



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

By

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A Dissertation

Submitted in fulfilment for the degree of

DOCTOR OF PHILOSOPHY

In

Faculty of Environmental and Information Engineering

University of Technology, Sydney

New South Wales, Australia

December 2015

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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ACKNOWLEDGEMENTS

It is my great pleasure to express my sincere gratitude to my supervisors, Prof. Huu Hao Ngo and Dr Wenshan Guo. As my principal supervisor, Prof. Ngo has been supporting me since the beginning of my PhD study by giving me the wonderful research topic and providing his research knowledge and experience. Without his invaluable guidance and supports, I will not be able to finish my study within the scheduled time. He has always inspired me whenever I confronted with difficulties and lost my confidence. In addition, he has also helped me to overcome my drawbacks and limitations in English writing and research approach. Although Dr Wenshan Guo is my alternative supervisor, the time and the efforts she has dedicated to my study is equal to that done by Prof. Ngo. Dr Wenshan Guo trained me for the experimental-setup and the sample analyses. She consistently encouraged me with her great patience. She always provided me with very helpful suggestions and advice for my experiments. Moreover, her great helps in revising the papers as well as improving my English writing to a better level with logical and systematic manner.

I would also like to thank the joint University of Technology Sydney–China Scholarship Council (UTS–CSC) Doctor of Philosophy (Ph.D.) Scholarship for supporting my study during my stay in Australia. Besides, I am also deeply grateful for the financial support from Prof. Ngo’s MBR research project, which covered the expenses of consumables for my experiments.

My earnest thanks go to M.E. Johir, who is UTS Environmental Engineering Laboratories Manager, for his kindly assistance on the operating of the analysis instruments. I am very grateful to Tram for her sincere help with binding this thesis.

Finally, I would like to expand my gratitude to my parents and friends who were very considerate of me and gave me their love and encouragement throughout my entire PhD study.

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Abbreviations

Symbol	Description
AC	Acylase
AFM	Atomic force microscopy
AnMBR	Anaerobic membrane bioreactor
AOB	Ammonium oxidizing bacteria
AOMBR	Anaerobic-oxic membrane bioreactor
A/O	Anoxic/oxic
A²/O	Anaerobic/anoxic/oxic
BAP	Biomass-associated products
BNR	Biological nutrient removal
BOD	Biological oxygen demand
BPC	Biopolymer clusters
BPC_C	Polysaccharides in biopolymer clusters
BPC_P	Proteins in biopolymer clusters
CA	Cellulose acetate
CEB	Chemical enhanced backwashing
CEBs	Cell entrapping beads
CER	Cation exchange resin
CF	Concentration factor
CIOF	Combined inorganic-organic flocculant
CIP	Cleaning in place
CMBR	Conventional membrane bioreactor
COD	Chemical oxygen demand
C/N or COD/N	Carbon to nitrogen ratio
C/P	Carbon to phosphorus ratio
CST	Capillary suction time
DC	Direct current
DO	Dissolved oxygen
DOC	Dissolved organic carbon
DOC'	Deoxycholate
DOM	Dissolved organic matter

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

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

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

Nomenclatures

Symbol	Description
ADIPAP KD 452 or KD452	Cationic polymers
$\text{Al}_2(\text{SO}_4)_3$	Aluminium sulfate
$\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$	Calcium chloride
C_c	Concentration of potential cake forming particles in the bulk liquid (e.g. MLSS) which typically varies over time in membrane bioreactor
CGMS	Modified corn starch
$\text{C}_6\text{H}_{12}\text{O}_6$	Glucose
CO_2	Carbon dioxide
$\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$	Cobalt chloride
CPE	Organic cationic polyelectrolyte
C_{SMP}	Time-dependent concentration of soluble particles entering the pores
$\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$	Cupric sulphate
$d_{p,\text{used}}$	The initial pore diameter of the membrane in μm
$d\text{TMP}/dt$	Membrane fouling rate
f	The membrane's porosity
FeCl_3	Ferric chloride or Ferric chloride anhydrous
Fe-MBR	Electro-membrane bioreactor with stainless steel mesh as the anode
h_c	Variable depth of the cake layer expressed as a first order differential function in time, which relies on the attachment and detachment of cake layer
h_m	The membrane's effective thickness
H_2O_2	Hydrogen peroxide
H_2SO_4	Sulphuric acid
J	The permeate flux
k	The factor representing the detachment of the cake layer from the membrane surface
kDa	Unified atomic mass unit

K₂Cr₂O₇	Potassium dichromate
KH₂PO₄	Potassium phosphate
MLSS_e	The MLSS concentration in the aqueous phase after emulsification
MLSS_i	The initial MLSS concentration of the mixed liquor sample
m_{d,o}	Outer membrane diameter
m_{d,i}	Inner membrane diameter
Mg	Magnesium
MGMS	Modified corn starch
MgSO₄·7H₂O	Magnesium sulphate
MPE	Organic flocculant
MPE50	Organic flocculant (cationic polymers)
MPL30	Cationic polymers
MnCl₂·7H₂O	Manganese chloride
NALCO MPE50	Cationic polymers
n_c	Cake fouling factor to explain the typically observed exponential rise of TMP due to the cake layer resistance especially at the final stage of operation of an MBR system
n_p	Pore fouling factor to explain the typically observed exponential rise of TMP due to the pore fouling resistance especially at the final stage of operation of an MBR system
NaCl	Sodium chlorite
NaClO	Sodium hypochlorite
NaHCO₃	Sodium hydrogen carbonate
Na₂MoO₄·2H₂O	Sodium molybdate dehydrate
NH₂OH	Hydroxylamine
NH₄-N or NH₄⁺	Ammonia nitrogen
(NH₄)₂SO₄	Ammonium sulphate
NO₂-N or NO₂⁻	Nitrite
NO	Nitric oxide
N₂	Nitrogen gas
N₂O	Nitrous oxide
NO₃-N or NO₃⁻	Nitrate
PAM-MGMS	Polyacrylamide-starch composite flocculant

pH	Power of hydrogen or potential hydrogen
PM30	A kind of polyethersulfone membrane
PO₄-P	Orthophosphate
Poly-1	Cationic polymers
Poly-2	Modified cationic polymers
r_p	Membrane pore radius
R80	Membrane bioreactor with 80 nm pore-sized ceramic membrane
R100	Membrane bioreactor with 100 nm pore-sized ceramic membrane
R200	Membrane bioreactor with 200 nm pore-sized ceramic membrane
R300	Membrane bioreactor with 300 nm pore-sized ceramic membrane
R²	Squared value of correlation coefficient
R_C	Cake layer resistance
R_M	Intrinsic membrane resistance
R_{IR}	Irreversible fouling resistance
R_P	Pore blocking resistance
R_T	Total fouling resistance
SMP_{total}	Total soluble microbial products
t	The filtration time
T	Temperature
TATE & LYLE Mylbond 168	Starch
Ti-MBR	Electro-membrane bioreactor with titanium anodes
TN	Total nitrogen
ZnSO₄·7H₂O	Zinc sulphate
x	Independent variable
y	Variable
ΔMLSS/Δt	Biomass growth rate
ΔP	Transmembrane pressure gradient
ΔTMP/Δt	Membrane fouling rate
YW 30	A kind of regenerated cellulose membrane

Greek symbols

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

PhD DISSERTATION ABSTRACT

Author: Lijuan Deng
Date: 18 December 2015
Thesis title: Development of specific integrated membrane bioreactors (MBRs) for membrane fouling control during wastewater treatment for reuse
Faculty: Environmental and Information Technology
School: Civil and Environmental Engineering
Supervisors: Prof. Dr. Huu Hao Ngo (Principal supervisor)
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Abstract

In recent years, membrane fouling has become a critical issue of membrane bioreactor (MBR) in wastewater treatment. To resolve this obstacle, introducing biomass carriers or flocculants into submerged MBR (SMBR) has become one of the effective technologies for membrane fouling control. This study aims to provide an in-depth analysis on membrane fouling behaviour in SMBRs with sponge and/or the patented green bioflocculant by considering the properties of activated sludge and cake layer. A new functional media (sponge modified plastic carrier) was also developed to enhance the performance of integrated moving bed biofilm reactor-membrane bioreactor (MBBR-MBR) systems. The results suggested that sponge addition in a SMBR (SSMBR) or bioflocculant addition in a SMBR (MBR-G) reduced cake layer formation and limited pore blocking, thus effectively minimizing membrane fouling. Better sludge characteristics were obtained in both of the SSMBR and the MBR-G due to less soluble microbial products (SMP), lower biomass growth and sludge viscosity, higher protein to polysaccharide ratio in extracellular polymeric substances, higher zeta potential, greater relative hydrophobicity, larger floc size and better flocculation ability. The presence of sponge or bioflocculant in the SMBR also eliminated extracellular polymeric substances (EPS), SMP and/or biopolymer clusters (BPC) on membrane surface. Consequently, cake layer (R_C) and pore blocking resistance (R_P) were decreased in the SSMBR and the MBR-G. A modified resistance-in-series model

proposed for the SMBR with and without biofloculant could quantitatively demonstrate the impacts of sludge characteristics on membrane fouling. In the SSMBR, a longer hydraulic retention time (HRT) of 6.67 h permitted more considerably fouling reduction comparing to shorter HRTs (5.33 and 4.00 h). Moreover, lower R_p and R_c at the prolonged HRT were mainly ascribed to the elevated protein to polysaccharide ratio in SMP (SMP_p/SMP_c) of mixed liquor, together with the declined EPS and BPC in cake layer. SMP was not the primary membrane foulant when the SSMBRs were operated at different HRTs. Biofloculant addition at the optimum HRT of 6.67 h further mitigated fouling in the SSMBR by improving activated sludge and cake layer characteristics. The integrated MBBR-MBR with the sponge modified plastic carriers showed better removal of DOC, NH_4-N , T-N and PO_4-P than the MBBR-MBR with plastic carriers only. Furthermore, the sponge modified plastic carriers also eliminated SMP of mixed liquor, and reduced SMP and BPC on membrane surface, which ameliorated membrane fouling, R_p and R_c as compared to the plastic carriers.

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

