UNIVERSITY
OF TECHNOLOGY
SYDNEY

Addition of powdered activated carbon to anaerobic membrane bioreactor for fouling control

by Weonjung Sohn

Thesis submitted in fulfilment of the requirements for the degree of

Master of Engineering

under the supervision of Prof. Wenshan Guo
and the co-supervision of Prof. Huu Hao Ngo

University of Technology Sydney
Faculty of Engineering and Information Technology

June 2021

CERTIFICATION OF ORIGINAL AUTHORSHIP

I, Weonjung Sohn declare that this thesis, is submitted in fulfilment of the requirements for the award of Master of Engineering, in the School of Civil and Environmental Engineering/Faculty of Engineering and Information Technology at the University of Technology Sydney.

This thesis is wholly my own work unless otherwise referenced or acknowledged. In addition, I certify that all information sources and literature used are indicated in the thesis.

This document has not been submitted for qualifications at any other academic institution.

This research is supported by the Australian Government Research Training Program.

Production Note:
Signature: Signature removed prior to publication.

Date: 08. 06. 2021

ACKNOWLEDGEMENTS

I would like to express my deepest gratitude to my principle supervisor Prof. Wenshan Guo for her patient guidance, and endless support. She also provided constant encouragement and mentorship for me all the way from the beginning of my master degree to the end of this degree. It was my pleasure to study under her supervision. I would like to appreciate my co-supervisor Prof. Huu Hao Ngo for his valuable encouragement and comments. I feel very fortunate to have been in his research group. I would like to extend my gratitude to my groupmates who always gave me their kindness, and especially to Dora for her great help to my experiment. Apart from my groupmates, I appreciate Allie for her great help in microbial community study, and Seongchul Ryu for his valuable life advice and help whenever I had difficulties. I am very grateful to my friends in Sydney who always support and encourage me. I was not able to successfully finish my study without their help to getting through the tough times during my study period. Finally, I would like to express my sincere gratitude to my family for financial support and endless love during my whole study time.

It was a big challenge for me to start an experiment, organizing data, and writing this master thesis. However, I was able to complete my master thesis as a result with my supervisor Prof. Wenshan guo's constant guidance.

TABLE OF CONTENTS

CHAPTER 1 Introduction

CHAPTER 2 Literature review

CHAPTER 3 Experimental investigations

CHAPTER 4 Optimization of powdered activated carbon dosage to anaerobic membrane bioreactor

CHAPTER 5 Long term experimental investigation of powdered activated carbon addition to anaerobic membrane bioreactor

CHAPTER 6 Recommendations and conclusions

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

Symbol	**Description**
AnMBRs	Anaerobic membrane bioreactors
AnFMBR	Anaerobic fluidized membrane bioreactor
AnCMBR	Anaerobic ceramic membrane bioreactor
AC	Activated carbon
AFCMBR	Anaerobic fluidized bed ceramic membrane bioreactor
AgNPs	Silver nanoparticles
AOX	Adsorbable organic halogen
ASVs	Amplicon sequence variants
BAC	Biologically activated carbon
BC	Biochar
BOD_5	Biological oxygen demand
PAC	Powdered activated carbon
Cbz	Carbamazepine
COD	Chemical oxygen demand
CSTR	Continuously stirred tank reactor
Dcf	Diclofenac
DIET	Direct interspecies electron transfer
DOM	Dissolved organic matter
DOC	Dissolved organic carbon
EGSB	Expanded granular sludge bed
ENMs	Engineered nanomaterials
EPS	Extracellular polymeric substances
ETS	Erythromycin-tetracycline-sulfamethoxazole
FO	Forward osmosis
F/M ratio	Food to microorganisms ratio
GAC	Granular activated carbon
HRT	Hydraulic retention time
IAFMBR	Integrated anaerobic fluidized-bed membrane bioreactor
LCWW	Low-grade coal wastewater
LCFA	Long chain fatty acids

MBRs	Membrane bioreactors
MLSS	Mixed liquor suspended solids
MD	Membrane distillation
MF	Microfiltration
MW	Molecular weight
NF	Nanofiltration
NOM	Natural organic matter
OCPs	Organochloride pesticides
OLR	Organic loading rate
ORP	Oxidation-reduction potential
PAC	Powder activated carbon
PC-A	Polycitrate-Alumoxane
PE	Polyethylene
PET	Polyethylene terephthalate
PES	Polyethersulfone
PP	Polypropylene
PVA	Polyvinyl alcohol
PVDF	Polyvinylidene difluoride
PhACs	Pharmaceutically active compounds
PHAs	Polycyclic aromatic hydrocarbons
SAF-MBR	Staged anaerobic fluidized membrane bioreactor
SAF-CMBR	Staged anaerobic fluidized bed ceramic membrane bioreactor
SCFA	Short chain fatty acids
SS	Suspended solids
SRT	Solids retention time
SMs	Sulfonamide antibiotics
SMP	Soluble microbial product
SMX	Sulfamethoxazole
ST	Erythromycin-tetracycline
SMZ	Sulfamethazine
RO	Reverse osmosis
TC	Tetracycline
Tcs	Triclosan

TMP	Transmembrane pressure
Tmp	Trimethoprim
TOC	Total organic carbon
TS	Total solids
TP	Total phosphorous
UASB	Up-flow anaerobic sludge blanket
US CDC	United State Centre for Disease Control and Prevention
UF	Ultrafiltration
WWTPs	Wastewater treatment plants
VFAs	Volatile fatty acids
VSS	Volatile suspended solids
VS	Volatile solids
YW	Yeast wate
ZVI	Zero-valent iron

LIST OF SYMBOLS

Symbol	**Description**
$Al(OH)_3$	Aluminum hydroxide
$AlCl_3$	Aluminum chloride
$Al_2(SO_4)_3$	Aluminum sulfate
Al_2O_3	Aluminum oxide
$C_6H_{12}O_6$	Glucose
Ca^{2+}	Calcium
$CaCl_2$	Calcium chloride
CO_2	Carbon dioxide
CO	Carbon monoxide
$CuSO_4 \cdot 5H_2O$	Cupric sulphate
Fe	Iron
$FeCl_3$	Ferric chloride anhydrous
FeS	Ferrous sulfide
H^+	Proton
H_2	Hydrogen
H_2O	Water
H_2S	Hydrogen sulfide
H_2SO_4	Sulphuric acid
K^+	Potassium
KOH	Potassium hydroxide
KH_2PO_4	Potassium dihydrogen phosphate
LMH	$L/m^2 \cdot h$
Mg^{2+}	Magnesium
$MgSO_4 \cdot 7H_2O$	Magnesium sulphate
N_2	Nitrogen gas
Na^+	Sodium
NaClO	Sodium hypochlorite
$NaNO_3$	Sodium nitrate
$NaHCO_3$	Sodium bicarbonate

$NaOH$	Sodium hydroxide
NH_4^+	Ionized ammonia
NH_4^+-N	Ammonium nitrogen
NH_4Cl	Ammonium chloride
NO^{2-}	Nitrite
NO^{3-}	Nitrate
NO_2-N	Nitrite nitrogen
NO_3-N	Nitrate nitrogen
O_2	Oxygen gas
O_3	Ozone
OH^-	Hydroxyl
PO_4^{3-}-P	Hydrogen phosphate phosphorus
TiO_2	Titanium dioxide
ZrO_2	Zirconium dioxide
$ZnSO_4 \cdot 7H_2O$	Zinc sulphate

ABSTRACT

Author:	Weonjung Sohn
Date:	08. 06. 2021
Thesis title:	Addition of powdered activated carbon to anaerobic membrane bioreactor for fouling control
Faculty:	Faculty of Environmental and Information Technology
School:	Civil and Environmental Engineering
Supervisors:	Prof. Wenshan Guo (Principal supervisor)
	Prof. Huu Hao Ngo (Co-supervisor)

Abstract

Membrane fouling remains a challenging issue which is the main obstacle of wider applications of anaerobic membrane bioreactors (AnMBRs). This is because the cake layer in AnMBRs is thicker and less removable than that in aerobic membrane bioreactors due to the different sludge characteristics. Pretreatment of feed wastewater can efficiently control membrane fouling by altering the feed water properties with the addition of fouling reduction enhancers, such as adsorbents and flocculants. Activated carbon, such as powdered activated carbon (PAC), which is comparatively simple and effective adsorbent, has been largely applied as an adsorbent to aerobic membrane bioreactors for membrane fouling mitigation. However, only a limited amount of research has focused on the application of PAC to AnMBRs. Although some studies have reported the effects of PAC addition on pollutants removal performance and fouling control of AnMBR system, few studies discussed the overall nutrient and organics removal performance as well as detailed membrane fouling behaviour regarding PAC addition at an ambient temperature. In addition, no study has been carried out on the effect of PAC on microbial community in AnMBR. The long-term effects of the optimal PAC dosage, which was found to be 5 g/L from the short-term experiments, on conventional AnMBR performance with a hollow fiber membrane were investigated in this study. The optimal one-off PAC dosing could increase average COD and total organic carbon (TOC) removal rates up to 15.71% and 15.69%, respectively. The PAC addition exhibited not only lower transmembrane pressure (TMP) increase rate, but also reduction of soluble microbial product (SMP) and extracellular polymeric substances (EPS). High protein fraction of SMP as well as increased zeta potential in AnMBR with PAC addition could enhance hydrophobicity, which improved the flocculation ability of sludge. In addition, lower total membrane resistance and pore blocking resistance indicated that PAC addition could prevent both severe pore blocking and irreversible fouling, due to the lower amount of polysaccharide of SMP in cake layer. PAC addition could reduce the abundance of fouling-related bacteria such as *Cloacibacterium* and *Smithella*, which contribute to the biofouling development due to the high affinity to attach on membrane surface. This is because the high adsorption capacity of PAC could enabled 1.7-4.7 times lower abundance of those bacteria by inhibiting proliferation of them. These results demonstrated that PAC addition to AnMBR could effectively mitigate membrane pore blocking and irreversible membrane fouling as well as improve microbial community.

Keywords: Anaerobic membrane bioreactor; Membrane fouling; Enhancers; Powdered activated carbon

CHAPTER 1

Introduction

1.1 Background

1.1.1 The necessity of wastewater treatment

Water is the most essential part for life. However, because of the limited amount of water on the Earth, a great portion of the world's population is facing the huge threat of water scarcity. It is reported that almost 3.5 billion people will be living in water-scarce regions by 2025, while billions of people are already suffering from water shortage issue (Jo, 2017). Due to the fast population growth and the subsequent increase in demand for fresh water, research on alternative water sources is vital. In addition, while the quantity of water greatly contributes to water scarcity issue, the quality of water also significantly contributes to the water shortage. Along with the continuous use of surfactants, pesticides, and industrial chemicals, the emergence of contaminants such as dissolved organics, heavy metals, and pharmaceuticals has limited the reuse of wastewater. This water quality degradation has posed a challenge to the wastewater treatment technologies. Anaerobic membrane bioreactor has been proved its efficiency in achieving high-quality effluent with excellent removal of chemical oxygen demand (COD) and suspended solids (SS) enabling effective wastewater reuse. In addition, the effluent from AnMBR with mineralized ammonia and orthophophate facilitates its direct reuse for agricultural purpose (Ozgun et al., 2013). Thus, it is necessary to understand and apply adequate technologies for high-quality water reuse as well as the production of different water quality for various usage purposes.

1.1.2 Anaerobic membrane bioreactor

Anaerobic digestion, which consists of four steps of hydrolysis, acidogenesis, acetogenesis, and methanogenesis for biomass to convert into biogas, is one of the most commonly applied processes for wastewater treatment. Although it has some drawbacks such as sensitivity to temperature, it can offer energy recovery by the generation of methane as a final product, as well as hydrogen and volatile fatty acids (VFA) as intermediate products (Khan, 2019). An anaerobic membrane bioreactor (AnMBR) is a technology which integrates anaerobic digestion and membrane technology. It has been increasingly used due to several benefits over an aerobic membrane bioreactor, such as long solid retention time (SRT), low sludge production, and potential energy usage. While the anaerobic digestion requires a mesophilic range of temperature, AnMBR can be operated in lower temperatures by extension of SRT. Although anaerobic process has been mainly applied for the treatment of industrial and high strength wastewater, AnMBR technology can be favorable for the treatment of low strength organic loads such as municipal wastewater by achieving slow-growth biomass retention with short hydraulic retention time (HRT) (Lin et al., 2013a).

1.1.3 Membrane fouling

One of the biggest obstacles in both aerobic and anaerobic membrane bioreactors is membrane fouling, since it can deteriorate the reactor performance and shorten the lifespan of the membrane. The membrane fouling normally occurs due to the initial attachment of sludge components on the surface of membrane and the subsequent blockage of membrane pores and cake layer formation. Once soluble and particulate matter start to accumulate on the membrane surface and inside the pores, physicochemical and biological interactions occur between the membrane and foulants. Thus, membrane fouling should be controlled by regular physical and chemical membrane cleaning as well as membrane replacement, which accounts for a high amount of the operation costs. Membrane fouling can be categorized into three fouling types according to the different cleaning requirements, which are removable, irremovable, and irreversible fouling. It also can be divided into organic, inorganic, and biofouling according to the foulant components.

Membrane fouling can be mitigated by changing feed water properties which can influence the formation as well as the compactness of the cake layer. There are several different pretreatment methods, including alkaline, acid, ozone pretreatment, or the addition of fouling reduction enhancers. The addition of enhancers, such as adsorbents, carriers and other chemical agents can effectively alter feed water characteristics leading to significant alleviation of membrane fouling. Adsorbents, such as activated carbon and zeolite, are able to promote organic compounds to adsorb and heavy metal ions to ion-exchange. Coagulants including ferric chloride and polyaluminum chloride can promote coagulation, while suspended carriers can assist flocculation. In consequence, enlarged floc particle size and decreased soluble organic matter in the supernatant can alleviate membrane fouling (Drews, 2010; Lin et al., 2013a). Powdered activated carbons have been broadly applied in membrane bioreactors due to their high adsorption capability, improvement of biodegradation, and subsequent removal of refractory pollutants (Skouteris et al., 2015).

1.2 Objectives of the study

To date, there are a number of investigations of fouling mitigation effect in aerobic MBRs by adding powdered activate carbon (PAC) as an adsorbent. However, only a limited number of studies has been done to investigate the impact of PAC addition in AnMBRs. Moreover, although previous studies have investigated the effects of PAC addition into AnMBRs, they mainly focused on removal performance of particular substances (Baêta et al., 2016; Xiao et al., 2017). For example, one study only focused on the removal of organic compounds like dyes by adsorbing toxic byproducts like aromatic amines, while the other focused on pharmaceutical compounds like antibiotics. Several studies have reported membrane fouling control along with removal performances, however they only paid attention to parameters such as flux change, concentration of soluble microbial product (SMP) and extracellular polymeric substances (EPS) in mixed liquor, and sludge floc size. To the best of our knowledge, no previous research has examined the impact of PAC addition on microbial diversity in AnMBR as well. In addition, the currently available studies were carried out at around 35 °C in submerged AnMBRs, despite the operating temperature affecting membrane fouling aggravation. Studies have reported that excessive PAC dosage could have adverse results on fouling behaviour (Akram and Stuckey, 2008). Therefore, the optimization of dosage is necessary for the best removal performance as well as fouling control before conducting a long term experiment. Hence, this study firstly examined the optimization of PAC dose to submerged AnMBR in regards to nutrient and organic carbon removals as well as transmembrane pressure (TMP) change. Then, it was focused on the overall effects of PAC addition to submerged AnMBR at room temperature (22 ± 1 °C) during longer term. The main objectives of this research are:

1) To determine an optimal dose of PAC to a conventional submerged AnMBR system in terms of nutrients (NO_3-N, NH_4^+-N, PO_4^3-P, and NO_2-N), total organic carbon (TOC) and chemical oxygen demand (COD) removals as well as TMP development regarding membrane fouling control;

2) To evaluate the long-term effect of the optimal PAC dose on the removal efficiencies including nutrient, TOC and COD removals in AnMBR; and

3) To investigate the membrane fouling behaviour with the optimal PAC addition in terms of zeta potential, variation of TMP development and filtration resistances, and variation of SMP and EPS concentration in both mixed liquor and cake layer over the whole operation

period;

4) To explore the effect of PAC addition on the microbial community in conventional AnMBR.

1.3 Scope of the study

1.3.1 Optimization of powdered activated carbon addition to anaerobic membrane bioreactor

Three sets of experiments were conducted with three different PAC doses, including 1, 3, and 5 g/L, using a synthetic wastewater. Each experiment was operated for 25 days in a submerged AnMBR, including one chemical membrane cleaning in each experiment. These experiments were conducted to investigate the optimal concentration of PAC dose by the examination of membrane fouling behaviour in terms of TMP change, as well as nutrients, TOC and COD removal.

1.3.2 Long-term experiment of powdered activated carbon addition

A detailed experiment study was conducted using a submerged AnMBR with PAC addition and the other submerged AnMBR without PAC addition in parallel operation. A submerged AnMBR was dosed with optimal PAC concentration which was investigated from the previous experiment, and operated for 67 days including two chemical membrane cleanings. The other AnMBR was operated without PAC addition for 63 days including three chemical membrane cleanings. The experiment was conducted to study the long-term effects of optimal PAC dose in AnMBR, such as nutrients, TOC and COD removal performance and membrane fouling mitigation. Membrane fouling behaviour was analysed by TMP development and zeta potential of the mixed liquor. The variation of membrane fouling resistance during the operation time was also evaluated using the resistance-in-series model (Choo and Lee, 1996). In addition, the variations of concentration and compositions of SMP and EPS in both mixed liquor and cake layer were measured throughout the whole operation time. Finally, the difference of microbial community between two reactors was investigated.

1.4 Organization and major contents of the thesis

The thesis consists of six chapters as follows:

Chapter 1 includes an importance of wastewater treatment, AnMBR and membrane fouling, and briefly introduces fouling control strategies. The overview of this thesis including study objectives and scope of the study are also presented in this chapter.

Chapter 2 reviews the current circumstance of water scarcity and wastewater reuse, and general information about wastewater treatment technologies. The fundamentals of membrane bioreactors, including aerobic and anaerobic membrane bioreactors, as well as membrane fouling were given in this chapter. Finally, the strategies for fouling control in membrane bioreactor were introduced, and studies of different fouling reduction enhancers including powdered activated carbon were reviewed.

Chapter 3 presents the details about experimental materials and methodologies used in this study. It presents the characteristics of anaerobic sludge, composition of synthetic wastewater, and analytical methods including nutrient, COD, TOC, SMP, and EPS analysis, and membrane resistance measurement. The experimental setup and operating conditions for each set of experiments are also included in this chapter.

Chapter 4 shows the optimization of PAC dosage in AnMBR with short-term operations of 25 days. The three different PAC dosages were selected and the operation of each dosage was conducted one by one. It highlights the removal performance of TOC, COD, and nutrients, and membrane fouling behaviour regarding TMP change.

Chapter 5 demonstrates the effects of the optimal PAC dose on the removal performance of AnMBR and also membrane fouling mitigation. This chapter investigates the removal efficiency of nutrients, TOC, and COD, and membrane fouling behaviours in terms of TMP change, membrane resistance, concentration of SMP and EPS, as well as microbial diversity change of AnMBR with optimal PAC dose, compared to no PAC addition.

Chapter 6 summarizes the major results and findings of this study. Additionally, the recommendations for future study are given in this chapter.

CHAPTER 2

Literature review

This chapter is based on the journal paper entitled “A review on membrane fouling control in anaerobic membrane bioreactors by adding performance enhancers”.

2. Literature review

2.1 Wastewater treatment

2.1.1 Limitation of water resource and wastewater reuse

One of the most essential ingredients for life is water. However, since the amount of water resources on the Earth is limited, the water scarcity issue is rapidly being exacerbated due to the increasing population growth and water demand. In fact, almost one quarter of the children in the world are expected to experience extremely severe water shortage issue by 2040 (UNICEF, 2017). As the water resources are recycled through the circulation process of the atmosphere on the Earth, the fresh water availability is limited despite the increasing demand for fresh water. Therefore, it is inevitable to research alternative water sources.

The fast industrialization has produced large amount of industrial wastewater from textile, pulp and paper, chemical, manufacturing, and food industries containing synthetic substances which can be toxic to biological environment. The presence of trace pollutants like pharmaceutically active compounds (PhACs) in municipal wastewater has also emerged serious environmental and health concerns. The industrial wastewater is usually categorized as a high strength wastewater, while the municipal wastewater is low organic strength wastewater with 300–350 mg/L of COD (Khan, 2019; Lin et al., 2013a). According to the United Nations (UN), there is no adequate wastewater treatment for over 80% of worldwide wastewater (Connor et al., 2017). Moreover, the world's population living in cities will increase up to 70% by 2050. Most cities are not able to manage wastewater efficiently and sustainably due to the lack of proper infrastructure (United Nations and Social Affairs, 2014). However, the risks of water quality deterioration can be sustainably reduced by the measures of wastewater reuse. Reclaimed and recycled water have become attractive options as reliable water resources due to their sustainability. For effective wastewater reuse, that features high water usage efficiency, it is necessary to understand and apply suitable treatment techniques for different purposes with different qualities. Furthermore, the presence of chemicals or inorganic matters such as personal care products, pharmaceuticals, high salinity, and heavy metals, have been increasingly found in reclaimed wastewater and limiting the reuse of wastewater. Some organic pollutants in water, such as organochloride pesticides (OCPs), polycyclic aromatic hydrocarbons (PHAs), and bisphenol A, can cause severe impacts by disrupting endocrine of aquatic organisms and human bodies, even with tiny amount of exposure. These toxic and carcinogenic contaminants can so easily bioaccumulate in the food chain causing a long-term harmful effect (Khan et al., 2018). Therefore, the application of adequate treatment technologies is required for suitable quality of wastewater reuse.

2.1.2 Technologies for wastewater treatment

Wastewater treatment systems are likely to be based on the natural processes, and different facilities can differentiate the application of technologies and their intensity for diverse purposes. Physical processes like settling, chemical processes like coagulation and flocculation, biological processes and any combination of these processes, including physicochemistry and biochemistry can be applied. In terms of microbiological processes, they can be categorized into two technologies according to the type of microbial growth, which are fixed and suspended. In fixed technologies such as trickle filters and rotating biological contactors, microorganisms can grow on a surface of solid material to form a biofilm. On the other hand, in the activated sludge process or membrane bioreactor, microorganisms are in suspension for the removal of pollutants (Salgot and Folch, 2018; Waqas et al., 2020; Zhang et al., 2018).

A conventional activated sludge process is the most commonly used biological wastewater treatment in many municipal wastewater treatment plants (WWTPs). The process uses microorganisms to degrade and transform organic matters and nutrients into basic substances and a stable biomass. The wastewater is firstly settled in the sedimentation tank and settleable inorganic and organic solids are removed. The wastewater then flows through the aerobic reactor, which has 2-3 mg/L of dissolved oxygen and 1000-10,000 mg VSS/L of total suspended solids, while aerobic microorganisms can degrade the organic substances and nitrogen and phosphorus removal. After the degradation, microorganisms are settled in the secondary clarifier, which has usually 4000-12,000 mg VSS/L, and recycled back to biological reactor. For the further removal of nutrients and toxic materials from the effluent from the secondary treatment, tertiary treatment is required to dispose or provide reusable water (Salgot and Folch, 2018; Zhou et al., 2018).

Membrane technologies, which can effectively separate solids or concentrate liquids from wastewater, have been playing an essential role in both industrial and municipal wastewater treatment. As the membrane technology is a novel purification and separation technology, it is widely applied for the purposes of changing the quality of water from surface runoff, brackish water, well water and seawater (Jo, 2017).

Membrane separation process is based on a pressure-driven force relying on the membrane pore size in order to selectively separate components accordingly. The membrane process can be sorted into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) according to the membrane pore sizes. The pore size of the membrane decreases from MF to NF and then finally RO, and the operating pressure increases as the membrane pore size decreases. Since the pore sizes of MF and UF operations ranges between 0.1-10 μm and 2-100 nm, respectively, they can reject macromolecules such as suspended solids, organic colloids, and bacteria. In contrast to MF, UF is able to reject virus, while both MF and UF allow dissolved ions to pass through the membrane. When the pore size is between 0.5-1 nm, the transition from pore flow to diffusion of solution occurs. Since NF and RO membranes have smaller pore sizes than MF and UF, they involve diffusion, partitioning and sorption. While ideal NF membrane can remove multivalent and divalent ions, ideal RO membrane can reject every ionic species and only allows water molecules to penetrate the membrane (Jo, 2017; Munirasu et al., 2016).

In terms of membrane material, polymeric and inorganic (metallic and ceramic) membranes are commonly used for membrane technologies especially for membrane bioreactors. Inorganic membranes, such as stainless steel, aluminium, and titanium oxide, provides superior resistance to abrasion, corrosion, and high temperature, showing high permeate flux and fouling recovery. However, polymeric membranes such as polyvinylidene difluoride (PVDF) and polyethersulfone (PES) have gained more popularity due to the lower cost (Lin et al., 2013a). In recent studies, the modification of membrane surface or fabrication of nanocomposite membranes has gained great attention in order to decline the biofouling tendency by enhancing the hydrophilicity of membrane surface. Various kinds of nanoparticles such as silver nanoparticles (AgNPs), polycitrate-Alumoxane (PC-A) nanoparticles can be combined with polymers to make the nanocomposite membranes (Pirsaheb et al., 2019; Zhang et al., 2020b).

Membrane-based technologies have been increasingly used in wastewater treatment to produce and supply high quality water for drinking and industrial purposes. Membrane distillation (MD) is one of the thermal desalination technologies which can completely separate macromolecules, inorganic ions as well as other non-volatile components using hydrophobic membrane. The process is based on the vapour pressure difference across membrane as a driving force, allowing transportation of only vapour molecules from the feed. MD can be applied not only for wastewater treatment, but also seawater desalination and dairy fluids processing. Forward osmosis (FO) is another emerging membrane technology. In the RO process, higher pressure than osmotic pressure of feed water is required to allow water molecules to pass through the membrane. Meanwhile, the FO process can occur spontaneously due to permeate containing high osmotic solution and the natural occurrence of osmotic pressure differences between the feed and permeate. The combination of MD and FO has also been studied for refinery wastewater treatment (Alkhudhiri et al., 2012; Munirasu et al., 2016).

2.2 Membrane bioreactor

2.2.1 Aerobic membrane bioreactor

The membrane bioreactor (MBR), which is the combination of conventional activated sludge processes and membrane technology, is one of the most prospective technologies for wastewater treatment. MF or UF membranes are normally used in MBR systems and the module configuration can be either plate or hollow fibre membranes. In contrast to conventional activated sludge processes which include a secondary clarifier to remove biomass, in the MBR process, the biomass can be filtered through the membrane during the aerobic degradation. Therefore, the MBR system provides considerable benefits such as small footprint, excellent performance with high mixed liquor suspended solids (MLSS) concentration, lower sludge production, high and consistent effluent quality, and independent manipulation of HRT and SRT. However, there are also some disadvantages such as the relatively high cost of installation and operation, additional cost for membrane cleaning and replacement, and limitations of operation parameters requirements for membrane tolerances (Jo, 2017).

2.2.2 Anaerobic membrane bioreactor

Anaerobic digestion is one of the significant processes in wastewater treatment because it can perform removal of pollutants and energy generation at the same time. Even though it has some problematic issues such as sensitivity to temperature, the necessity of long SRTs, and post-treatment for the effluent that is hard to discharge directly, there are many remarkable benefits

(Smith et al., 2012). Compared to aerobic systems, it is energy efficient as it does not require aeration and produces less sludge to be treated. Therefore, the costs of aeration and sludge disposal can be much lower than with aerobic treatment. Moreover, the generation of biogas which can be recovered and used as energy such as methane is also one of the best advantages in anaerobic digestion.

Anaerobic digestion process involves four major phases of hydrolysis, acidogenesis, acetogenesis and methanogenesis. Many hydrolytic microorganisms, such as *Bacterides*, *Clostridia*, *Micrococci*, and *Streptococcus*, facilitate conversion of suspended polysaccharides, proteins, and fats into amino acids, sugars and fatty acids in the hydrolysis stage. At the acidogenesis stage, the products from the previous stage are converted into hydrogen, carbon dioxide, acetates, and VFAs. Then, acetogenesis occurs, which includes the conversion of VFAs into acetate, hydrogen, and carbon dioxide. Finally, methanogenesis mainly occurs with acetotrophic and hydrogenotrophic methanogens. While acetotrophic methanogens can convert the acetate into both methane and carbon dioxide, the hydrogenotrophic methanogens can transform hydrogen and carbon dioxide into methane (Khan et al., 2019).

AnMBR technology, which combines anaerobic digestion and membrane filtration, has been gaining increasing popularity. AnMBRs have the same benefits as aerobic MBRs, such as a footprint reduction and superior permeate quality. AnMBRs also can provide several advantages over the aerobic processes, including long SRT, low sludge production and potential energy use (Lin et al., 2013a). Moreover, as AnMBR is the integration of anaerobic digestion process with membrane separation, these processes can provide benefits as well. Throughout the anaerobic digestion process, organic materials are biodegraded into VFAs and hydrogen as intermediate products and methane as a final product (Khan et al., 2016). All these products can make the AnMBR an energy neutral or even energy positive. In addition, unlike anaerobic digestion that requires mesophilic ranges from 35 ℃ to 37 ℃, AnMBR can be operated in room temperature or even in cold temperatures by expanding SRT, which is both cost and energy-efficient (Robles et al., 2018). Ceramic and metallic membranes have been widely applied in the early research of AnMBRs, as they resulted in better permeate flux, thermal resistance as well as fouling recovery. However, due to their high costs, polymeric membranes like PVDF, polyethylene (PE), and polypropylene (PP) have gained more attention for recent studies (Lin et al., 2013a). Along with the polymeric membranes, MF and UF membranes of flat sheet, tubular or hollow fiber configuration have been applied to AnMBRs, while the hollow fiber module has been the most commonly used for submerged AnMBRs due to its high packing density (Lin et al., 2013a).

However, there are some issues that need further attention in AnMBR technology. One of the most challenging issues is membrane fouling, which deteriorates the performance of membrane and shortens membrane lifespan. Membrane fouling generally occurs when the components of sludge interact with membrane material, causing an initial pore blocking and cake layer formation. It is reported that membrane fouling in AnMBR can have more severe impacts compared to the aerobic system in terms of pollutant removal efficiency, sludge characteristics, even using the same membrane material (Lin et al., 2013a). Meng et al. (2009) also mentioned that the cake layer formed with anaerobic sludge might have comparatively lower removability than that with aerobic sludge.

There are two main configurations in AnMBR in terms of whether the membrane is located inside or outside of the bioreactor. These two configurations have different approaches to operating under pressure or under vacuum and using gravity. When the operation is under high pressure, membrane should be separated from the bioreactor, so this is usually called an external cross-flow or sidestream configuration. A pump is required to this configuration, which can cause high energy consumption. In this case, high-strength wastewater containing a large amount of organic components can be managed. So-called submerged or immersed membrane configurations, which are operated under vacuum and use gravity to membrane filtration, can be divided into two configurations again: a directly immersed form into bioreactors, and a separated form from the bioreactors. These submerged configurations can usually be applied to low-strength wastewater management. The separated chamber configuration is generally used for full-scale AnMBRs as the isolation of the membrane chamber is easier to clean the membrane without removing it from the reactor (Robles et al., 2018). Figure 2-1 below is describing a schematic outline of the three AnMBR configurations as well as the AnMBR used in this study.

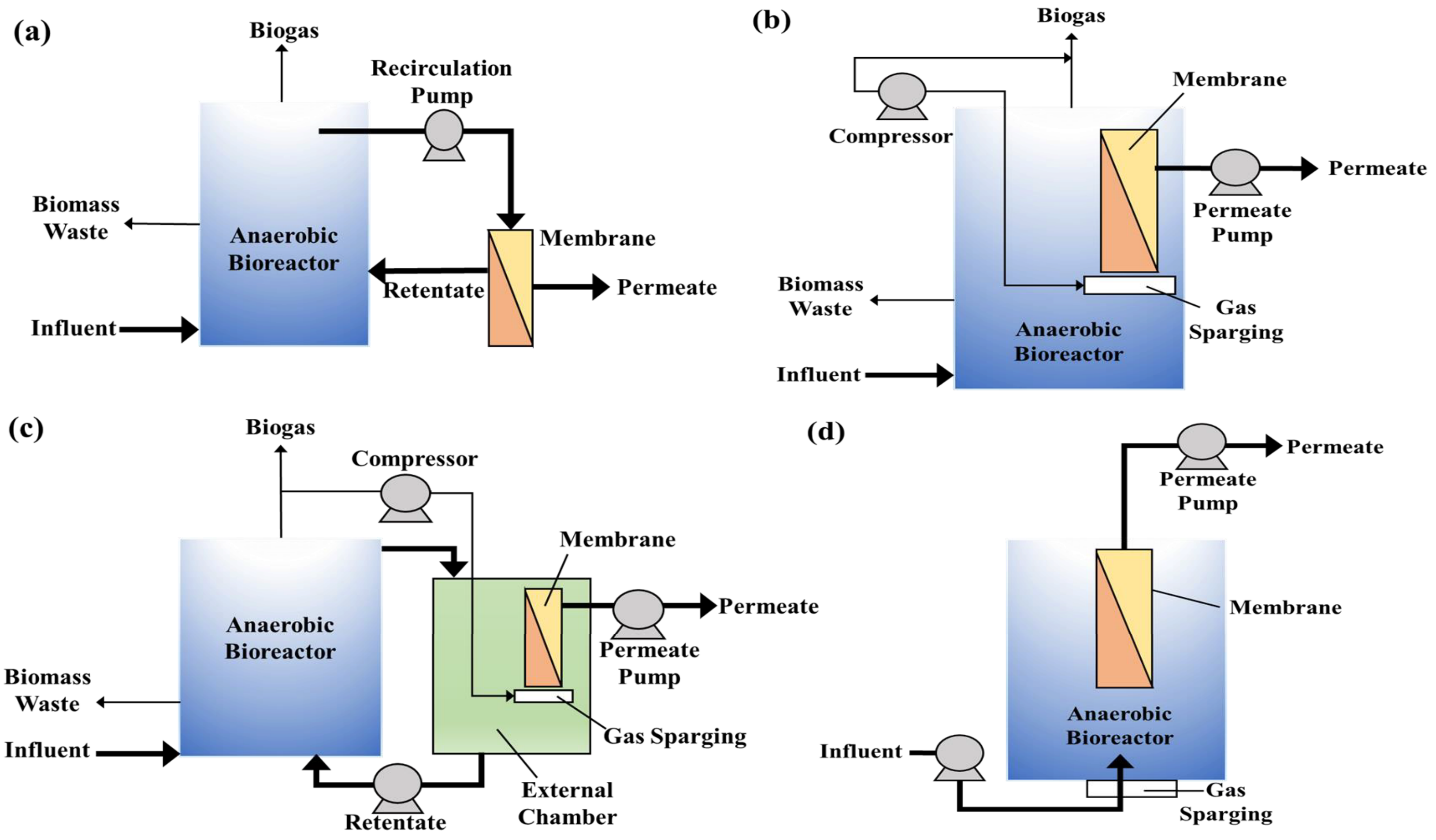

Figure 2-1. Schematic outline of AnMBR configurations. (a) External cross-flow configuration. (b) Submerged configuration with directly immersed membrane into the bioreactor. (c) Submerged configuration with membrane immersed in an external chamber. (d) Configuration of AnMBR used in this study.

2.3 Membrane fouling

Membrane fouling is one of the biggest unresolved obstacles of every membrane technology, including aerobic and anaerobic MBRs. Many studies have been carried out and the increasing number of researchers are still focusing on fouling and cleaning, because membrane fouling reduces the performance of the membrane which needs to be cleaned physically and chemically. When the membrane bioreactor continues to be operated and various contaminants are filtrated, membrane fouling occurs and can consequently lead to performance degradation. The definition of membrane fouling can be the unwanted deposition and aggregation of components such as colloids, solutes and microorganisms. It is basically due to the communication between the membrane itself and the compounds from the sludge suspension, and therefore causes a decrease in operation productivity, periodic membrane cleaning and replacement, and reduction of membrane lifespan (Guo et al., 2012; Lin et al., 2013a).

Classifications of the membrane fouling have slightly different definitions in each publication although they can basically be divided into two groups of reversible and irreversible fouling. Reversible fouling includes two types of fouling, removable and irremovable fouling. Removable fouling means that membrane performance can be easily recovered through simple physical cleaning such as backwashing and relaxation because the foulants are not attached very tightly. On the other hand, irremovable fouling is scarcely removed by physical cleaning and hence chemical cleaning is necessary. In general, removable and irremovable fouling result from cake layer formation and pore blocking, respectively. Irreversible fouling, in contrast, cannot be eliminated by any methods and is caused by precipitation of both organic and inorganic substances into the membrane pores from the initial stage and subsequent deposition and multiplication of cells, producing EPS (Lin et al., 2013a).

2.3.1 Organic and inorganic fouling

Membrane fouling can also be classified into three different groups according to foulant constituents. Both organic and inorganic compounds are influential to membrane fouling, which are called ‘organic fouling’ and ‘inorganic fouling’ respectively. Organic fouling occurs as a result of deposited biopolymers of which the main components are proteins and polysaccharides, and also dissolved organic matter (DOM) which is mostly constituted of SMP (Guo et al., 2012). It contains three layers and these layers have all different characteristics. The upper layer consists of insecurely tied sludge flocs and SMP, while the intermediate layer, much denser than the upper layer, is composed by SMP and aggregation of bacteria as well as polysaccharides due to soluble carbohydrate accumulation. The lower layer, the pore fouling layer, contains predominantly SMP and EPS, and this layer is irremovable since it is strongly bound to the membrane and the densest among the three layers (Iorhemen et al., 2016). As a consequence, SMP and EPS are the basis of organic fouling and have a significant influence on fouling development. On the other hand, inorganic fouling mainly results from chemical precipitation due to hydrolysis and oxidation. The mineral scale formed by the deposition of inorganic salts is mainly caused by crystallization and particulate fouling during the scaling on membrane surface. Thus, when the salt concentration is higher in the feed water, more inorganic scaling tends to occur. The main substances of inorganic fouling are commonly Ca, Mg, Fe, carbonate, sulfate and silica due to their low solubility in water (Guo et al., 2012).

2.3.2 Biofouling

The main determinants of biofouling is microbial metabolites such as SMP and EPS. Once they are accumulated and adsorbed on the surface or inside pores of membrane, it is called biological fouling, and this might occur when bacteria cells or flocs adhere to the membrane, growing while metabolism takes place (Guo et al., 2012). The biofilm can be formed firstly by the adhesion of planktonic cells on the surface and the subsequent formation of colonies and their development of biofilms. The major components of biofilms are cell biomass and EPS. EPS can be subdivided into soluble EPS and bound EPS. The soluble EPS is biodegradable and a product of bound EPS which is dissolved by bacterial hydrolysis. Both EPS consist of various organic macromolecules such as proteins, polysaccharides, lipids and other polymeric compounds. Proteins and polysaccharides are normally considered to be main contributors to fouling. When the EPS are released from the microbial aggregates into water, they are called SMP.

2.3.3 Membrane fouling in AnMBR

As the membrane is one of the major donors to capital as well as operational costs in AnMBRs, membrane fouling can be an obstacle that limits scaling-up and wider use of AnMBRs. Although membrane fouling occurrence generally characterised by pore blocking and subsequent cake layer formation in both aerobic and anaerobic MBRs, the properties of membrane fouling can be significantly different. It is reported that the cake layer removability in AnMBR was lower than that in aerobic MBR due to the difference of sludge characteristics (Lin et al., 2013a). Compared to the cake layer having reversible features in an aerobic MBR, the cake layer formed in AnMBRs was irremovable by relaxation or back flushing due to the consolidation. It was also found that the thickness of the cake layer in a submerged AnMBR was much higher than that in aerobic MBR, which were 1900-2100 and 20-200 μm, respectively (Gao et al., 2011). In addition, due to a relatively high concentration of carbonate and bicarbonate from anaerobic digestion, AnMBR might have more propensity for the inorganic fouling than aerobic MBR (Ramos et al., 2014). The membrane fouling in AnMBRs can be significantly affected by temperature as well. When the two submerged AnMBRs were operated under different temperature conditions with 55 °C and 37 °C, the higher temperature resulted in higher filtration resistance. As the temperature increased, the floc size became smaller and greatly deposited on membrane surface building into a cake. Moreover, higher operating temperature could lead to more production of SMP and EPS causing more membrane fouling (Charfi et al., 2012).

Table 2-1. Classification of membrane fouling types

Criteria	**Fouling types**	**Description (Main foulants and mechanisms)**	
Cleaning methods	Reversible	Removable	Easy recovery after physical cleaning like backwashing and relaxation.
		Irremovable	Chemical cleaning is needed to remove foulants.
	Irreversible		Cannot be recovered by any cleaning methods due to precipitation of organic and inorganic substances in the pores and multiplication of cells, producing EPS.
Foulant composition	Colloidal	Organic or inorganic particulates or colloids initially block membrane pores and develop a cake layer.	
	Organic	Dissolved organic matter (DOM) such as SMP attach on the membrane surface and block pores by adsorption.	
	Inorganic	Dissolved inorganic components, like calcium, magnesium, iron and silica, precipitate on the membrane by hydrolysis and oxidation as well as crystallization.	
	Biofouling	Microorganisms deposit on the membrane by bioadhesion and bioadsorption, and metabolism of bacteria forms microcolony developing biofilm.	

2.4 The strategies for fouling control in MBR

2.4.1 Optimization of operation conditions

The operation conditions of MBR, such as HRT, organic loading rate (OLR), temperature, food to microorganisms (F/M) ratio, and upflow velocity, have significant impact on membrane fouling. Since the bound EPS production is mainly influenced by HRT and OLR, which govern biomass concentration and F/M ratio, these parameters are closely related to membrane fouling propensity. As the HRT decreased, the membrane flux increased and thus fouling tended to increase. In addition, the increase of OLR and F/M ratio promoted the generation of bound EPS, which caused more severe membrane fouling (Wu et al., 2013). Moreover, it was found that the decrease of SRT from 100 days and 30 days to 20 days and 10 days, respectively, resulted in more membrane fouling (Huang et al., 2011b).

In the case of anaerobic condition, temperature change significantly affects methanogenic activity. The temperature increase results in decrease of liquid viscosity, leading to membrane flux improvement. Similar to aerobic MBR, HRT plays an important role for fouling control in AnMBR, showing that shorter HRTs can lead to more membrane fouling (Maaz et al., 2019). The upflow velocity of AnMBR also plays a vital role while having contrary results. The increase of upflow velocity facilitated a better mixing of substrate and biomass, whereas it could also break the sludge aggregation into fine particles due to strong shear force, leading to more generation of microbial byproducts and more fouling (Ozgun et al., 2015).

2.4.2 Membrane modification

Another strategy for fouling control is modification of membrane characteristics, including material, pore size, and hydrophilicity. In terms of membrane material, there are three types of membrane, which are polymer, metallic, and ceramic membranes. Among these three types of membranes, polymeric membranes have lower permeability than ceramic membranes. As the pore size of membrane considerably influences permeate flux, it was found that larger membrane pore size could lead to higher fouling rates compared to smaller pore sizes (Maaz et al., 2019). In addition, it was studied that the increase of hydrophilicity by the addition of engineered nanomaterials (ENMs), such as silver nanoparticles and titanium dioxide nanoparticles, could effectively mitigate the membrane fouling because of the high packing density of membranes in aerobic MBR (Meng et al., 2017).

2.4.3 Pretreatment of feed water

2.4.3.1 Alkaline pretreatment

The variations of pH can significantly influence the mixed liquor properties and microbial activities and the following membrane fouling behaviours. Sodium hypochlorite (NaClO) is a common membrane cleaning reagent for effective cleaning of organic foulants. The addition of different NaClO dosage between 5 and 50 mg/g SS to MBR could significantly affect microorganisms in terms of metabolism behaviours and cell lysis in activated sludge (Han et al., 2016). Furthermore, alkaline treatment of pH 11 under anaerobic condition also showed efficient disintegration of organic compounds and enhancement of VFA production (Yu et al., 2013).

2.4.3.2 Acid pretreatment

Acid pretreatment could also be effective in fouling control by changing microbial community and feed water properties. The 0.5 mM of nitric oxide in aerobic MBR resulted in effective reduction of membrane fouling by the reduction of biofilm growth as well as delay of a new biofilm formation on the membrane. This could be caused by a decrease in the abundance of *Proteobacteria*, which was dominant in biofilm formation, from 61% to 44% with the addition of nitric oxide (Jo et al., 2019). Furthermore, the formation of EPS in the mixed liquor significantly caused membrane fouling under pH 5.5 condition in aerobic MBR (Sanguanpak et al., 2015).

2.4.3.3 Ozone pretreatment

Ozone is one of the powerful oxidizing agents which can oxidize various recalcitrant organic matters without the addition of other chemicals. Previous studies have shown the impacts of the ozonation on the membrane fouling in MBRs. Ozonation was able to effectively reduce the membrane fouling by improving the biodegradability of the organic components. Natural organic matter (NOM) such as humic substances and polysaccharides, is one of the major problems causing membrane fouling. Ozone oxidation of feed water before membrane filtration could change chemical structures of NOM and resulted in significantly reduced membrane fouling (Van Geluwe et al., 2011). Xue et al. (2016) also investigated the effects of ozone pretreatment of an anoxic-aerobic MBR for treatment of oil-sands-process affected water. The result showed that 30 mg O_3/L of mild ozonation was able to enhance the degradation of organic compounds and change the structure of microbial community. The abundance of microorganisms excreting EPS and forming biofilm, such as *Rhodocyclales* and *Cytophagale*, remarkably lowered after the ozonation. As a result, not only EPS production but also the EPS protein/polysaccharides ratio decreased, facilitating the reduction of membrane fouling (Xue et al., 2016; Zhu et al., 2017). Recent study showed that ozone pretreatment after algal treatment of reverse osmosis concentrate before the microfiltration process efficiently removed nutrients as well as caffeine and carbamazepine. This combination of ozone and algal pretreatment could also alleviate flux decline while enhancing the reversibility of the MF fouling (Woo et al., 2019).

2.4.3.4 Addition of fouling reduction enhancers

a. Activated carbon

Activated carbon (AC) has been widely applied in MBRs due to its high adsorption capability, enhancement of biodegradation, and subsequent removal of recalcitrant pollutants. The addition of AC can also efficiently mitigate membrane fouling, as it might enhance membrane flux as well as the removal performance of COD and recalcitrant pollutants (Skouteris et al., 2015; Zhang et al., 2019a). Moreover, the use of AC in anaerobic digestion process has been gaining more attention because it facilitates the alleviation of organic shock loading, the enrichment of microorganisms and the improvement of anaerobic digestion stability (Zhang et al., 2019a).

There are two types of AC, namely powdered activated carbon (PAC) and granular activated carbon (GAC). PAC has high porosity and large surface area, which can lead to high adsorption capacity, while removing odour, colour and taste (Zhang et al., 2019a). Compared to PAC, GAC has larger size, which is more easily retained in the reactor and more economical when it is used continuously, because GAC can be regenerated by thermal process. Due to the larger size, GAC also has stronger physical interactions with membrane surface (Wu et al., 2017). The application of GAC to AnMBRs will be focused in this section, and the fouling reduction mechanism of PAC and its addition to aerobic and anaerobic MBRs will be discussed in the following section of 2.5.

In recent years, the addition of GAC has been extensively applied in anaerobic digestion processes to enhance both reactor efficiency and microorganisms. The anaerobic digestion process contains electron exchange between fermentative bacteria and methanogens in the form of metabolites, such as acetate, H_2 and methanol. Previous studies have demonstrated that conductive additives like GAC enabled direct electron exchange instead of metabolites, which could eventually enhance methanogenic conversion of short-chain fatty acids, such as acetate, butyrate, and propionate, and subsequent improvement of methane production (Barua et al., 2019; Florentino et al., 2019; Park et al., 2018). Zhang et al. (2017d) showed that GAC remarkably promoted methanogenesis by enhancing direct interspecies electron transfer between fermentative bacteria, *Geobacteraceae*, and methanogens, *Methanosaetaceae.* Another study has also concluded that surface modified GAC with magnetite stimulated enrichment of electroactive bacteria, such as *Shewanella, Pseudomonas, Geobacter and Desulfuromonas*, enhancing the methane production by a degradation of propionate to acetates and electrons that can be utilized by methanogens (Barua et al., 2019).

As a membrane fouling mitigation strategy, methods of inducing unsteady-state shear on the membrane surface, such as bubbling and vibration have been applied to MBRs. Particle fluidization has recently been presented as an alternative to bubbling, as it could have the same effect on membrane fouling reduction with at least ten times lower energy requirements than bubbling. Particularly, the fluidization of GAC has gained significant attention, because larger GAC is more effective in the longer term of operation (Wang et al., 2016a; Wang et al., 2016b). Therefore, previous studies have reported that GAC fluidization resulted in significant alleviation of membrane fouling in anaerobic fluidized membrane bioreactor (Aslam et al., 2018; Gao et al., 2014a; Gao et al., 2014b; Shin et al., 2014; Wu et al., 2015a; Yoo et al., 2012). In the studies of integrated anaerobic fluidized-bed membrane bioreactor, high amount of protein was adsorbed resulting in remarkable improvement of membrane filtration (Gao et al., 2014a; Gao et al., 2014b). The effect of GAC fluidization has been demonstrated with its energy efficient and effective advantages, unlike the popular air-sparging method which requires comparatively high energy costs (Wang et al., 2018c). As GAC fluidization is one way to induce unsteady-state shear on membrane, which has been identified as cost-effective, it eventually lower the energy requirement in the process (Zamani et al., 2015). As a case in point, the electrical energy requirement for anaerobic fluidized-bed ceramic membrane bioreactor operation was estimated to be 0.039 kW h/m^3, which was only 17% of electrical energy that can be generated from produced methane (Aslam et al., 2017). The average energy consumption of GAC fluidization is generally reported as 0.15 kW h/m^3, whereas that of gas sparging is twice higher, which is 0.31 kW h/m^3 including pumping and mixing (Evans et al., 2019). The table 2-2 summarizes the effects of GAC in anaerobic fluidized membrane bioreactor.

In the research of Ding et al. (2014), they added 50 g/L of GAC to an expanded membrane-coupled granular sludge bed (EGSB) and showed a remarkable enhancement of COD removals (80% vs 62% without PAC) and decrease in SMP concentration. The cake layer resistance, which was the main fouling mechanism in membrane-coupled EGSB process, was also decreased by 53.5%. Another research by Wang et al. (2018b) treating wastewater containing phenol and quinolone reported that a 2 g/L of GAC could not only remove COD and SMP by adsorption, but also enhance the degradation of phenol and quinolone. The high adsorption capacity of GAC could capture some fouling-causing compounds like SMP prior to attachment on membrane surface. Meanwhile, GAC could scour the foulants from the membrane surface and prevent the accumulation of foulants. Hence, the use of GAC as suspended medium effectively mitigated irreversible fouling (Johir et al., 2011). As a case in point, scouring effect of GAC with flux of 16 LMH in a two-stage anaerobic fluidized membrane bioreactor was able to mitigate membrane fouling, along with effective removal of 20 common pharmaceuticals (i.e. ibuprofen, caffeine, and sulfamethoxazole, etc.) through adsorption and biodegradation (Dutta et al., 2014a; Lim et al., 2019).

Although having positive effects on membrane fouling control, Wu et al. (2015a) suggested that the behaviour and characteristics of GAC might have a harmful influence on membrane performance. Due to the fine carbon particles that are released from GAC, the fouling could be aggravated by blocking the pores and forming a thin cake layer. GAC abrasion also led to a partial loss from the initial membrane quality. The reduction in adsorption capacity of GAC over time was also a major limitation. Thus, the exhausted GAC needs to be replaced or regenerated by thermal process to recover the adsorption capability (Korotta-Gamage and Sathasivan, 2017).

Furthermore, the energy requirements for fluidization and membrane fouling mitigation were significantly different depending on the particle size of GAC as well as adsorption capacity. When the adsorption of fresh GAC predominantly took place, comparatively small GAC particles with had greater effect on fouling reduction due to large surface area, along with less energy consumption. However, after the adsorption capacity was exhausted, dominant process for fouling reduction became the scouring effect. Then, the relatively large GAC particles were more effective in fouling reduction, but more energy is required for fluidization (Aslam et al., 2014). Charfi et al. (2017) showed that 2-3 mm of GAC particles acted better as a method of fouling reduction by removing accumulated cake layer, whereas small particles from 0.18 mm to 0.5 mm rather intended to accumulate on membrane surface. It was also found that, although large particles were more effective in scouring due to inertial forces, the energy requirement on fluidizing the particle was also higher (Wang et al., 2016a). Thus, further studies are necessary for a better understanding of production of fine carbon particles, GAC abrasion and choosing suitable particle sizes for mitigating membrane fouling.

Table 2-2. Effects of GAC on performance of fluidized AnMBRs

Reactors[a]	GAC properties and Packing ratio	Feed water	Operating condition	Effects of GAC on performance	References
SAF-MBR	• 139 kg • Calgon F300, USA • 0.8-1mm • 25%	Primary-settled domestic wastewater	• SRT : 485 d • HRT : 4.5–6.8 h • Temperature : 8–30 ℃ • Average effluent COD : ~23 mg/L	• No chemical membrane cleaning	Shin et al. (2014)
SAF-MBR	• >0.85mm • 25% in AFBR and 50% in AFMBR	Synthetic wastewater	• HRT : 8.7 h • Feed COD: 150 mg/L • Flux : 5 L/m^2·h • Temperature : 25 ℃	• Complete removals of diclofenac, ibuprofen and sulfamethoxazole	Lim et al. (2019)
SAF-MBR	• 30 g • 10 × 30 mesh • MRX-M, Calgon Carbon Corp., Pittsburgh	Municipal wastewater primary-clarifier effluent	• HRT : 2.3 h • Flux : 6–11 L/m^2·h • OLR : 1.0–3.5 kgCOD/m^3·d • Temperature : 25 ℃	• No requirement of other fouling control process • Lower electrical energy requirements	Yoo et al. (2012)
SAF-CMBR	• 300g • 10 × 30 mesh • MRX-M, Calgon • Carbon Corp., Pittsburg, PA • 50%	Domestic wastewater	• HRT : 1.3–2.1 h • Feed COD: 250 mg/L • Flux : 22 L/m^2·h • Temperature : 25 ℃ • pH : 7.3 -7.6	• Lower biosolids production • Low energy requirement (10% of energy converted form methane)	Aslam et al. (2018)
AFCMBR	• 50%	Dilute wastewater	• HRT : 1.3–2.1 h • Feed COD: 300mg/L • Flux : 22 L/m^2·h • Temperature : 25 ℃	• Lower energy requirement • No adverse effect of maintenance cleaning on organic removal	Aslam et al. (2017)

Two-stage AnFMBR	• 10 × 30 mesh • Bulk density: 500–1000 m^2/g • specific gravity: 0.85 and 2 g/cm^3 • 25% in AFBR, 50% in AFMBR	Municipal wastewater	• HRT : 1.28 h • OLR : 5.65 kgCOD/m^3·d • Feed COD: 250mg/L	• Effective removals of pharmaceuticals • No requirement of other fouling control process	Dutta et al. (2014a)
AnFMBR	• >2 mm, • 0.85-2 mm • 0.5-0.85 mm • 0.18-0.5 mm • <0.18 mm • 10%, 30%, 50%	Synthetic wastewater	• Flux : 50 L/m^2·h • Feed COD: 250 mg/L	• Energy requirement increased with particle size • The higher the packing ratio, the greater the fouling reduction	Aslam et al. (2014)
AnFMBR	• N/A	Screened domestic wastewater	• SRT : 12 d • HRT : 4 h • Flux : 7.6–7.9 L/m^2·h • OLR : 1.3–1.4 kgCOD/m^3·d • Temperature : 13–32 °C	• Similar BOD$_5$ and COD removal efficiencies (around 85%) achieved when operating at a 65% shorter HRT than gas-sparing system (removals of around 90%)	Evans et al. (2019)
AnFMBR	• Lignite coal (0.42-0.85 mm, 650 m^2/g) • Peat bog (0.85-2.4 mm, 600-800 m^2/g)	Synthetic wastewater	• Flux : 14 L/m^2·h	• Fouling reduction and detrimental effects on membrane	Wu et al. (2015a)

	- Peat bog (2.4-4.6 mm, 600-800 m^2/g)				
IAFMBR	- 40 g - 10 × 30 mesh	Domestic wastewater	- HRT : 6 h - Flux : 7.1 $L/m^2 \cdot h$ - Feed COD: 247-449 mg/L - Permeation : 23.2 L/d - Temperature : 35, 25, 15 ℃ - pH : 7.18–7.99	- High protein adsorption by GAC	Gao et al. (2014a)
IAFMBR	- 200-300 g - 10 × 30 mesh	Domestic wastewater	- HRT : 4, 6, 8 h - Flux : 0.27 $m^3/m^2 \cdot d$ - Feed COD: 300 mg/L - Temperature : 35 ± 2 ℃ - pH : 7.5 ± 0.21	- Adsorption of protein in cake layer by GAC - Improved membrane filtration	Gao et al. (2014b)

[a] AFCMBR, anaerobic fluidized bed ceramic membrane bioreactor; AnFMBR, anaerobic fluidized membrane bioreactor; SAF-MBR, staged anaerobic fluidized membrane bioreactor; IAFMBR, integrated anaerobic fluidized-bed membrane bioreactor; SAF-CMBR, staged anaerobic fluidized bed ceramic membrane bioreactor.

b. Biochar

Biochar is a porous and carbonaceous residue obtained from thermal decomposition of biomass in an oxygen deleted environment, or other processes such as pyrolysis, hydrothermal carbonisation, gasification and torrefaction. It is usually produced at a lower temperature under 700 °C, because reaction above 900 °C causes the destruction of walls between pores, which results in widening pores of biochar (Luo et al., 2015; Qambrani et al., 2017). Unlike AC, it is produced without any activation, and this non-activation makes the specific surface area of biochar less efficient compared to AC. However, the production cost of biochar is one tenth cheaper than that of AC (Luo et al., 2015; Masebinu et al., 2019; Pan et al., 2019). After a series of reactions of biomass such as dehydration, depolymerisation and carbonisation during thermal decomposition, three products, namely condensable liquid (bio-oil), non-condensable gases (syngas) and biochar are produced, which depends on the type of biomass used and process conditions (i.e. temperature and residence time). Biochar usually consists of fixed carbon, labile carbon and other volatile compounds, as well as moisture and ash. Fast pyrolysis aims at liquid oil production, whereas the goal of slow pyrolysis is biochar production, as the slow evaporation of water and release of volatile components can result in an increase of relatively fixed carbon content of the solid (Masebinu et al., 2019; Weber and Quicker, 2018). The heterogeneous surface of biochar, which has both carbonised and non-carbonised fractions, accommodates several adsorption mechanisms. Physical adsorption, surface precipitation and the pore-filling are the major routes of adsorption. Moreover, for positively charged organic compounds, hydrophobic effect and hydrogen bonding of biochar surface are the important adsorption routes. On the other hand, the removal of inorganic compounds largely depends on electrostatic attraction, precipitation, and ion exchange (Fagbohungbe et al., 2017; Masebinu et al., 2019).

The addition of biochar on anaerobic digestion process has shown to be effective in terms of biogas production and selectively enrich microbial groups. The biochar addition was able to enhance VFA production and degradation, and improve both hydrogen and methane production (Luo et al., 2015; Sunyoto et al., 2016). Some studies demonstrated that better biogas production could be resulted from enhanced direct interspecies electron transfer (DIET) process by enrichment of electrogenic *Geobacter* and *Bacteroidetes*, which are potential direct interspecies electron transfer partners during anaerobic digestion (Lü et al., 2016; Wang et al., 2018a). In addition, since biochar contains redox active moieties such as quinines, phenolics and phenazines, they can catalyse the electron transfer between biochar and outer membrane cytochromes during redox reactions (Shanmugam et al., 2018; Wang et al., 2020). Moreover, biochar addition significantly enhanced methanogenesis by facilitating the enrichment of *Methanosarcina* even in high ammonium stress, and also favoured anaerobic sludge granulation, due to the ability of promoting biofilm formation and reducing the inhibition behaviour of ammonia (Mumme et al., 2014; Shen et al., 2020). Similar to the biogas yield enhancement, biochar could facilitate hydrogen production via enrichment of hydrogen-producing bacteria (Zhang et al., 2017a). The alleviation of sulphide toxicity during anaerobic treatment of sulphate-rich wastewater using biochar was investigated and biochar promoted reactor stability by adsorption of H_2S from biogas (Oliveira et al., 2020). Due to the adsorption capability and functional groups on the surface, biochar was also able to adsorb EPS and enhance sludge granulation, which could lead to significant mitigation of membrane fouling in aerobic MBRs (Jiang et al., 2020; Ming et al., 2020; Sima et al., 2017).

Some previous studies have demonstrated that biochar could be beneficial to anaerobic membrane bioreactor. Bamboo charcoal, one kind of biochar, was able to enhance the removal performance of AnMBR as well as mitigate membrane fouling. In this study, two AnMBRs treating bamboo industry wastewater were analysed with and without bamboo charcoal addition. The result showed that COD removal efficiency increased about 5% after the addition of bamboo charcoal, as well as reduced membrane fouling owing to the decrease of both concentration of SMP and resistance of the fouling layer. Meanwhile, the methane yield became higher as a result of greater microbial activity of dominant microorganisms in methane production, such as *Methanosaeta, Methanospirillum* and *Methanobacterium*, occurring additionally inside the pores of the bamboo charcoal (Mei et al., 2016). More recent study showed that membrane fouling was effectively reduced in a biochar-amended AnMBR along with 56% of decreased TMP rising rate and decreased proteins of EPS. In addition, *Arcobacter*, one of the bio-foulants that is involved in membrane biofouling, was hardly accumulated due to the presence of biochar (Chen et al., 2020).

c. Waste yeast

Waste yeast is traditionally used as a protein supplement in animal feed or alimentary substrate for the food processing industry. The brewing industry is the major source of spent yeast, which also produces other residues in addition to brewery wastewater, such as methanogens and small cellulosic particles (Zupančič et al., 2012). Due to the high degradation capacity of yeast, it was favourable in treatment of landfill leachate, which contains high amount of recalcitrant compounds like phenolic compounds as well as toxic substances such as halogenated and heavy metals (Reis et al., 2017). Yeast has lower tendency to adhere on membrane surfaces than other microorganisms, so that its application in MBR can be beneficial in membrane fouling control and system operation (Reis et al., 2017). The presence of yeast in aerobic MBR could significantly remove not only COD, colour and EPS, but also refractory substances including polyacrylamide (Brito et al., 2019; Silva et al., 2019; Song et al., 2019; Zhang, 2020).

Anaerobic co-digestion, which balances the nutrient component of different residues, is widely applied as a way to dilute potential toxic compounds and enhance biogas production (Neira and Jeison, 2010). Some previous studies have demonstrated that additional co-substrates in anaerobic wastewater treatment increased methane production by maintaining a pH level within the methanogenesis range between 7.0 and 7.5, while improving the degradation of low biodegradable substrates (Agler et al., 2010; Zupančič et al., 2012). The research on the supplementation of yeast in brewery wastewater treatment as a co-substrate in a upflow anaerobic sludge blanket (UASB) reactor showed enhanced biogas production by 50%, while no significant changes in COD removal efficiency and accumulation of VFAs were observed up to 1.1 (v/v)% of brewery yeast concentration (Neira and Jeison, 2010; Zupančič et al., 2012). These results indicated that the additional waste yeast could be a feasible substrate in anaerobic digestion in terms of high biodegradability and biogas yield.

The supplementation of yeast wastes as co-substrate in an AnMBR can have positive effects on membrane fouling control as well. A research by Yun et al. (2019) investigated the effects of yeast addition to AnMBR treating low-grade coal wastewater (LCWW). Compared to no methane production in the absence of yeast wastes, AnMBR with yeast wastes gradually increased COD removal efficiency as well as methane production. In addition, the presence of yeast wastes showed the significant growth of some microorganisms such as *Methanococcus* and *Methanosarcina* which were responsible for the degradation of LCWW and biogas production. However, due to the metabolism of these bacteria, the fraction of SMPs and aromatic group with high molecular weight (> 1 kDa) also increased. Thus, the addition of yeast wastes could be a potential alternative as an additive to AnMBR due to their positive effects on biodegradation of LCWW and growth of microorganisms, but further research should be carried out to find out the effect on fouling control.

d. Iron

Iron, which is the most abundant transition metal on the Earth, is also an essential component for the growth of most living organisms. Although iron itself is a non-toxic and electron donor in redox reactions, the presence of iron in anaerobic environment plays an important role in the electron cycling and metabolic activity of microorganisms (Baek et al., 2019; Wei et al., 2018). The Fe(II) and Fe(III), which are generated from several iron compounds, can be provided as nutrients for microbial activity or as redox mediators to facilitate the conversion of organic matters to methane (Baek et al., 2019). One of the strong reductants, zero-valent iron (ZVI), is an active anode material in electrocoagulation, and electrically produces Fe(II) ions which promote coagulation and effectively decrease the soluble and colloidal particular matters. Although the generation of Fe(II) ions from ZVI in aerobic process needs to be triggered by the electric field, it can occur spontaneously in the anaerobic digestion process. The protons, which are released by acidogens during the acidification, can help spontaneous generation of Fe(II) ions without any drive of electric field (Zhang et al., 2020a).

Many previous studies have investigated the addition of iron or ZVI into anaerobic digestion which could significantly increase methane production and improve COD removal (Kong et al., 2018; Ruan et al., 2017; Suanon et al., 2017; Wu et al., 2015b). When iron was added to anaerobic aquatic environment, hydrogen was produced by iron corrosion. This hydrogen evolution can benefit methane production by enhancing both hydrogenotrophic methanogenesis and homoacetogenesis. In addition, iron was able to serve as an electron donor to reduce oxidation-reduction potential (ORP), and led to the decrease in propionic-type fermentation and subsequent enhancement of methanogenesis. As the accumulation of propionate destroyed the pH balance between acidogenesis and methanogenesis as well as hindered the methanogenesis of acetate, it should be reduced during the anaerobic process

(Feng et al., 2014; Liu et al., 2012). Although anaerobic digestion can be limited by low efficiency of hydrolysis and acidification, ZVI could intensify the activities of enzymes related to hydrolysis and acidification, such as protease which is responsible for catalysing hydrolysis of polysaccaharide to monoses (Feng et al., 2014). Moreover, the presence of ZVI stimulated the growth of hydrogen-consuming microorganisms such as homoacetogens and hydrogenotrophic methanogens, thereby enhancing acetate or methane production (Feng et al., 2014; Xu et al., 2019). In addition, iron could also effectively eliminate odorous H_2S gas by precipitation of FeS. Likewise, the iron might be used for phosphate recovery in the form of compounds of iron and phosphate such as vivianite (Wei et al., 2018). It was also found that the supplementation of iron salts to anaerobic digestion could be potentially advantageous to membrane fouling by supporting granulation and stabilisation. When ferrous iron was supplied to UASB reactor, anaerobic bacteria and EPS tended to adhere to iron in order to form a more stable structure with 56 % of enlarged granule diameter. Moreover, inorganic precipitates such as ferrous sulphide could contribute to the stability of granules.

The iron addition to AnMBR provided remarkable benefits on membrane fouling mitigation in several previous studies. Dong et al. (2015) showed the influence of $FeCl_3$ as an additive in long-term operation of an AnMBR treating municipal sewage. The performance of the AnMBR, including removal efficiencies of COD and BOD_5, was enhanced by adding 26 mg/L of $FeCl_3$. Furthermore, even though the addition of $FeCl_3$ caused the increase of MLSS concentration and a more thickened cake layer, membrane fouling has been mitigated due to the more porous cake layer formation and increased filterability of mixed liquor. Zhang et al. (2017c) also investigated the addition of $FeCl_3$ to AnMBR, which effectively reduced membrane fouling by increasing both sludge floc size and the colloids as well as decreasing SMPs. Since iron remained in the reactor as a precipitate, resulting in minimal concentration of iron in the effluent or supernatant, it was expected to be advantageous for a long-term operation. In a recent study, ZVI has been applied into AnMBR with and without electric field. Although ZVI with electric field facilitated the increase of iron releasing rate of ZVI by 12 times and enhanced removal performances of COD and total phosphorous (TP) by about 3% and 50%, respectively, it resulted in more severe fouling due to the high density of Fe-rich fouling layer. However, ZVI without electric field significantly mitigated membrane fouling rate by 20% through the enhancement of mixed liquor filterability (Zhang et al., 2020a).

e. Calcium

Calcium can be another special additive to alleviate fouling and enhance characteristics of granular sludge by enhancing bioflocculation. EPS, which is known to be the main substance affecting membrane fouling, typically contains negatively charged functional groups such as hydroxyl and carboxyl. Due to negatively charged EPS, cations play an important role in sludge flocculation. Divalent cations including calcium ions tend to combine preferentially with carboxylic functional groups of EPS and form bridges between the EPS molecules. This bridge formation promotes the improvement of bioflocculation, enlarges flocs and mitigates fouling (Chen et al., 2011; Zhang et al., 2016; Zhang et al., 2017b; Zhao et al., 2015a). Since the cost of calcium salts are relatively low, they have been widely used in aerobic process as an additive, which improve the properties of mixed liquor (Barros et al., 2020; Zhang et al., 2016). However, the decline in permeability and subsequent inorganic fouling occurred with high concentration of calcium of 830 mg/L, due to the precipitation of calcium carbonate. Therefore, more research is necessary to have a better understanding of effects of calcium addition and find out the optimal calcium concentration (Zhou et al., 2014).

The addition of calcium can also positively affect anaerobic processes. Some previous studies have been conducted to evaluate the influence of calcium addition and the most effective dosage for anaerobic digestion. When five different concentrations of calcium chloride ($CaCl_2$), which are 0, 1, 3, 5, and 7 g/L, were added to anaerobic digestion process, 3 g/L of calcium concentration was optimal for the best performance of anaerobic digestion and biogas production (Ahn et al., 2006). Similarly, according to a study of Ahmad et al. (2011), calcium oxide (CaO) in the UASB reactor enhanced granulation and the accumulation of biomass as well as the degradation of butyrate and acetate acid. Since the addition of calcium on anaerobic digestion process significantly increased the abundance of *Methanosaeta* as the dominant methanogen, the methane production could be improved (Salama et al., 2020). However, an overdose of calcium from 5 to 7 g/L of concentration, which may lead to precipitation and limit mass transfer between microbes and organic compounds, further inhibit anaerobic process. When the precipitates such as calcium carbonate were formed on the surface or within the granules, they can cause sludge washout, as well as the declined methanogenic activity and diffusion limitation (Ahmad et al., 2011).

Due to the positive effects of calcium addition on anaerobic process, the use of calcium as an additive in membrane bioreactors can also be a promising way to reduce membrane fouling (Ding et al., 2015). An investigation for the effects of calcium addition (0, 50 and 100 mg/L of calcium) was conducted in three sequencing batch reactors with external dead-end microfiltration. The result showed that the highest dosage of calcium was able to enhance the reduction of fine particles, EPS and colloids in supernatant, leading to the mitigation of membrane fouling (Liu and Sun, 2012). This significant reduction of membrane fouling was mainly due to calcium promoted bioflocculation, which achieved high volumetric organic removal and increased methane production rate. Furthermore, the enlarged size of anaerobic sludge granules by calcium addition was also reported in some studies using membrane-coupled expanded granular sludge bed (EGSB) reactor or UASB reactors (Cunha et al., 2018; Faria et al., 2019; Gagliano et al., 2020). When calcium chloride was added to the EGSB reactor, the membrane fouling was alleviated effectively and the concentration of SMP decreased (Ding et al., 2015).

f. Polyaluminum chloride

One of the aluminum salts, polyaluminum chloride (PAC), generally consists of various polynuclear aluminum hydrolysis products, including Al monomers such as $Al(OH)^{2+}$, dimer ($Al_2(OH)_2^{4+}$), trimer ($Al_3(OH)_4^{5+}$), Al_{13} ($AlO_4Al_{12}(OH)_{24}(H_2O)_{12}^{7+}$) and aluminum hydroxide ($Al(OH)_3$). Due to the presence of these products, PAC is superior to the traditional aluminum coagulants, such as $AlCl_3$ and $Al_2(SO_4)_3$, for removing organic matters (Wu et al., 2019). The behaviour of aluminum coagulants can be greatly affected by basicity values (B), which is the molar ratio of OH/Al^{3+}, because the dominant hydrolysis products are different under different basicity conditions. PAC with high basicity value ($B = 2.4$) resulted in increased membrane fouling propensity, as well as higher DOC removal efficiency and zeta potential of flocs, compared to PAC with lower basicity ($B = 2.0$ and $B = 1.6$). This phenomenon might be related to the different dominant mechanisms of coagulation according to the content of Al species. As the percentage of Al_{13} increased along with the basicity value increase, it could provide a larger amount of positive charges for charge neutralization rather than adsorption bridge effect. As the flocs produced from charge neutralization are smaller than those from adsorption bridge effect, it could result in more severe membrane fouling (Zhao et al., 2015b).

Many previous studies have reported high charge neutralization capacity of PAC, which can lead to enlarged floc size and better filtration performance. The dose of PAC and subsequent hydrolysis can provide positive charge, which can neutralize the negatively charged sludge flocs and colloids. This neutralization results in weaker repulsion among flocs and colloids, and easier formation of large particles. In addition, SMP and EPS in mixed liquor can be compressed and removed from membrane surface by the charge neutralization and adsorption of PAC (Wang et al., 2019c). When PAC was added in anaerobic digestion process, it facilitated the reduction of SMP and improved sludge filterability. However, a high concentration of PAC over 500 mg/L in anaerobic digestion could inhibit short chain fatty acids (SCFA) production as well as anaerobic process such as hydrolysis, acidogenesis and methanogenesis by decreasing the ratio of bioavailable nutrient, especially phosphorous (Chen et al., 2018b; Yu et al., 2015).

The positive effects of PAC dosing into AnMBR as an inorganic coagulant on membrane fouling control have been studied in some previous research. The addition of PAC could influence microbial characteristics as well as cake layer structure in the anaerobic digestion process. The abundance of anaerobic microorganisms, especially *Cloacimonetes* and *Smithella*, was significantly enriched in AnMBR (Zheng et al., 2018). Moreover, PAC was able to increase the hydrogen yield by washing out hydrogen consumers, including *Acetoanaerobium* and *Desulfobulbus* (Wu et al., 2019). It was also reported that the cake layer on the membrane surface became more porous and looser when PAC was added, which could provide better filterability. A result from a study showed that PAC dosing could lower the composition rate of carbohydrate in SMP and EPS, as well as compress the concentration of EPS. This resulted in reduction of adherence capacity of sludge and more substantially porous cake layer (Wang et al., 2019c; Yao et al., 2015).

g. Zeolite

Zeolite is a porous substance with high crystallinity, which mainly consists of aluminium, oxygen, and metals such as titanium, tin, and zinc. While natural zeolite can be normally found in rocks near volcanoes all over the world, it can also be synthesized or modified in order to improve properties for different applications. Both natural and modified zeolites can be used for adsorption and ion exchange. The presence of cations like Na^+, Ca^{2+}, K^+ and Mg^{2+} on the porous surface of zeolite facilitates ion exchange from a contact solution. Thus, the use of zeolite can be applied in both aerobic and anaerobic biological processes including nitrification and denitrification, activated sludge, and anaerobic digestion. In aerobic processes, zeolite can act as an ion exchanger as well as a biomass carrier, whilst in anaerobic processes, zeolite can also act as an inhibitor of ammonia and heavy metals by ion exchange (Montalvo et al., 2020; Wijesinghe et al., 2018; Yuniarto et al., 2013).

Zeolite has been reported to improve the performance of anaerobic processes as a porous microbial carrier as well as ion-exchanger. Its high ion-exchange capacity can contribute to enhance NH_4^+ removal which is known as an inhibitor of anaerobic digestion. Indeed, Lin et al. (2013b) used modified zeolite to reduce NH_4^+ concentration by ion exchange with Na^+ and Ca^{2+} as dominant ions for NH_4^+ adsorption. Additionally, the application of zeolite also showed remarkable improvement in methane production and COD removal (Dutta et al., 2014b; Sudibyo et al., 2018; Wang et al., 2019a). These improvements of anaerobic digestion can be attributed to ion exchange of NH_4^+, cations like Ca^{2+} and Mg^{2+} and long chain fatty acids (LCFA) (Nordell et al., 2013; Wang et al., 2019a). Another researcher focused on the microbial communities apart from ion exchange, and suggested that zeolite could specifically preserve the growth and immobilisation of microorganisms, especially *Methanosarcina* and *Methanobacteriums* (Poirier et al., 2017).

Zeolite has been widely applied to aerobic and anaerobic membrane bioreactor for membrane fouling reduction. This is because it can improve the settlement of sludge as well as the removal of nitrogen and phosphorous. As zeolite has high porosity and large surface area, it provides a stable environment for bacterial attachment, and substantial microbial aggregation can enhance the settleability of sludge (Wei et al., 2013). As a result, membrane fouling can be alleviated by forming rigid sludge flocs and enhancing membrane permeability. Likewise, the application of zeolite as a carrier showed effective removal of COD and suspended solid (SS) which could facilitate better membrane performance and less fouling in anaerobic fluidized membrane bioreactor (Chen et al., 2019a; Li et al., 2020). When an anaerobic fluidized membrane bioreactor was operated with natural zeolites as carriers, the removal rate of SS significantly improved by 22%. It was also observed that the anaerobic microorganisms were able to attach on the surface of zeolites with remarkable growth. Thus, no membrane fouling was observed due to the low COD and SS concentrations (Chen et al., 2019a).

h. Beads

The use of granular media fluidizing in AnMBR has been gaining attention along with the development of anaerobic fluidized bed membrane bioreactor. Previous studies showed that fluidization of glass beads and polyethylene terephthalate (PET) beads can act as turbulence promotors and scouring media, respectively, which effectively controlled membrane fouling. Polymer-based gel beads have also been proved to be an ideal microbial carrier, based on their cost effectiveness, high bio-compatibility, strong stability for long-term use, as well as porous structure for microbial attachment and aggregation. Moreover, controlling the size and density of the beads can be achievable by changing the synthesis conditions (Pandey and Sarkar, 2017; Wang et al., 2019b). Polyvinyl alcohol (PVA), which is water soluble polymer, can form gel beads by cross-linking with other materials like sodium alginate and chitosan (Cho et al., 2017). Moreover, when PVA and chitosan form more stable structure through covalent bonds with metal ions, they can be applied in different fields as adsorbent materials, antibacterial agents, or biocarriers (Gutha et al., 2017; Jiao et al., 2017). A research by Wang et al. (2019b) showed the effect of PVA/chitosan gel beads and PVA/chitosan/iron gel beads on anaerobic sludge. Both gel beads favoured the adhesion and aggregation of methanogens, mainly *Mathanospirillum*, *Methanosaeta* and *Methanobacterium*.

According to Düppenbecker et al. (2017b), the use of fluidized glass beads in AnMBR with external tubular membrane could be a promising option for alleviating membrane fouling in AnMBRs. The optimal diameter of 1.5-mm fluidized glass beads reduced the fouling despite the membrane has been damaged by abrasion. Moreover, the COD removal rate was remained between 77% and 83%, and methane production increased by around 30% as well. The same research group also evaluated the fouling behaviour by three different ceramic membranes including ZrO_2 and Al_2O_3 ultrafiltration membranes and TiO_2 microfiltration membrane. The presence of fluidized glass beads was able to reduce the fouling rate by around 95% for all three membranes. Although all types of membranes were damaged by abrasion of glass beads, Al_2O_3 microfiltration membrane showed the least abrasion in a clean water filtration test (Düppenbecker et al., 2017a). Similarly, the fluidization of PET beads with bigger size and lower density also demonstrated significant fouling reduction by scouring the membrane (Charfi et al., 2018a; Charfi et al., 2018b). These fluidized beads can mitigate membrane fouling by two mechanisms. Firstly, the mixing action of particles can lead to increase in turbulence and thus concentration polarization can be decreased. Secondly, scouring effect on the previously deposited foulants can also alleviate fouling (Düppenbecker et al., 2017b).

Table 2-3. Effects of other enhancers on AnMBR performance

Enhancer type	Enhancer details	Dose	Configuration of reactor[a]	Feed water	Operating conditions	Effects of other enhancers on performance	References
Biochar	• Bamboo charcoal • Watson Bamboo Charcoal Products Company in Anji. BC • Particle size of 0.5 mm	19.01 g/L	External AnMBR	Bamboo industry wastewater	• SRT : 150 d • HRT : 3 d • OLR : 6 kgCOD/m^3·d • Temperature : 32 ± 2 °C	• Increase in COD removal efficiency by 5% • Increase in biogas production and methane yield • Declined SMP concentration and cake layer resistance • Increased microbial diversity and activity of methanogens	Mei et al. (2016)
	• Biochar with adsorption capacity of biopolymers • Particle size of 0.25-0.4 mm	4 g/L	Biochar-amended external AnMBR	Pharmaceutical wastewater	• SRT : 120 d • HRT : 24 h • VSS : 14.67 g/L • OLR : 7 kgCOD/m^3·d • Temperature : 32 ± 2 °C	• Effective removal of adsorbable organic halogen (AOX) (average 61.5% vs 56.2% without biochar) • Decrease in TMP rising rate by 56% • Slower TMP jump • Reduced proteins of EPS • Decrease in abundance of biofoulant, mainly *Arcobacter*	Chen et al. (2020)

Waste yeast	N/A	LCWW to YW of 50:50	Submerged AnMBR	Low-grade coal wastewater	• HRT : 1 d • COD : 2 g/L • OLR : 1 kgCOD/m^3·d • Temperature : 35 ± 1 °C • pH : 7.8–8.0	• High COD removal efficiency (58%) and methane production (182 CH_4 mL/g COD); • Improved degradation of LCWW	Yun et al. (2019)
Iron	N/A	26 mg/L of $FeCl_3$	Pilot scale AnMBR	Municipal wastewater	• HRT : 8.5 h • SRT : 70 d • Flux : 17 LMH • COD : 383 ± 113 mg/L • pH : 6.7–6.8 • Temperature : 23 ± 1 °C	• Improved COD and BOD_5 removals by 13.8% and 10.8%, respectively • Increased filterability of mixed liquor with reduced colloidal matter • Increased porosity of fouling layer	Dong et al. (2015)
	N/A	150 mg/L of $FeCl_3$	AnMBR	Synthetic sewage	• Flux : 15 LMH • Feed COD : 500 mg/L • VSS : 5.0-5.5 g/L • Temperature : 35 °C • pH : 6–7	• Increased sludge floc size and colloids particle size • Reduced thickness of fouling layer • Decreased levels of COD and proteins	Zhang et al. (2017c)
	Two pairs of electrodes with flat ZVI anodes (90 cm × 5 cm × 0.3 cm) and titanium		AnMBR	Municipal wastewater	• HRT : 10 h • Flux : 15 LMH • Temperature : 35 °C • COD : 483 ± 16 mg/L	• Enhanced COD removal by 3%, and TP removal by 50%, and H_2S removal (> 500 ppm with electric field vs < 60	Zhang et al. (2020a)

	cathodes (90 cm × 5 cm)					ppm without electric field) • Membrane fouling mitigation by the improvement of mixed liquor filterability	
Calcium	367.5 mg/L of $CaCl_2$	100 mg/L	Dead-end microfiltration	Granular sludge mixed liquors from SBR	• HRT : 4 h • SRT : 96 d • SS : 5000 mg/L • TMP : 5 kPa	• Reduction of fine particles, colloids and SMP • Limited deposition of fine particles and colloids on the membrane as cake layer served as a prefilter	Liu and Sun (2012)
	2.5 mM of $CaCl_2$	100 mg/L	Membrane-coupled EGSB	Synthetic wastewater	• HRT : 4 h • COD : 310–360 mg/L • pH : 7.0–7.5 • TMP : 30 kPa • Flow rate : 0.75 L/h	• Decrease in SMP concentration in the effluent by 47.7 - 60.7% • Decline in cake layer resistance by 42.8% • Delayed transition from pore blocking to cake filtration	Ding et al. (2015)
Polyaluminum chloride	N/A	500 mg/L	AnMBR	Anaerobic sludge	• HRT : 3.4 d • SRT : 40 d • TS : 30.16 g/L • VS : 1.2 g/L • Temperature : 35 °C	• Improved filterability of mixed sludge liquor • Decreased concentration of SMP and zeta potential of sludge • Reduced TMP increase rate	Yu et al. (2015)

	N/A	200 mg/L	AnCMBR	Phenol- and quinoline-containing wastewater	• HRT : 48 h • Flux : 4.32 LMH • MLSS : 35.77 g/L • MLVSS : 20 g/L • Temperature : 35 ± 1℃	• Changes in the structure of bulk sludge and cake layer • Changes in the component of SMP and EPS • Reduced specific resistance to sludge filtration	Wang et al. (2019c)
	N/A	500 mg/L	AnMBR	Excess anaerobic sludge	• HRT : 3.4 d • SRT : 40 d • TS : 30.16 g/L • VS : 13.22 g/L • Temperature : 35 ℃	• Enrichment of anaerobic microorganisms such as *Cloacimonetes* and *Smithella* • Slightly reduced ratio of VS/TS	Zheng et al. (2018)
Zeolite	Natural zeolite (0.2 – 1 nm of pore size, 3 of hardness, 2.1 g/cm^3 of density)	350 mL	AnFMBR	Campus domestic wastewater	• HRT : 2.5 h • COD : 130 ± 38 mg/L • pH : 7.2 ± 0.2 • Flux : 10 LMH • Temperature : 20 - 35 ℃	• Increase in SS removal rate by 22 % • TMP < 0.2 bar • Significant growth of anaerobic microbes on the surface of zeolite	Chen et al. (2019a)
Beads	Glass beads (soda-lime glass, 2500 kg/m^3 of density, Worf Glaskugeln, Germany)	4 mm as support layer	AnFMBR	Municipal wastewater	• HRT : 1.7 h • SRT : 46 d • COD : 369 ± 98 mg/L • pH : 7.0–7.5 • TMP : 30 kPa	• Increase in methane production by around 30% • Reduction of the fouling rate by around 95%	Düppenbecker et al. (2017a); Düppenbecker et al. (2017b)

					Temperature : 20 ℃ Upflow velocity : 24–37 m/h	Least abrasion by Al_2O_3 microfiltration membrane	
	Polyethylene terephthalate beads (3 mm of size and 1.3 of specific gravity)	0.4 v/v of packing ratio	AnFMBR	Synthetic wastewater	HRT : 3.75 h SRT : 37.5 d COD : 250 mg/L pH : 7.0–7.5 Temperature : 25 ℃ Flux : 10 LMH	Higher effectiveness when applying PET beads with bigger size and lower density Significant fouling mitigation by souring	Charfi et al. (2018b)

[a] AnCMBR, anaerobic ceramic membrane bioreactor; AnFMBR, anaerobic fluidized membrane bioreactor; AnMBR, anaerobic membrane bioreactor; EGSB, expanded granular sludge bed;

2.5 Powdered activated carbon (PAC) as a fouling reduction enhancer

2.5.1 Effects of PAC on membrane fouling

PAC can reduce the membrane fouling through three mechanisms. Firstly, at the initial stage of PAC addition, adsorption of organic matters occurs, which greatly removes the DOM from wastewater. Secondly, after initially adsorbing organic matter and becoming saturated, microorganisms aggregate on the porous surfaces of PAC particles as the supporting medium for attached bacterial growth (Chong, 2015). Finally, the formation of biologically activated carbon (BAC) promotes the degradation of pollutants and modifies the sludge properties, which is the most important mechanism. After the colonization of microorganisms, the planktonic microorganisms transform into biofilm. Once attached bacteria produce EPS, it helps not only the attachment of microorganisms to biofilm, but also the stabilisation of the biofilm structure. The active biofilm continues to biodegrade organic compounds as well as reduce the attachment of microorganisms on the membrane surface so that it can relieve the membrane biofouling (Gao et al., 2016; Korotta-Gamage and Sathasivan, 2017). In addition, the scouring effect of PAC also alleviates membrane fouling, as it can remove the deposited cake layer on the membrane surface while limiting the accumulation of foulants (Chong, 2015). Figure 2-2 illustrates the mechanism of fouling reduction when activated carbon is added.

More recent studies have confirmed that PAC has positive effects on sludge morphology, aggregation ability and microbial properties, and thus the pollutants removal mechanism can be enhanced. The study by Zhang et al. (2017c) reported that PAC addition in a submerged AnMBR was able to form larger floc size of the sludge compared to AnMBR without PAC. However, in case of long-term operation over 140 days, the sludge diameter rather decreased from 20.66 μm to 17.00 μm, which is a contrary result from previous research of PAC in aerobic MBR (Lei et al., 2019). This may have resulted from the fact that PAC enabled the generation of free living filamentous microbes after the long term operation and prevented the large floc size formation in mixed liquor. In addition, the sludge aggregation ability was able to be assessed using the total interaction energy which is a function of separation distance between the sludge surfaces. It could be calculated by summing Lifshitz-van der Waals energy, Lewis acid-base energy and repulsive or attractive electrostatic double layer energy (Yu et al., 2019). PAC addition showed highly negative value of total interaction energy per unit area of sludge, which indicated that the characteristics of sludge surface has transformed into hydrophobicity, and thus sludge cells adherence could be strengthened by stronger attractive interaction. This improvement of aggregation ability led to more stable sludge flocs and potentially reduced the EPS release, which mitigated the pore blocking and irremovable membrane fouling (Hu et al., 2014). The increased bacterial diversity and evolution of the bacterial community were also attributed to PAC addition creating additional microbial environment in the form of BAC, which promoted the enrichment and growth of some special functional bacteria. The enrichment of *Acinetobacter, Comamonas, Flavobacterium and Pseudomonas*, which can contribute to the formation of sludge flocs and degradation of organics, were highly promoted (Yu et al., 2019). When PAC was applied in anaerobic batch biofilm reactors as a biofilm carrier for the enhancement of refractory compounds degradation, it increased the abundance of *Methanothrix, Methanomassiliicoccus, and Methanobacterium* which were favourable for the methane production (Shi et al., 2019).

2.5.2 Use of PAC in aerobic MBR

The addition of PAC to aerobic MBR has been broadly used for the removal of organic and inorganic pollutants. The presence of 2 g/L of PAC in aerobic MBR treating pharmaceutical wastewater provided high COD removal rates of up to 98% as well as 99% removal of etodolac, which is one of the pharmaceutical active compounds (Kaya et al., 2016). Another study of MBR with 4 g/L of PAC treating coal gasification wastewater investigated enhancements of total phenols and ammonium removal efficiencies in addition to higher COD removal (Jia et al., 2014). Furthermore, MBR with PAC could effectively remove inorganic pollutants from groundwater, such as iron, manganese, and ammonia. The concentration of Fe^{2+}, Mn^{2+}, and NH^{4+}-N significantly decreased from 15 mg/L to 0.2 mg/L, 1.2 mg/L to 0.1 mg/L, and 2.2 mg/L to 0.05 mg/L, respectively (Du et al., 2017). Other studies showed that the PAC addition to MBR system reduced trihalomethane formation potential from the effluent, and effectively attemuated bacterial aerosols, which have negative effects on human health and also require excessice aeration during MBR processes (Gao et al., 2016; Noh et al., 2019). Another recent bit of research applied PAC to sulfur-based carriers for the removal of nitrate in MBR process. The result showed that the removal of nitrate improved due to the increased denitrification efficiency by adding PAC (Woo et al., 2020). These results demonstrated that the PAC addition could improve the biodegradability of both organic and inorganic matters.

At the same time, PAC addition was able to improve the diversity and richness of bacterial community. Recent study investigated the improvement in abundance of *Comamonas* and *Methanomethylovorans*, which can degrade the recalcitrant micropollutants, as well as *Cyanobacteria* and *Verrucomicrobia*, which play important roles in the nitrification process (Asif et al., 2020). MBR system with PAC also improved the manganese oxidizing bacteria, such as *Leptothrix*, *Pseudomonas*, and *Planctomyces*, and ammonia oxidizing bacteria, such as *Nitospira* and *Nitrosomonas*, which could efficiently remove inorganic pollutants (Du et al., 2017).

Many previous studies have applied PAC to aerobic MBR as an adsorbent to mitigate membrane fouling. Adsorbents provide the opportunity to adsorb disolved organic polymers like SMPs, hence it can reduce the propensity of membrane fouling (Iorhemen et al., 2016). When the 4 g/L of PAC was added to MBR, the TMP rise rate significantly decreased and promoted coagulation of colloids and small particles to transform into larger particles (Jia et al., 2014). In addition, the cake layer resistance, which is the predominant fraction in the total membrane resistance, could decrease with the PAC addition (Jamal Khan et al., 2012). Another study showed that the EPS concentration in the sludge could not be affected by PAC addition, while the EPS composition could be altered. The EPS composition, in terms of the protein and polysaccharide ratio, resulted in higher ratio which can lead to lower fouling potential (Mohamadi et al., 2020).

2.5.3 Use of PAC in AnMBR

As the usage of PAC has been demonstrated to be effective for the removal performance and membrane fouling mitigation, many researchers have also applied the PAC addition to AnMBRs. Since the adsorption capability of PAC was able to modify the sludge properties as well as remove organic and inorganic matters, it could efficiently reduce the membrane fouling and improve the performance of the AnMBR. In a study of Hu and Stuckey (2007), 1.7 g/L of PAC addition to submerged AnMBR showed significant benefits on the removal of fine colloidal particles as well as membrane flux improvement and TMP reduction. It was also found that PAC addition showed 22.4% of increase rate in COD removal, while GAC showed no significant increase. The reason why PAC was more efficient than GAC in terms of COD removal, might be due to the greater surface area per mass than GAC. Another study, which applied 1.7 g/L of PAC as well, showed 30% increased dissolved organic carbon (DOC) removal and decrease in SMP concentration (Vyrides and Stuckey, 2009). On the other hand, when the more PAC concentration of 4 g/L was added, the SMP was rather accumulated while having excellent COD and colour removal efficiency ((Baêta et al., 2016; Baêta et al., 2013). Several studies have revealed that the PAC addition not only decreased turbidity and colour, but also removed potential foulants such as fine colloids and SMP, which could lead to the reduction of fouling layer thickness (Baêta et al., 2012; Chong, 2015; Zhang et al., 2017c). Table 2-4 summarizes the effects of PAC addition on membrane fouling control in submerged AnMBRs.

PAC can also be beneficial to AnMBRs in terms of the removal of antibiotics, which have negative impact on membrane fouling. In fact, the existence of antibiotics in AnMBRs has worsened performance and membrane fouling, since it could lead to a decrease of the floc size and pH, and an increase in the secretion of EPS and SMP. Moreover, it could facilitate the development of microbial communities which had the most contribution to membrane fouling, such as *Firmicutes*, *Proteobacteria* and *Chloroflexi* (Cheng et al., 2018; Juntawang et al., 2017). According to a review by Cheng et al. (2018), the addition of antibiotics and combined antibiotics to anaerobic reactors, such as sulfamethoxazole (SMX), tetracycline (TC), erythromycin-tetracycline-sulfamethoxazole (ETS), and erythromycin-tetracycline (ST), could cause negative effects on pH, COD removal efficiency, and biogas production. Both the pH value and COD removal efficiency in anaerobic sequencing batch reactors significantly decreased when the high concentration of antibiotics, such as 45 mg/L of SMX, 8.5 mg/L of TC, and 46 mg/L of ETS, were added (Aydin et al., 2015; Cetecioglu et al., 2013; Cetecioglu et al., 2016). Biogas generation, which is inherently related to COD removals under anaerobic conditions, was inhibited as well, and the reason for this might be the methanogenesis process was sensitive to the presence of antibiotics in anaerobic processes. Likewise, 100 µg/L of SMX and TC in anaerobic/aerobic-MBR accelerated the rate of TMP rise and decreased the membrane fouling cycle from 25 days to 8 days. In addition, the fouling layer became denser and thicker with 20 µm of thickness. Furthermore, higher concentration of two antibiotics, 1000 µg/L of SMX and TC, resulted in further decrease in membrane fouling cycle to 4 days and 40 µm of the fouling layer thickness (Zhu et al., 2018). However, the addition of activated carbon could remediate these negative results. As a case in point, the addition of PAC into AnMBR increased removal efficiencies of five different pharmaceuticals including SMX, trimethoprim (Tmp), carbamazepine (Cbz), diclofenac (Dcf), and triclosan (Tcs) by approximately 5%-92%, as the adsorption of pharmaceuticals to PAC thermodynamically enhanced their biotransformation (Xiao et al., 2017).

Although the optimal dosage of PAC results in significant alleviation in membrane fouling, overdosing might have contrary results due to its potential to become a foulant. Akram and Stuckey (2008) proposed that appropriate amount of PAC should be added for the best improvement of performance of AnMBR and membrane fouling amelioration. In their research, PAC concentration of 1.67 g/L highly improved the flux by more than four times, while 3.4 g/L of PAC caused a decrease of flux and adsorption incapacity of PAC to higher concentration of biomass. The excessive dosage of PAC could result in poor membrane filtration due to the increased sludge viscosity caused by the presence of more extracellular polymers. Moreover, small PAC particles (8-35 µm) at high concentration in suspension increased turbidity of mixed liquor and caused more membrane pore blockage and abrasion (Skouteris et al., 2015). Therefore, the optimal PAC dosage is effective for flux improvement and adsorption of fine solutes, and regular replacement of aged PAC with fresh PAC is necessary.

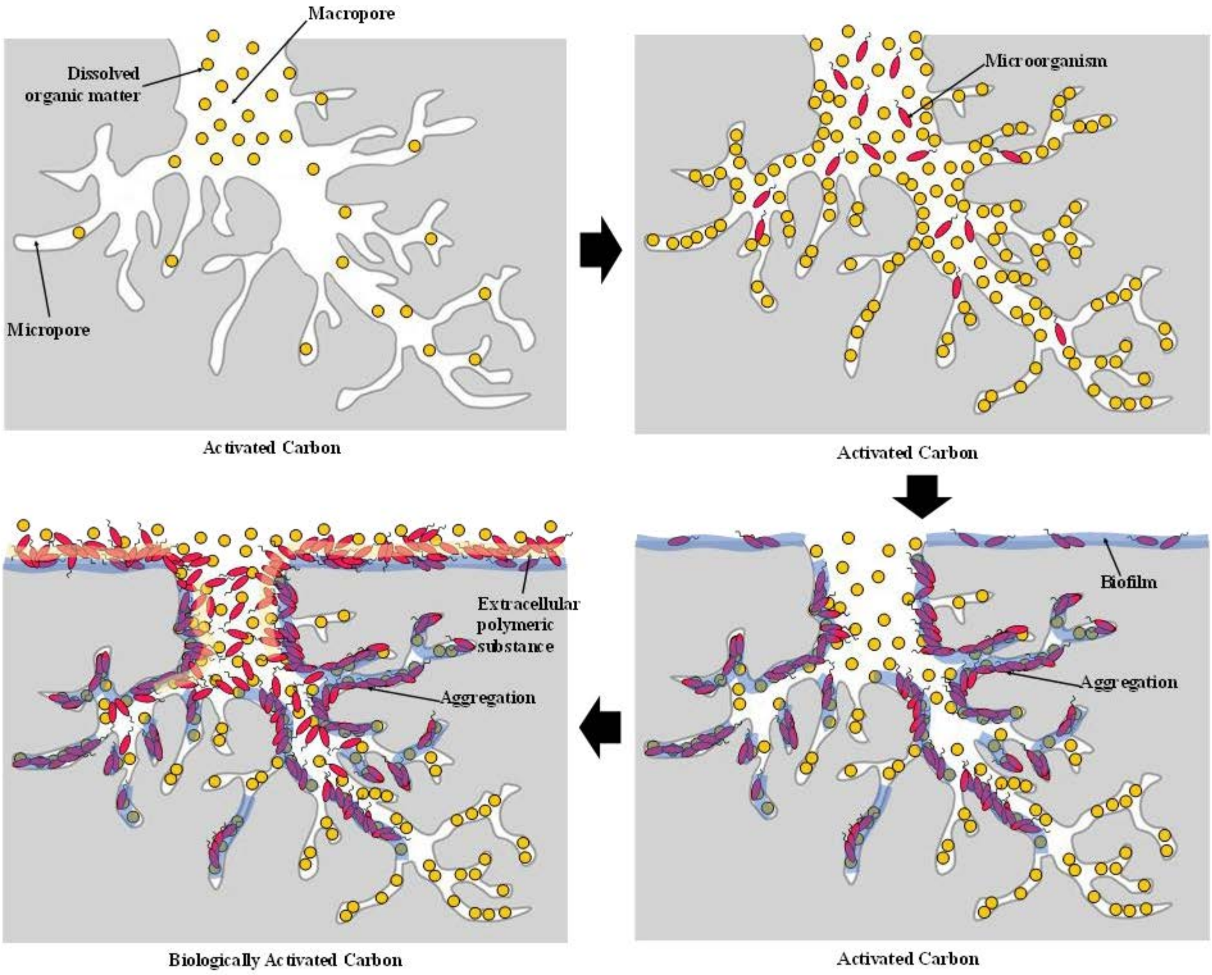

Figure 2-2. Mechanism of fouling reduction performance of activated carbon

Table 2-4. Effects of PAC addition on performance of submerged AnMBRs

AC supplier	Dose of AC	Feed water	Operating conditions	Effects on performance[a]	References
• Norits-Super • Total surface area of 1300 m^2/g	1.7 g/L	Saline sewage	• SRT : 250 d • HRT : 8, 20 d • OLR : 2 gCOD/L·d • Flux : 5–8 $L/m^2·h$ • Salinity : 0-35 gNaCl/L • Temperature : 35 ± 1 °C	• Decrease in TMP by 0.070 bar • Increase in dissolved organic carbon (DOC) removal by 30% in the reactor and 5% in effluent • Reduction of high MW compounds by 70% • Reduction of large flocs attached to the biofilm • Decrease in SMP	Vyrides and Stuckey (2009)
• Norit, Singapore • BET surface area of 925 m^2/g • average particle size of 22 µm	1 g/L	Synthetic sewage	• SRT : 213 d • HRT : 6 h • Flux : 5 $L/m^2·h$ • Feed COD : ~500 mg/L • Total nitrogen : ~100 mg/L • Temperature : 35 °C	• Enhanced removal of five selected pharmaceuticals (Tmp, Smx, Cbz, Dcf, Tcs) by about 5-92% • Increased biotransformation of Tmp by 4.5%, Smx by 18.8% and Tcs by 34.8%	Xiao et al. (2017)
• Norit, UK • Average particle size : 15-25 µm	400 mg/L	Synthetic sewage	• Flux : 15 $L/m^2·h$ • Feed COD : 500 mg/L • VSS : 5.0-5.5 g/L • Temperature : 35 °C • pH : 6–7	• Reduction in supernatant supra-colloidal particles, colloids and SMPs • Reduced thickness of fouling layer reduced • Declined levels of COD by 13% and proteins	Zhang et al. (2017c)
• Norit, UK	1.67, 3.4 g/L	Synthetic wastewater	• SRT : 250 d • HRT : 6 h • OLR : 16 gCOD/L·d • Feed COD : 4 g/L • Temperature : 35 ± 1 °C	• Enhanced performance during start-up period (i.e. shortened start-up duration, increased COD removal, declined SMP level,	Akram and Stuckey (2008)

			• Neutral pH	increased concentration of biomass and enrichment of microorganisms) • Adsorption of biodegradable low and high MW residual COD by PAC • Adsorption of fine colloids and dissolved organics by PAC • Improvement in flux (i.e, increase in flux from 2 to 9 L/m^2·h with 1.67 g/L PAC)	
• Synth®	4 g/L	Textile wastewater	• HRT : 24 h • Temperature : 35 ℃ • pH : 6.8–7.2 • Feed COD : 670 mg/L • COD:N:P : 350:5:1	• Enhanced removals of COD, VFA, turbidity and colour by about 11%, 8%, 43% and 69%, respectively • Increased reactor stability • Enhanced membrane permeability (higher critical flux) • Adsorption of toxic compounds, aromatic amines and VFA by PAC • Increased accumulation of SMP	Baêta et al. (2016); Baêta et al. (2012)
• Synth®	4 g/L	Domestic sewage	• HRT : 24 d • COD:N:P : 350:5:1 • OLR : 0.53 kg/m^3·d • Temperature : 35 ℃ • pH : 6.5–7.5	• Enhanced COD and colour removal by about 9% and 6.7-12.7%, respectively • Increased reactor stability • Less accumulation of VFA • Adsorption of aromatic amines by PAC	Baêta et al. (2013)
• Extra pure charcoal powdered activated carbon	1, 3, 5 g/L	Palm Oil Mill Effluent (POME)	• SRT : 30 d • HRT : 6 d • Feed COD: 4.74 ± 1 g/L • Temperature : 35 ℃ • pH : 7-8	• Increased COD removal efficiency at higher PAC dosage • Increased floc size at higher PAC dosage	Chong (2015)

			• At mesophilic condition	• More reduction of EPS concentration and membrane fouling at higher PAC dosage	
• Darco KB-B, Norit, US	5 g/L	Synthetic wastewater	• SRT : 67 d • HRT : 8.75 h • Flux : 5 L/m^2·h • Feed COD : 550 ± 10 mg/L • MLSS : 5 ± 0.1 g/L • Temperature : 22 ± 1 °C • pH : 7	• Increased nutrient, COD and TOC removal • Reduced TMP rise rate • Decreased SMP and EPS concentration, while increased protein ratio • Increased zeta potential of the sludge • Reduced the abundance of filamentous bacteria	This study

[a]Cbz, carbamazepine; Dcf, diclofenac; Smx, sulfamethoxazole; Tcs, triclosans; Tmp, trimethoprim

Table 2-5. Advantages and disadvantages of fouling reduction enhancers

Enhancers	Advantages	Disadvantages
PAC	• High adsorption capacity • Increase removal efficiency of COD, fine colloids, colour, and antibiotics • Improve aggregation ability leading to stable sludge flocs • Large surface area than GAC	• Decrease sludge particle size in long-term operation • Overdosing can increase membrane fouling due to the PAC being a potential foulant
GAC	• Improve methane production • Enhance COD, SMP and pharmaceuticals removal • GAC fluidization has lower energy requirement than gas sparging • Scouring effect • Recover can be done by thermal process after adsorption capacity exhausted	• The abrasion can aggravate fouling • Large particles require more energy for fluidization
Biochar	• Improve hydrogen and methane production by enrichment of microorganisms • Decrease SMP and proteins of EPS • Enhance COD removal efficiency and sludge granulation	• Less efficient specific surface area than AC due to non-activation
Waste yeast	• High degradation capacity of phenolic and toxic compounds • Low tendency to adhere on membrane surface • Enhance biogas production	• No significant change in VFA accumulation • Increase SMP and aromatic group with high molecular weight
Iron	• Enhance sludge granulation and stabilization • Improve methane production and COD removal • Eliminate H_2S gas • Enhance mixed liquor filterability • Advantageous for long-term operation due to the formation of a precipitate	• Formation of thick cake layer • Cause more severe membrane fouling due to the high density of Fe-rich fouling layer
Calcium	• Enhance bioflocculation • Improve accumulation of biomass and degradation of butyrate and acetate acid • Reduce fine particles, SMP,	• Overdosing can lead to precipitation, declined methanogenic activity and inorganic fouling

	EPS and colloids	
Polyaluminum chloride	• High charge neutralization capacity can lead to enlarged floc size and high filtration performance • Improve the abundance of anaerobic microorganisms and hydrogen yield • Reduce the composition rate of carbohydrate in SMP and EPS • The cake layer become more porous and looser	• High concentration can inhibit SCFAs production and decrease phosphorous
Zeolite	• High ion exchange capacity can enhance ammonia and heavy metals removal • Improve methane production, COD, nitrogen and phosphorous removal • Enhance sludge settlement by bacterial attachment and aggregation	
Beads	• Fluidization of PET beads can act as turbulence promotors and scouring media • Polymer-based gel beads have cost effectiveness, high bio-compatibility, high stability for long-term use, and porous structure for microbial attachment and aggregation • The size and density can be controlled by different synthesis conditions • Increase COD removal rate and methane production	• Fluidization of glass beads can damage membrane by abrasion

CHAPTER 3

Experimental investigations

3.1 Introduction

The first study was carried out with synthetic wastewater and to find out an optimal dosage of PAC to conventional AnMBR with a hollow fiber membrane. The second study was conducted to investigate the long-term effects of the optimal PAC dosage on AnMBR, and the performance was also compared to that of an AnMBR without PAC addition. The synthetic wastewater, which has 550 mg/L of COD and C: N: P ratio of 100:5:1 containing trace contaminants, was used for the two experiments. Thus, this chapter includes the characteristics of the sludge and synthetic wastewater in the form of chemical mixed solvent, experimental set-up, operating conditions, as well as materials and analysis methods during the whole experiments.

3.2 Wastewater and sludge used in the experiments and their characteristics

3.2.1 Anaerobic sludge and wastewater

The mixed liquor seed sludge used in this study was obtained from a domestic sewage treatment plant in Sydney, Australia. The sludge during the start-up of this study had a MLSS concentration of 7-8 g/L. The nitrogen has been purged into the system using the diffused aeration tubes on a regular basis. The sludge was acclimatized for 90 days until the stable removal rates of total organic carbon and nutrient were gained.

The synthetic wastewater was used in this experiment, which has feed COD concentration of 550 mg/L and the C: N: P ratio of 100:5:1. Glucose ($C_6H_{12}O_6$), sodium nitrate ($NaNO_3$), and potassium phosphate (KH_2PO_4) were used as the major sources of carbon, nitrogen, and phosphorus respectively, and 80% of $NaNO_3$ and 20% of NH_4Cl were mixed for nitrogen source. The specific concentration of the nutrients in the feed solution is summarized in table

1. The following components have been used with the specified concentrations to make the stock solution for synthetic feed wastewater. This composition was based on the synthetic wastewater used in the previous study (Khan, 2019).

Table 3-1. Composition of different components in synthetic feed wastewater

Compounds	Concentration (mg/L)
Organics and nutrients	
Glucose ($C_6H_{12}O_6$)	514
Sodium Nitrate ($NaNO_3$)	49.90
Ammonium chloride (NH_4Cl)	7.85
Potassium phosphate (KH_2PO_4)	9.03
Trace nutrients	
Magnesium sulphate ($MgSO_4 \cdot 7H_2O$)	9.31
Sodium molybdate dehydrate ($Na_2MoO_4 \cdot 2H_2O$)	2.31
Calcium chloride ($CaCl_2 \cdot 2H_2O$)	0.68
Manganese chloride ($MnCl_2 \cdot 7H_2O$)	0.50
Zinc sulphate ($ZnSO_4 \cdot 7H_2O$)	0.81
Ferric chloride anhydrous ($FeCl_3$)	2.66
Cupric sulphate ($CuSO_4 \cdot 5H_2O$)	0.72
Cobalt chloride ($CoCl_2 \cdot 6H_2O$)	0.77
Yeast extract	55

3.3 Analytical methods

3.3.1 Mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS)

The measurement of MLSS and MLVSS concentrations were conducted by sieving a well-mixed sample with 1.2µm filter paper. The remaining residue on the filter was dried in an oven at 105 ºC for 2 h. The increment in weight of the filter paper indicated the MLSS in the sample. Subsequently, the filter paper was ignited in a furnace at 550 ºC for 20 min. The weight reduced during ignition represented the MLVSS in the sample.

3.3.2 Other measurements

Nutrients measurement, such as ammonium (NH_4^+-N), nitrate (NO_3—N), nitrite (NO_2—N), and phosphate (PO_4^3—P), were carried out using the test kit (Merck Millipore, Burlington, USA) and a photometer (Spectroquant NOVA 60, Merck). The pH of the reactor was measured by using pH meter every day (HI9025, Hanna Instruments, Limena, Italy).

Total organic carbon (TOC) concentration of the influent and effluent was analysed using a TOC analyzer (Analytik Jena Multi N/C 2000). The chemical oxygen demand (COD) of influent and effluent was analysed using medium range COD reagent test kit (HI93754B-25, Hanna Instruments Australia, Melbourne, Australia) and DR/2000 spectrophotometer (HACH) according to Standard Methods.

Membrane fouling resistance was measured to examine the filtration characteristics by using the resistance-in-series model, when J is the permeate flux, ΔP is the TMP, μ is the permeate viscosity, and R is membrane resistance (Deng et al., 2015):

$J = \Delta P/\mu R$ .... (1)

The total resistance, R_t, consists of three components, which are R_m, R_c, and R_p. It can be described as follows (Deng et al., 2014):

$R_t = R_m + R_c + R_p$ .... (2)

R_m is an inherent membrane resistance, R_c is a cake layer resistance which came from the deposited sludge particles on the membrane surface, and R_p is a pore blocking resistance which was brought about the adsorption of dissolved matters and colloids with similar size to membrane pore size (Deng et al., 2014). R_m was measured by using various fluxes with distilled water with a cleaned membrane before the operation. At the end of each operation, R_t was obtained with fouled membrane by the same methods as R_m. The cake layer was then removed using brush and distilled water from the membrane surface and $R_m + R_p$ was determined. R_c was calculated by deducting $R_m + R_p$ from R_t, and thus R_p could be calculated by deducting R_m from $R_m + R_p$.

The 10 mL of the mixed liquor was extracted from each reactor at the end of the experiment for the measurement of zeta potential. Zetasizer (nano instrument ZS Zen3600, UK) was used to analyze the zeta potential of the sludge.

3.3.3 Extracellular polymeric substances (EPS) and soluble microbial products (SMP) analysis

The concentrations of proteins and polysaccharides in SMP and EPS from the mixed liquor were analysed based on the methods as described below. The 30 mL of mixed liquor was first centrifuged at 3000 rpm for 30 min after 30-min storage in the fridge. The supernatant was again centrifuged at 3000 rpm for 30 min, while the pellet was resuspended in 30 mL of phosphorous buffer solution. The second supernatant was sieved by 0.45 μm syringe filter, which was measured towards protein and polysaccharide of SMP. The amount of cation exchange resin (Dowex^{TM} Marathon^{TM} C, Na^+ form, Sigma-Aldrich, Bellefonte, PA) was calculated according to the MLVSS of the sample and then added on the resuspended pellet. This mixture was centrifuged at 2000 rpm for 2 hours, and the following supernatant was filtered by 1.2 μm syringe filter for the measurement of protein and polysaccharide of EPS. Protein analysis were made according to modified Lowry method (Sigma, Australia), and polysaccharides were analysed based on Anthrone–sulfuric acid method (Deng et al., 2014).

The membrane module was taken out from the reactor after finishing the experiment for the analysis of SMP and EPS of the cake layer. The cake layer was collected by brushing the membrane surface, and then was dissolved in 30 mL of distilled water. Subsequently, the SMP and EPS of the collected cake layer were analysed by the same method as above.

3.3.4 DNA extraction and quality monitoring

The samples were taken out from each reactor and duplicated after the entire operation time for microbial community profiling. Samples were mixed with 100% ethanol (1:1 v/v) and stored at -20 ºC before DNA extraction. Genomic DNA extraction was conducted using QIAamp DNA Stool Mini Kit (Qiagen) by following the manuals. The concentration of the extracted DNA, its integrity and purity were analysed by NanoDrop® spectrophotometer. DNA concentrations of all samples were normalized to 20 ng/μL using DNase/Pyrogen-Free Water before the samples were sent to the sequencing facility.

3.3.5 Amplicon sequencing and bioinformatics analysis

The universal primer set Pro341F (5’-CCTAYGGGRBGCASCAG-3’) and Pro806R (5’-GGACTACNNGGGTATCTAAT-3’) was used to target both bacterial and archaeal 16S rRNA V3–V4 regions for depiction of the whole microbial community (Takahashi et al., 2014). Paired-end amplicon sequencing (2 x 300 bp) was conducted on the Illumina MiSeq platform (UTS Next Generation Sequencing Facility, Sydney, Australia), using next-generation sequencing (NGS) method. Raw sequence data were produced with the Illumina *bcl2fastq* pipeline.

Raw reads were brought into Quantitative Insights into Microbial Ecology (QIIME) 2 (version 2020.11.1) for computational analysis (Bolyen et al., 2019). Quality filtering, denoising (primer and read trimming), paired-end reads merging, dereplication, chimera filtering and amplicon sequence variants (ASVs) clustering (≥ 97% similarity) were performed using the *q2-dada2 denoise-paired* plugin (Callahan et al., 2016). Reverse reads sequences were truncated at position 240 in the 3’ end due to decrease in quality. Reads were mapped back to ASVs with a minimum identity of 97% to obtain the number of reads in each ASV.

Taxonomy was assigned to ASVs using the *q2-feature-classifier* (Bokulich et al., 2018) *classify-sklearn* Naïve Bayes taxonomy classifier against the SILVA database (release 132) (Glöckner et al., 2017; Quast et al., 2012; Yilmaz et al., 2013) with a confidence of 0.7. All ASVs were aligned with mafft (Katoh et al., 2002) and used to construct phylogenetics tree with FastTree2 (Price et al., 2010) via the *q2-phylogeny align-to-tree-mafft-fasttree* pipeline. Alpha-diversity metrics including Observed ASVs and Shannon index (Shannon, 1948), beta diversity metrics including weighted UniFrac (Lozupone et al., 2007), unweighted UniFrac (Lozupone and Knight, 2005), Jaccard distance, and Bray-Curtis dissimilarity were estimated using *q2-diversity core-metrics-phylogenetic* pipeline after samples were rarefied (subsampled without replacement) to 18,000 sequences per sample.

3.4 Optimization of PAC dosage to AnMBR

3.4.1 Characteristics of powdered activated carbon

PAC (Darco KB-B, Norit, US) was used in this study. Its particle size was from 100 to 325 mesh, and had 500–1000 m^2/g of surface area. The cost of PAC was 0.15 AUD/g, which was lower than other chemical additives. PAC was first washed for three times with milli-Q water and dried at 105 °C for overnight before use. While 2.5 of the PAC to biomass ratio with 5 g/L of PAC and 2 g/L of MLSS in the previous study showed the best flux improvement, the range of PAC dosages in recent studies regarding PAC addition to AnMBR was 0.4–5 g/L which had less than 1 of PAC to biomass ratio (Chong, 2015; Park et al., 1999; Zhang et al., 2017c). Thus, the three PAC dosages for the optimisation were determined to be 1, 3, and 5 g/L, within the range of MLSS of the AnMBRs. PAC was dosed only at the initial stage of the experiment. For 1 g/L of PAC, 3.5 g of PAC were added, and for 3 g/L of PAC, 10.5 g of PAC were added. In case of 5 g/L of PAC, 17.5 g of PAC were added. PAC was dosed only at the initial stage of each operation.

3.4.2 Experimental conditions

The continuous AnMBR system, which has working volume of 3.5 L, and hollow fibre membrane (PVDF, Pore size 0.07–0.1 μm) with 1.00 mm of the inner diameter and 2.20 mm of outer diameter, and 0.08 m^2 of surface area, was used for this study. Before the PAC addition, the initial MLSS concentration were adjusted to 5 g/L, and MLVSS/MLSS ratio was over 0.8 in each experiment. No excess sludge withdrawal was applied during each set of operation. The pH value and the temperature were kept constant at around 7 and 22 ± 0.1 °C, respectively. A constant pH value was maintained at around 7 by adding $NaHCO_3$ or H_2SO_4 to the feed solution. The AnMBR was operated at 5 LMH of flux. The synthetic wastewater was transported into each reactor using an influent pump at 6.67 mL/min of flow rate and the effluent pump was used as well at 8.33 ml/min with circulation of 8 minutes pumping and 2 minutes break. As the flow rate of influent was kept constant at 6.67 mL/min, the dilution rate in this study was 0.114 h^{-1}. The determination of these experimental conditions was based on the previous study, which demonstrated that the highest VFA yield with simultaneous production of methane were achieved at 8 hrs of HRT, 550 g/L of COD and pH 7 (Khan, 2019). Details of operating conditions are presented in table 3-2, and a simple schematic diagram of AnMBR is shown in Figure 3-1.

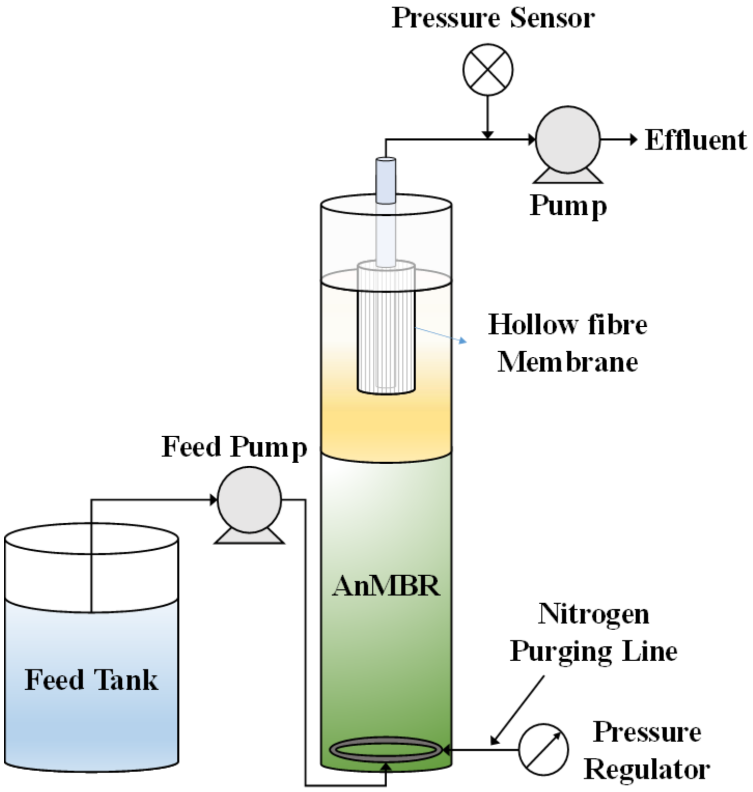

Figure 3-1. Schematic diagram of experimental set up of AnMBR

12L of the feed wastewater was prepared every day for one-day feeding and was fed using the feed pump after 15-20 minutes of nitrogen purging. In the meantime, both influent line and effluent line were cleaned every other day to prevent blocking. In addition, the reactor was purged with nitrogen once a week to take oxygen or unexpected air out. For nutrient analysis, sampling was recorded every 3 days during the whole operation. For COD and TOC analysis, sampling was also recorded every 3 days and assessed after acidified and refrigerated. The reactor was first continuously operated until the transmembrane pressure (TMP) reached over 35 kPa, and after the chemical cleaning of membrane, the operation continued until the TMP reached over 35 kPa again, until the total 25 days of operation. For the chemical cleaning of membrane, membrane was first placed in 0.5% of citric acid for 6 hours for removing inorganic foulant, and then put in 0.4% of sodium hydroxide for 6 hours to remove organic substances. Finally, it was put in 0.8% of sodium hypochlorite for another 6 hrs for removal of microorganisms, bacteria, and algae. As soon as the membrane was separated from the reactor for the cleaning, nitrogen gas was purged for 5 minutes to remove unexpected oxygen and maintain the anaerobic condition in the reactor. While the membrane was immersed in the cleaning solution, the reactor was kept feeding with synthetic wastewater under anaerobic condition so that anaerobiosis can be maintained.

Table 3-2. AnMBR operating conditions

Operating parameter	**Value**
MLSS (g/L)	5 ± 0.1
MLVSS (g/L)	4.5 ± 0.1
COD in feed solution (mg/L)	550 ± 10
HRT (h)	8.75
SRT (d)	25
Flux ($L/m^2 \cdot h$)	5
Temperature (°C)	22 ± 1
pH	7.0 ± 0.1
PAC dosage (g/L)	1, 3, and 5

3.5 Long term experimental investigation of PAC addition to AnMBR

3.5.1 Experimental conditions

The two AnMBR systems, which have working volume of 3.5 L and the same membrane module described in section 3.3.2, were operated at the same time in the continuous mode. Both reactors had the same operation conditions as the previous study, except for the PAC dosage, which is expressed in table 3-3, and the same schematic diagram of the reactors is the same as figure 3-1. The 8-minute suction and 2-minute relaxation mode was also applied for the two AnMBRs. The same characteristics of PAC of was added with the concentration of 5 g/L to one of the AnMBRs only at the beginning of the operation. No excess sludge withdrawal was applied during the operation.

Two buckets of 12 L feed wastewater were prepared every day for one-day feeding of the two reactors and nitrogen was sparged for 15-20 minutes. In the meantime, every-day cleaning of both influent line and effluent line, and nitrogen sparging once a week were also applied for both reactors. For nutrient, TOC and COD analysis, sampling was recorded every 3 days until it became stable and then was recorded every 5 days during the rest of the operation. For SMP and EPS analysis of mixed liquor, sampling was also recorded at the first day and at the last day of each operation set. The SMP and EPS of cake layer was also measured after the end of each operation set. For the analysis of microbial community, duplicated samples from each reactor were collected at the end of the whole operation period. In this study, the COD removal was measured with inclusion of SMP fraction as an indicator of treatment and SMP was measured as an indicator of membrane fouling propensity. The two reactors were operated until the TMP reached at 35 kPa, and after the chemical cleaning of membrane, the operation continued until the TMP reached over 35 kPa again. This value was based on the previous study which reported that the membrane required chemical cleaning when the TMP reached at

35 kPa (Deng et al., 2015). The membrane was chemically cleaned as the same process described in section 3.3.2.

Table 3-3. Operating conditions of two AnMBRs

Operating parameter	**AnMBR with PAC**	**AnMBR without PAC**
MLSS (g/L)	5 ± 0.1	5 ± 0.1
MLVSS (g/L)	4.5 ± 0.1	4.5 ± 0.1
COD in feed solution (mg/L)	550 ± 10	550 ± 10
HRT (h)	8.75	8.75
SRT (d)	67	63
Flux ($L/m^2 \cdot h$)	5	5
Temperature (°C)	22 ± 1	22 ± 1
pH	7.0 ± 0.1	7.0 ± 0.1
PAC dosage (g/L)	5	0

CHAPTER 4

Optimization of powdered activated carbon dosage to anaerobic membrane bioreactor

4.1 Materials and methods

4.1.1 Experimental set-up

A single stage AnMBR having 3.5 L of working volume was operated continuously using synthetic wastewater for this study. Three sets of experiments were conducted with different PAC dosage, which are 1, 3, and 5 g/L. Each set of experiment was operated for a total of 25 days, including one chemical membrane cleaning during each operation. The membrane was chemically cleaned whenever the TMP increased over 35 kPa. 3.5 g, 10.5 g, and 17.5 g of solid powdered PAC (Darco KB-B, Norit, US) were dosed only at the initial stage of each operation with no sludge withdrawal.

Details on the experiment materials and methods are given in Chapter 3.

4.2 Results and discussion

4.2.1 Nutrient removal performance

During the entire 25-day operation of each experiment, the nitrate removal rate remained steadily at around 77% of removal rate despite slight fluctuation. Compared to 1 g/L and 3 g/L of PAC addition, 5 g/L of PAC showed approximately 1% higher nitrate removal rate as well as remarkable increase after 13 days of operation. This could be attributed to the fact that the microorganisms had enough time to contact the feed wastewater continuously and the denitrification had been processed efficiently. In terms of phosphate removal rate which was expected to be below 10% in the ideal anaerobic process, they significantly fluctuated, due to the replacement of new anaerobic sludge before starting each experiment. The sludge replacement might have caused the increase of phosphate removal rate due to the biomass growth. Although there were significant fluctuations in the experiments of three dosages, 5 g/L of PAC addition was able to lead to far less fluctuation compared to the other two dosages of addition. Moreover, while the highest removal efficiency reached up to 74.29 % and 46.26% with 1 g/L and 3 g/L of PAC, respectively, 31.06% was the highest rate in 5 g/L experiment. Additionally, the 5 g/L of PAC addition showed the lowest phosphate removal rate between 11.45% and 13.01% during the final 5 days of operation. The nitrate and phosphate removal performances of AnMBR are presented in figure 4-1 and 4-2.

The ammonia removal rate had large fluctuations, but ended up remaining around 0–10.34% of removal rate during the final 7 days of operation of every experiment. This means that the oxygen remaining in the reactor has been removed compared to the initial operation time, and the ammonia removal rate was overall lower than AnMBR with 1 g/L of PAC. Additionally, nitrite was hardly removed during the whole operation of each experiment, as there was little difference between the nitrite concentration in the influent and effluent. Overall, the highest dosage of PAC (5 g/L) resulted in the improvement of nitrate removal rate and the lowest phosphate removal rate, which well describes the anaerobic process. Figure 4-3 and 4-4 illustrate the removal performances of ammonia and nitrite during the operations, while table 4-1 shows the average concentration of influent and effluent, and removal efficiency with different PAC dosages.

Table 4-1. Average concentration of influent and effluent, and removal efficiency with different PAC dosages

PAC dosage	**Average concentration**	**COD**	**TOC**	**NO_3-N**	**NH_4^+-N**	**PO_4^{3}-P**	**NO_2-N**
0 g/L	Influent (mg/L)	555.54 ± 56.24	77.46 ± 12.93	10.21 ± 1.45	4.59 ± 1.87	2.06 ± 0.37	0.05 ± 0.02
	Effluent (mg/L)	252.69 ± 31.17	30.44 ± 3.82	2.79 ± 0.70	3.98 ± 1.58	1.68 ± 0.32	0.04 ± 0.03
	Removal efficiency (%)	54.39 ± 4.65	59.90 ± 6.91	72.57 ± 5.98	10.54 ± 5.37	18.07 ± 4.21	9.95 ± 14.08
1 g/L	Influent (mg/L)		51.55 ± 2.15	9.81 ± 0.45	3.68 ± 1.55	2.64 ± 0.70	0.02 ± 0.02
	Effluent (mg/L)		24.70 ± 4.94	2.24 ± 0.42	2.79 ± 0.67	1.65 ± 0.28	0.01 ± 0.00
	Removal efficiency (%)		52.22 ± 8.41	77.18 ± 3.76	20.77 ± 11.81	33.24 ± 18.89	12.96 ± 20.03
3 g/L	Influent (mg/L)	516 ± 21.28	75.33 ± 14.53	9.56 ± 2.42	3.03 ± 1.23	2.44 ± 0.56	0.02 ± 0.01
	Effluent (mg/L)	230 ± 30.21	22.54 ± 5.64	2.24 ± 1.18	2.71 ± 1.09	1.74 ± 0.14	0.02 ± 0.00
	Removal efficiency (%)	55.24 ± 7.45	69.20 ± 8.53	77.73 ± 7.12	9.67 ± 7.58	26.38 ± 12.18	5.56 ± 16.67
5 g/L	Influent (mg/L)	574.75 ± 45.88	76.71 ± 18.43	9.41 ± 0.44	3.27 ± 1.57	2.62 ± 0.20	0.02 ± 0.00
	Effluent (mg/L)	222.75 ± 10.5	26.96 ± 6.01	2.04 ± 0.56	2.49 ± 0.74	2.06 ± 0.3	0.02 ± 0.01
	Removal efficiency (%)	61.07 ± 3.54	64.30 ± 5.85	78.29 ± 5.86	17.91 ± 15.40	21.82 ± 7.23	11.11 ± 22.05

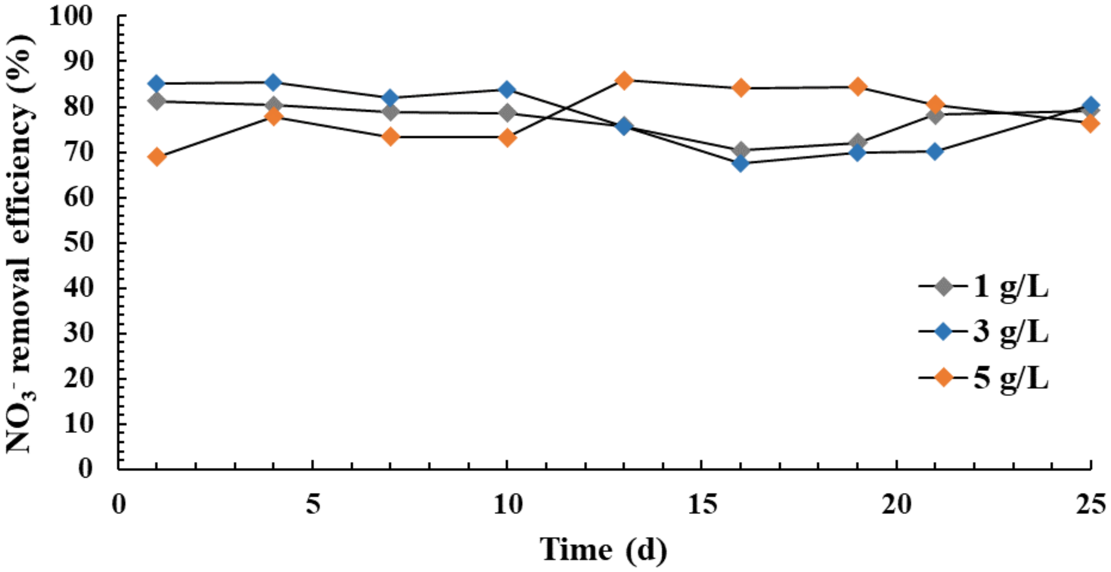

Figure 4-1. Nitrate removal performance of AnMBR

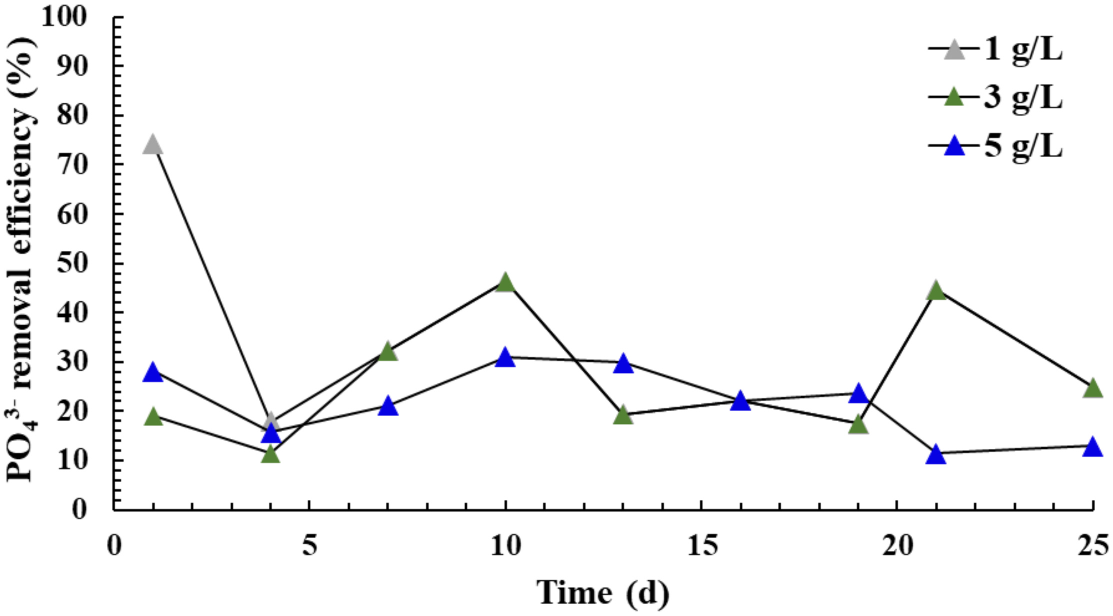

Figure 4-2. Phosphate removal performance of AnMBR

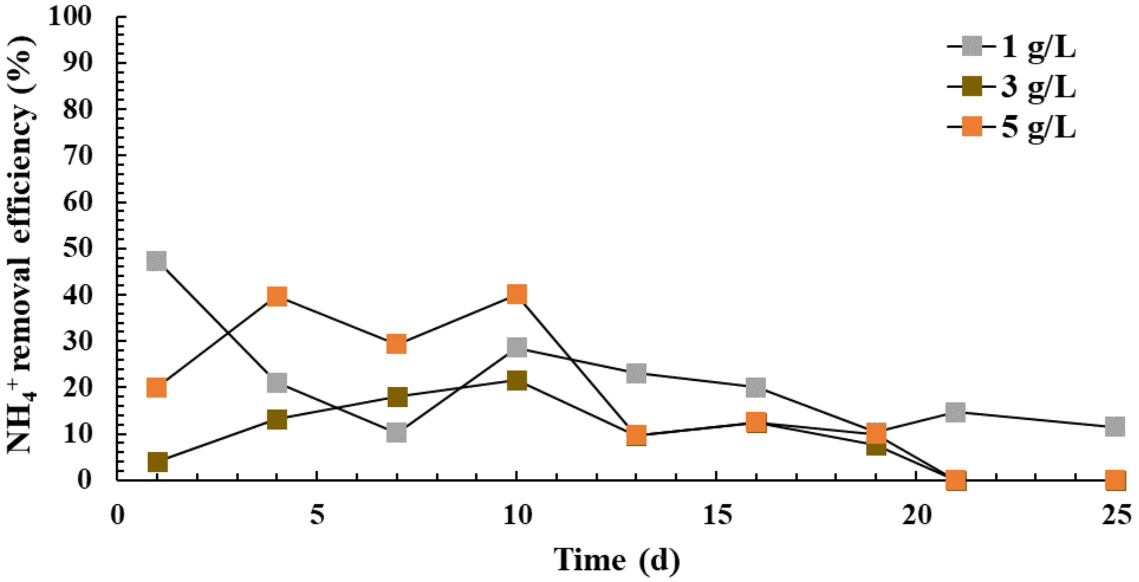

Figure 4-3. Ammonium removal performance of AnMBR

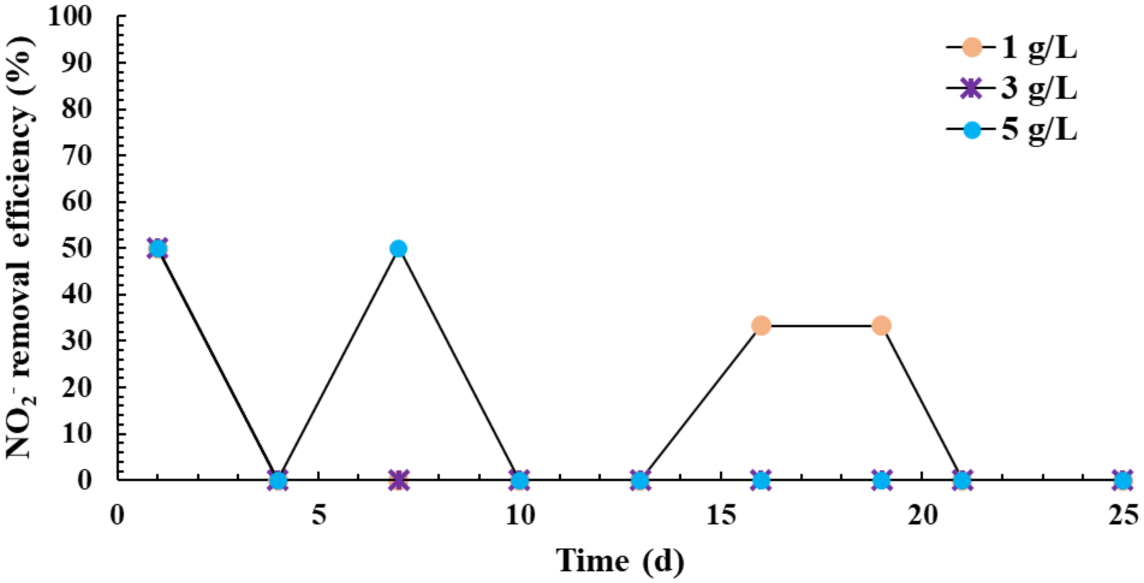

Figure 4-4. Nitrite removal performance of AnMBR

4.2.2 TOC and COD removal performance and membrane fouling

During the experiment of 3 g/L and 5 g/L of PAC, TOC removal rate were on average 69.20% and 64.30%, respectively, while the experiment with 1 g/L of PAC showed 52.22% of removal rate. In terms of COD removal rate, 5 g/L of PAC resulted in 7 % higher COD removal rate than 3 g/L of PAC. This result showed that higher concentration of PAC addition facilitated significant removal of organic matter by effective adsorption and biodegradation. The TOC and COD removal performances of AnMBR are shown in figure 4-5 and 4-6.

The membrane fouling behaviour can be influenced by the type of membrane, sludge properties, feed water properties, and operating conditions (Guo et al., 2012). In these three experiments, the membrane module, characteristics of sludge and feed water, and operating conditions were kept constant, except for the PAC dosage. Although only the PAC dosages were different, the fouling pattern changed along with the TMP changes. The variations of TMP during the operation of AnMBR were described in figure 4-7. Every experiment with a different PAC dose started with around 2 kPa of TMP. In the first week of every operation, no significant TMP development was observed staying under 10 kPa. After the first week, the TMP development started to increase and varied according to the PAC concentration. It indicated that soluble organics and nutrients in the feed started to attach and deposit on the membrane surface (Khan et al., 2019). However, there was a significant difference in time to reach up to 35 kPa. In the experiment with 1 g/L of PAC, the TMP reached at 36.03 kPa after 16 days of operation, and then the membrane was cleaned chemically. After the membrane cleaning, TMP started again from 2.55 kPa and increased to 35.29 kPa within 9 days. TMP in the AnMBR with 3 g/L of PAC resulted in the highest increase rate compared to the other two dosages. It took 13 days to reach at 36.03 kPa and after membrane cleaning, it took 12 days to be 37.68 kPa. On the other hand, the TMP with 5 g/L of PAC addition resulted in 37.5 kPa at the 15th day, and after membrane cleaning, it started from 2.41 kPa and reached to 27.36 kPa at the 25th day of operation, which was the slowest TMP increase rate. Previous study showed that the higher PAC dosage (5 g/L) resulted in larger floc size in AnMBR compared to the lower PAC dosage (1 g/L), accommodating more adsorbed microorganisms (Chong, 2015). This implied that 5 g/L of PAC might have enlarged the sludge particle size by adsorption of PAC, resulting in the most effective membrane fouling mitigation. Thus, 3 g/L of PAC addition could result in the worst membrane fouling, as the TMP increase rate was the highest compared to other dosages.

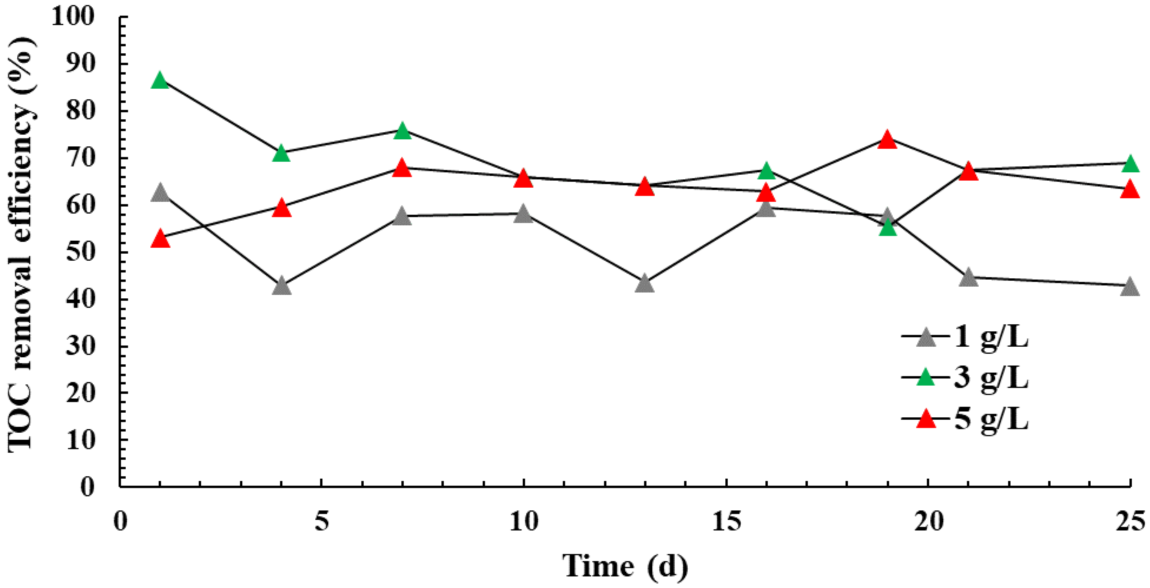

Figure 4-5. TOC removal performance of AnMBR

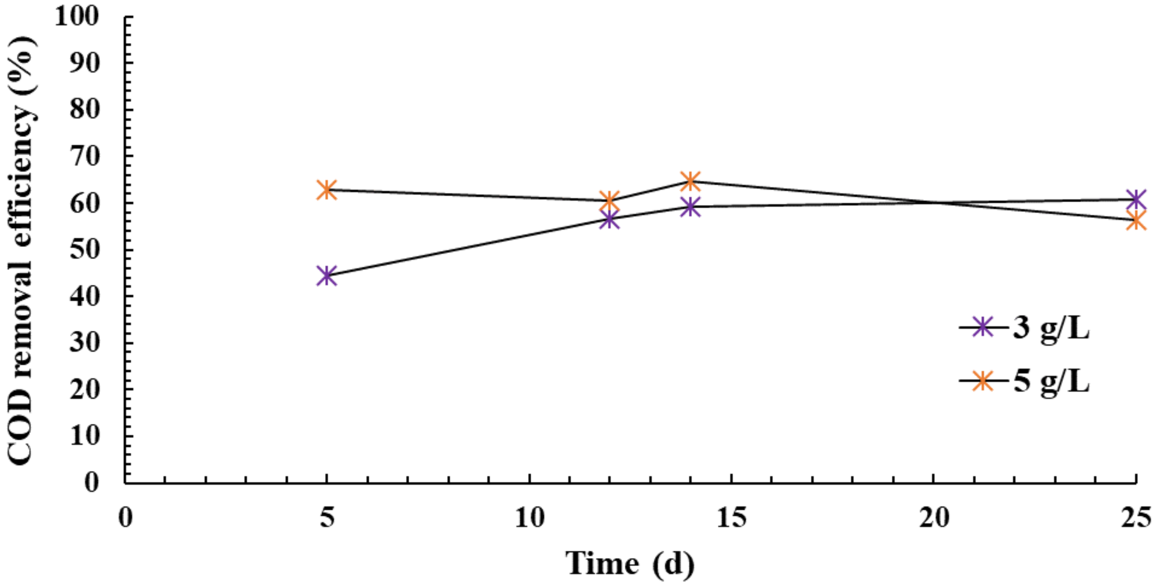

Figure 4-6. COD removal performance of AnMBR

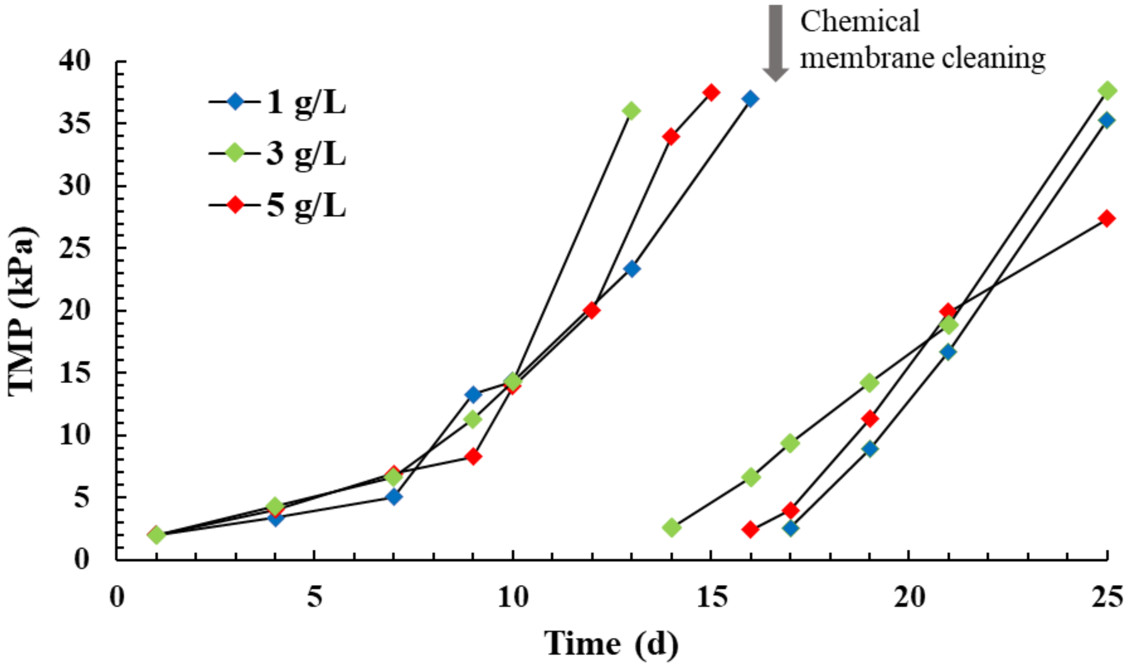

Figure 4-7. The variations of TMP during operation of AnMBR

4.3 Conclusions

Three experiments were conducted with 1 g/L, 3 g/L, and 5 g/L of PAC addition to conventional AnMBR. Each experiment was conducted for 25 days to investigate the effects of different PAC dosage on removal performance of conventional AnMBR in terms of TOC, COD and nutrient removal, as well as TMP change related to membrane fouling. In every experiment, the nitrate, nitrite and ammonium removal rate were close to the ideal rate in anaerobic process, while phosphate removal showed slightly higher removal rate during the whole operation, which is ideally expected to be 0–10% of removal rate. 5 g/L of PAC addition, particularly, showed the highest nitrate removal rate and the lowest phosphate removal rate. On the other hand, TOC removal performance showed the best removal rate in 3 g/L of PAC, remaining at average 69.20% despite a slight fluctuation. However, the average COD removal rate was the best in the 5 g/L experiment (61.01%) with the least fluctuation, while that of the 3 g/L experiment showed an average 55.24%. In terms of TMP development, 1 g/L and 3 g/L of PAC showed more rapid TMP increase and reached up to 35 kPa in shorter time compared to the highest amount of PAC addition. This could mean that membrane fouling has not been reduced due to the insufficient PAC doses. However, with 5 g/L PAC, the TMP increase rate significantly decreased after membrane cleaning, indicating that more PAC dose facilitated more stable sludge properties by effective adsorption and subsequently enlarged floc size.

Overall, compared to the experiments with lower PAC dosages, 5 g/L of PAC showed significantly improved performance in terms of nutrient and COD removal. Moreover, although the TMP increase rate was similar to the experiment with 1 g/L PAC in the first 15 days, it showed the lowest increase rate after the chemical membrane cleaning. These results might indicate that the membrane fouling was mitigated with 5 g/L of PAC due to the formation of large sludge floc size through the adsorption of PAC and microbial aggregation on the PAC.

In conclusion, 5 g/L of PAC is the optimal dosage for AnMBR and it was necessary to find out the long-term effects on removal performance along with membrane fouling control by carrying out the study of 5 g/L of PAC added to AnMBR.

CHAPTER 5

Long term experimental investigation of powdered activated carbon addition to anaerobic membrane bioreactor

This chapter is based on the journal paper entitled “Powdered activated carbon addition for fouling control in anaerobic membrane bioreactor”.

5.1 Materials and methods

5.1.1 Experimental set-up

A single stage AnMBR having 3.5 L of working volume was operated continuously using synthetic wastewater for this study. Two AnMBRs were conducted in parallel with and without the optimal PAC dosage, which was investigated as 5 g/L from the previous study. The conventional AnMBR with the optimal PAC dosage was operated for a total of 67 days including two chemical membrane cleanings with no sludge withdrawal. The membrane was chemically cleaned after the TMP reaches over 35 kPa in each cycle. The first cycle maintained for 24 days, then 22 days in the second cycle, and the last cycle was operated for 21 days. On the other hand, AnMBR without PAC addition was operated for a total of 63 days including three chemical membrane cleanings with no sludge withdrawal. It maintained for 18 days in the first cycle, 18 days in the second cycle, 14 days in the third cycle and 13 days in the last cycle. Each operation cycle was followed by a one-week of recovery period after the membrane cleaning to stabilize the pH level and temperature of the reactor so that a steady state could be remained in the next operation cycle. For the AnMBR with PAC, 17.5 g of solid powdered PAC were dosed only at the initial stage of each operation.

Details on the experiment materials and methods are given in Chapter 3.

5.2 Results and discussion

5.2.1 Nutrient removal performance

The average nutrient concentrations of influent and effluent, and removal efficiencies of AnMBRs with PAC and without PAC are given in table 5-1. The nitrate removal efficiencies of both AnMBRs during the first 7 days of operation showed a rather low removal rate around 60-75%, but they significantly increased up to 85.39% and 80.99% of removal efficiency afterwards, in AnMBR, with and without PAC respectively. Compared to the reactor with no PAC addition, 5 g/L of PAC showed approximately 7% higher nitrate removal rate on average, and less fluctuation over the whole operation time. In terms of phosphate removal rates which are expected to be below 10 % in the ideal anaerobic process, they significantly fluctuated in both reactors. The AnMBR with PAC showed slightly higher phosphate removal during the initial 16 days of operation, which might have been caused by the attached biomass growth on the PAC surface. However, during the rest of the operation days, AnMBR with PAC showed more stable and lower phosphate removal efficiency than that of AnMBR without PAC. This indicated that the PAC addition might have facilitated aggregation and abundance of denitrifying bacteria. Both nutrients removal efficiency in two reactors became stable from the middle of the operation days, meaning that the microorganisms had enough time to contact the feed wastewater continuously and the denitrification had been processed efficiently (Khan, 2019). The nitrate removal efficiency and phosphate removal efficiency of both reactors over the operation time are presented in figure 5-1 and 5-2, respectively.

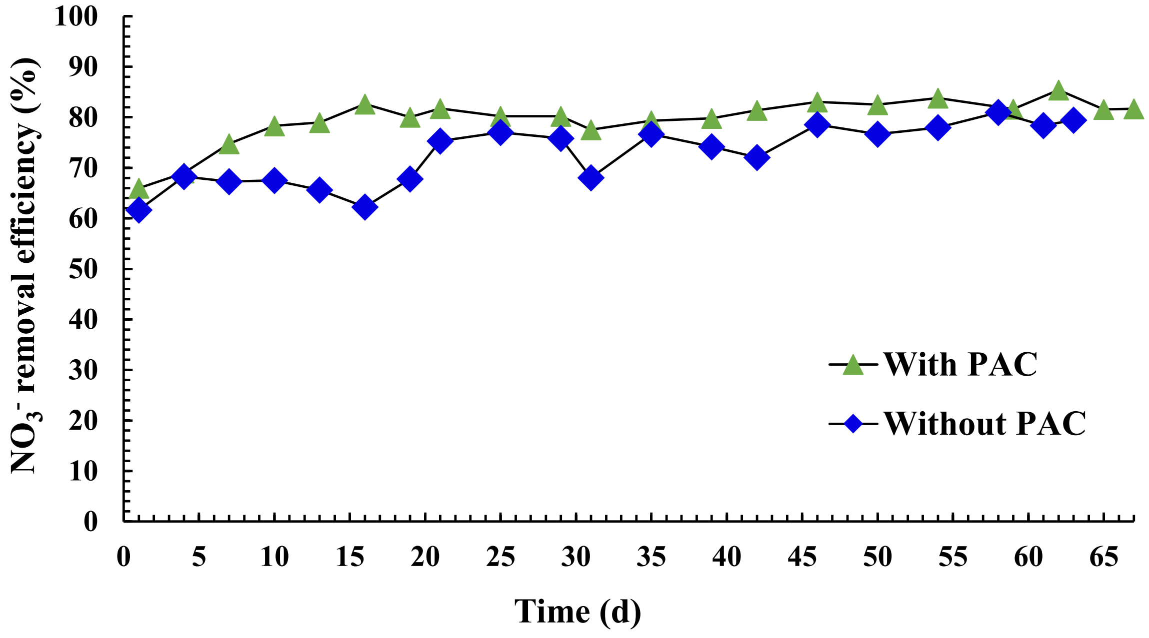

Figure 5-1. Nitrate removal performance of two AnMBRs

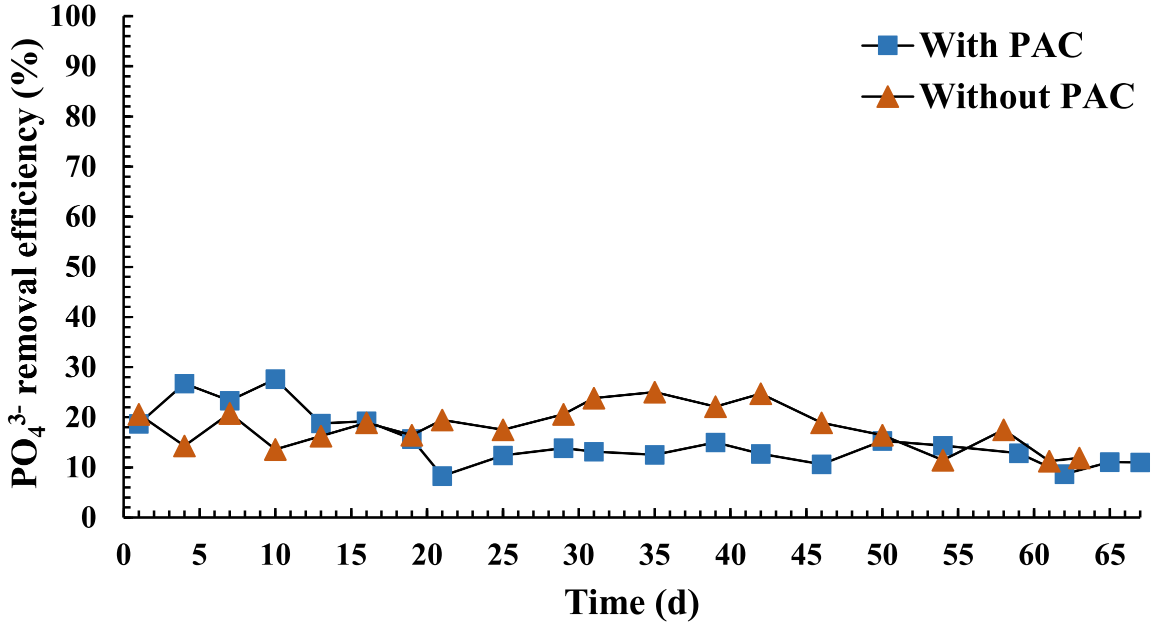

Figure 5-2. Phosphate removal performance of two AnMBRs

The ammonium removal rate fluctuated, but remained at around 0–8.33% of removal rate during the final 7 days of operation of both reactors. This means that oxygen in the reactor has been removed compared to the beginning of the operation. The ammonium removal rate in the reactor with PAC was overall lower than AnMBR without PAC. During the entire operation of the two AnMBRs, the nitrite removal rate fluctuated and the average removal efficiency of AnMBR with PAC resulted in an almost 6% lower removal efficiency than AnMBR without PAC, showing 3.33 ± 6.22% and 9.95 ± 14.08%, respectively. Additionally, nitrite was hardly removed during the last 10 days of operation of each reactor, as there was little difference between the nitrite concentration in the influent and effluent. The removal efficiency of ammonium and nitrite in both reactors over the operation time are shown in figure 5-3 and 5-4, respectively. Overall, the 5 g/L of PAC addition resulted in the more efficient nutrient removal rate including higher nitrate removal rate and the lowest phosphate removal rate, which well describes the anaerobic process.

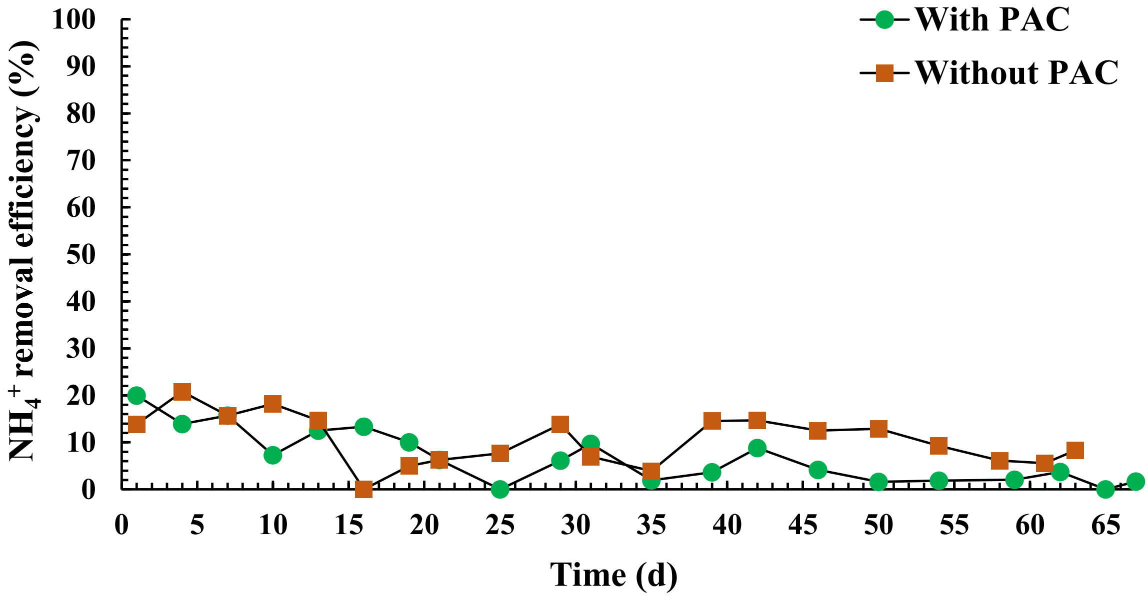

Figure 5-3. Ammonium removal performance of two AnMBRs

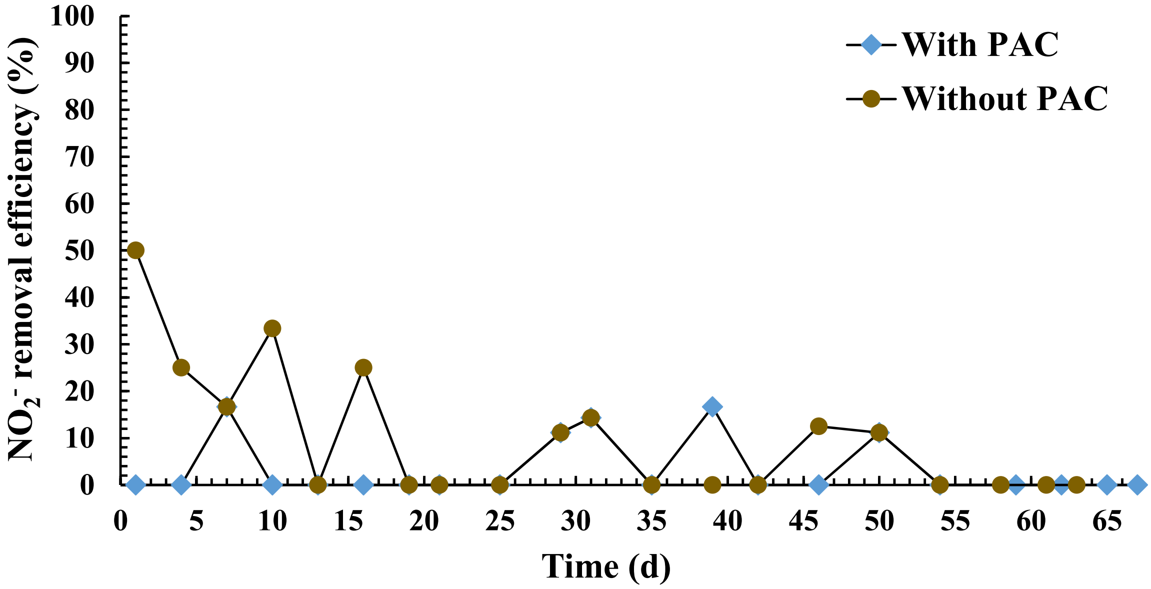

Figure 5-4. Nitrite removal performance of two AnMBRs

Table 5-1. Average concentration of influent and effluent, and removal efficiencies of AnMBRs with PAC and without PAC

AnMBR type	**Average concentration**	**COD**	**TOC**	**NO_3-N**	**NH_4^+-N**	**PO_4^{3}-P**	**NO_2-N**
With 5g/L PAC	Influent (mg/L)	558.70 ± 47.11	77.95 ± 13.07	10.54 ± 1.76	4.40 ± 2.03	1.99 ± 0.33	0.05 ± 0.03
	Effluent (mg/L)	167.54 ± 21.82	18.89 ± 4.23	2.17 ± 0.70	4.09 ± 1.87	1.68 ± 0.30	0.04 ± 0.03
	Removal efficiency (%)	70.10 ± 2.01	75.59 ± 5.21	79.50 ± 4.63	6.87 ± 5.68	15.31 ± 5.36	3.33 ± 6.22
Without PAC	Influent (mg/L)	555.54 ± 56.24	77.46 ± 12.93	10.21 ± 1.45	4.59 ± 1.87	2.06 ± 0.37	0.05 ± 0.02
	Effluent (mg/L)	252.69 ± 31.17	30.44 ± 3.82	2.79 ± 0.70	3.98 ± 1.58	1.68 ± 0.32	0.04 ± 0.03
	Removal efficiency (%)	54.39 ± 4.65	59.90 ± 6.91	72.57 ± 5.98	10.54 ± 5.37	18.07 ± 4.21	9.95 ± 14.08

5.2.2 TOC and COD removal performance and biomass concentration

During the whole experiment of two AnMBR operations with infinite SRT, one-off PAC dosing at the beginning resulted in remarkably higher TOC removal rate than in the reactor with no PAC dosing, with the average rate of 75.59 ± 5.21% and 59.90 ± 6.91%, respectively. Although it had the lowest removal efficiencies in AnMBRs with PAC and without PAC at the initial day of the operation, showing 66.39% and 48.83%, respectively, TOC removal rate increased in both reactors up to around 79.72% and 69.92% at the end of the operation. It was noticeable that the TOC removal rate significantly increased to 82.51% with the PAC addition during the initial 5 days, which may have resulted from the high adsorption capacity of PAC on organic matter. In terms of COD removal rate, the addition of PAC resulted in around 16% higher COD removal rate than no PAC addition, as shown in table 5-1. Figure 5-5 and 5-6 present the TOC and COD removal rate of both AnMBRs, respectively. In terms of biomass concentration, although the mixed liquor volatile suspended solids (MLVSS) concentration kept constant at 4.26 g/L before the operation, a significant increase was observed in both AnMBRs. As soon as the PAC addition, the MLVSS concentration increased up to 9.24 g/L and at the end of operation, it was 11.43 g/L. The biomass concentration in AnMBR without PAC increased to 5.26 g/L after the entire operation. The PAC facilitated more biomass growth as it offered enough space to microorganisms to attach and thrive, thereby the biomass settleability was able to be improved.

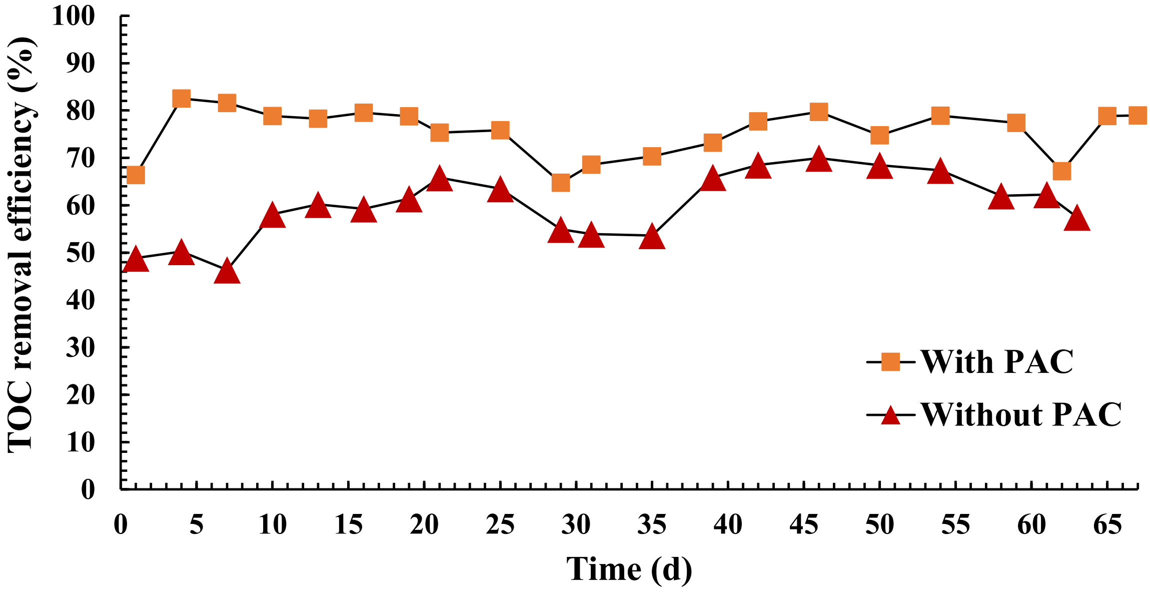

Figure 5-5. TOC removal performance of two AnMBRs

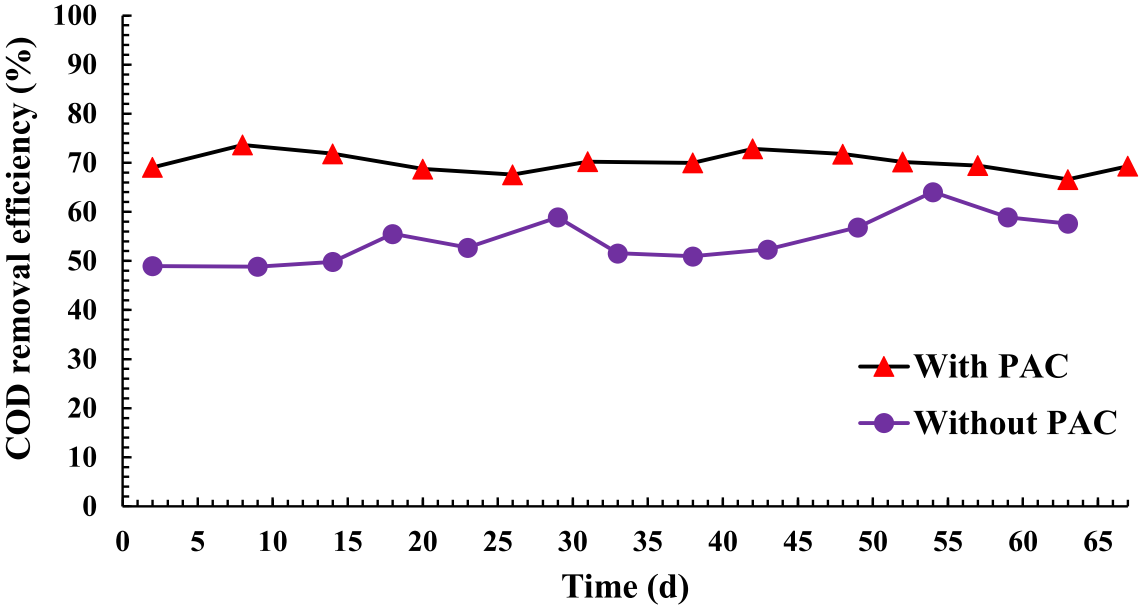

Figure 5-6. COD removal performance of two AnMBRs

5.2.3 Transmembrane pressure (TMP) development

TMP development is closely related to membrane fouling. Figure 5-7 presents the TMP development of the two reactors. During the parallel operation of two reactors, the membrane was chemically cleaned whenever the TMP increases over 35 kPa, and then used again for the next cycle of operation. Both AnMBRs started with 2.15 kPa and 2.55 kPa of TMP. However, there was a significant difference in time to reach up to 35 kPa. The AnMBR with PAC was operated for a total of 67 days including twice receiving chemical membrane cleanings, while AnMBR without PAC was operated for 63 days including three times receiving chemical membrane cleanings. The first cycle of the continuous operation was 18 and 24 days for the reactor with and without PAC respectively, which means PAC addition provided 33% longer operation without chemical membrane cleaning. In the absence of PAC, the TMP increased rapidly with a sharp TMP jump within 5 days in each operation cycle. On the other hand, the presence of PAC showed a slow rise and TMP jump took place after 15 days of the operation cycle. The rapid TMP increase might have resulted by the accumulation of fine particles on membrane as well as pore clogging by biomass at the beginning of the operation (Chen et al., 2013; Pradhan et al., 2011). Moreover, after the chemical membrane cleaning, the TMP in AnMBR without PAC dropped to 3.11 kPa, whereas the TMP in AnMBR with PAC dropped to nearly 2.00 kPa, indicating an efficient restoration of membrane permeability. In terms of TMP increase trend for the last cycle in each reactor, it was shown that the rapid TMP increase occurred after 12 days with PAC addition while the increase occurred within 3 days without PAC addition. In addition, as the last cycle was 3rd and 4th cycle for AnMBR with and without PAC respectively, the operation time persisted for 21 days in AnMBR with PAC, which was 8 days longer than the reactor without PAC. Moreover, the operation time for the last cycle of AnMBR with PAC was shorter than the previous cycles, because some amount of exhausted PAC might have acted as a foulant due to the one-off PAC dose at the first operation day. Thus, further investigation regarding the replenishment ratio as well as the different partial replacement ratios of PAC in long term operation can be carried out for the future study. Overall, TMP increase in the AnMBR with optimal dosage of PAC resulted in a lower increase rate than AnMBR without PAC. It means that 5 g/L of PAC addition could mitigate the membrane fouling due to enlarged particle size as well as changing the sludge properties.

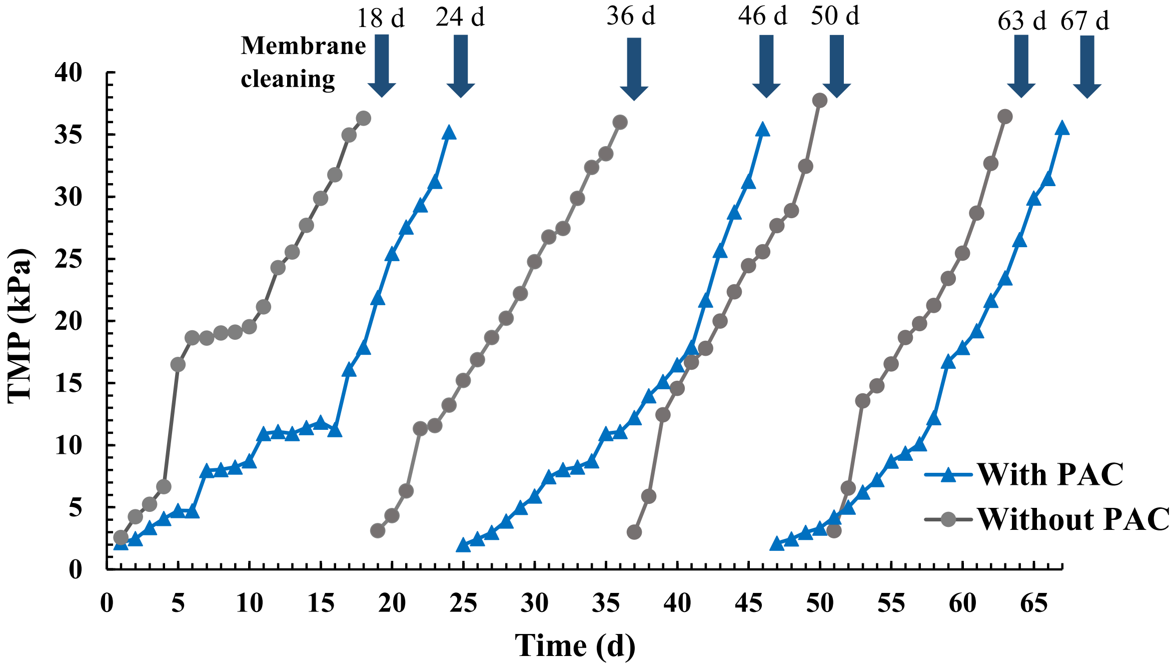

Figure 5-7. The variations of TMP during operation of two AnMBRs

5.2.4 Zeta potential in mixed liquor

It was investigated that hydrophobicity and surface charge could significantly influence the flocculation ability of sludge (Chen et al., 2018a). Previous studies found that increased zeta potential and increased hydrophobicity enhanced flocculation of the mixed liquor and enlarged suspended flocs, thereby mitigating membrane fouling (Huang and Wu, 2008; Wu and Huang, 2010). In this study, the zeta potential value of anaerobic sludge in AnMBR with PAC was -

11.60 ± 0.46 mV, while that in AnMBR without PAC was -14.40 ± 0.44 mV. The result showed that the PAC addition to AnMBR had higher zeta potential value than the conventional AnMBR, indicating that the negative charged sludge flocs could be reduced or that charge neutralization has been caused by PAC. This was because it was reported that the charge neutralization by addition of flocculants could enhance the sludge flocculation ability (Deng et al., 2015; Ji et al., 2014). Hence, increase of zeta potential by PAC addition could attribute to improvement of biomass flocculation ability.

5.2.5 SMP and EPS in mixed liquor

The main fractions of SMP and EPS are mainly considered as proteins and polysaccharides. Table 5-2 presents the compositions of SMP and EPS in the mixed liquor in both AnMBRs as well as protein to polysaccharide ratio at different operating periods. During the first 7 days of operation, the conventional AnMBR had higher total SMP and EPS values than AnMBR with PAC addition, which are 20.02 ± 0.83 mg/L and 22.94 ± 0.62 mg/L, respectively. While the AnMBR without PAC showed a similar value of protein and polysaccharide in SMP, polysaccharide concentration in SMP was much higher than the protein in AnMBR with PAC. The total EPS concentration was twice as high in AnMBR without PAC as the one with PAC addition. After that, the SMP and EPS concentration in AnMBR with PAC during the 14 days of the operation decreased. This is because the addition of PAC, which has a high surface area, could initially reduce the concentration of both SMP and EPS by effective adsorption.

However, after the decrease of SMP and EPS concentration in the second week of operation, the EPS concentration in AnMBR with PAC remarkably increased up to 21.86 ± 2.00 mg/L. This might be attributed to the fact that PAC become saturated with organic matter and the subsequent attachment and abundant growth of microorganisms could release more EPS as microbial products. It was reported that EPS could enhance the flocculation ability of the mixed liquor by polymer entanglement, contributing to enlarged sludge floc size (Chen et al., 2018a). Thus, this increase of EPS concentration in AnMBR with PAC could enhance the formation of larger floc size as well as sludge flocculation ability. Although it was reported that a high amount of EPS could accelerate the cake layer formation and fouling process, excellent sludge flocculation and enlarged flocs might have rather formed a porous cake layer in this experiment (Chen et al., 2018a). During the rest of the operation time, EPS concentration was slightly reduced to 18.32 ± 0.87 mg/L, by the concurrent effects of biologically activated carbon (BAC) including the adsorption and biodegradation of attached microorganisms. Overall, the total EPS concentration in AnMBR with PAC was kept in lower values than the reactor without PAC. Regarding SMP concentration, it is reported that infinite SRT in submerged AnMBR could result in high SMP concentration (Huang et al., 2011). Although AnMBR with PAC was operated with infinite SRT, the SMP concentration showed steady reduction over the whole operation period, indicating that PAC addition could effectively reduce SMP through the simultaneous adsorption and biodegradation.

On the other hand, in the AnMBR without PAC, the total concentration of SMP showed minor difference during the entire time, while there was a significant change in its composition. The amount of polysaccharide in SMP significantly increased from 9.00 ± 0.26 mg/L to 17.01 ± 0.06, while the amount of protein decreased. This could lead to severe pore blocking on the membrane due to the soluble state of SMP as well as the hydrophilic nature of polysaccharide (Chen et al., 2017). The total EPS concentration incredibly increased, while the composition showed minor change during the whole time. The higher amount of EPS could cause higher sludge viscosity as well as acceleration of cake formation, thereby deterioration of membrane permeability (Chen et al., 2017).

Previous research found that the protein to polysaccharides ratio of SMP and EPS (PN/PS ratio) played an significant role in membrane fouling, since the ratio might be closely related to properties of sludge flocs such as hydrophobicity and surface charge (Arabi and Nakhla, 2008b; Tian et al., 2011). The higher PN/PS ratio of EPS was found in AnMBR with PAC compared to AnMBR without PAC after 21 days of operation. This higher amount of protein might have induced higher hydrophobicity as well as higher zeta potential of sludge, which enabled more agglomeration of sludge flocs. The PN/PS ratio of SMP in remarkably increased up to 2.80, while that in AnMBR without PAC lowered after 14 days and stayed at a lower value. This high protein fraction of SMP in AnMBR with PAC also induced better settleability by the hydrophobic nature of protein. The result of previous research showed that high PN/PS ratio of SMP had beneficial impact on fouling control, such as less irreversible fouling, and the results in this study also corresponded to this previous result (Yao et al., 2011). Figure 5-8 and 5-9 show the time variation of SMP and EPS concentration as well as composition in both AnMBRs. SMP_p and SMP_c indicate protein and polysaccharide in SMP, respectively, while EPS_p and EPS_c refer protein and polysaccharide in EPS, respectively.

Table 5-2. SMP and EPS compositions in mixed liquor in two AnMBRs at different times during the operation period

		SMP				**EPS**			
		PN[a] (mg/L)	PS[b] (mg/L)	PN/PS ratio	SMP (mg/L)	PN (mg/L)	PS (mg/L)	PN/PS ratio	EPS (mg/L)
AnMBR with PAC	1–7 days	1.05 ± 0.22	13.93 ±0.78	0.08	14.99 ± 0.14	5.57 ± 0.44	6.09 ± 0.11	0.91	11.66 ± 0.55
	8–14 days	2.64 ± 0.06	10.68 ± 0.04	0.25	13.32 ± 0.11	2.20 ± 0.03	7.01 ± 0.49	0.31	9.20 ± 0.52
	15–21 days	5.74 ± 0.08	7.50 ± 0.10	0.77	13.23 ±0.18	8.59 ± 0.12	5.65 ± 0.13	1.52	14.24 ± 0.01
	22–28 days	4.03 ± 0.35	8.55 ± 0.01	0.47	12.59 ± 0.33	12.07 ± 0.59	9.79 ± 1.42	1.23	21.86 ± 1.00
	29–35 days	4.07 ± 0.03	5.13 ± 0.03	0.79	9.20 ± 0.01	10.80 ± 1.23	9.23 ± 0.12	1.17	20.03 ± 0.85
	36–42 days	5.38 ± 0.40	4.81 ± 0.07	1.12	10.19 ± 0.47	13.43 ± 0.09	6.25 ± 0.12	2.15	19.68 ± 0.21
	43–49 days	5.98 ± 0.54	2.13 ± 0.13	2.80	8.21 ± 0.67	13.28 ± 0.17	6.08 ± 0.07	2.19	19.35 ± 0.24
	50–56 days	5.33 ± 0.32	4.01 ± 0.23	1.32	9.34 ± 0.32	13.34 ± 0.43	6.53 ± 0.11	2.04	19.87 ± 0.54
	50–67 days	2.90 ± 0.10	3.75 ± 0.18	0.77	6.65 ± 0.08	13.16 ± 0.72	5.15 ± 0.15	2.55	18.32 ± 0.87
AnMBR without PAC	1–7 days	10.61 ± 0.57	9.00 ± 0.26	1.17	20.02 ± 0.83	12.25 ± 1.04	10.69 ± 1.18	1.15	22.94 ± 0.62
	8–14 days	9.60 ± 0.86	6.20 ± 0.39	1.55	15.20 ± 0.47	12.33 ± 1.19	7.21 ± 0.01	1.70	19.53 ± 0.49
	15–21 days	3.23 ± 0.68	9.55 ± 0.06	0.34	18.39 ± 0.80	12.98 ± 0.87	8.10 ± 0.06	1.60	21.08 ± 0.94
	22–28 days	4.46 ± 0.73	12.37 ± 0.01	0.36	16.02 ± 0.85	9.02 ± 0.98	9.18 ± 0.05	0.98	18.20 ± 0.86
	29–35 days	1.28 ± 0.07	14.92 ± 0.08	0.09	16.20 ± 0.16	9.79 ± 0.15	10.16 ± 0.08	0.96	19.94 ± 0.26
	36–42 days	1.88 ± 0.04	12.84 ± 0.04	0.15	14.71 ± 0.01	13.55 ± 0.07	10.71 ± 0.63	1.27	24.26 ± 0.70
	43–49 days	1.09 ± 0.05	17.74 ± 0.03	0.06	18.82 ± 0.07	15.02 ± 0.02	15.87 ± 0.64	0.94	30.88 ± 0.66
	50–56 days	2.25 ± 0.02	16.87 ± 0.11	0.13	19.12 ± 0.12	18.78 ± 0.11	13.56 ± 0.44	1.38	32.34 ± 0.54
	57–63 days	1.96 ± 0.01	17.01 ± 0.06	0.12	18.97 ± 0.07	23.48 ± 0.31	13.29 ± 1.20	1.77	36.77 ± 0.92

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

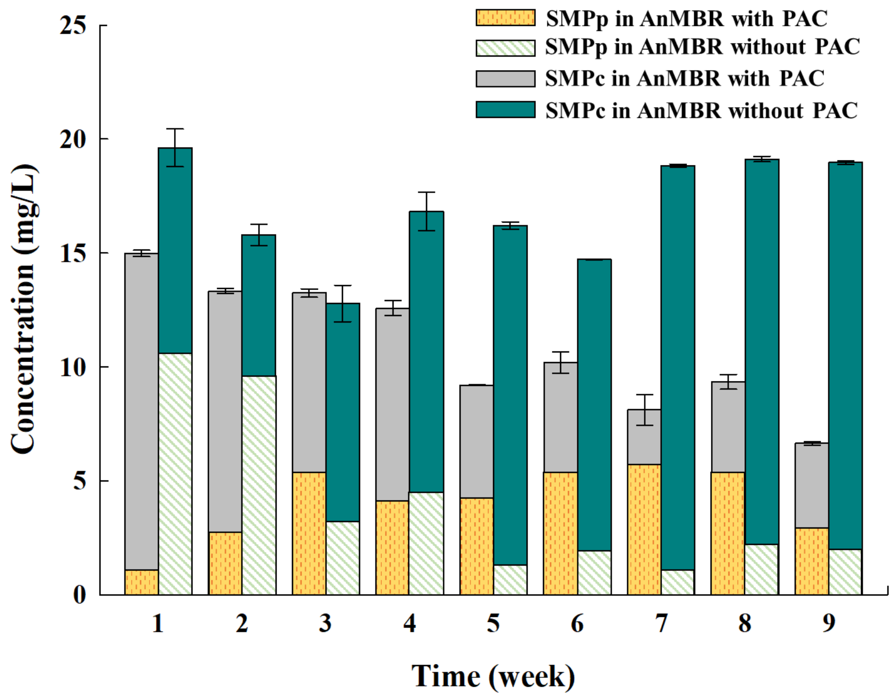

Figure 5-8. The variations of composition in SMP in two AnMBRs

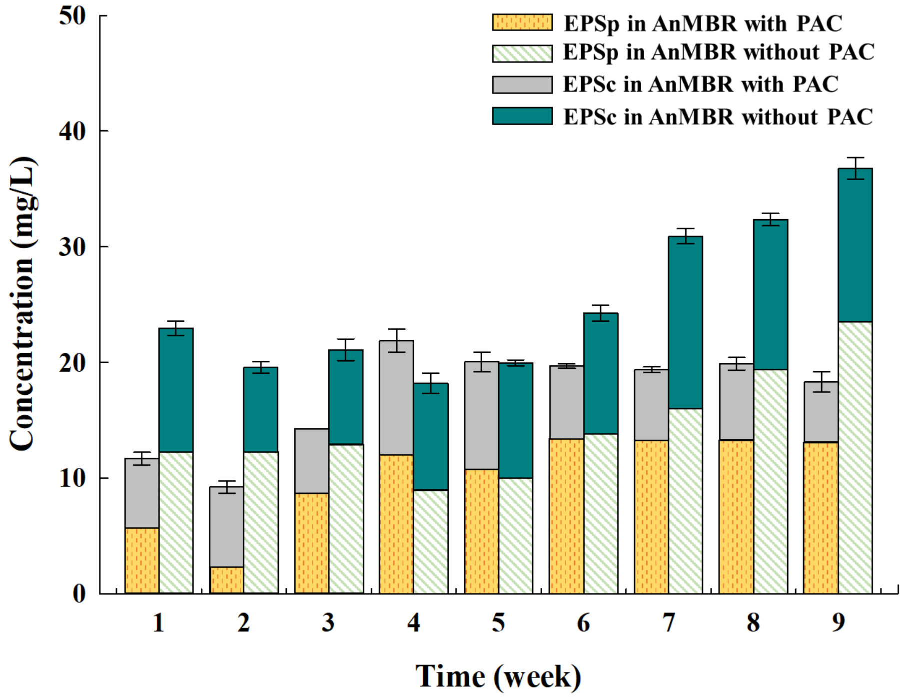

Figure 5-9. The variations of composition in EPS in two AnMBRs

5.2.6 Membrane fouling resistance

The membrane fouling resistance of both AnMBRs in each cycle are presented in table 5-3. Membrane fouling resistances were measured before and after each cycle when the chemical membrane cleaning was performed. Although the initial resistance of cleaned membrane (R_m) was similar in both reactors, the total resistance (R_t) was significantly higher in the AnMBR without PAC, indicating that the PAC addition could effectively reduce R_t. Moreover, R_t of both reactors showed gradually increasing trends, while that of AnMBR without PAC increased with a larger magnitude than the one with PAC addition, which was from 1.52×10^{13} m^{-1} to 1.93×10^{13} m^{-1}. Since the ratio of cake layer resistance (R_c) to R_t in both reactors was between 76.17% and 86.44%, the predominant fouling mechanism in both AnMBRs might be due to a cake layer formation. However, R_c in AnMBR without PAC was much higher than that in AnMBR with PAC. The pore blocking resistance (R_p) of AnMBR with PAC showed minor difference after all three cycles. However, R_p of AnMBR without PAC had a remarkable increase from 4.02×10^{11} m^{-1} to 2.08×10^{12} m^{-1} after four cycles, accounting for 10.78% of R_t after the entire operation. As discussed in 5.3.5, comparatively high SMP concentration in AnMBR without PAC could result in severe pore blocking due to the soluble form of SMP fractions (Chen et al., 2017). In addition, it was also noticeable that R_m of AnMBR without PAC increased after every membrane cleaning, while AnMBR with PAC remained at similar value, indicating that irreversible fouling has occurred without PAC addition. The larger amount of polysaccharide in SMP in mixed liquor might have induced more pore clogging due to the hydrophilic property of polysaccharide which enables it to penetrate the cake layer and then cause irreversible pore blocking (Deng et al., 2014). It was found that small sludge particle size could lead to serious pore clogging and cake layer formation due to its high tendency to adhere on membrane surface and pore walls (Cheng et al., 2020). On the other hand, higher flocculation ability as well as PN/PS ratio of SMP induced by PAC addition brought about larger sludge flocs and subsequently less deposition on membrane surface, because of proportionally increased shear-induced diffusion and inertial lift force according to particle size (Pan et al., 2010). Therefore, PAC addition could prevent pore clogging and irreversible fouling on the membrane surface. The variations of membrane resistance of two AnMBRs are presented in figure 5-10.

Table 5-3. Comparison of membrane fouling resistance values in two AnMBRs.

Membrane resistance[a] (m^{-1})		**With 5 g/L PAC**		**Without PAC**	
			% of R_t		**% of R_t**
1st cycle	R_t	9.25×10^{12}		1.52×10^{13}	
	R_m	1.60×10^{12}	17.29	1.66×10^{12}	10.92
	R_p	3.22×10^{11}	3.48	4.02×10^{11}	2.64
	R_c	7.34×10^{12}	79.35	1.34×10^{13}	86.44
2nd cycle	R_t	1.05×10^{13}		1.65×10^{13}	
	R_m	1.77×10^{12}	16.86	1.80×10^{12}	10.90
	R_p	4.16×10^{11}	3.96	6.51×10^{11}	3.95
	R_c	7.83×10^{12}	74.57	1.41×10^{13}	85.45
3rd cycle	R_t	1.15×10^{13}		1.72×10^{13}	
	R_m	1.79×10^{12}	15.56	1.91×10^{12}	11.10
	R_p	4.56×10^{11}	3.97	1.42×10^{12}	8.26
	R_c	9.24×10^{12}	80.35	1.39×10^{13}	80.81
4th cycle	R_t			1.93×10^{13}	
	R_m			2.20×10^{12}	11.40
	R_p			2.08×10^{12}	10.78
	R_c			1.47×10^{13}	76.17

[a] R_t, total resistance; R_m, cleaned membrane resistance; R_p, pore blocking resistance; R_c, cake resistance.

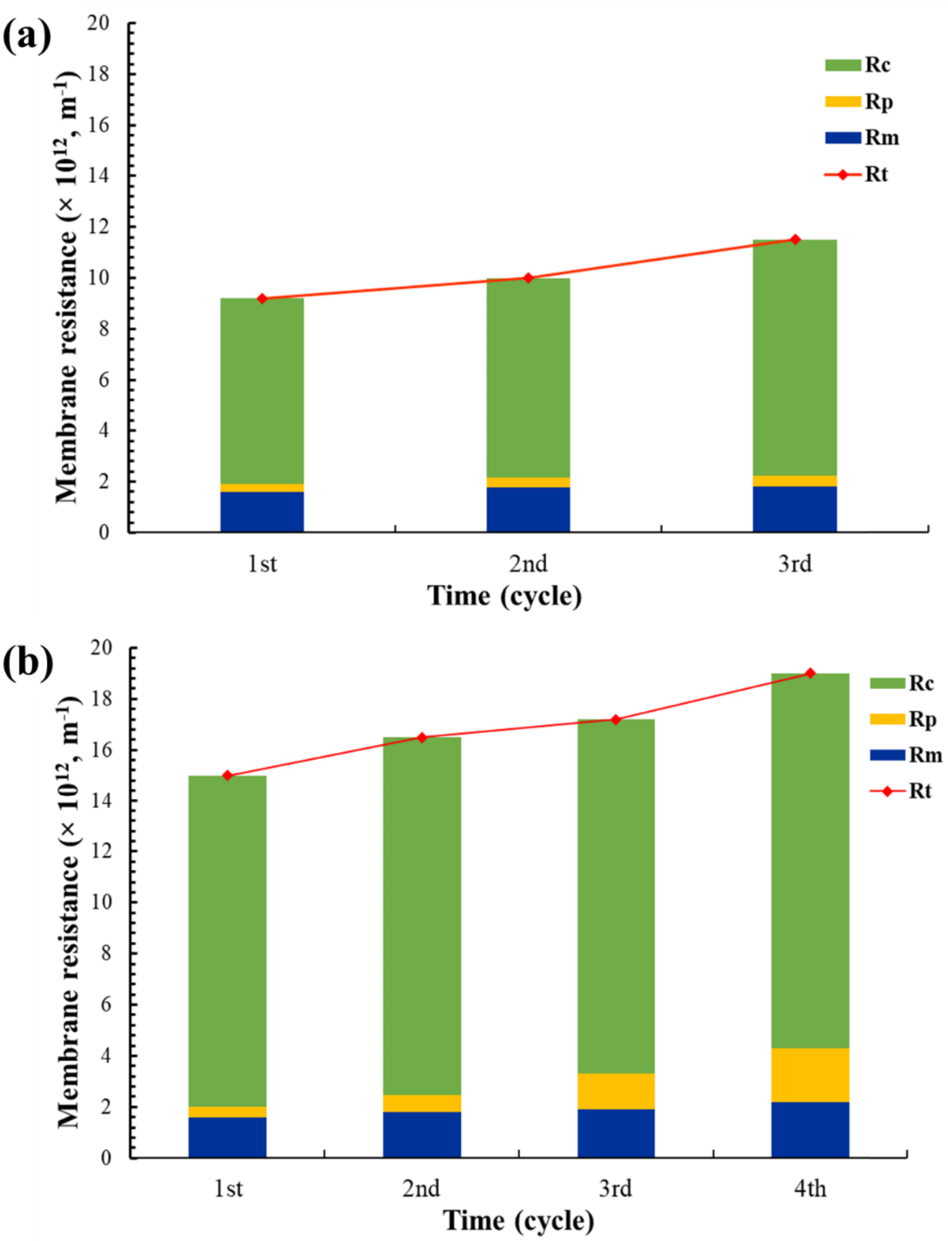

Figure 5-10. The variations of membrane fouling resistance of two AnMBRs: (a) AnMBR with PAC; and (b) AnMBR without PAC.

5.2.7 SMP and EPS in cake layer

As the cake layer was the largest fraction of the total fouling resistance, the cake layer compositions of SMP and EPS were analysed. The protein and polysaccharide composition of EPS in the cake layer were similar in AnMBR with PAC and AnMBR without PAC in every cycle. On the other hand, the total SMP concentration was remarkably higher with the absence of PAC (77.19 ± 7.37 mg/L) compared to the presence of PAC (17.43 ± 1.59 mg/L). In the SMP composition of the cake layer in AnMBR without PAC, the main fraction was protein at the first cycle, accounting 51.78 ± 7.28 mg/L. From the second cycle, polysaccharide had a larger composition than protein, which was the main factor of cake layer formation as well as pore blocking, both leading to deterioration in membrane filterability. This result elucidated that the cake layer resistance of AnMBR without PAC was mainly due to the high SMP concentration on the membrane surface. In AnMBR without PAC, large amount of SMP was able to be adsorbed or attached on membrane surface because of the drag force from the effluent pump along with a rapid TMP increase (Deng et al., 2014). However, the PAC addition effectively reduced both protein and polysaccharide of SMP in the cake layer, which might be resulted from the adsorption of SMP on the PAC and biodegradation by attached microorganisms prior to attachment on membrane surface. Figure 5-11 and 5-12 present variation of SMP and EPS compositions in cake layers.

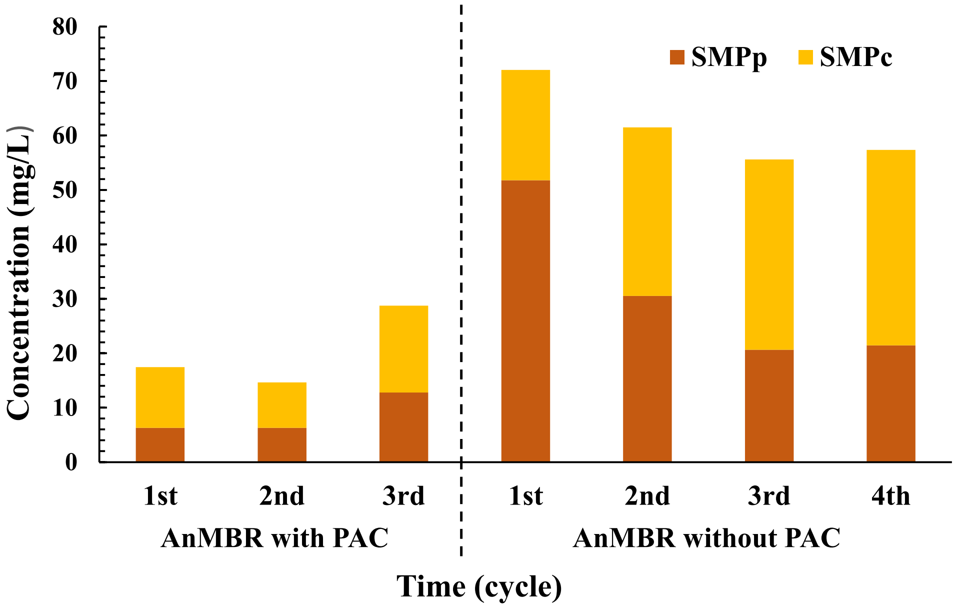

Figure 5-11. The variation of SMP composition in cake layers

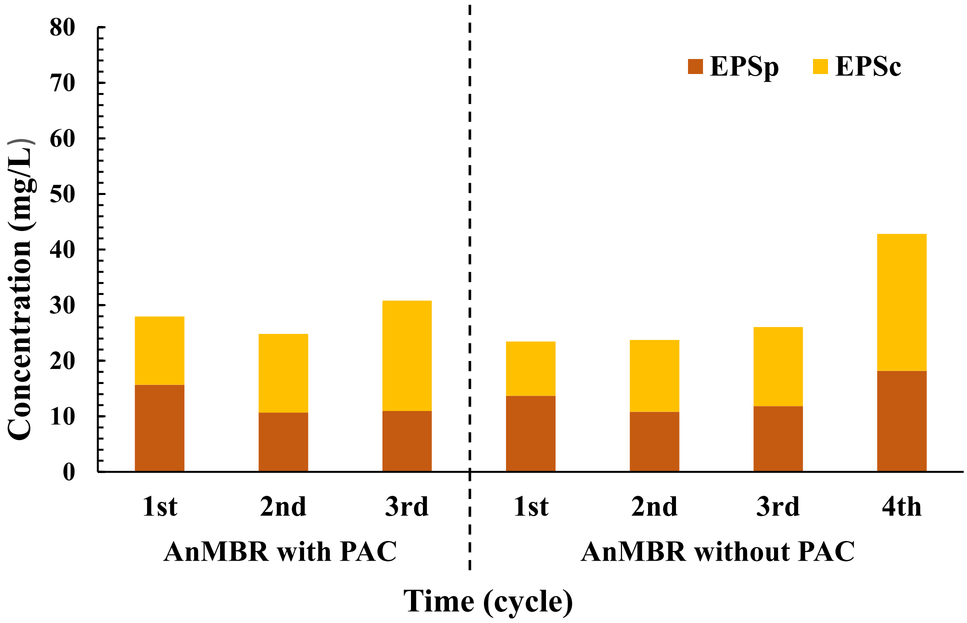

Figure 5-12. The variation of EPS composition in cake layers

5.2.8 Impact of PAC addition on microbial community

5.2.8.1 Overview of sequencing data

Paired-end Illumina sequencing generated 203,054 sequences from 4 samples. At least 49.4% of input sequences of each sample passed quality filtering, 82.6% of filtered sequences were merged, and 99% of merged sequences were non-chimeric. The minimum number of sequences per denoised samples was 18,643, and the maximum number was 34,899. In total, 106,153 denoised sequences were clustered to a total of 751 ASVs with a mean frequency of 141.3. Rarefaction curves of Observed ASVs at maximum sequencing depth of 18,643 showed that all samples approached a saturation plateau at sequencing depth of about 17,000, confirming the sufficient sequencing depth in this study (Figure 5-13).

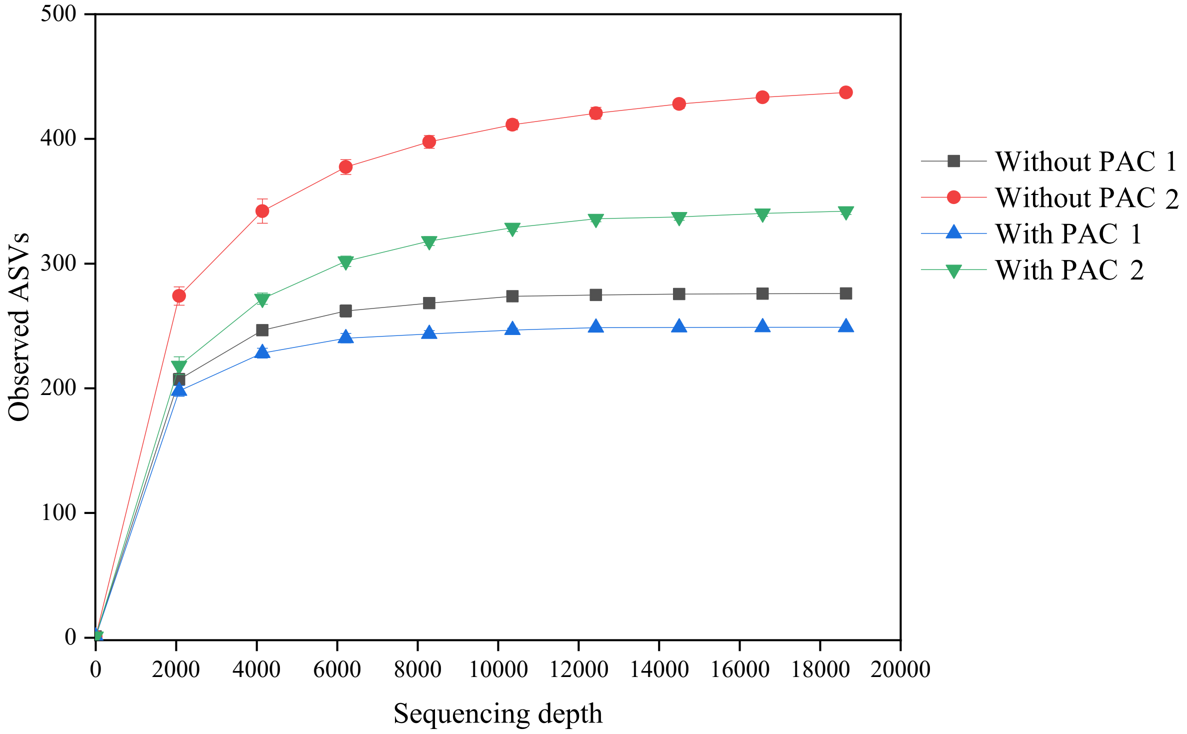

Figure 5-13. Rarefaction curves of 16S rRNA marker gene amplicon sequences at maximum depth of 18,643.

5.2.8.2 Impact of PAC addition on microbial diversity

AnMBR with PAC addition has a slightly lower diversity level compared to the reactor without PAC addition (Table 5-4). This is true for both species richness (number of observed ASVs) and species evenness (Shannon index). Low species richness and evenness indicate that PAC addition created a selective force, and some species were enriched that they became predominant microbes. To the best of our knowledge, the impact of PAC on microbial diversity in AnMBR has not been reported previously. A review of the literature shows that PAC addition can increase microbial diversity in conventional and dynamic aerobic MBRs (Asif et al., 2020; Hu et al., 2017). It is noteworthy that the microbial community in aerobic MBRs are very different from that in an anaerobic environment. Thus, further research is needed to elucidate more clearly how PAC affect microbial diversity in AnMBRs.

Table 5-1. Microbial diversity of two AnMBRs with and without PAC

Diversity index	Without PAC	With PAC
Number of observed ASVs	354.5 ± 79.5	296 ± 47
Shannon index	6.08 ± 0.23	5.92 ± 0.03

5.2.8.3 Impact of PAC addition on microbial composition

Microbial composition analysis revealed key functional groups in the anaerobic digestion process of both AnMBRs (Figure 5-14). *Lactococcus* and unassigned *Enterobacteriaceae* were the two most abundant bacterial genera in all samples (24.8–30.6%). *Enterobacteriaceae* participates in hydrolysis and acidognesis while *Lactococcus* is a lactate producer (Detman et al., 2018). Other dominant hydrolytic and fermentative bacteria included unassigned *Saccharimonadaceae* (AnMBR without PAC only), *Brooklawnia* (family *Propionibacteriaceae*), *Paludibacter*, *Thermovirga* (amino acid degradation), *Enterobacter*, *Lactobacillus*, and uncultured *Anaerolineaceae*. *Smithella* and uncultured *Synergistaceae* convert lactate and other volatile fatty acids to acetate – the precursor for methanogenesis performed by *Methanosaeta* (Liu et al., 1999; Nguyen et al., 2019).

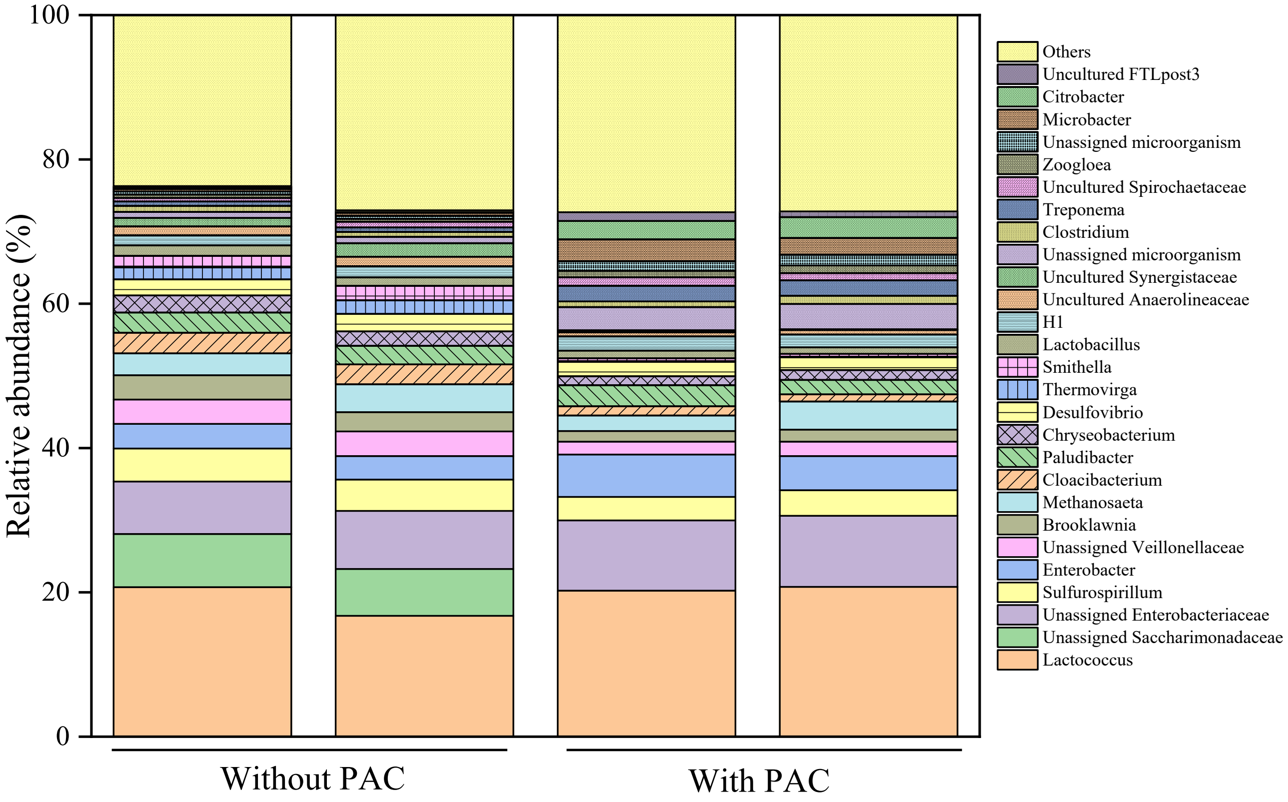

Figure 5-14. Microbial composition in AnMBRs with and without PAC addition. Data are presented at genus level. Only genera with relative abundance > 1% in at least one sample are shown individually, the rest are grouped into "Others".

PAC addition exerted strong impact on the microbial composition in AnMBR (Figure 5-14 and 5-15), especially on filamentous and fouling-related microorganisms, which could reduce fouling propensity. Filamentous bacteria play an important role in wastewater treatment, as they can benefit the formation of flocs and subsequent settling by offering a base structure for other microorganisms to attach. However, excessive growth of filamentous bacteria can have negative effect on membrane fouling. Sludge viscosity increases due to the abundance of filamentous bacteria, which leads to the formation of non-porous cake layer on the membrane. More production of SMP by excess filamentous bacteria can also accelerate severe membrane fouling (Deng et al., 2014). Filamentous microorganisms including uncultured *Anaerolineaceae*, *Chryseobacterium* and *Methanosaeta* can contribute to the initial attachment step of biofouling development due to their high affinity for attachment (Harb et al., 2015; Yamada et al., 2006). In fact, *Chryseobacterium* has been identified as one of three pioneer bacteria involving membrane biofilm development (Piasecka et al., 2012). Specifically, PAC addition could lead to lower relative abundance of filamentous microorganism in the reactor. The PAC addition might inhibit proliferation of filamentous bacteria and its high adsorption capacity could mitigate fouling by reducing the attachment of filamentous bacteria on the membrane surface. The relative abundance of these filamentous microbes in AnMBR without PAC was 1.7–4.7 times higher than that of AnMBR with PAC addition. PAC addition also resulted in lower relative abundance of previously identified fouling-associated bacteria such as *Cloacibacterium* and *Paludibacter* (Lei et al., 2019; Sun et al., 2016; Xu et al., 2020). *Cloacibacterium* was repeatedly detected in membrane biofilm, suggesting its high potential attachment on the membrane and membrane fouling propensity (Sun et al., 2016). Meanwhile, Lei et al. (2019) reported *Paludibacter* as one of the predominant bacteria in the fouling layer of AnMBR. On the other hand, the relative abundance of these fouling-associated bacteria was only 8.7–59.4% in AnMBR with PAC.

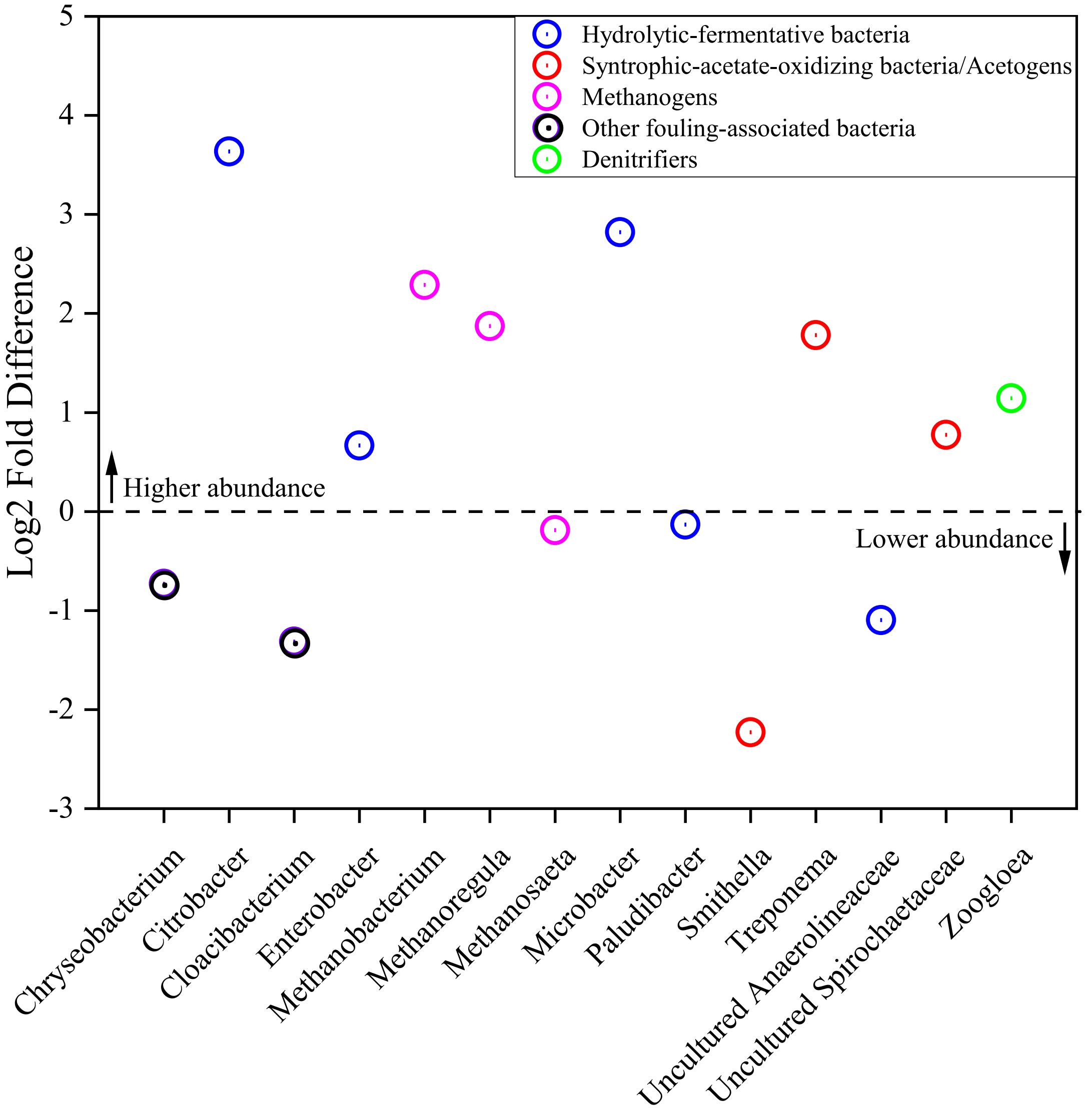

Figure 5-15. Impact of PAC addition on microbial composition represented by log2-fold difference in the relative abundances at genera level, with genera coloured by functional groups.

In AnMBR with PAC addition, different non-filamentous microorganisms were enriched to fulfil functional roles of the above mentioned filamentous and fouling-related genera (Figure 5-15). The role of uncultured *Anaerolineaceae* in hydrolysis and fermentation was met by *Microbacter* (relative abundance 7.1 times higher than AnMBR without PAC) (Harb et al., 2015), while *Treponema* (relative abundance 3.4 times higher) instead of *Smithella* was responsible for *acetogenesis* (Wang et al., 2013). PAC addition also resulted in a higher relative abundance of hydrogen/formate-utilizing methanogens *Methanoregula* (Bräuer et al., 2011) and *Methanobacterium* (Chen et al., 2019b), as well as hydrogen-producing bacteria including *Enterobacter* (Xiao et al., 2013), uncultured *Spirochaetaceae* (Nguyen et al., 2019) and *Citrobacter* (Cabrol et al., 2017; Thompson et al., 2008). This indicates a higher contribution of hydrogenotrophic methanogenesis pathway to compensate for the lower abundance of acetoclastic methanogen *Methanosaeta*.

AnMBR with PAC showed an enhanced balance between archaeal and bacterial groups, with higher ratio between total relative abundance of methanogenic archaeal orders and bacterial genera (A/B) (0.083 compared to 0.075 in AnMBR without PAC). Higher balance among functional groups has been shown to be essential for stable and efficient process performance in AnMBR (Cheng et al., 2021). These results implied for better COD and TOC removals observed with PAC addition. Genus *Zoogloea*, consisting of denitrifying bacteria (Huang et al., 2015) – was also enriched in AnMBR with PAC (Figure 5-15), contributing to the higher nitrate removal efficiency observed. This is in agreement with findings from Asif et al. (2020) who reported an improved abundance of *Zoogloea* along with lower nitrate concentration in PAC-MBR compared to MBR without PAC.

5.3 Conclusion

The two conventional AnMBRs were set in operation at the same time to compare the effects of PAC addition in terms of nutrient, TOC, and COD removal performance as well as membrane fouling mitigation. One AnMBR, which was dosed with 5 g/L of PAC only at the first day of operation without any replacement, was operated for 67 days including three operating cycles. The other AnMBR without PAC addition was operated for 63 days including four operating cycles. Both two AnMBRs were operated under infinite SRT conditions. The addition of PAC showed overall better nutrient removal efficiencies as well as significantly higher TOC and COD removal efficiency, due to the high adsorption capacity of PAC as well as the enrichment of *Methanoregula*, *Methanobacterium*, *Enterobacter*, uncultured *Spirochaetaceae*, *Citrobacter*, and genus *Zoogloea*. AnMBR without PAC showed more rapid TMP rise and reached 35 kPa in shorter time, indicating that more serious membrane fouling occurred compared to the AnMBR with PAC.

Overall, the total SMP and EPS concentration in the AnMBR with 5 g/L of PAC resulted in lower concentrations over the whole operating period compared to the AnMBR without PAC, indicating lower fouling potential. Although the EPS concentration showed increase in the middle of the operation, this could enhance flocculation and aggregation, leading to enlarged flocs and porous cake layer. The total SMP concentration was remarkably reduced with the PAC addition, especially the polysaccharide fraction decreased. This could lead to the higher PN/PS ratio of SMP in AnMBR with PAC, facilitating effective membrane fouling mitigation. On the other hand, the SMP_c significantly increased with no PAC addition, leading to severe pore blocking and deterioration of membrane permeability.

The membrane fouling resistance values showed that the main fraction of fouling resistance was cake layer formation in both AnMBRs, however increase of pore resistance and cleaned membrane resistance indicated the occurrence of irreversible fouling in the absence of PAC. Moreover, the analysis of cake layer composition indicated that the main contributor of membrane fouling in this study was SMP_c, inducing severe pore clogging and irreversible fouling. Thus, the optimal PAC addition to AnMBR could effectively reduce SMP_c in the cake layer by the adsorption and biodegradation of PAC, thereby lowered membrane fouling propensity. The lower abundance of filamentous and fouling-related microorganisms such as uncultured *Anaerolineaceae*, *Chryseobacterium*, *Methanosaeta Smithella*, *Cloacibacterium* and *Paludibacter*, which have high affinity to attach on membrane, could also reduce fouling potential. To sum up, the optimal PAC addition to conventional AnMBR enhanced overall removal performance and effectively mitigated membrane fouling.

CHAPTER 6

Conclusions and recommendations

6.1 Conclusions

The first study for the optimisation of PAC dose was conducted with 1 g/L, 3 g/L, and 5 g/L in conventional AnMBRs. In every sets of the experiment, the nitrate, nitrite and ammonium removal rate were close to the ideal rate in anaerobic process, while phosphate removal showed a slightly higher removal rate during the whole operation than the fact that it is expected to be 0-10% of removal rate. 5 g/L of PAC addition, particularly, showed the highest nitrate removal rate and the lowest phosphate removal rate, indicating the abundance of denitrifying bacteria caused by PAC adsorption. Moreover, the average COD removal rate was the best in the 5 g/L experiment (61.01%), describing the effective organic matter removal by the PAC adsorption and biodegradation. In terms of membrane fouling behaviour regarding the TMP rise, its increasing rate significantly decreased after membrane cleaning with the presence of 5 g/L PAC. As a consequence, the optimal PAC dose was 5 g/L, showing the highest overall removal performance as well as the lowest fouling rate.

For the investigation of long-term effect of 5 g/L PAC, the two conventional AnMBR systems were operated in parallel to compare the effects of PAC addition in terms of nutrient, TOC, and COD removal performance as well as membrane fouling mitigation. The one-off dosing of the optimal PAC showed overall better nutrient removal efficiency as well as 15.71% and 15.69% higher TOC and COD removal efficiencies, respectively, due to the high adsorption capacity of PAC. It also resulted in much slower TMP rise showing shorter time to reach at 35 kPa, indicating that less membrane fouling occurred compared to the AnMBR with PAC. In addition, the total SMP and EPS concentration in the AnMBR with 5 g/L of PAC resulted in lower concentrations over the whole operating period compared to the AnMBR without PAC, indicating lower fouling propensity. Although the EPS concentration showed increase in the middle of the operation, this was able to enhance flocculation and aggregation, leading to enlarged flocs and porous cake layer. This enhancement of flocculation ability was also proved by high zeta potential value of sludge. The remarkable reduction of total SMP concentration as well as higher PN/PS ratio of SMP by PAC addition facilitated effective membrane fouling mitigation by improving hydrophobicity and settleability of sludge. On the other hand, the absence of PAC could lead to severe pore blocking and deterioration of membrane permeability, mainly caused by the high amount of SMP_c in mixed liquor. Although the dominant mechanism for both reactors were cake formation, the increase of R_p and R_m in the AnMBR without PAC proved the occurrence of severe pore clogging and irreversible fouling. The analysis of cake layer composition showed that SMP_c was the main contributor of membrane fouling to AnMBR without PAC. Although PAC addition lowered microbial diversity level, it could effectively change the microbial composition not only by selective enrichment of hydrogen/formate-utilizing methanogens, hydrogen-producing bacteria, and denitrifying bacteria, but also by reduction of filamentous and fouling-related microorganisms.

In conclusion, the optimal PAC addition enhanced nutrient, TOC and COD removal performance. This result implied that PAC could act as supporting media for anaerobic microorganism growth and enrichment of methanogens and denitrifying microorganisms, and also effectively adsorb and biodegrade dissolved and colloidal organic matter. Although the main cause of membrane fouling in conventional AnMBR was pore blocking and irreversible fouling induced by SMP, the PAC addition could effectively prevent pore clogging as well as irreversible fouling by not only improving flocculation ability and hydrophobicity of sludge, but also lowering the abundance of fouling-associated microorganisms.

6.2 Recommendations

The research has investigated an optimum dosage of PAC to AnMBR treating synthetic wastewater, and then the long-term effect of the optimal PAC addition on the removal performance as well as membrane fouling behaviour compared to conventional submerged AnMBR. Further research regarding this topic can provide a better understanding of the optimisation of PAC dose to conventional AnMBRs as well as removal performance and membrane fouling control. Some recommendations for future study are summarized as following:

- Overdosed or exhausted PAC can cause poorer membrane fouling as well as degradation of pollutant removal performances, due to its potential to be fouled. Unfortunately, the replenishment ratio of PAC in AnMBR remains as a challenging issue, while that of optimal PAC for effective fouling reduction in aerobic MBR was reported as 1.67 % (Zhang et al., 2019b). Thus, further investigation regarding the optimal dosage and replenishment ratio of PAC can be carried out. Moreover, the requirement of partial replacement can be investigated in long term operation, and the effect of different replacement ratios of PAC can also be analysed for not only the best enhancement of performance, but also control of membrane fouling.
- Since PAC is one of the particulate enhancers, comparatively large particle size could result in abrasions and damages to membrane. Thus, more studies on the membrane lifespan with PAC is necessary for better understanding of the membrane performance. Additionally, the optimisation of PAC particle size is also essential for the operation of conventional AnMBR.
- More studies with regard to the long-term effect of PAC addition for wastewater treatment can be carried out.

- As the operating conditions such as pH level, HRT, and OLR highly affects the performance of anaerobic digestion as well as the fouling propensity, further studies under different conditions can be investigated.
- Further studies on different kinds of novel enhancers, such as waste yeast, zeolite, and beads, are required, since there are fewer number of studies compared to those on activated carbon. The optimal conditions for each enhancer can be analysed for a better insight into the effect of each enhancer compared to PAC.
- The improvement of methane-containing biogas production with PAC addition to AnMBR can be investigated for the analysis of energy efficiency.
- In spite of several studies and applications having been conducted in AnMBR with enhancer addition, most of the studies were confined to bench-scale or lab-scale experiments. The major barrier limiting scaling-up and broader application of AnMBRs can be membrane fouling, since membrane is one of the largest fractions to the capital and operational costs in AnMBRs. The capital cost of a full-scale submerged AnMBR system was reported to be about 800 USD/m^3/day. It was also estimated that around 72.3% of capital cost accounted for membrane fraction with the assumed capacity of 20,000 m^3/day, which was higher than that of aerobic MBR system (25–60%) (Lin et al., 2011; Lin et al., 2013a). On the other hand, the operational cost including gas scouring, pumping and sludge disposal for submerged AnMBR was reported to be around a third of that of the aerobic MBR system, which was about USD 235,000/year and USD 822,741/year, respectively (Lin et al., 2011). Thus, the full-scale AnMBR system could be economic feasibility by adopting some solutions to reduce membrane costs. In regard to activated carbon's relatively cheap cost (0.6–20 USD/kg) compared to other chemical enhancers, full-scale AnMBR with PAC addition is also feasible

(Chiappero et al., 2020). Thus, careful examination and further evaluation of PAC addition for the scale-up should be conducted.

Reference

Agler, M.T., Aydinkaya, Z., Cummings, T.A., Beers, A.R., Angenent, L.T. 2010. Anaerobic digestion of brewery primary sludge to enhance bioenergy generation: A comparison between low-and high-rate solids treatment and different temperatures. Bioresource technology, 101(15), 5842-5851.

Ahmad, A., Ghufran, R., Wahid, Z.A. 2011. Role of calcium oxide in sludge granulation and methanogenesis for the treatment of palm oil mill effluent using UASB reactor. Journal of Hazardous Materials, 198, 40-48.

Akram, A., Stuckey, D.C. 2008. Flux and performance improvement in a submerged anaerobic membrane bioreactor (SAMBR) using powdered activated carbon (PAC). Process Biochemistry, 43(1), 93-102.

Alkhudhiri, A., Darwish, N., Hilal, N. 2012. Membrane distillation: A comprehensive review. Desalination, 287, 2-18.

Arabi, S., Nakhla, G. 2008b. Impact of protein/carbohydrate ratio in the feed wastewater on the membrane fouling in membrane bioreactors. Journal of Membrane Science, 324(1), 142-150.

Asif, M.B., Ren, B., Li, C., Maqbool, T., Zhang, X., Zhang, Z. 2020. Powdered activated carbon – Membrane bioreactor (PAC-MBR): Impacts of high PAC concentration on micropollutant removal and microbial communities. Science of the Total Environment, 745, 141090.

Aslam, M., McCarty, P.L., Bae, J., Kim, J. 2014. The effect of fluidized media characteristics on membrane fouling and energy consumption in anaerobic fluidized membrane bioreactors. Separation and Purification Technology, 132, 10-15.

Aslam, M., McCarty, P.L., Shin, C., Bae, J., Kim, J. 2017. Low energy single-staged anaerobic fluidized bed ceramic membrane bioreactor (AFCMBR) for wastewater treatment. Bioresource Technology, 240, 33-41.

Aslam, M., Yang, P., Lee, P.-H., Kim, J. 2018. Novel staged anaerobic fluidized bed ceramic membrane bioreactor: Energy reduction, fouling control and microbial characterization. Journal of Membrane Science, 553, 200-208.

Aydin, S., Ince, B., Cetecioglu, Z., Arikan, O., Ozbayram, E.G., Shahi, A., Ince, O. 2015. Combined effect of erythromycin, tetracycline and sulfamethoxazole on performance of anaerobic sequencing batch reactors. Bioresource Technology, 186, 207-214.

Baek, G., Kim, J., Lee, C. 2019. A review of the effects of iron compounds on methanogenesis in anaerobic environments. Renewable and Sustainable Energy Reviews, 113, 109282.

Baêta, B.E.L., Lima, D.R.S., Silva, S.Q., Aquino, S.F. 2016 Influence of the applied organic load (OLR) on textile wastewater treatment using submerged anaerobic membrane bioreactors (SAMBR) in the presence of redox mediator and powdered activated carbon (PAC). Brazilian Journal of Chemical Engineering, 33, 817-825.

Baêta, B.E.L., Luna, H.J., Sanson, A.L., Silva, S.Q., Aquino, S.F. 2013. Degradation of a model azo dye in submerged anaerobic membrane bioreactor (SAMBR) operated with powdered activated carbon (PAC). Journal of Environmental Management, 128, 462-470.

Baêta, B.E.L., Ramos, R.L., Lima, D.R.S., Aquino, S.F. 2012. Use of submerged anaerobic membrane bioreactor (SAMBR) containing powdered activated carbon (PAC) for the treatment of textile effluents. Water Science and Technology, 65(9), 1540-1547.

Barros, A.R.M., de Sousa Rollemberg, S.L., de Carvalho, C.d.A., Moura, I.H.H., Firmino, P.I.M., dos Santos, A.B. 2020. Effect of calcium addition on the formation and maintenance of aerobic granular sludge (AGS) in simultaneous fill/draw mode sequencing batch reactors (SBRs). Journal of environmental management, 255, 109850.

Barua, S., Zakaria, B.S., Lin, L., Dhar, B.R. 2019. Magnetite doped granular activated carbon as an additive for high-performance anaerobic digestion. Materials Science for Energy Technologies, 2(3), 377-384.

Bokulich, N.A., Kaehler, B.D., Rideout, J.R., Dillon, M., Bolyen, E., Knight, R., Huttley, G.A., Gregory Caporaso, J. 2018. Optimizing taxonomic classification of marker-gene amplicon sequences with QIIME 2's q2-feature-classifier plugin. Microbiome, 6(1), 90.

Bolyen, E., Rideout, J.R., Dillon, M.R., Bokulich, N.A., Abnet, C.C., Al-Ghalith, G.A., Alexander, H., Alm, E.J., Arumugam, M., Asnicar, F. 2019. Reproducible, interactive, scalable and extensible microbiome data science using QIIME 2. Nature biotechnology, 37(8), 852-857.

Bräuer, S.L., Cadillo-Quiroz, H., Ward, R.J., Yavitt, J.B., Zinder, S.H. 2011. Methanoregula boonei gen. nov., sp. nov., an acidiphilic methanogen isolated from an acidic peat bog. International Journal of Systematic and Evolutionary Microbiology, 61(1), 45-52.

Brito, G.C.B., Lange, L.C., Santos, V.L., Amaral, M.C.S., Moravia, W.G. 2019. Long-term evaluation of membrane bioreactor inoculated with commercial baker's yeast treating landfill leachate: pollutant removal, microorganism dynamic and membrane fouling. Water Science and Technology, 79(2), 398-410.

Cabrol, L., Marone, A., Tapia-Venegas, E., Steyer, J.-P., Ruiz-Filippi, G., Trably, E. 2017. Microbial ecology of fermentative hydrogen producing bioprocesses: useful insights for driving the ecosystem function. FEMS Microbiology Reviews, 41(2), 158-181.

Callahan, B.J., McMurdie, P.J., Rosen, M.J., Han, A.W., Johnson, A.J.A., Holmes, S.P. 2016. DADA2: High-resolution sample inference from Illumina amplicon data. Nature Methods, 13(7), 581-583.

Cetecioglu, Z., Ince, B., Gros, M., Rodriguez-Mozaz, S., Barceló, D., Orhon, D., Ince, O. 2013. Chronic impact of tetracycline on the biodegradation of an organic substrate mixture under anaerobic conditions. Water Research, 47(9), 2959-2969.

Cetecioglu, Z., Ince, B., Orhon, D., Ince, O. 2016. Anaerobic sulfamethoxazole degradation is driven by homoacetogenesis coupled with hydrogenotrophic methanogenesis. Water Research, 90, 79-89.

Charfi, A., Amar, N.B., Harmand, J. 2012. Analysis of fouling mechanisms in anaerobic membrane bioreactors. Water research, 46(8), 2637-2650.

Charfi, A., Aslam, M., Kim, J. 2018a. Modelling approach to better control biofouling in fluidized bed membrane bioreactor for wastewater treatment. Chemosphere, 191, 136-144.

Charfi, A., Aslam, M., Lesage, G., Heran, M., Kim, J. 2017. Macroscopic approach to develop fouling model under GAC fluidization in anaerobic fluidized bed membrane bioreactor. Journal of Industrial and Engineering Chemistry, 49, 219-229.

Charfi, A., Park, E., Aslam, M., Kim, J. 2018b. Particle-sparged anaerobic membrane bioreactor with fluidized polyethylene terephthalate beads for domestic wastewater treatment: Modelling approach and fouling control. Bioresource Technology, 258, 263-269.

Chen, C., Guo, W., Ngo, H., Chang, S., Nguyen, D., Zhang, J., Liang, S., Guo, J., Zhang, X. 2018a. Effects of C/N ratio on the performance of a hybrid sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor for municipal wastewater treatment. Bioresource technology, 247, 340-346.

Chen, K., Wang, X., Li, X., Qian, J., Xiao, X. 2011. Impacts of sludge retention time on the performance of submerged membrane bioreactor with the addition of calcium ion. Separation and Purification Technology, 82, 148-155.

Chen, L., Cheng, P., Ye, L., Chen, H., Xu, X., Zhu, L. 2020. Biological performance and fouling mitigation in the biochar-amended anaerobic membrane bioreactor (AnMBR) treating pharmaceutical wastewater. Bioresource Technology, 302, 122805.

Chen, R., Nie, Y., Hu, Y., Miao, R., Utashiro, T., Li, Q., Xu, M., Li, Y.-Y. 2017. Fouling behaviour of soluble microbial products and extracellular polymeric substances in a submerged anaerobic membrane bioreactor treating low-strength wastewater at room temperature. Journal of Membrane Science, 531, 1-9.

Chen, W.-H., Tsai, C.-Y., Chen, S.-Y., Sung, S., Lin, J.-G. 2019a. Treatment of campus domestic wastewater using ambient-temperature anaerobic fluidized membrane bioreactors with zeolites as carriers. International Biodeterioration & Biodegradation, 136, 49-54.

Chen, X., Ottosen, L.D.M., Kofoed, M.V.W. 2019b. How Low Can You Go: Methane Production of Methanobacterium congolense at Low CO2 Concentrations. Frontiers in Bioengineering and Biotechnology, 7(34).

Chen, X., Suwarno, S.R., Chong, T.H., McDougald, D., Kjelleberg, S., Cohen, Y., Fane, A.G., Rice, S.A. 2013. Dynamics of biofilm formation under different nutrient levels and the effect on biofouling of a reverse osmosis membrane system. Biofouling, 29(3), 319-330.

Chen, Y., Wu, Y., Wang, D., Li, H., Wang, Q., Liu, Y., Peng, L., Yang, Q., Li, X., Zeng, G., Chen, Y. 2018b. Understanding the mechanisms of how poly aluminium chloride inhibits short-chain fatty acids production from anaerobic fermentation of waste activated sludge. Chemical Engineering Journal, 334, 1351-1360.

Cheng, D., Ngo, H.H., Guo, W., Chang, S.W., Nguyen, D.D., Nguyen, Q.A., Zhang, J., Liang, S. 2021. Improving sulfonamide antibiotics removal from swine wastewater by supplying a new pomelo peel derived biochar in an anaerobic membrane bioreactor. Bioresource Technology, 319, 124160.

Cheng, D., Ngo, H.H., Guo, W., Liu, Y., Chang, S.W., Nguyen, D.D., Nghiem, L.D., Zhou, J., Ni, B. 2018. Anaerobic membrane bioreactors for antibiotic wastewater treatment: Performance and membrane fouling issues. Bioresource Technology, 267, 714-724.

Cheng, H., Li, Y., Guo, G., Zhang, T., Qin, Y., Hao, T., Li, Y.-Y. 2020. Advanced methanogenic performance and fouling mechanism investigation of a high-solid anaerobic membrane bioreactor (AnMBR) for the co-digestion of food waste and sewage sludge. Water Research, 187, 116436.

Chiappero, M., Norouzi, O., Hu, M., Demichelis, F., Berruti, F., Di Maria, F., Mašek, O., Fiore, S. 2020. Review of biochar role as additive in anaerobic digestion processes. Renewable and Sustainable Energy Reviews, 131, 110037.

Cho, K., Choi, M., Jeong, D., Lee, S., Bae, H. 2017. Comparison of inoculum sources for long-term process performance and fate of ANAMMOX bacteria niche in poly(vinyl alcohol)/sodium alginate gel beads. Chemosphere, 185, 394-402.

Chong, C.T. 2015. Performance of anaerobic membrane bioreactors (AnMBRs) with different dosages of powdered activated carbon (PAC) at mesophilic regime in membrane fouling control, UTAR.

Choo, K.-H., Lee, C.-H. 1996. Membrane fouling mechanisms in the membrane-coupled anaerobic bioreactor. Water Research, 30(8), 1771-1780.

Connor, R., Renata, A., Ortigara, C., Koncagül, E., Uhlenbrook, S., Lamizana-Diallo, B.M., Zadeh, S.M., Qadir, M., Kjellén, M., Sjödin, J. 2017. The United Nations world water development report 2017. Wastewater: The untapped resource. The United Nations World Water Development Report.

Cunha, J.R., Schott, C., van der Weijden, R.D., Leal, L.H., Zeeman, G., Buisman, C. 2018. Calcium addition to increase the production of phosphate granules in anaerobic treatment of black water. Water Research, 130, 333-342.

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

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

Detman, A., Mielecki, D., Pleśniak, Ł., Bucha, M., Janiga, M., Matyasik, I., Chojnacka, A., Jędrysek, M.-O., Błaszczyk, M.K., Sikora, A. 2018. Methane-yielding microbial communities processing lactate-rich substrates: a piece of the anaerobic digestion puzzle. Biotechnology for Biofuels, 11(1), 116.

Ding, A., Liang, H., Qu, F., Bai, L., Li, G., Ngo, H.H., Guo, W. 2014. Effect of granular activated carbon addition on the effluent properties and fouling potentials of membrane-coupled expanded granular sludge bed process. Bioresource Technology, 171, 240-246.

Ding, A., Pronk, W., Qu, F., Ma, J., Li, G., Li, K., Liang, H. 2015. Effect of calcium addition on sludge properties and membrane fouling potential of the membrane-coupled expanded granular sludge bed process. Journal of Membrane Science, 489, 55-63.

Dong, Q., Parker, W., Dagnew, M. 2015. Impact of FeCl3 dosing on AnMBR treatment of municipal wastewater. Water Research, 80, 281-293.

Drews, A. 2010. Membrane fouling in membrane bioreactors—Characterisation, contradictions, cause and cures. Journal of Membrane Science, 363(1), 1-28.

Du, X., Liu, G., Qu, F., Li, K., Shao, S., Li, G., Liang, H. 2017. Removal of iron, manganese and ammonia from groundwater using a PAC-MBR system: The anti-pollution ability, microbial population and membrane fouling. Desalination, 403, 97-106.

Düppenbecker, B., Engelhart, M., Cornel, P. 2017a. Fouling mitigation in Anaerobic Membrane Bioreactor using fluidized glass beads: Evaluation fitness for purpose of ceramic membranes. Journal of Membrane Science, 537, 69-82.

Düppenbecker, B., Kale, S., Engelhart, M., Cornel, P. 2017b. Fluidized glass beads reduce fouling in a novel anaerobic membrane bioreactor. Water Science and Technology, 76(4), 953-962.

Dutta, K., Lee, M.-Y., Lai, W.W.-P., Lee, C.H., Lin, A.Y.-C., Lin, C.-F., Lin, J.-G. 2014a. Removal of pharmaceuticals and organic matter from municipal wastewater using two-stage anaerobic fluidized membrane bioreactor. Bioresource Technology, 165, 42-49.

Dutta, K., Tsai, C.-Y., Chen, W.-H., Lin, J.-G. 2014b. Effect of carriers on the performance of anaerobic sequencing batch biofilm reactor treating synthetic municipal wastewater. International Biodeterioration & Biodegradation, 95, 84-88.

Evans, P.J., Parameswaran, P., Lim, K., Bae, J., Shin, C., Ho, J., McCarty, P.L. 2019. A Comparative Pilot-Scale Evaluation of Gas-Sparged and Granular Activated Carbon-Fluidized Anaerobic Membrane Bioreactors for Domestic Wastewater Treatment. Bioresource Technology.

Fagbohungbe, M.O., Herbert, B.M.J., Hurst, L., Ibeto, C.N., Li, H., Usmani, S.Q., Semple, K.T. 2017. The challenges of anaerobic digestion and the role of biochar in optimizing anaerobic digestion. Waste Management, 61, 236-249.

Faria, C., Souza, D., Pontes, T., Amaral, M., Fonseca, F. 2019. Strategies of anaerobic sludge granulation in an EGSB reactor. Journal of environmental management, 244, 69-76.

Feng, Y., Zhang, Y., Quan, X., Chen, S. 2014. Enhanced anaerobic digestion of waste activated sludge digestion by the addition of zero valent iron. Water research, 52, 242-250.

Florentino, A.P., Sharaf, A., Zhang, L., Liu, Y. 2019. Overcoming ammonia inhibition in anaerobic blackwater treatment with granular activated carbon: The role of electroactive microorganisms. Environmental Science: Water Research and Technology, 5(2), 383-396.

Gagliano, M.C., Sudmalis, D., Temmink, H., Plugge, C.M. 2020. Calcium effect on microbial activity and biomass aggregation during anaerobic digestion at high salinity. New Biotechnology.

Gao, D.-W., Hu, Q., Yao, C., Ren, N.-Q. 2014a. Treatment of domestic wastewater by an integrated anaerobic fluidized-bed membrane bioreactor under moderate to low temperature conditions. Bioresource Technology, 159, 193-198.

Gao, D.-W., Hu, Q., Yao, C., Ren, N.-Q., Wu, W.-M. 2014b. Integrated anaerobic fluidized-bed membrane bioreactor for domestic wastewater treatment. Chemical Engineering Journal, 240, 362-368.

Gao, W.J., Lin, H.J., Leung, K.T., Schraft, H., Liao, B.Q. 2011. Structure of cake layer in a submerged anaerobic membrane bioreactor. Journal of Membrane Science, 374(1), 110-120.

Gao, Y., Ma, D., Yue, Q., Gao, B., Huang, X. 2016. Effect of powdered activated carbon (PAC) on MBR performance and effluent trihalomethane formation: At the initial stage of PAC addition. Bioresource Technology, 216, 838-844.

Glöckner, F.O., Yilmaz, P., Quast, C., Gerken, J., Beccati, A., Ciuprina, A., Bruns, G., Yarza, P., Peplies, J., Westram, R., Ludwig, W. 2017. 25 years of serving the community with ribosomal RNA gene reference databases and tools. Journal of Biotechnology, 261, 169-176.

Guo, W., Ngo, H.-H., Li, J. 2012. A mini-review on membrane fouling. Bioresource technology, 122, 27-34.

Gutha, Y., Pathak, J.L., Zhang, W., Zhang, Y., Jiao, X. 2017. Antibacterial and wound healing properties of chitosan/poly (vinyl alcohol)/zinc oxide beads (CS/PVA/ZnO). International journal of biological macromolecules, 103, 234-241.

Han, X., Wang, Z., Wang, X., Zheng, X., Ma, J., Wu, Z. 2016. Microbial responses to membrane cleaning using sodium hypochlorite in membrane bioreactors: Cell integrity, key enzymes and intracellular reactive oxygen species. Water Research, 88, 293-300.

Harb, M., Xiong, Y., Guest, J., Amy, G., Hong, P.-Y. 2015. Differences in microbial communities and performance between suspended and attached growth anaerobic membrane bioreactors treating synthetic municipal wastewater. Environmental Science: Water Research & Technology, 1(6), 800-813.

Hu, A.Y., Stuckey, D.C. 2007. Activated Carbon Addition to a Submerged Anaerobic

Membrane Bioreactor: Effect on Performance, Transmembrane Pressure, and Flux. Journal of Environmental Engineering, 133(1), 73-80.

Hu, J., Shang, R., Deng, H., Heijman, S.G.J., Rietveld, L.C. 2014. Effect of PAC dosage in a pilot-scale PAC–MBR treating micro-polluted surface water. Bioresource Technology, 154, 290-296.

Hu, Y., Yang, Y., Wang, X.C., Hao Ngo, H., Sun, Q., Li, S., Tang, J., Yu, Z. 2017. Effects of powdered activated carbon addition on filtration performance and dynamic membrane layer properties in a hybrid DMBR process. Chemical Engineering Journal, 327, 39-50.

Huang, T.-L., Zhou, S.-L., Zhang, H.-H., Bai, S.-Y., He, X.-X., Yang, X. 2015. Nitrogen Removal Characteristics of a Newly Isolated Indigenous Aerobic Denitrifier from Oligotrophic Drinking Water Reservoir, Zoogloea sp. N299. International journal of molecular sciences, 16(5), 10038-10060.

Huang, X., Wu, J. 2008. Improvement of membrane filterability of the mixed liquor in a membrane bioreactor by ozonation. Journal of Membrane Science, 318(1), 210-216.

Huang, Z., Ong, S.-L., Ng, H. 2011a. Submerged Anaerobic Membrane Bioreactor for Low-Strength Wastewater Treatment: Effect of HRT and SRT on Treatment Performance and Membrane Fouling. Water research, 45, 705-13.

Huang, Z., Ong, S.L., Ng, H.Y. 2011b. Submerged anaerobic membrane bioreactor for low-strength wastewater treatment: Effect of HRT and SRT on treatment performance and membrane fouling. Water Research, 45(2), 705-713.

Iorhemen, O.T., Hamza, R.A., Tay, J.H. 2016. Membrane bioreactor (MBR) technology for wastewater treatment and reclamation: membrane fouling. Membranes, 6(2), 33.

Jamal Khan, S., Visvanathan, C., Jegatheesan, V. 2012. Effect of powdered activated carbon (PAC) and cationic polymer on biofouling mitigation in hybrid MBRs. Bioresource Technology, 113, 165-168.

Ji, J., Li, J., Qiu, J., Li, X. 2014. Polyacrylamide–starch composite flocculant as a membrane fouling reducer: Key factors of fouling reduction. Separation and Purification Technology, 131, 1-7.

Jia, S., Han, H., Hou, B., Zhuang, H., Fang, F., Zhao, Q. 2014. Treatment of coal gasification wastewater by membrane bioreactor hybrid powdered activated carbon (MBR–PAC) system. Chemosphere, 117, 753-759.

Jiang, B., Zeng, Q., Liu, J., Hou, Y., Xu, J., Li, H., Shi, S., Ma, F. 2020. Enhanced treatment performance of phenol wastewater and membrane antifouling by biochar-assisted EMBR. Bioresource Technology, 306, 123147.

Jiao, X., Gutha, Y., Zhang, W. 2017. Application of chitosan/poly(vinyl alcohol)/CuO (CS/PVA/CuO) beads as an adsorbent material for the removal of Pb(II) from aqueous environment. Colloids and Surfaces B: Biointerfaces, 149, 184-195.

Jinsong, Z., Chuan, C.H., Jiti, Z., Fane, A. 2006. Effect of sludge retention time on membrane bio-fouling intensity in a submerged membrane bioreactor. Separation Science and Technology, 41(7), 1313-1329.

Jo, Y. 2017. Membrane hybrid systems in wastewater treatment.

Jo, Y., Johir, M.A.H., Cho, Y., Naidu, G., Rice, S.A., McDougald, D., Kandasamy, J., Vigneswaran, S., Sun, S. 2019. A comparative study on nitric oxide and hypochlorite as a membrane cleaning agent to minimise biofilm growth in a membrane bioreactor (MBR) process. Biochemical Engineering Journal, 148, 9-15.

Johir, M.A.H., Aryal, R., Vigneswaran, S., Kandasamy, J., Grasmick, A. 2011. Influence of supporting media in suspension on membrane fouling reduction in submerged membrane bioreactor (SMBR). Journal of Membrane Science, 374(1), 121-128.

Juntawang, C., Rongsayamanont, C., Khan, E. 2017. Entrapped cells-based-anaerobic membrane bioreactor treating domestic wastewater: Performances, fouling, and bacterial community structure. Chemosphere, 187, 147-155.

Katoh, K., Misawa, K., Kuma, K.-i., Miyata, T. 2002. MAFFT: a novel method for rapid multiple sequence alignment based on fast Fourier transform. Nucleic acids research, 30(14), 3059-3066.

Kaya, Y., Bacaksiz, A.M., Golebatmaz, U., Vergili, I., Gönder, Z.B., Yilmaz, G. 2016. Improving the performance of an aerobic membrane bioreactor (MBR) treating pharmaceutical wastewater with powdered activated carbon (PAC) addition. Bioprocess and biosystems engineering, 39(4), 661-676.

Khan, M.A. 2019. Optimization and performance improvement of Anaerobic Membrance Biocreactor (AnMBR) for volatile fatty acid and biohydrogen production.

Khan, M.A., Ngo, H.H., Guo, W., Liu, Y., Chang, S.W., Nguyen, D.D., Nghiem, L.D., Liang, H. 2018. Can membrane bioreactor be a smart option for water treatment? Bioresource Technology Reports, 4, 80-87.

Khan, M.A., Ngo, H.H., Guo, W., Liu, Y., Nghiem, L.D., Chang, S.W., Nguyen, D.D., Zhang, S., Luo, G., Jia, H. 2019. Optimization of hydraulic retention time and organic loading rate for volatile fatty acid production from low strength wastewater in an anaerobic membrane bioreactor. Bioresource Technology, 271, 100-108.

Khan, M.A., Ngo, H.H., Guo, W.S., Liu, Y.W., Zhou, J.L., Zhang, J., Liang, S., Ni, B.J., Zhang, X.B., Wang, J. 2016. Comparing the value of bioproducts from different stages of anaerobic membrane bioreactors. Bioresource Technology, 214, 816-825.

Kong, X., Yu, S., Xu, S., Fang, W., Liu, J., Li, H. 2018. Effect of Fe0 addition on volatile fatty acids evolution on anaerobic digestion at high organic loading rates. Waste Management, 71, 719-727.

Korotta-Gamage, S.M., Sathasivan, A. 2017. A review: Potential and challenges of biologically activated carbon to remove natural organic matter in drinking water purification process. Chemosphere, 167, 120-138.

Lei, Z., Yang, S., Li, X., Wen, W., Huang, X., Yang, Y., Wang, X., Li, Y.-Y., Sano, D., Chen, R. 2019. Revisiting the effects of powdered activated carbon on membrane fouling mitigation in an anaerobic membrane bioreactor by evaluating long-term impacts on the surface layer. Water Research, 167, 115137.

Li, Y., Sim, L.N., Ho, J.S., Chong, T.H., Wu, B., Liu, Y. 2020. Integration of an anaerobic fluidized-bed membrane bioreactor (MBR) with zeolite adsorption and reverse osmosis (RO) for municipal wastewater reclamation: Comparison with an anoxic-aerobic MBR coupled with RO. Chemosphere, 245, 125569.

Liao, B.-Q., Kraemer, J.T., Bagley, D.M. 2006. Anaerobic membrane bioreactors: applications and research directions. Critical Reviews in Environmental Science and Technology, 36(6), 489-530.

Lim, M., Ahmad, R., Guo, J., Tibi, F., Kim, M., Kim, J. 2019. Removals of micropollutants in staged anaerobic fluidized bed membrane bioreactor for low-strength wastewater treatment. Process Safety and Environmental Protection, 127, 162-170.

Lin, H., Chen, J., Wang, F., Ding, L., Hong, H. 2011. Feasibility evaluation of submerged anaerobic membrane bioreactor for municipal secondary wastewater treatment. Desalination, 280(1), 120-126.

Lin, H., Peng, W., Zhang, M., Chen, J., Hong, H., Zhang, Y. 2013a. A review on anaerobic membrane bioreactors: Applications, membrane fouling and future perspectives. Desalination, 314, 169-188.

Lin, L., Lei, Z., Wang, L., Liu, X., Zhang, Y., Wan, C., Lee, D.-J., Tay, J.H. 2013b. Adsorption mechanisms of high-levels of ammonium onto natural and NaCl-modified zeolites. Separation and Purification Technology, 103, 15-20.

Liu, Y.-J., Sun, D.D. 2012. Membrane fouling mechanism in dead-end microfiltration of denitrifying granular sludge mixed liquors developed in SBRs at different calcium concentrations. Journal of Membrane Science, 396, 74-82.

Liu, Y., Balkwill, D.L., Aldrich, H.C., Drake, G.R., Boone, D.R. 1999. Characterization of the anaerobic propionate-degrading syntrophs Smithella propionica gen. nov., sp. nov. and Syntrophobacter wolinii. International Journal of Systematic and Evolutionary Microbiology, 49(2), 545-556.

Liu, Y., Zhang, Y., Quan, X., Li, Y., Zhao, Z., Meng, X., Chen, S. 2012. Optimization of anaerobic acidogenesis by adding Fe0 powder to enhance anaerobic wastewater treatment. Chemical engineering journal, 192, 179-185.

Lozupone, C., Knight, R. 2005. UniFrac: a new phylogenetic method for comparing microbial communities. Applied and environmental microbiology, 71(12), 8228-8235.

Lozupone, C.A., Hamady, M., Kelley, S.T., Knight, R. 2007. Quantitative and qualitative beta diversity measures lead to different insights into factors that structure microbial communities. Applied and environmental microbiology, 73(5), 1576-1585.

Lü, F., Luo, C., Shao, L., He, P. 2016. Biochar alleviates combined stress of ammonium and acids by firstly enriching Methanosaeta and then Methanosarcina. Water Research, 90, 34-43.

Luo, C., Lü, F., Shao, L., He, P. 2015. Application of eco-compatible biochar in anaerobic digestion to relieve acid stress and promote the selective colonization of functional microbes. Water Research, 68, 710-718.

Maaz, M., Yasin, M., Aslam, M., Kumar, G., Atabani, A.E., Idrees, M., Anjum, F., Jamil, F., Ahmad, R., Khan, A.L., Lesage, G., Heran, M., Kim, J. 2019. Anaerobic membrane bioreactors for wastewater treatment: Novel configurations, fouling control and energy considerations. Bioresource Technology, 283, 358-372.

Masebinu, S.O., Akinlabi, E.T., Muzenda, E., Aboyade, A.O. 2019. A review of biochar properties and their roles in mitigating challenges with anaerobic digestion. Renewable and Sustainable Energy Reviews, 103, 291-307.

Mei, X., Wang, Z., Miao, Y., Wu, Z. 2016. Recover energy from domestic wastewater using anaerobic membrane bioreactor: Operating parameters optimization and energy balance analysis. Energy, 98, 146-154.

Melin, T., Jefferson, B., Bixio, D., Thoeye, C., De Wilde, W., De Koning, J., van der Graaf, J., Wintgens, T. 2006. Membrane bioreactor technology for wastewater treatment and reuse. Desalination, 187(1), 271-282.

Meng, F., Chae, S.-R., Drews, A., Kraume, M., Shin, H.-S., Yang, F. 2009. Recent advances in membrane bioreactors (MBRs): membrane fouling and membrane material. Water research, 43(6), 1489-1512.

Meng, F., Zhang, S., Oh, Y., Zhou, Z., Shin, H.-S., Chae, S.-R. 2017. Fouling in membrane bioreactors: An updated review. Water Research, 114, 151-180.

Ming, J., Wang, Q., Yoza, B.A., Liang, J., Guo, H., Li, J., Guo, S., Chen, C. 2020. Bioreactor performance using biochar and its effect on aerobic granulation. Bioresource Technology, 300, 122620.

Mohamadi, S., Hazrati, H., Shayegan, J. 2020. Influence of a new method of applying adsorbents on membrane fouling in MBR systems. Water and Environment Journal, 34, 355-366.

Montalvo, S., Huiliñir, C., Borja, R., Sánchez, E., Herrmann, C. 2020. Application of zeolites for biological treatment processes of solid wastes and wastewaters – A review. Bioresource Technology, 301, 122808.

Mumme, J., Srocke, F., Heeg, K., Werner, M. 2014. Use of biochars in anaerobic digestion. Bioresource Technology, 164, 189-197.

Munirasu, S., Haija, M.A., Banat, F. 2016. Use of membrane technology for oil field and refinery produced water treatment—A review. Process Safety and Environmental Protection, 100, 183-202.

Neira, K., Jeison, D. 2010. Anaerobic co-digestion of surplus yeast and wastewater to increase energy recovery in breweries. Water Science and Technology, 61(5), 1129-1135.

Nguyen, A.Q., Nguyen, L.N., Phan, H.V., Galway, B., Bustamante, H., Nghiem, L.D. 2019.

Effects of operational disturbance and subsequent recovery process on microbial community during a pilot-scale anaerobic co-digestion. International Biodeterioration & Biodegradation, 138, 70-77.

Noh, J.H., Choi, H., Kim, H.Y., Choi, S., Maeng, S.K. 2019. Reducing bacterial aerosol emissions from membrane bioreactors: The impact of SRT and the addition of PAC and calcium. Water Research, 156, 58-70.

Nordell, E., Hansson, A.B., Karlsson, M. 2013. Zeolites relieves inhibitory stress from high concentrations of long chain fatty acids. Waste management, 33(12), 2659-2663.

Oliveira, F.R., Surendra, K.C., Jaisi, D.P., Lu, H., Unal-Tosun, G., Sung, S., Khanal, S.K. 2020. Alleviating sulfide toxicity using biochar during anaerobic treatment of sulfate-laden wastewater. Bioresource Technology, 301, 122711.

Ozgun, H., Dereli, R.K., Ersahin, M.E., Kinaci, C., Spanjers, H., van Lier, J.B. 2013. A review of anaerobic membrane bioreactors for municipal wastewater treatment: Integration options, limitations and expectations. Separation and Purification Technology, 118, 89-104.

Ozgun, H., Gimenez, J.B., Ersahin, M.E., Tao, Y., Spanjers, H., van Lier, J.B. 2015. Impact of membrane addition for effluent extraction on the performance and sludge characteristics of upflow anaerobic sludge blanket reactors treating municipal wastewater. Journal of Membrane Science, 479, 95-104.

Pan, J., Ma, J., Zhai, L., Luo, T., Mei, Z., Liu, H. 2019. Achievements of biochar application for enhanced anaerobic digestion: A review. Bioresource Technology, 292, 122058.

Pan, J.R., Su, Y.-C., Huang, C., Lee, H.-C. 2010. Effect of sludge characteristics on membrane fouling in membrane bioreactors. Journal of Membrane Science, 349(1), 287-294.

Pandey, S., Sarkar, S. 2017. Anaerobic treatment of wastewater using a two-stage packed-bed reactor containing polyvinyl alcohol gel beads as biofilm carrier. Journal of Environmental Chemical Engineering, 5(2), 1575-1585.

Park, H., Choo, K.-H., Lee, C.-H. 1999. Flux enhancement with powdered activated carbon addition in the membrane anaerobic bioreactor. Separation Science and Technology, 34(14), 2781-2792.

Park, J.-H., Park, J.-H., Je Seong, H., Sul, W.J., Jin, K.-H., Park, H.-D. 2018. Metagenomic insight into methanogenic reactors promoting direct interspecies electron transfer via granular activated carbon. Bioresource Technology, 259, 414-422.

Piasecka, A., Souffreau, C., Vandepitte, K., Vanysacker, L., Bilad, R.M., De Bie, T., Hellemans, B., De Meester, L., Yan, X., Declerck, P., Vankelecom, I.F.J. 2012. Analysis of the microbial community structure in a membrane bioreactor during initial stages of filtration. Biofouling, 28(2), 225-238.

Pirsaheb, M., Hossein Davood Abadi Farahani, M., Zinadini, S., Zinatizadeh, A.A., Rahimi, M., Vatanpour, V. 2019. Fabrication of high-performance antibiofouling ultrafiltration membranes with potential application in membrane bioreactors (MBRs) comprising polyethersulfone (PES) and polycitrate-Alumoxane (PC-A). Separation and Purification Technology, 211, 618-627.

Poirier, S., Madigou, C., Bouchez, T., Chapleur, O. 2017. Improving anaerobic digestion with support media: Mitigation of ammonia inhibition and effect on microbial communities. Bioresource Technology, 235, 229-239.

Pradhan, M., Aryal, R., Vigneswaran, S., Kandasamy, J. 2011. Application of air flow for mitigation of particle deposition in submerged membrane microfiltration. Desalination and

Water Treatment, 32(1-3), 201-207.

Price, M.N., Dehal, P.S., Arkin, A.P. 2010. FastTree 2 – Approximately Maximum-Likelihood Trees for Large Alignments. PLOS ONE, 5(3), e9490.

Qambrani, N.A., Rahman, M.M., Won, S., Shim, S., Ra, C. 2017. Biochar properties and eco-friendly applications for climate change mitigation, waste management, and wastewater treatment: A review. Renewable and Sustainable Energy Reviews, 79, 255-273.

Quast, C., Pruesse, E., Yilmaz, P., Gerken, J., Schweer, T., Yarza, P., Peplies, J., Glöckner, F.O. 2012. The SILVA ribosomal RNA gene database project: improved data processing and web-based tools. Nucleic Acids Research, 41(D1), D590-D596.

Ramos, C., García, A., Diez, V. 2014. Performance of an AnMBR pilot plant treating high-strength lipid wastewater: Biological and filtration processes. Water Research, 67, 203-215.

Reis, B.G., Silveira, A.L., Tostes Teixeira, L.P., Okuma, A.A., Lange, L.C., Amaral, M.C.S. 2017. Organic compounds removal and toxicity reduction of landfill leachate by commercial bakers' yeast and conventional bacteria based membrane bioreactor integrated with nanofiltration. Waste Management, 70, 170-180.

Robles, Á., Ruano, M.V., Charfi, A., Lesage, G., Heran, M., Harmand, J., Seco, A., Steyer, J.-P., Batstone, D.J., Kim, J. 2018. A review on anaerobic membrane bioreactors (AnMBRs) focused on modelling and control aspects. Bioresource technology.

Ruan, R., Cao, J., Li, C., Zheng, D., Luo, J. 2017. The influence of micro-oxygen addition on desulfurization performance and microbial communities during waste-activated sludge digestion in a rusty scrap iron-loaded anaerobic digester. Energies, 10(2), 258.

Salama, E.-S., Jeon, B.-H., Kurade, M.B., Patil, S.M., Usman, M., Li, X., Lim, H. 2020.

Enhanced anaerobic co-digestion of fat, oil, and grease by calcium addition: Boost of biomethane production and microbial community shift. Bioresource technology, 296, 122353.

Salgot, M., Folch, M. 2018. Wastewater treatment and water reuse. Current Opinion in Environmental Science & Health, 2, 64-74.

Sanguanpak, S., Chiemchaisri, C., Chiemchaisri, W., Yamamoto, K. 2015. Influence of operating pH on biodegradation performance and fouling propensity in membrane bioreactors for landfill leachate treatment. International Biodeterioration & Biodegradation, 102, 64-72.

Shanmugam, S.R., Adhikari, S., Nam, H., Kar Sajib, S. 2018. Effect of bio-char on methane generation from glucose and aqueous phase of algae liquefaction using mixed anaerobic cultures. Biomass and Bioenergy, 108, 479-486.

Shannon, C.E. 1948. A Mathematical Theory of Communication. Bell System Technical Journal, 27(3), 379-423.

Shen, R., Jing, Y., Feng, J., Luo, J., Yu, J., Zhao, L. 2020. Performance of enhanced anaerobic digestion with different pyrolysis biochars and microbial communities. Bioresource Technology, 296, 122354.

Shi, J., Han, Y., Xu, C., Han, H. 2019. Enhanced biodegradation of coal gasification wastewater with anaerobic biofilm on polyurethane (PU), powdered activated carbon (PAC), and biochar. Bioresource Technology, 289, 121487.

Shin, C., McCarty, P.L., Kim, J., Bae, J. 2014. Pilot-scale temperate-climate treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor (SAF-MBR). Bioresource Technology, 159, 95-103.

Silva, N.C.M., Moravia, W.G., Amaral, M.C.S., Figueiredo, K.C.S. 2019. Evaluation of fouling mechanisms in nanofiltration as a polishing step of yeast MBR-treated landfill leachate. Environmental Technology, 40(27), 3611-3621.

Sima, X.-F., Wang, Y.-Y., Shen, X.-C., Jing, X.-R., Tian, L.-J., Yu, H.-Q., Jiang, H. 2017. Robust biochar-assisted alleviation of membrane fouling in MBRs by indirect mechanism. Separation and Purification Technology, 184, 195-204.

Skouteris, G., Saroj, D., Melidis, P., Hai, F.I., Ouki, S. 2015. The effect of activated carbon addition on membrane bioreactor processes for wastewater treatment and reclamation – A critical review. Bioresource Technology, 185, 399-410.

Smith, A.L., Stadler, L.B., Love, N.G., Skerlos, S.J., Raskin, L. 2012. Perspectives on anaerobic membrane bioreactor treatment of domestic wastewater: A critical review. Bioresource Technology, 122, 149-159.

Song, L., Shao, Y., Shi, S., Tan, L. 2019. Continuously Biodegrading High Concentration of Acid Red B Under Hypersaline Conditions in a Membrane Bioreactor Bioaugmented by a Halotolerant Yeast Pichia occidentalis G1 and Microbial Community Dynamics. Environmental Engineering Science, 36(11), 1412-1420.

Sonune, A., Ghate, R. 2004. Developments in wastewater treatment methods. Desalination, 167, 55-63.

Suanon, F., Sun, Q., Li, M., Cai, X., Zhang, Y., Yan, Y., Yu, C.-P. 2017. Application of nanoscale zero valent iron and iron powder during sludge anaerobic digestion: Impact on methane yield and pharmaceutical and personal care products degradation. Journal of hazardous materials, 321, 47-53.

Sudibyo, H., Shabrina, Z.L., Wondah, H.R., Hastuti, R.T., Purnomo, C.W., Budhijanto, W.

2018. Anaerobic digestion of landfill leachate with natural zeolite and sugarcane bagasse fly ash as the microbial immobilization media in packed bed reactor. Acta Polytechnica, 58(1), 57-68.

Sun, Y., Fang, Y., Liang, P., Huang, X. 2016. Effects of online chemical cleaning on removing biofouling and resilient microbes in a pilot membrane bioreactor. International Biodeterioration & Biodegradation, 112, 119-127.

Sunyoto, N.M.S., Zhu, M., Zhang, Z., Zhang, D. 2016. Effect of biochar addition on hydrogen and methane production in two-phase anaerobic digestion of aqueous carbohydrates food waste. Bioresource Technology, 219, 29-36.

Takahashi, S., Tomita, J., Nishioka, K., Hisada, T., Nishijima, M. 2014. Development of a prokaryotic universal primer for simultaneous analysis of Bacteria and Archaea using next-generation sequencing. PloS one, 9(8), e105592.

Thompson, L.J., Gray, V.M., Kalala, B., Lindsay, D., Reynolds, K., von Holy, A. 2008. Biohydrogen production by Enterobacter cloacae and Citrobacter freundii in carrier induced granules. Biotechnology Letters, 30(2), 271-274.

Tian, Y., Chen, L., Zhang, S., Zhang, S. 2011. A systematic study of soluble microbial products and their fouling impacts in membrane bioreactors. Chemical Engineering Journal, 168(3), 1093-1102.

UNICEF. 2017. Thirsting for a future: water and children in a changing climate. UNICEF.

United Nations, D.o.E., Social Affairs, P.D. 2014. World Urbanization Prospects: The 2014 Revision, Highlights (ST/ESA/SER. A/352), UN New York, NY.

Van Geluwe, S., Braeken, L., Van der Bruggen, B. 2011. Ozone oxidation for the alleviation of membrane fouling by natural organic matter: A review. Water Research, 45(12), 3551-3570.

Vyrides, I., Stuckey, D.C. 2009. Saline sewage treatment using a submerged anaerobic membrane reactor (SAMBR): Effects of activated carbon addition and biogas-sparging time. Water Research, 43(4), 933-942.

Wang, C., Liu, Y., Gao, X., Chen, H., Xu, X., Zhu, L. 2018a. Role of biochar in the granulation of anaerobic sludge and improvement of electron transfer characteristics. Bioresource Technology, 268, 28-35.

Wang, H., Xu, J., Sheng, L., Liu, X., Zong, M., Yao, D. 2019a. Anaerobic digestion technology for methane production using deer manure under different experimental conditions. Energies, 12(9), 1819.

Wang, J., Liang, J., Sun, L., Gao, S. 2019b. PVA/CS and PVA/CS/Fe gel beads' synthesis mechanism and their performance in cultivating anaerobic granular sludge. Chemosphere, 219, 130-139.

Wang, J., Wu, B., Liu, Y., Fane, A.G., Chew, J.W. 2018b. Monitoring local membrane fouling mitigation by fluidized GAC in lab-scale and pilot-scale AnFMBRs. Separation and Purification Technology, 199, 331-345.

Wang, J., Wu, B., Yang, S., Liu, Y., Fane, A.G., Chew, J.W. 2016a. Characterizing the scouring efficiency of Granular Activated Carbon (GAC) particles in membrane fouling mitigation via wavelet decomposition of accelerometer signals. Journal of Membrane Science, 498, 105-115.

Wang, J., Zamani, F., Cahyadi, A., Toh, J.Y., Yang, S., Wu, B., Liu, Y., Fane, A.G., Chew, J.W. 2016b. Correlating the hydrodynamics of fluidized granular activated carbon (GAC) with membrane-fouling mitigation. Journal of Membrane Science, 510, 38-49.

Wang, P., Peng, H., Adhikari, S., Higgins, B., Roy, P., Dai, W., Shi, X. 2020. Enhancement of biogas production from wastewater sludge via anaerobic digestion assisted with biochar amendment. Bioresource Technology, 309, 123368.

Wang, S., Ma, C., Pang, C., Hu, Z., Wang, W. 2018c. Membrane fouling and performance of anaerobic ceramic membrane bioreactor treating phenol- and quinoline-containing wastewater: granular activated carbon vs polyaluminum chloride. Environmental Science and Pollution Research.

Wang, S., Ma, C., Pang, C., Hu, Z., Wang, W. 2019c. Membrane fouling and performance of anaerobic ceramic membrane bioreactor treating phenol-and quinoline-containing wastewater: granular activated carbon vs polyaluminum chloride. Environmental Science and Pollution Research, 26(33), 34167-34176.

Wang, W., Xie, L., Luo, G., Zhou, Q., Angelidaki, I. 2013. Performance and microbial community analysis of the anaerobic reactor with coke oven gas biomethanation and in situ biogas upgrading. Bioresource Technology, 146, 234-239.

Waqas, S., Bilad, M.R., Man, Z., Wibisono, Y., Jaafar, J., Indra Mahlia, T.M., Khan, A.L., Aslam, M. 2020. Recent progress in integrated fixed-film activated sludge process for wastewater treatment: A review. Journal of Environmental Management, 268, 110718.

Weber, K., Quicker, P. 2018. Properties of biochar. Fuel, 217, 240-261.

Wei, D., Xue, X., Chen, S., Zhang, Y., Yan, L., Wei, Q., Du, B. 2013. Enhanced aerobic granulation and nitrogen removal by the addition of zeolite powder in a sequencing batch reactor. Applied microbiology and biotechnology, 97(20), 9235-9243.

Wei, J., Hao, X., van Loosdrecht, M.C., Li, J. 2018. Feasibility analysis of anaerobic digestion of excess sludge enhanced by iron: a review. Renewable and Sustainable Energy Reviews, 89, 16-26.

Wijesinghe, D.T.N., Dassanayake, K.B., Scales, P.J., Sommer, S.G., Chen, D. 2018. Effect of Australian zeolite on methane production and ammonium removal during anaerobic digestion of swine manure. Journal of Environmental Chemical Engineering, 6(1), 1233-1241.

Woo, H., Yang, H.S., Timmes, T.C., Han, C., Nam, J.-Y., Byun, S., Kim, S., Ryu, H., Kim, H.-C. 2019. Treatment of reverse osmosis concentrate using an algal-based MBR combined with ozone pretreatment. Water Research, 159, 164-175.

Woo, Y.C., Lee, J.J., Jeong, A., Song, J., Choi, Y., Kim, H.-S. 2020. Removal of nitrogen by a sulfur-based carrier with powdered activated carbon (PAC) for denitrification in membrane bioreactor (MBR). Journal of Water Process Engineering, 34, 101149.

Wu, B., Kitade, T., Chong, T.H., Uemura, T., Fane, A.G. 2013. Impact of membrane bioreactor operating conditions on fouling behavior of reverse osmosis membranes in MBR–RO processes. Desalination, 311, 37-45.

Wu, B., Wong, P.C.Y., Fane, A.G. 2015a. The potential roles of granular activated carbon in anaerobic fluidized membrane bioreactors: effect on membrane fouling and membrane integrity. Desalination and Water Treatment, 53(6), 1450-1459.

Wu, B., Zamani, F., Lim, W., Liao, D., Wang, Y., Liu, Y., Chew, J.W., Fane, A.G. 2017. Effect of mechanical scouring by granular activated carbon (GAC) on membrane fouling mitigation. Desalination, 403, 80-87.

Wu, D., Zheng, S., Ding, A., Sun, G., Yang, M. 2015b. Performance of a zero valent iron-based anaerobic system in swine wastewater treatment. Journal of hazardous materials, 286, 1-6.

Wu, J., Huang, X. 2010. Use of ozonation to mitigate fouling in a long-term membrane bioreactor. Bioresource Technology, 101(15), 6019-6027.

Wu, Y., Wang, D., Liu, X., Xu, Q., Chen, Y., Yang, Q., Li, H., Ni, B. 2019. Effect of poly aluminum chloride on dark fermentative hydrogen accumulation from waste activated sludge. Water research, 153, 217-228.

Xiao, L., Deng, Z., Fung, K.Y., Ng, K.M. 2013. Biohydrogen generation from anaerobic digestion of food waste. International Journal of Hydrogen Energy, 38(32), 13907-13913.

Xiao, Y., Yaohari, H., De Araujo, C., Sze, C.C., Stuckey, D.C. 2017. Removal of selected pharmaceuticals in an anaerobic membrane bioreactor (AnMBR) with/without powdered activated carbon (PAC). Chemical Engineering Journal, 321, 335-345.

Xu, R., Qin, W., Zhang, B., Wang, X., Li, T., Zhang, Y., Wen, X. 2020. Nanofiltration in pilot scale for wastewater reclamation: Long-term performance and membrane biofouling characteristics. Chemical Engineering Journal, 395, 125087.

Xu, R., Xu, S., Zhang, L., Florentino, A.P., Yang, Z., Liu, Y. 2019. Impact of zero valent iron on blackwater anaerobic digestion. Bioresource technology, 285, 121351.

Xue, J., Zhang, Y., Liu, Y., Gamal El-Din, M. 2016. Effects of ozone pretreatment and operating conditions on membrane fouling behaviors of an anoxic-aerobic membrane bioreactor for oil sands process-affected water (OSPW) treatment. Water Research, 105, 444-455.

Yamada, T., Sekiguchi, Y., Hanada, S., Imachi, H., Ohashi, A., Harada, H., Kamagata, Y. 2006. Anaerolinea thermolimosa sp. nov., Levilinea saccharolytica gen. nov., sp. nov. and Leptolinea tardivitalis gen. nov., sp. nov., novel filamentous anaerobes, and description of the new classes

Anaerolineae classis nov. and Caldilineae classis nov. in the bacterial phylum Chloroflexi. International Journal of Systematic and Evolutionary Microbiology, 56(6), 1331-1340.

Yao, M., Ladewig, B., Zhang, K. 2011. Identification of the change of soluble microbial products on membrane fouling in membrane bioreactor (MBR). Desalination, 278(1), 126-131.

Yao, M., Nan, J., Chen, T., Zhan, D., Li, Q., Wang, Z., Li, H. 2015. Influence of flocs breakage process on membrane fouling in coagulation/ultrafiltration process—Effect of additional coagulant of poly-aluminum chloride and polyacrylamide. Journal of Membrane Science, 491, 63-72.

Yilmaz, P., Parfrey, L.W., Yarza, P., Gerken, J., Pruesse, E., Quast, C., Schweer, T., Peplies, J., Ludwig, W., Glöckner, F.O. 2013. The SILVA and "All-species Living Tree Project (LTP)" taxonomic frameworks. Nucleic Acids Research, 42(D1), D643-D648.

Yoo, R., Kim, J., McCarty, P.L., Bae, J. 2012. Anaerobic treatment of municipal wastewater with a staged anaerobic fluidized membrane bioreactor (SAF-MBR) system. Bioresource Technology, 120, 133-139.

Yu, H., Wang, Z., Wang, Q., Wu, Z., Ma, J. 2013. Disintegration and acidification of MBR sludge under alkaline conditions. Chemical Engineering Journal, 231, 206-213.

Yu, Z., Hu, Y., Dzakpasu, M., Wang, X.C., Ngo, H.H. 2019. Dynamic membrane bioreactor performance enhancement by powdered activated carbon addition: Evaluation of sludge morphological, aggregative and microbial properties. Journal of Environmental Sciences, 75, 73-83.

Yu, Z., Song, Z., Wen, X., Huang, X. 2015. Using polyaluminum chloride and polyacrylamide to control membrane fouling in a cross-flow anaerobic membrane bioreactor. Journal of

Membrane Science, 479, 20-27.

Yun, Y.-M., Kim, M., Kim, H., Kim, D.-H., Kwon, E.E., Kang, S. 2019. Increased biodegradability of low-grade coal wastewater in anaerobic membrane bioreactor by adding yeast wastes. Journal of Environmental Management, 234, 36-43.

Yuniarto, A., Noor, Z.Z., Ujang, Z., Olsson, G., Aris, A., Hadibarata, T. 2013. Bio-fouling reducers for improving the performance of an aerobic submerged membrane bioreactor treating palm oil mill effluent. Desalination, 316, 146-153.

Zamani, F., Chew, J.W., Akhondi, E., Krantz, W.B., Fane, A.G. 2015. Unsteady-state shear strategies to enhance mass-transfer for the implementation of ultrapermeable membranes in reverse osmosis: A review. Desalination, 356, 328-348.

Zhang, H., Fan, X., Wang, B., Song, L. 2016. Calcium ion on membrane fouling reduction and bioflocculation promotion in membrane bioreactor at high salt shock. Bioresource technology, 200, 535-540.

Zhang, J., Chua, Q.W., Mao, F., Zhang, L., He, Y., Tong, Y.W., Loh, K.-C. 2019a. Effects of activated carbon on anaerobic digestion – Methanogenic metabolism, mechanisms of antibiotics and antibiotic resistance genes removal. Bioresource Technology Reports, 5, 113-120.

Zhang, J., Fan, C., Zang, L. 2017a. Improvement of hydrogen production from glucose by ferrous iron and biochar. Bioresource Technology, 245, 98-105.

Zhang, L. 2020. Advanced treatment of oilfield wastewater by a combination of DAF, yeast bioreactor, UASB, and BAF processes. Separation Science and Technology, 1-10.

Zhang, M., Lin, H., Shen, L., Liao, B.-Q., Wu, X., Li, R. 2017b. Effect of calcium ions on fouling properties of alginate solution and its mechanisms. Journal of Membrane Science, 525, 320-329.

Zhang, Q., Singh, S., Stuckey, D.C. 2017c. Fouling reduction using adsorbents/flocculants in a submerged anaerobic membrane bioreactor. Bioresource Technology, 239, 226-235.

Zhang, S., Chang, J., Lin, C., Pan, Y., Cui, K., Zhang, X., Liang, P., Huang, X. 2017d. Enhancement of methanogenesis via direct interspecies electron transfer between Geobacteraceae and Methanosaetaceae conducted by granular activated carbon. Bioresource Technology, 245, 132-137.

Zhang, S., Xiong, J., Zuo, X., Liao, W., Ma, C., He, J., Chen, Z. 2019b. Characteristics of the sludge filterability and microbial composition in PAC hybrid MBR: Effect of PAC replenishment ratio. Biochemical Engineering Journal, 145, 10-17.

Zhang, S., Zhao, Y., Yang, K., Liu, W., Xu, Y., Liang, P., Zhang, X., Huang, X. 2020a. Versatile zero valent iron applied in anaerobic membrane reactor for treating municipal wastewater: Performances and mechanisms. Chemical Engineering Journal, 382, 123000.

Zhang, X., Guo, Y., Wang, T., Wu, Z., Wang, Z. 2020b. Antibiofouling performance and mechanisms of a modified polyvinylidene fluoride membrane in an MBR for wastewater treatment: Role of silver@silica nanopollens. Water Research, 176, 115749.

Zhang, Y., Wei, S., Hu, Y., Sun, S. 2018. Membrane technology in wastewater treatment enhanced by functional nanomaterials. Journal of Cleaner Production, 197, 339-348.

Zhao, F., Xu, K., Ren, H., Ding, L., Geng, J., Zhang, Y. 2015a. Combined effects of organic matter and calcium on biofouling of nanofiltration membranes. Journal of Membrane Science, 486, 177-188.

Zhao, S., Gao, B., Sun, S., Yue, Q., Dong, H., Song, W. 2015b. Coagulation efficiency, floc properties and membrane fouling of polyaluminum chloride in coagulation–ultrafiltration system: The role of magnesium. Colloids and Surfaces A: Physicochemical and Engineering Aspects, 469, 235-241.

Zheng, W., Yu, Z., Xia, Y., Wen, X. 2018. Influence of polyaluminum chloride on microbial characteristics in anaerobic membrane bioreactors for sludge digestion. Applied Microbiology and Biotechnology, 102(2), 1005-1017.

Zhou, H., Li, X., Xu, G., Yu, H. 2018. Overview of strategies for enhanced treatment of municipal/domestic wastewater at low temperature. Science of the Total Environment, 643, 225-237.

Zhu, H., Han, Y., Ma, W., Han, H., Ma, W. 2017. Removal of selected nitrogenous heterocyclic compounds in biologically pretreated coal gasification wastewater (BPCGW) using the catalytic ozonation process combined with the two-stage membrane bioreactor (MBR). Bioresource Technology, 245, 786-793.

Zhu, Y., Wang, Y., Zhou, S., Jiang, X., Ma, X., Liu, C. 2018. Robust performance of a membrane bioreactor for removing antibiotic resistance genes exposed to antibiotics: role of membrane foulants. Water research, 130, 139-150.

Zupančič, G.D., Škrjanec, I., Logar, R.M. 2012. Anaerobic co-digestion of excess brewery yeast in a granular biomass reactor to enhance the production of biomethane. Bioresource technology, 124, 328-337.

Appendix

List of publication

Sohn, W., Guo, W., Ngo, H.H., Deng, L., Cheng, D., Zhang, X. 2021. A review on membrane fouling control in anaerobic membrane bioreactors by adding performance enhancers. Journal of Water Process Engineering, 40, 101867.

Sohn, W., Guo, W., Ngo, H.H., Deng, L., Cheng, D. 2021. Powdered activated carbon addition for fouling control in anaerobic membrane bioreactor. Bioresource Technology Reports, 15, 100721.