



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

Cheng Chen

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In

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CERTIFICATION OF ORIGINAL AUTHORSHIP

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

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

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LIST OF ABBREVIATIONS

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

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

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

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

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

LIST OF SYMBOLS

B - A	The number of days that the system is operated
C ₂	Acetic acid
C ₃	Propionic acid
C ₄	Butyric acid
C ₅	Valeric acid
Ca	Calcium
CaCl ₂ ·2H ₂ O	Calcium chloride
CH ₄	Methane
C ₆ H ₁₂ O ₆	Glucose
Co	Cobalt
CoCl ₂	Cobalt chloride
CO ₂	Carbon dioxide
CuSO ₄ ·5H ₂ O	Cupric sulphate
Fe	Iron
Fe ²⁺	Ironized ferrous
FeCl ₃	Ferric chloride anhydrous
FeSO ₄	Ferrous sulfate
H ₂	Hydrogen
H ₂ S	Hydrogen sulfide
H ₂ SO ₄	Hydrogen sulfate
i-C ₄	Iso-butyric acid
i-C ₅	Iso-valeric acid
IC ₅₀	50% activity inhibitory concentration
J	Permeation flux
KH ₂ PO ₄	Potassium phosphate
Mg	Magnesium
MgSO ₄ ·7H ₂ O	Magnesium sulphate
MLSS _A	MLSS concentration of granular sludge when operation is started
MLSS _{A1}	MLSS concentration of sponge-attached biosolids when operation is initiated

MLSS _B	MLSS concentration of granular sludge when operation is terminated
MLSS _{B1}	MLSS concentration of biosolids attached on the sponge when operation is terminated
MnCl ₂ ·7H ₂ O	Manganese chloride
N ₂	Nitrogen gas
Na ⁺	Ionized sodium
NaCl	Sodium chloride
NaClO	Sodium hypochlorite
NaHCO ₃	Sodium biocarbonate
Na ₂ MoO ₄ ·2H ₂ O	Sodium molybdate dehydrate
NaOH	Sodium hydroxide
(NH ₄) ₂ SO ₄	Ammonium sulphate
NH ₃	Free ammonia
NH ₄ ⁺	Ionized ammonia
NH ₄ -N	Ammonia nitrogen
Ni	Nickel
NiSO ₄	Nickel sulphate
NO ₂ -N	Nitrite nitrogen
NO ₃ -N	Nitrate nitrogen
PO ₄ -P	Orthophosphate
R _C	Cake layer resistance
R _M	Intrinsic membrane resistance
R _P	Pore blocking resistance
R _T	Total resistance
SO ₄ -S	Sulfate sulfur
SO ₄ ²⁻	Ionized sulfate
Sw	The weight of sponge
TMP _A	The transmembrane pressure value (kPa) when operation is started
TMP _B	The transmembrane pressure value (kPa) when operation is terminated
μ	Dynamic viscosity of the permeate
U _v	Upflow velocity
V _T	Total volatile fatty acids

$\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$	Zinc sulphate
$\Delta\text{MLSS}/\Delta t$	Biomass growth rate
$\Delta\text{MLSS}/S_w$	Attached biomass growth rate
ΔP	Transmembrane pressure
$\Delta\text{TMP}/\Delta t$	Membrane fouling rate

PhD DISSERTATION ABSTRACT

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Faculty: FEIT

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Supervisors: Dr. Wenshan Guo (Principal supervisor)
Dr. Huu Hao Ngo (Alternative supervisor)

Abstract

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

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

settleability. The EG-AnMBR demonstrated less SMP and EPS in the mixed liquor and cake layer, which might reduce the cake layer resistance and lower fouling rate.

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

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

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