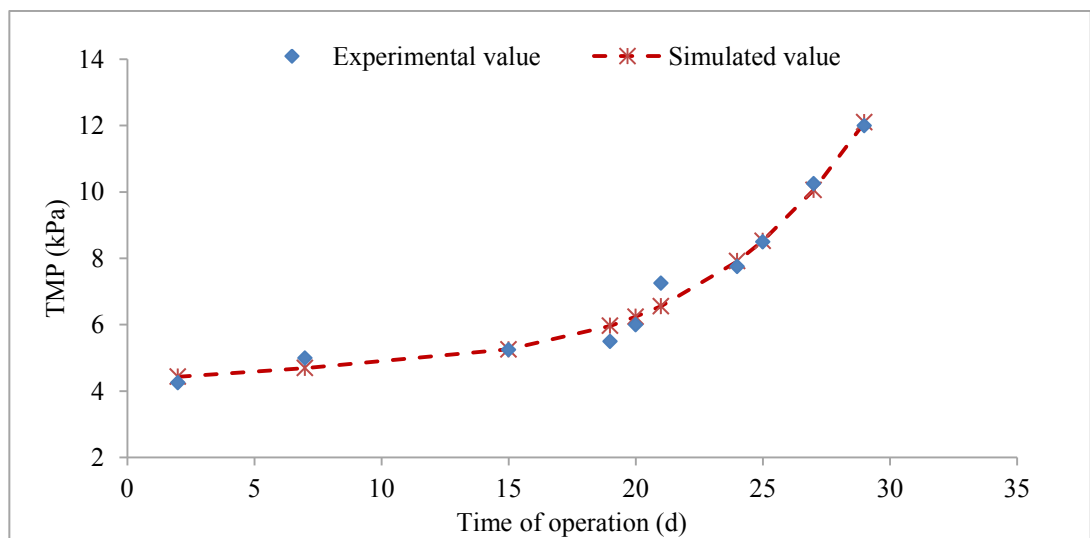


Figure 7.3 Comparison of the experimentally measured TMP and the TMP calculated from mathematical model (for the first 32 days of SSMBR operation)

However, the unknown parameters and constants determined from the combined mathematical modelling of total internal resistances ( $R_T=R_m+R_p+R_c$ ) against experimental TMP are meaningless according to their definition in the model equations (e.g.  $k \gg 1$ ). Although the total resistance ( $R_T$ ) could be predicted fairly accurately, the boundary values of  $R_p$  and  $R_c$  as determined from these coefficients do not satisfy the values that were experimentally measured at the end of operation of the SSMBR.

Therefore, the calibration of the mathematical model of membrane fouling was done separately for the two dynamic components of fouling resistances,  $R_p$  and  $R_c$ . The initial and final boundary values of  $R_p$  used for the model calibration are zero and  $3.5 \cdot 10^{12}$  /m, respectively. Figure 7.4 shows simulated results of  $R_p$  with different assumed values of effective initial porosity as 7%, 10%, 15% and 25% of the membrane. The Matlab program files for the simulation for different porosities are attached in Appendix 2(b).

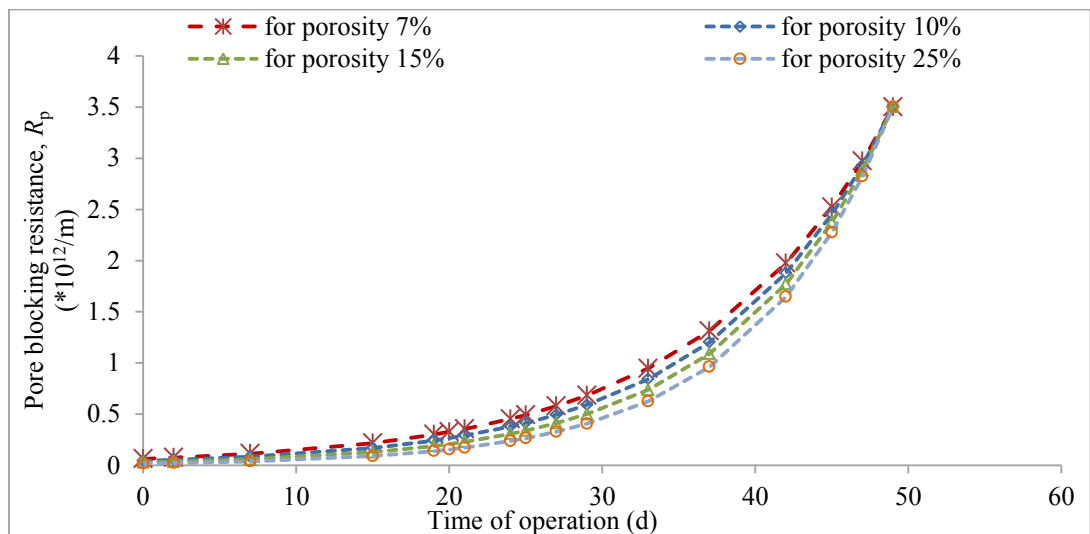


Figure 7.4 Simulated  $R_p$  for various initial porosities of membrane

The values of the coefficients and constants as determined from the model simulations for  $R_p$  are then used for further simulations for TMP against  $R_T$  to determine other coefficients and constants (Appendix 2(c)). Table 7.1 shows the calculated values of all the model parameters obtained from the mathematical model simulations using the data for the first 32 days of operation of the SSMBR system. The respective

calculated values of cake layer resistance ( $R_c$ ) and the pore fouling resistance ( $R_p$ ) at the end of day 49 of the SSMBR's operation are also included in Table 7.1 disregarding the fact that a fraction of the activated sludge was withdrawn to avoid the instable operation of the SSMBR system when the rapid rise of TMP was observed. Table 7.1 shows that the calculated values of coefficients are meaningful and reasonable when the initial porosities of membrane are below 15%. The calculated rate of cake layer detachment (the value of the coefficient,  $k$ ), for example, should be a positive number with a value less than 1 which could only be found when the initial porosities of the membrane was assumed to be between 7% and 15%. At the same time, the assumed porosity between 7% and 15% is also a reasonable assumption for the microfiltration membrane.

The cake layer resistance  $R_c$ , as determined by the model simulation with assumed effective initial porosity of 15%, is found to  $1.205 \cdot 10^{13}$ /m. The measured cake layer resistance ( $1.07 \cdot 10^{13}$  /m) at day 49, however, was less than that determined from the model simulations. This is acceptable considering the fact that a fraction of the sludge was withdrawn at day 32 of the SSMBR's operation. An even better agreement with the experimentally measured pore fouling resistance ( $R_p$ ) at day 49 could be found by the model simulation results as shown in Table 7.1. Therefore, an effective initial porosity of 15% seems to be reasonable for the membrane of the SSMBR considering the better agreement for both the boundary values of  $R_p$  and  $R_c$ .

*Table 7.1* Parameters and model simulation results with various porosities of membrane

Parameters	Initial porosities (%)			
	7	10	15	18
$K$	0.07	0.067	0.025	-0.007
$n_p$	0.081	0.089	0.097	0.107
$n_c$	0.231	0.118	0.065	0.065
$R_c$ (/m)	$1.230 \cdot 10^{13}$	$1.204 \cdot 10^{13}$	$1.205 \cdot 10^{13}$	$9.830 \cdot 10^{14}$
$R_p$ (/m)	$3.5 \cdot 10^{12}$	$3.5 \cdot 10^{12}$	$3.5 \cdot 10^{12}$	$3.5 \cdot 10^{12}$

*Table 7.2* Calibrated model parameters and coefficients used in simulations

Parameter	Description	Value
$K$	rate of detachment of cake layer due to the combined effects of backwash and aeration (%)	0.025
$\alpha_c$	specific cake resistance (m/Kg)	$1 \times 10^{14}$
$\alpha_f$	membrane porosity reduction coefficient (m <sup>2</sup> /Kg)	3.25
$\alpha_p$	pore size reduction coefficient (m <sup>3</sup> /Kg)	0.000943
$n_c$	exponential parameter (used in cake layer resistance)	0.065
$n_p$	exponential parameter (used in pore resistance)	0.097
$R_m$	membrane's intrinsic resistance (/m)	$7.43 \times 10^{11}$

### *C. Comparison between experimental and simulated results*

The model of membrane fouling described in this chapter has introduced two new parameters  $n_p$  and  $n_c$ , respectively to account for the exponential rise of dynamic resistances  $R_p$  and  $R_c$  that are typically comparable with the exponential rise of TMP especially after the initial stage of operation of any MBR. The model could not be calibrated against the boundary values of  $R_p$  and  $R_c$  without using these exponential parameters. Figure 7.5, for example, shows the model simulation results for  $R_p$  with and without using the parameter  $n_p$  in the expression for  $R_p$ . It is evident from the simulation results that the rise of pore fouling resistance without exponential parameter in the model is very insignificant which does not fit the typical observations of the increase of  $R_p$  in any MBR system which is also comparable to the pattern of TMP rise. Meaningful values of the model parameters and coefficients can only be calculated by the calibration of the model separately for the two dynamic components of fouling resistances,  $R_p$  and  $R_c$ .

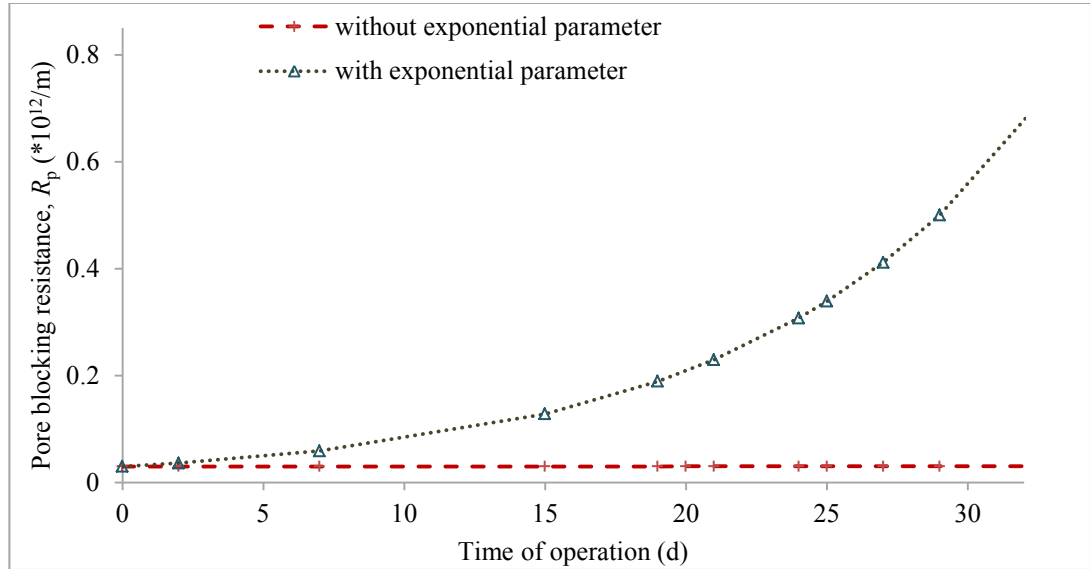


Figure 7.5 Simulated  $R_p$  with and without using the parameter  $n_p$  (for porosity 15%)

The final boundary values of  $R_p$  and  $R_c$  as determined by the calibrated model agree reasonably well with the experimentally determined  $R_p$  and  $R_c$  although only few selected experimental measures of TMP have been used for the calibration of the model. Figure 7.6(a) shows that the experimental  $R_p+R_c$  also compares well against the daily variations of simulated  $R_p+R_c$  particularly for the period when exponential increase of fouling resistances/ TMP occurred. Figure 7.6(b) shows the experimental results of TMP against the daily variations of simulated TMP according to Eq. (7.1) followed by the determination of  $R_c$  and  $R_p$  (the flowchart shown in Figure 7.7). The Matlab program files for the simulations are attached in Appendix 2. Therefore, the mathematical model can be effectively used to predict separate components of the dynamic fouling resistances along with the prediction of total fouling resistances or TMP differences.

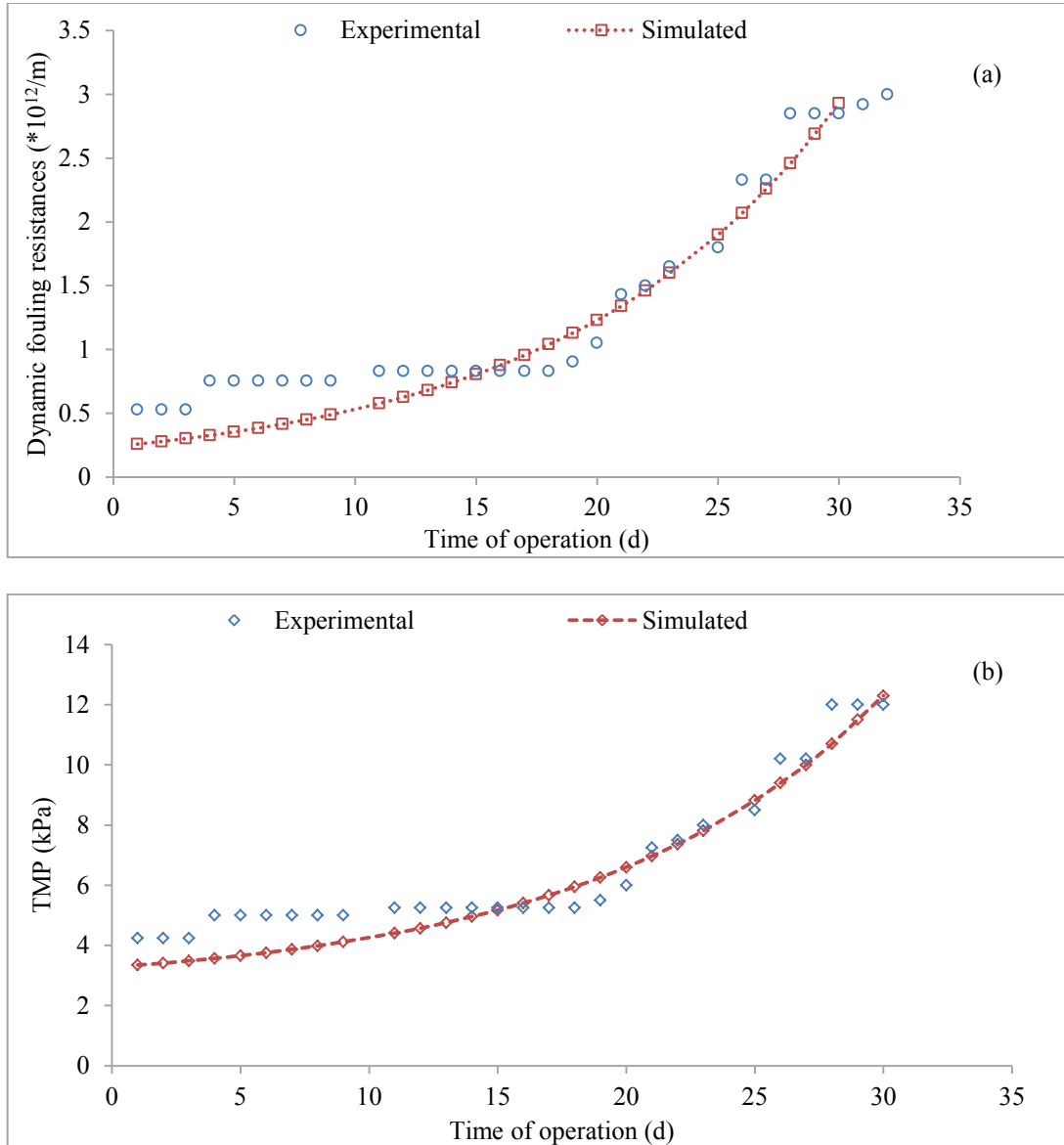


Figure 7.6 Comparison of model simulation results with experimental results of SSMBR (a)  $R_p + R_c$ ; (b) TMP

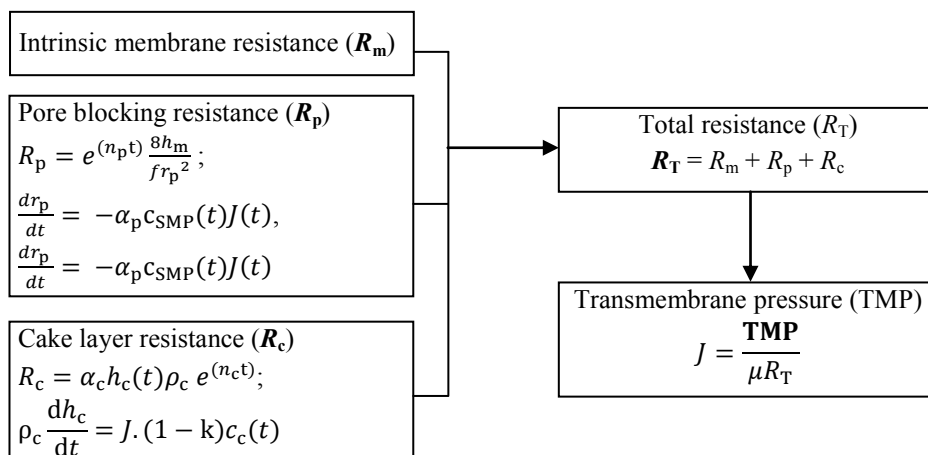


Figure 7.7 Flowchart for the calculation of TMP

The model simulation was also carried out to compare the rise of TMP in a conventional MBR system (CMBR, without sponges in the bioreactor) of the same type that was run with a reduced constant flux ( $10 \text{ L/m}^2/\text{hr}$ ) but with nearly the same initial MLSS concentrations of the sludge ( $5.7 \text{ g/L}$ ) as that of the SSMBR. The wastewater characteristics and the volume of the reactors are same in both the MBR systems. The mean pore size of the membrane of the CMBR is  $0.2 \mu\text{m}$ . The model simulation has been done by using the calibrated model parameters and coefficients of the SSMBR, but changing the parameters relevant to the operational parameters of the CMBR (e.g.  $J$ ). Figure 7.8 shows the model simulation results of TMP of the CMBR as compared to the experimentally measured results.

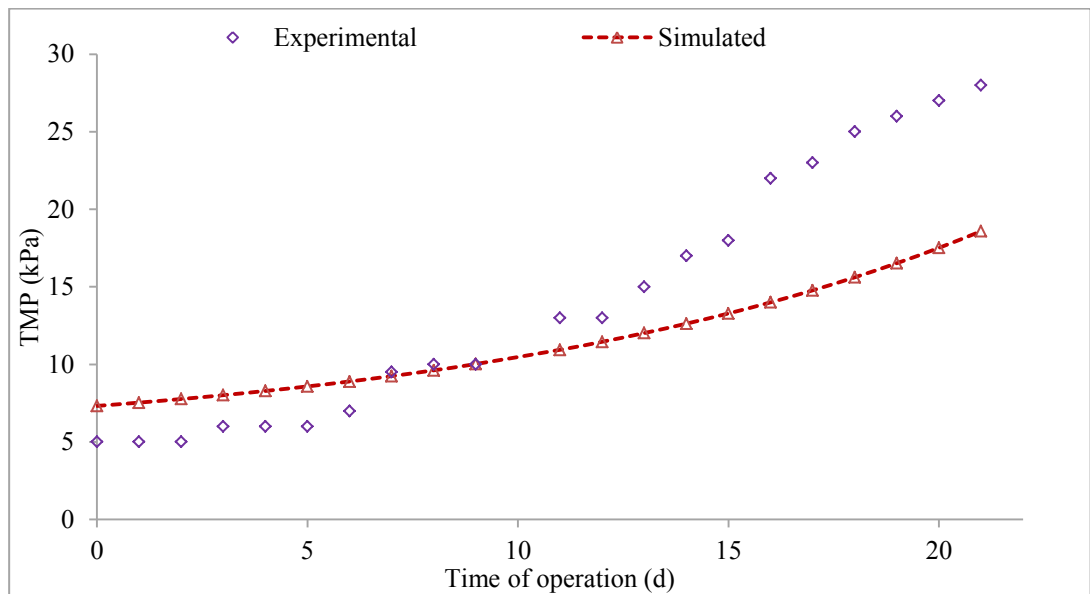
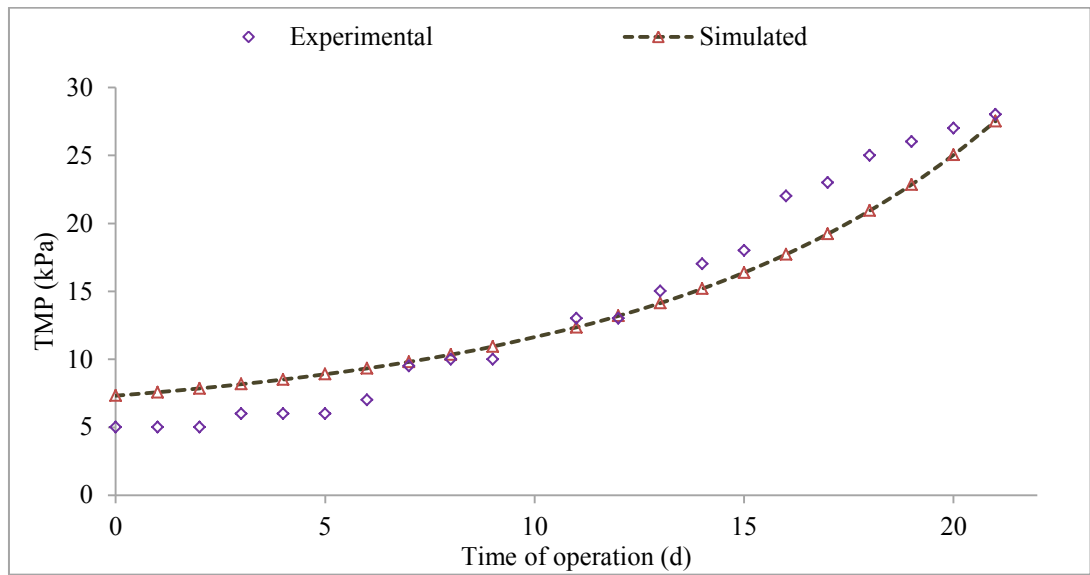


Figure 7.8 Comparison of simulated TMP with experimental TMP of the CMBR

From the experimental observation for the CMBR, the exponential rise of TMP cannot be simulated well (Figure 7.8) without changing the exponential parameter ( $n_c$ ) for the cake layer resistance ( $R_c$ ). The fundamental difference between the operation of CMBR and SSMBR system is that the sponges in the bioreactor of a SSMBR act against the cake layer growth and hence reduce the exponential rise of TMP due to the formation of the cake layer. The fact is also evident in the model simulation results of

the CMBR where the cake layer resistance ( $R_c$ ) cannot be simulated accurately with the same value of the coefficient  $n_c$  as used for the SSMBR ( $n_c = 0.065$ ). However, the cake layer resistance ( $R_c$ ) and hence the TMP in the CMBR can be simulated reasonably well by simply changing the value of  $n_c$  to 0.140 which accounts for more rapid rise of TMP in the CMBR. Figure 7.9 shows the model simulation results of TMP of the CMBR with the modified value of  $n_c = 0.140$  instead of 0.065.



*Figure 7.9* Comparison of simulated TMP with experimental TMP of the CMBR (with modified value of exponent coefficient  $n_c$  of the model)

## 7.5 Conclusion

A new and practical semi-empirical mathematical model of membrane fouling has been developed that accounts pore blocking and cake formation on membrane as the major processes of membrane fouling in an aerobic submerged MBR. Soluble microbial products (SMPs) are considered as the key components of pore fouling while mixed liquor suspended solids (MLSS) are assumed as contributors of foulants of cake layer including the biofilm. Constituent parameters of the model can be easily determined depending on the design and operating conditions of an MBR. The model requires input of the concentration of the cake forming particles (e.g. MLSS) and pore fouling



particles (e.g. soluble EPS) in bioreactor as best approximated empirical functions of time. The expression of dynamic cake layer formation in the mathematical model is based on net irreversible attachment of the cake layer on the membrane surface. A factor for the average rate of cake layer detachment is introduced in the model which is reasonably assumed to be constant over time for an MBR system run on continuous aeration with periodic backwashing. The pore fouling of the membrane is assumed to be dynamically built up due to the progressive reduction of the pore size and porosity of the membrane.

The simulation of the model for a laboratory scale submerged MBR system could predict reasonably well the development of transmembrane pressure at different days of operation of the MBR. The individual resistances due to the pore fouling and cake layer formation could also be predicted fairly accurately. With the incorporation of simplified parameters in the model, to account for the exponential increase of TMP, the simulated results showed better sensitivity while predicting fairly accurately the exponential rise of TMP, especially after the initial stage of operation of MBR. However, the calibrated values of the model coefficients may have dependency on the concentration of the cake/pore blocking particles in the suspension, which may also vary depending on the design and operating conditions of a particular MBR system. This phenomenon needs further verification of the mathematical model by operating the MBR system with different MLSS concentrations and at different operating conditions.



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## **CHAPTER 8**

# **Conclusions and Recommendations**

### 8.1 Summary of the major findings of the research

The major objective of the research is to develop simple but efficient mathematical models of biomass viability and membrane fouling that can be applied for integrated operational control of aerobic submerged MBR (SMBR) systems for treatability and fouling control. Based on the literature review and extended analytical study of state-of-the-art models, new conceptual models of biomass viability and fouling are proposed in Chapter 3. With a view to develop further improved mathematical models, experimental studies were conducted on two lab-scale SMBR systems and potential biomass parameters were analyzed for the effects on oxygen uptake of microorganisms and fouling resistances. The specific findings of this study are as follows:

- The parameters included in the conceptual model of biomass viability are the soluble and colloidal COD of the effluent along with the gross biomass parameters such as the concentrations of MLSS and MLVSS in the bioreactor. The easily measurable soluble and colloidal COD parameters indicate respectively the presence of soluble and colloidal (inert) microbial products in the bioreactor that affect biomass viability and membrane resistance.
- Empirical correlations may be established between the concentrations of soluble or colloidal COD of the effluent and the concentrations of SMP or bEPS in the bioreactor. It was shown from the analysis of test results in Chapter 5 that there existed correlations between the concentrations of soluble COD of the effluent and the concentrations of SMP in the bioreactor. Disregarding variable membrane rejection efficiency of soluble or colloidal compounds, the development of such correlations would help integrate models of biomass viability and membrane fouling for better operational control of MBR systems,

for example by controlling inhibitory effects of SMP or bEPS on biomass viability or membrane fouling.

- The proposed conceptual models may beneficially serve to estimate SMP/EPS concentration in the supernatant of the MBR avoiding their tedious and expensive measurement. Because of the common parameters in both the models, the proposed model of biomass viability can be easily linked to the model of membrane fouling.
- Analyses of experimental results expose that the soluble and colloidal microbial products had adverse effects on microbial oxygen uptake rate in the experimental SSMBR systems which as a consequence had effect of reducing membrane permeability as well. Some correlations and indicative parameters are suggested by means of which any instability of the bioprocess operation can be tracked, and remedial measures can be taken to avoid further problems or instability in the system's operation.
- The conceptual models do not incorporate the dynamic membrane rejection efficiency of soluble/colloidal particles and the rate of transport of these particles back into the bioreactor. Based on correlations of experimental data with parameters indicative of oxygen uptake rate and that of fouling resistances, improved models of biomass viability and fouling are developed and validated in Chapter 6 and Chapter 7 respectively. The formulation and validation of the model of biomass viability are based on the experimental results of an SSMBR for its operation of about 50 days. One of the critical findings from the analysis of test results is that the bEPS, SMP, MLSS and MLVSS significantly affected the SOUR and their relative influence on SOUR was  $EPS > bEPS > SMP > MLVSS/MLSS$ . The progressive change of SMP and bEPS

within the bioreactor of the SSMBR consistently maintained exponentially inverse correlation with SOUR. Based on the statistical analysis of test results and trends of SOUR as affected by characteristic biomass parameters, two independent mathematical models were developed which express SOUR as a function of MLSS, MLVSS, bEPS and SMP. The EPS represented as a lumped parameter of bEPS and SMP in the model still showed good correlation with SOUR in the model simulations.

- The mathematical model of membrane fouling requires input of the concentration of the cake forming particles (e.g. MLSS) and pore fouling particles (e.g. SMP) in the bioreactor as best approximated empirical functions in time. With the inputs for dynamic variations of these parameters, the dynamic cake layer formation and the pore fouling can be determined easily by model simulations. The mathematical model simulation for the laboratory scale SMBR systems could predict reasonably well the development of transmembrane pressure (TMP) at different days of operation of the MBR. The model calibration was done using experimental data obtained from the lab-scale SSMBR, and further verification of the model was done for additional data of the SSMBR and that of a conventional SMBR. As new exponential terms are included in the model to account for the exponential increase of TMP, the simulated results showed better sensitivity while predicting fairly accurately the exponential rise of TMP especially after the initial stage of operation of the MBR systems. The individual resistances due to the pore fouling and cake layer formation could also be predicted reasonably well. The major application of this fouling model is tracking any instability in the operation of an MBR system by

the rise of TMP, and the model will help devise operational control, for example by changing the concentration of MLSS to minimize the fouling resistances.

## **8.2 Future perspectives**

The drawbacks of the bioprocess operations that affect the biomass viability have complex correlations with the process of membrane fouling as well. Moreover, all the bioprocesses that operate on bio-oxidation kinetics are more or less linked to biomass viability and subsequently to membrane fouling processes. This means very high complexity in the formulation of a complete mathematical model of biomass viability and fouling, both in terms of over parameterization in the models and the formulations of complex correlations among the parameters. In this regard, the major outcome of the research is the development of simple but efficient empirical models of biomass viability and fouling consisting of major biomass compounds that inhibit the biomass viability and contribute to the fouling resistances. The application of these models will easily help operational control by changing the gross biomass parameters such as the concentration of MLSS which subsequently will affect the concentration of SMP or bEPS to minimize their effects on biomass viability and fouling.

However, the calibrated values of the model coefficients may have a dependency on the concentration of the cake/pore blocking particles in the suspension, which according to the formulation of the models, will vary depending on the design and operating conditions of a particular MBR system. This phenomenon needs further verification of the mathematical models by operating the MBR system with slightly different MLSS concentrations and at different operating conditions such as different aeration rates and backwashing frequency.

The initial concentration of MLSS, the rate of aeration and backwashing frequency in the MBR systems are usually maintained at or around optimum values depending on

the bioreactor volume, economic considerations and type of wastewater being treated by the MBR systems. When the bioreactor volume is changed to cope with the practical volume of wastewater to be treated, these design and operational requirements of the MBR will be changed as well. In order to find the calibrated values of model coefficients for future design changes, the application of the proposed models are recommended in different bioreactor volumes with different design and operational conditions of the SMBR systems.

The mathematical modelling study was conducted to characterize the biomass viability and fouling in a conventional lab-scale aerobic SMBR system. The disturbances due to temperature fluctuations and other environmental disturbances were strictly minimized to avoid their effects on biomass viability. However, practical MBR systems are exposed to these environmental disturbances. Consequently, calibration of models is suggested for experimental data of SMBR operated at different temperature conditions or other variable environmental settings.

Last but not least, the pore fouling resistances due to the soluble products of wastewater largely depend on the mean pore size of the membrane and also on the porosity of the membrane. The efficiency of an MBR system may be considerably increased with careful selection of membranes to minimize the pore fouling resistances. It is recommended that the modelling and the calibration protocol, as proposed in the study, is followed while choosing appropriate membrane sizes to minimize pore fouling resistances.



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# **Appendix**

## Appendix 1

### Program and output of simulations for SOUR model coefficients

#### *(a) Simulations considering bEPS and SMP*

##### Program

% objective function or targeted function

```
modelfun = @(a,x)(a(1) + exp(a(2) + a(3)*x(:,1)+a(4)*x(:,2)+ a(5)*x(:,3)));
```

% a(1), a(2), a(3), a(4) and a(5) are the coefficients, x(:,1) is MLVSS/MLSS (experimental); x(:,2) is bEPS<sub>i</sub>/bEPS<sub>0</sub> (experimental); x(:,3) is SMP<sub>i</sub>/SMP<sub>0</sub> (experimental); y(:,1) is SOUR (experimental);

```
Acc_Sponge_5gL = ...
```

```
[1 0.81300813 1 1 4.56165
```

```
2 0.827586207 1.104166667 1.148691768 4.02
```

```
5 0.83943662 1.4375 1.403956605 3.297010842
```

```
6 0.866498741 2.5 1.786853861 2.825570878
```

```
8 0.880952381 2.8125 2.042118698 2.475283544
```

```
11 0.86557377 3.25 1.723037652 2.34890417
```

```
13 0.88173913 2.4375 1.91448628 2.390142648
```

```
14 0.874576271 2.625 1.786853861 2.348454113
```

```
15 0.886855241 3.125 2.361199745 1.956304411
```

```
16 0.875404531 3.5625 2.488832163 1.927375695
```

```
20 0.853448276 3.041666667 2.233567326 2.368371818
```

```
21 0.844192635 3.125 1.91448628 2.360424262];
```

```
X1(:,1) = Acc_Sponge_5gL(:,2);% gives 'MLVSS/MLSS'
```

```
X1(:,2) = Acc_Sponge_5gL(:,3);% gives 'bEPSi/bEPS0'
```

```
X1(:,3) = Acc_Sponge_5gL(:,4); % gives 'SMPi/SMP0'
Y1(:,1) = Acc_Sponge_5gL(:,5); % gives 'SOUR'
beta0 = [.1 .1 .1 .1 .1];
mdl = fitnlm(X1,Y1,modelfun,beta0)
```

### Output

```
>> mdl =
```

Nonlinear regression model:

$$y \sim (a1 + \exp(a2 + a3*x1 + a4*x2 + a5*x3))$$

Estimated Coefficients:

	Estimate	SE	tStat	pValue
	-----	-----	-----	-----
a1	1.3121	0.64753	2.0263	0.082361
a2	5.6276	2.4761	2.2728	0.057238
a3	-4.6538	3.1924	-1.4578	0.18826
a4	-0.22043	0.14175	-1.5551	0.16388
a5	-0.46254	0.3928	-1.1775	0.27746

Number of observations: 12, Error degrees of freedom: 7

Root Mean Squared Error: 0.165

R-Squared: 0.974, Adjusted R-Squared 0.959

F-statistic vs. constant model: 65.4, p-value = 1.26e-05

### ***(b) Simulations considering $EPS = bEPS + SMP$***

#### Program

% objective function or targeted function

```
modelfun = @(a,x)(a(1) + exp(a(2) + a(3)*x(:,1)+a(4)*x(:,2)));
```

```

% a(1), a(2), a(3) and a(4) are the coefficients,
% x(:,1) is MLVSS/MLSS (experimental), and x(:,2) is EPSi/EPS0 (experimental)
% y(:,1) is SOUR (experimental)

Acc_Sponge_5gL = ...
[1 0.81300813 1 4.56165
2 0.827586207 1.126197242 4.02
5 0.83943662 1.420903063 3.297010842
6 0.866498741 2.147142406 2.825570878
8 0.880952381 2.431323019 2.475283544
11 0.86557377 2.494474266 2.34890417
13 0.88173913 2.17871803 2.390142648
14 0.874576271 2.210293653 2.348454113
15 0.886855241 2.747079255 1.956304411
16 0.875404531 3.031259867 1.927375695
20 0.853448276 2.641827176 2.368371818
21 0.844192635 2.526049889 2.360424262];

X1(:,1) = Acc_Sponge_5gL(:,2); % gives 'MLVSS/MLSS'
X1(:,2) = Acc_Sponge_5gL(:,3); % gives 'EPSi/EPS0'
Y1(:,1) = Acc_Sponge_5gL(:,4); % gives 'SOUR'

beta0 = [.1 .1 .1 .1]; % initialization of coefficients

mdl = fitnlm(X1,Y1,modelfun,beta0)

```

### Output

```
>> mdl =
```

Nonlinear regression model:

$$y \sim (a_1 + \exp(a_2 + a_3 * x_1 + a_4 * x_2))$$

## Estimated Coefficients:

	Estimate	SE	tStat	pValue
a1	1.2012	0.73964	1.624	0.14302
a2	6.0259	2.2713	2.653	0.029118
a3	-5.3009	2.8398	-1.8667	0.098915
a4	-0.5283	0.27301	-1.9351	0.089022

Number of observations: 12, Error degrees of freedom: 8

Root Mean Squared Error: 0.159

R-Squared: 0.972, Adjusted R-Squared 0.962

F-statistic vs. constant model: 93.9, p-value = 1.41e-06

## Appendix 2

### *(a) Program and output of simulation for TMP with six unknown parameters*

#### Program

```

modelfun = @(a,x)(3.33967e-9.*(7.43e11+0.00112.*exp(a(1).*x(:,1))./
    ((-1.42626e-7.*x(:,2)+a(2)).*(-4.13835e-11.*x(:,2)+a(3)).^2)+
    ((1-a(4))*exp(a(5).*x(:,1)).*(0.256.*(x(:,1)).^2+
    5.6005.*x(:,1)+a(6))).*3.33e8));

```

% a(1) is the unknown exponential coefficient for pore fouling resistance

% a(2) is the unknown porosity and a(3) is the unknown mean pore size of membrane

% a(4) is a coefficient for the rate of cake layer detachment by aeration

% a(5) is the unknown exponential coefficient for cake layer resistance

% a(6) is the integration constant in equation of cake layer resistance,

% and x(:,1) is day, and y(:,1) is the TMP (experimental)

Bioreactor = ...

[2 2 2.3537 4250

7 7 11.0563 5000

15 15 28.66122 5250

19 19 38.41524 5500

20 20 40.93608 6000

21 21 43.48725 7250

24 24 51.3121 7750

25 25 53.97436 8500

27 27 59.37489 10250

29 29 64.8720 12000];

```

X1(:,1) = Bioreactor(:,2); % gives 'day'
X1(:,2) = Bioreactor(:,3); % gives 'day^1.2391 as required by equation'
Y1(:,1) = Bioreactor(:,4); % gives 'TMP_experimental (Pa)'
beta0 = [0.1 0.10 0.0000001 0.85 0.07 300]; % initialization of coefficients'
mdl = fitnlm(X1,Y1,modelfun,beta0)

```

### Output

```
>> mdl =
```

Nonlinear regression model:

$$y \sim F(a,x)$$

Estimated Coefficients:

	Estimate	SE	tStat	pValue
a1	0.075236	0.00050477	149.05	4.5921e-15
a2	0.073989	7.8491e-06	9426.4	1.7966e-29
a3	9.4254e-08	9.6603e-10	97.568	1.3597e-13
a4	44.647	2.3842e-08	1.8726e+09	7.407e-72
a5	0.023446	0.00039561	59.265	7.2993e-12
a6	79.643	8.2865e-08	9.6111e+08	1.5382e-69

Number of observations: 10, Error degrees of freedom: 8

Root Mean Squared Error: 345

R-Squared: 0.983, Adjusted R-Squared 0.981

F-statistic vs. zero model: 2.4e+03, p-value = 7.72e-12



***(b) Program and output of simulation for pore resistance (exponential) coefficient***Program (for porosity 7%)

% objective function or targeted function

```
modelfun = @(a,x)(0.00112.*exp(a(1).*x(:,1))./((-1.42626e-7.*x(:,2)+0.07).*
    (-4.13835e-11.*x(:,2)+0.0000005).^2));
```

% a(1) is the unknown coefficient,

% and x(:,1) is day, x(:,2) is day<sup>1.2391</sup> (as required by equation) and

% y(:,1) is the pore resistance  $R_p$  (experimental)

Bioreactor = ...

```
[0 0 0 .35e11
```

```
49 49 124.2573 3.5e12];
```

```
X1(:,1) = Bioreactor(:,2); % gives 'day'
```

```
X1(:,2) = Bioreactor(:,3); % gives 'day1.2391'
```

```
Y1(:,1) = Bioreactor(:,4); % gives 'pore resistance(experimental)'
```

```
beta0 = 0.08; % initialization of the coefficient
```

```
mdl = fitnlm(X1,Y1,modelfun,beta0)
```

Output (for porosity 7%)

```
>> mdl =
```

Nonlinear regression model:

$y \sim F(a,x)$

Estimated Coefficients:

	Estimate	SE	tStat	pValue
a1	0.081239	0.00016909	480.44	0.0013251

Number of observations: 2, Error degrees of freedom: 1

Root Mean Squared Error: 2.9e+10

R-Squared: 1, Adjusted R-Squared 1

F-statistic vs. zero model: 1.46e+04, p-value = 0.00527

Program (for porosity 10%)

% objective function or targeted function

```
modelfun = @(a,x)(0.00112.*exp(a(1).*x(:,1))./((-1.42626e-7.*x(:,2)+0.10).*
    (-4.13835e-11.*x(:,2)+0.0000005).^2));
```

% a(1) is the unknown coefficient,

% x(:,1) is day, x(:,2) is day<sup>1.2391</sup> (as required by equation) and

% y(:,1) is the pore resistance  $R_p$  (experimental)

Bioreactor = ...

```
[0 0 0 .35e11
```

```
49 49 124.2573 3.5e12];
```

```
X1(:,1) = Bioreactor(:,2); % gives 'day'
```

```
X1(:,2) = Bioreactor(:,3); % gives 'day1.2391'
```

```
Y1(:,1) = Bioreactor(:,4); % gives 'pore resistance(experimental)'
```

```
beta0 = 0.08; % initialization of the coefficient
```

```
mdl = fitnlm(X1,Y1,modelfun,beta0)
```

Output (for porosity 10%)

```
>> mdl =
```

Nonlinear regression model:

$$y \sim F(a,x)$$

Estimated Coefficients:

	Estimate	SE	tStat	pValue
a1	0.08852	5.7142e-05	1549.1	0.00041096

Number of observations: 2, Error degrees of freedom: 1

Root Mean Squared Error: 9.8e+09

R-Squared: 1, Adjusted R-Squared 1

F-statistic vs. zero model: 1.28e+05, p-value = 0.00178

Program (for porosity 15%)

% objective function or targeted function

```
modelfun = @(a,x)(0.00112.*exp(a(1).*x(:,1))./((-1.42626e-7.*x(:,2)+0.15).*
(-4.13835e-11.*x(:,2)+0.0000005).^2));
```

% a(1) is the unknown coefficient,

% x(:,1) is day, x(:,2) is day<sup>1.2391</sup> (as required by equation) and

% y(:,1) is the pore resistance  $R_p$  (experimental)

Bioreactor = ...

```
[0 0 0 .35e11
```

```
49 49 124.2573 3.5e12];
```

```
X1(:,1) = Bioreactor(:,2); % gives 'day'
```

```
X1(:,2) = Bioreactor(:,3); % gives 'day1.2391'
```

```
Y1(:,1) = Bioreactor(:,4); % gives 'pore resistance (experimental)'
```

```
beta0 = 0.08; % initialization of the coefficient
```

```
mdl = fitnlm(X1,Y1,modelfun,beta0)
```

Output (for porosity 15%)

```
>> mdl =
```

Nonlinear regression model:

$$y \sim F(a,x)$$

Estimated Coefficients:

	Estimate	SE	tStat	pValue
a1	0.096796	2.9932e-05	3233.9	0.00019686

Number of observations: 2, Error degrees of freedom: 1

Root Mean Squared Error: 5.13e+09

R-Squared: 1, Adjusted R-Squared 1

F-statistic vs. zero model: 4.65e+05, p-value = 0.000934

***(c) Program and output of simulation for sludge cake resistance (exponential)  
coefficient and constant***

Program (for porosity 15%)

**% objective function or targeted function**

```
modelfun = @(a,x)(3.33967e-9.*(7.43e11+0.00112.*exp(0.096796.*x(:,1)))/
    ((-1.42626e-7.*x(:,2)+0.15).*(-4.13835e-11.*x(:,2)+0.0000005).^2)
    +((1-a(1))*exp(a(2).*x(:,1)).*(0.256.*(x(:,1)).^2
    +5.6005.*x(:,1)+a(3))).*3.33e8));
```

**% coefficient and constant determined from the equation of TMP or total resistance**

**(using the pore resistance coefficient value already determined for porosity 15%)**

**% a(1) is the unknown coefficient for cake layer detachment**

**% a(2) is a coefficient at exponent in equation of cake layer resistance**

```
% a(3) is the integration constant in equation of cake layer resistance,
% Equations solved for the total TMP (experimental)
% x(:,1) is day, x(:,2) is day^1.2391 and y(:,1) is the TMP experimental (Pa)
```

```
Bioreactor = ...
```

```
[2 2 2.3537 4250
```

```
7 7 11.0563 5000
```

```
15 15 28.66122 5250
```

```
19 19 38.41524 5500
```

```
20 20 40.93608 6000
```

```
21 21 43.48725 7250
```

```
24 24 51.3121 7750
```

```
25 25 53.97436 8500
```

```
27 27 59.37489 10250
```

```
29 29 64.8720 12000];
```

```
X1(:,1) = Bioreactor(:,2); % gives 'day'
```

```
X1(:,2) = Bioreactor(:,3); % gives 'day^1.2391'
```

```
Y1(:,1) = Bioreactor(:,4); % gives 'TMP(expt)'
```

```
beta0 = [0.05 0.08 100]; % initialization of coefficients
```

```
mdl = fitnlm(X1,Y1,modelfun,beta0)
```

*Output (for porosity 15%)*

```
>> mdl =
```

Nonlinear regression model:

$$y \sim F(a,x)$$

Estimated Coefficients:

	Estimate	SE	tStat	pValue
a1	0.025044	53.093	0.0004717	0.99964
a2	0.065003	0.71505	0.090906	0.93011
a3	644.97	34544	0.018671	0.98562

Number of observations: 10, Error degrees of freedom: 7

Root Mean Squared Error: 731

R-Squared: 0.933, Adjusted R-Squared 0.914

F-statistic vs. zero model: 354, p-value = 5.29e-08