Doctoral Thesis

Treatment Performance of Real Municipal Sewage by an Anaerobic Membrane Bioreactor

（嫌気性膜分離バイオリアクターによる実下水の処理性能）

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ABSTRACT

The Activated Sludge Process has been developed into a mature process and have been worldwide applied for treating sewage wastewater during the past one hundred years. However, during the treatment process, a great deal of energy is needed for aeration requirement, a large amount of greenhouse gas is discharged, and a big amount of waste sludge is produced which have a further demand for sludge treatment. In order to develop a new energy positive municipal sewage treatment process with high organic removal as well as low waste sludge generated, this thesis written on a study implemented a series of researches based on a long-term operated new designed mini-pilot AnMBR treating the real municipal sewage wastewater. The study was divided into three parts: start-up phase with the exploration of various conditions, treatment performance at room temperature, and effect of temperature and performance at low temperature.

The organic removal efficiency was achieved around 90% of COD removal, more than 90% of BOD removal and 100% of SS removal with low sludge yield. The biogas production rate was achieved as high as 0.30 L-gas/g-COD_{rem} and the highest methane yield was obtained 0.24 L-CH_4/g-COD_{rem} at 25°C during HRT 12h. The methane content in the produced biogas was obtained around 80%. COD conversion to CH_4 was obtained 83.4% in HRT 24h at 15°C low temperature. Sewage wastewater treatment capacity was achieved 6L-water/L-reactor/d and the biggest FLUX for membrane filtration was 0.34 m/d obtained in HRT 4h at room temperature. Effluent pH and ORP inside reactor was presented around 6.8 and -300 ~ -330 mV, respectively.

Detail structure was included 7 chapters consisted as this thesis.

Chapter 1. General introduction: introduces the background of innovation for sewage wastewater treatment as well as the structure of the thesis.

Chapter 2. Literature review on sewage treatment and AnMBR: introduces the development of process for sewage wastewater treatment and researches based on AnMBR treating the sewages or low strength wastewater by AnMBR.

Chapter 3. Effect of membrane pore size on start-up and long-term operation performance: two mini-pilot scale AnMBRs by different pore size membranes (0.4μm pore size MF, 0.05μm pore size UF) was installed in a wastewater treatment plant to evaluated more suitable membrane pore size for the sewage wastewater treatment by AnMBRs on the purpose of economical and reasonable operation as well as verify the feasibility for AnMBR start-up and treating the real sewage wastewater.

Chapter 4. Effect of HRT on treatment performance at room temperature: a mini-pilot AnMBR was operated in the WWTP at room temperature (25°C) and continued to dealing with the real sewage at operated HRTs ranged from 12, 8, 6 to 4 hours. The performance of sewages treated by AnMBR in each HRT condition was investigated.

Chapter 5. Effect of temperature on treatment performance of MF-MBR: The mini-pilot AnMBR was operated at HRT condition of 6 hours dealing with the real sewage by operated temperatures from 25°C, 20°C then 15°C (HRT 6 to 24 hours). Then the performance of sewages treated by AnMBR in each temperature condition and the different HRTs at low temperature condition was investigated.

Chapter 6. Conclusions and perspectives: the main conclusions were summarized based on the experimental results. The perspective for further investigation or utilization were also suggested.

CONTENTS

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Chapter 1

General introduction

1.1 Background

In the past one hundred years, the Activated Sludge Process (ASP) has been developed into a mature process and have been worldwide applied for treating sewage wastewater. However, during the ASP treatment, there are some issues presented, for example:

- ✧ a great deal of energy is needed for aeration requirement;
- ✧ a large amount of greenhouse gas is discharged;
- ✧ a big amount of waste sludge is produced.

On the other hands, sewage is considered to be the most abundant type of wastewater and a valuable resource containing water, nutrients and energy which is worthy of recovery and reuse. If recovery and reuse could be achieved in technology, it would be possible to build more sustainable sewage treatment plant and even become net suppliers if energy positive can be achieved (Khiewwijit et al., 2015; Ozgun et al., 2013). While the anaerobic process does not have those disadvantages and has drawn considerable attention for its ability to convert chemically bound energy in the organic pollutants to useful energy namely biogas (Shizas and Bagley, 2004). So there were many attempts for combining the anaerobic process to treat the sewage wastewater in recent years. However, there are two points have become the main obstacles to applying anaerobic digestion directly into the sewage treatment. Firstly, anaerobic sludge shows a trend of slowly grow, especially under the condition of low organic strength feeding. Secondly, it is hard to separate activated sludge and the treated water in traditional anaerobic digestion process and that appears even in the situation of treating a very big amount of wastewater which is one of the characters of the sewage wastewater. Therefore, the anaerobic membrane bio-reactor (AnMBR) was brought up. AnMBR integrates the anaerobic digestion process and the membrane technology so that created a new process which could provide with both the advantages of anaerobic digestion as well as the high efficiency of sludge-water separation due to the filtration by membranes. Usually, organic matters are highly removed dependent on the anaerobic digestion ability due to the four key steps to be known as hydrolysis, acidogenesis, acetogenesis and methoanogenesis during the anaerobic digestion process. The four steps are carried out by distinct consortia of bacteria, namely fermentative bacteria, syntrophic acetogens, homoacetogens, hydrogenetrophic methanogens and aceticlastic methanogens (Batstone et al., 2002). And normally, in the AnMBRs, SS could be removed to a large extent due to the micro- filtration or ultra-filtration by membrane module while on the other sides, pollutants like TN, ammonia and TP are incapable of removal by the anaerobic fermentation process because of the mechanisms of anaerobic digestion.

Up to now, AnMBR has been successfully applied in the treatment of industrial wastewater and there are plenty of studies related to AnMBR focused on high organic strength wastewater or industrial wastewater while still lacking of development for treating the municipal or domestic sewage wastewater. However, despite that, there are some studies which related to the low organic strength wastewater or sewage based on man-made synthetic wastewater or even real sewage wastewater in some case have be reported. Those studies have obtained some primary achievements of which are introduced in detail in next Chapter of this thesis. And this study is trying to develop the process of AnMBR treating the sewage wastewater based on a new-designed simple structure AnMBR system (figure 1.1) feeding with the raw real sewage wastewater. Thus, in order to promote the process of application for AnMBR to dealing with the sewage wastewater, the efforts of systematically application researches was implemented during this study based on the built mini-pilot AnMBR systems placed in a municipal sewage wastewater treatment plant (Sen-En WWTP, figure 1.2).

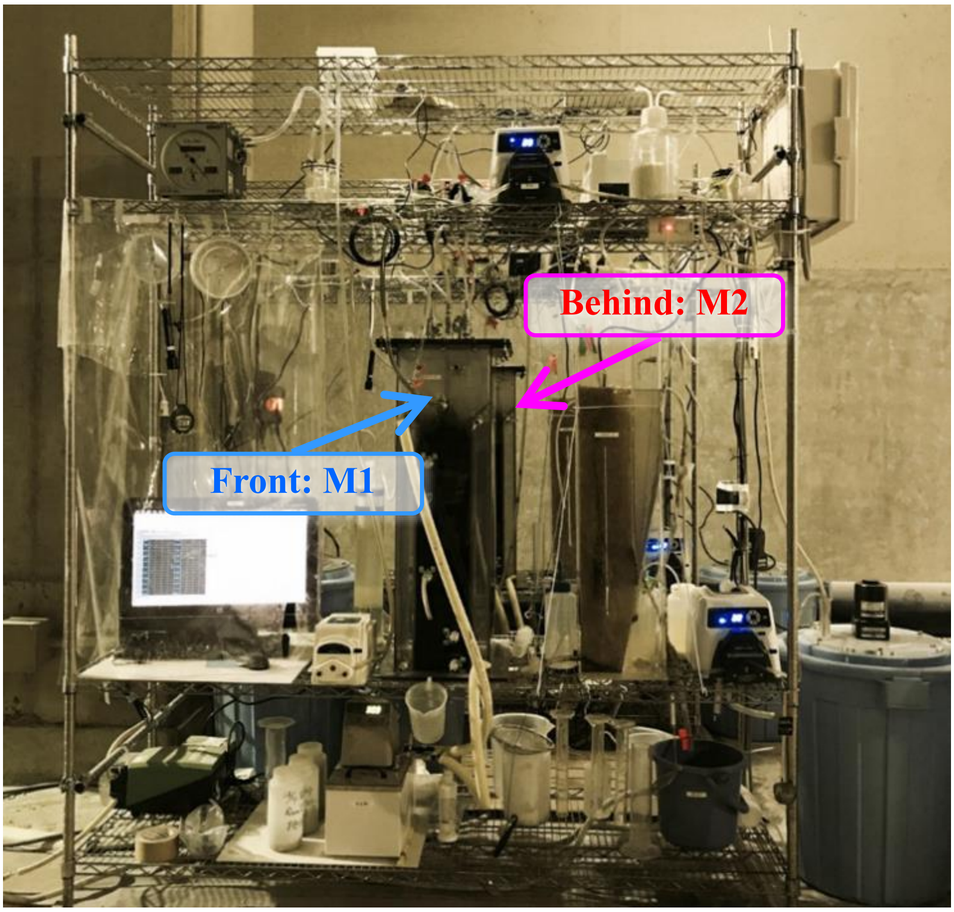


Fig. 1.1 Reactors and equipment in this research.

Fig. 1.2 Location of Sen-En WWTP in Japan.

1.2 Thesis structure

In this study, a series of studies was conducted on innovation of real sewage wastewater treatment by AnMBR. There are 7 chapters in this thesis and the structure was illustrated in figure 1.3. Kinds of data taken in each condition is shown in table 1.1.

Chapter 1 General introduction

This chapter introduces the background of sewage wastewater treatment for innovation as well as the structure of this thesis.

Chapter 2 Literature review on sewage treatment and AnMBR

This chapter introduces the development of process for sewage wastewater treatment and researches based on AnMBR treating the sewages or low strength wastewater by AnMBR. Moreover, the purpose of this study also be concluded.

Chapter 3 Effect of membrane pore size on start-up and long-term operation performance

In this chapter, two mini-pilot scale AnMBRs by different pore size membranes (one is 0.4μm pore size MF, the other one is 0.05μm pore size UF) was installed in the wastewater treatment plant to evaluated more suitable membrane pore size for the sewage wastewater treatment by AnMBRs on the purpose of economical and reasonable operation as well as verify the feasibility for AnMBR start-up and treating the real sewage wastewater.

Chapter 4 Effect of HRT on treatment performance at room temperature

In this chapter, a mini-pilot AnMBR was operated in the wastewater treatment plant at room temperature (25°C) and continued to dealing with the real sewage at operated HRTs ranged from 12, 8, 6 to 4 hours. The performance of sewages treated by AnMBR in each HRT condition was investigated in aspects on pollutant removal performance, gas yield, sludge yield, COD balance as well as the filtration performance of the membranes.

Chapter 5 Effect of temperature on treatment performance of MF-MBR

The mini-pilot AnMBR was operated in the wastewater treatment plant at HRT condition of 6 hours and continued to dealing with the real sewage by operated temperatures from 25°C, 20°C then 15°C (HRT 6 to 24 hours) in this Chapter. Then the performance of sewages treated by AnMBR in each temperature condition and the different HRTs at low temperature condition was investigated in aspects on pollutant removal performance, gas yield, sludge yield, COD balance as well as the filtration performance of the membranes.

Chapter 6 Conclusions and perspectives

The main conclusions were summarized based on the experimental results. The perspectives for further investigation or utilization were also suggested.

Furthermore, a chapter of appendix concluded abbreviation index used in this thesis and the supplement data/figure was also added as a part of this thesis.

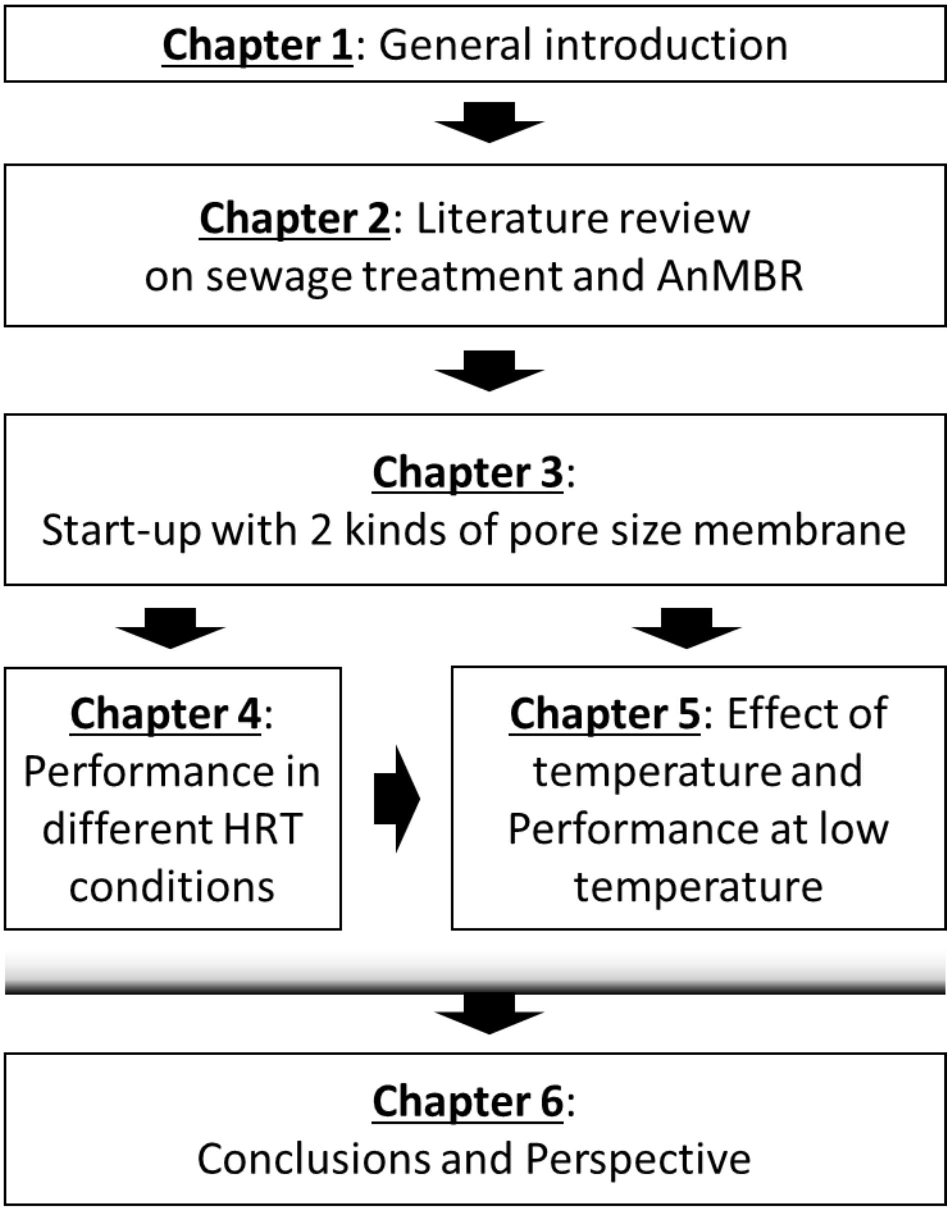


Fig. 1.3The structure of dissertation.

Table 1.1 Data taken in each condition**.**

Tem. (°C)	HRT (h)	Organic removal	Sludge yield	Biogas production	COD balance	TMP growth	SMP /EPS	Microbial analysis	SMA
25	24	★		★		★		★	
25	12	★	★	★	★	★		★	
25	8	★	★	★	★	★			
25	6	★	★	★	★	★	★		★
25	4	★	★	★	★	★			
20	6	★	★	★	★	★	★		★
15	24	★	★	★	★	★			★
15	16	★	★	★	★	★			★
15	12	★	★	★	★	★			★
15	6	★	★	★	★	★	★		★

REFRENCES:

Batstone, D.J., Keller, J., Angelidaki, I., Kalyuzhnyi, S. V, Pavlostathis, S.G., Rozzi, A., Sanders, W.T.M., Siegrist, H., Vavilin, V.A., 2002. The IWA Anaerobic Digestion Model No 1 (ADM1). Water Sci. Technol. 45, 65–73. https://doi.org/10.2166/wst.2002.0292

Khiewwijit, R., Temmink, H., Rijnaarts, H., Keesman, K.J., 2015. Energy and nutrient recovery for municipal wastewater treatment: How to design a feasible plant layout? Environ. Model. Softw. 68, 156–165. https://doi.org/https://doi.org/10.1016/j.envsoft.2015.02.011

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. Sep. Purif. Technol. https://doi.org/10.1016/j.seppur.2013.06.036

Shizas, I., Bagley, D.M., 2004. Experimental Determination of Energy Content of Unknown Organics in Municipal Wastewater Streams. J. Energy Eng. https://doi.org/10.1061/(ASCE)0733-9402(2004)130:2(45)

Chapter 2

Literature review on sewage treatment and AnMBR

2.1 Brief introduction of sewage wastewater treatment

2.1.1 History of sewage wastewater treatment development

Generally, sewage wastewater is generated by residential, institutional, commercial and even some industrial establishments. It includes household waste liquid as a main source from toilets, baths, kitchens, and sinks (for example, laundry sink) draining into sewers. In many areas, sewage also includes liquid waste or effluent from industry and commerce. As water is one of the most important resources on this planet, wastewater reuse has an ancient practice, which has been applied since the daybreak of human civilization history, with the connected to the development of sanitation purposes (Khouri et al., 1994).

In ancient times, reuse of untreated sewage wastewater has been practiced for many centuries with the objective of transferring human waste out of the urban residential and as one of the options of a relatively few technologies in the old times, land application of sewage wastewater has gone through different stages of development. Sewage wastewater was used for irrigation by Minoan Civilization since the Age of Bronze (Angelakis and Spyridakis, 1996). Thereafter, wastewater was used for not just irrigation, but also disposal and fertilization by Hellenic civilizations (Angelakis et al., 2005) and later by Romans (Tzanakakis et al., 2007). The above situation also occurred in Asia, China in one of the four ancient civilizations, use of human excreta for fertilizing agricultural crops has been applied from time immemorial (Ghneim., 2010). As there were no sewer systems were built in the ancient times, land application was played as an important role for the sewage wastewater treatment. While due to a fast development of heavy industrialization and urbanization, the sewers systems were started to build since the mid-nineteenth century as a reaction to the exacerbation of sanitary conditions. The huge amount of untreated sewage wastewater was simply piped to natural water systems away from the population centers at the beginning designed of the sewer systems led out various water pollution problems reported on the newspaper or magazine at that time.

For decades in the Mid-18th century, a sewage treatment method was diverting sewage for use of fertilizer to farms initially proposed by James Vetch was applied (Gordon, 1851) but brought out the problems of stinky, unhygienic and big cost for cleaning the heavier solids in the sewage transport channel built in farms (Cooper, 2007). Later, a method of cesspool, invented by L.H Mouras in the 1860s of France, in which the water was sealed off to prevent contamination and the solid waste was slowly liquefied by anaerobic action was applied while this method was difficult to dealing with a big amount water as the big extension time for wastewater. Then cesspool was improved as called septic tank which is still in worldwide using, especially in countryside place out of the large-scale sewage systems (Melosi, 2008). Cesspool or septic tank formed the rudiment of anaerobic biological treatment technology. Then in early 1910s, the activated sludge process, known as a type of wastewater treatment process for treating sewage or industrial wastewaters using aeration and a biological floc composed of bacteria and protozoa, was first discovered by two English engineers named Edward Ardern and W.T. Lockett during conducting research at Davyhulme Sewage Works for the Manchester Corporation Rivers Department (Alleman, 2005) and was considered as one of the most significant improvement in public health and the environment protection during the 20^{th} century and was become widely used in the world during the past 100 years' development.

2.1.2 Traditional activated sludge process

In the past one hundred years, the Activated Sludge Process (ASP) has been developed into a mature process for treating sewage wastewater from a single aeration tank. The overview for a ASP used for treating the sewage wastewater in wastewater treatment plant (WWTP) is shown as figure 2.1. Normally, it concluded with: primary treatment, secondary treatment and tertiary treatment (also be known as advanced treatment) for sewage treatment in addition with the treatment process for waste sludge by many ponds or tanks combined by physical, chemical and biological methods. In the process of sewage wastewater treatment, biological treatment method occupies a dominant position. Through the ASP in a WWTP, kinks of pollutant in sewage wastewater can be removed and reach to a high level water quality for discharging. With the improvement of sewage discharge standards in various countries and regions, activated sludge process has also been continuously improved and applied in WWTPs. However, during the ASP treatment, there are some issues still presented, for example:

- ✧ a great deal of energy is needed for aeration requirement;
- ✧ a large amount of greenhouse gas is discharged;
- ✧ a big amount of waste sludge is produced.

It is generally believed that these shortcomings are difficult to solve because of the activated sludge process own characteristics (Jenkins and Wanner, 2014).

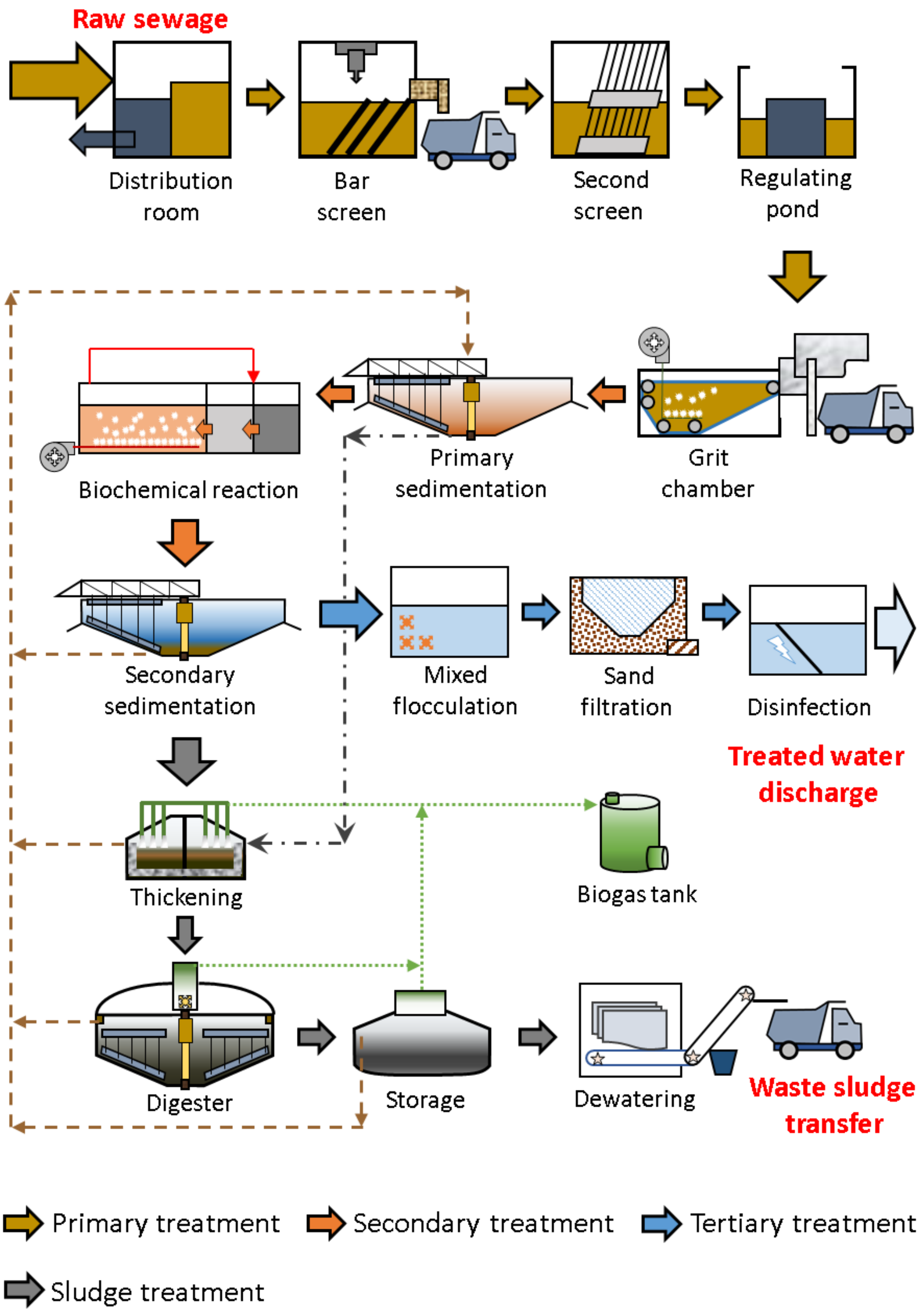


Fig. 2.1 A typical sewage treatment process in WWTP.

2.1.3 Innovation in sewage wastewater treatment process

The improvements have never stopped since the sewage treatment system was produced. In19th century, it was to achieve the removal of pollutants and the 20th century was to continuously improve the removal efficiency of pollutants in response to the increasing emission standards of each countries and regions. While, in 21st century now, the enforces are not only purify sewage wastewater but also try to recycling resources and recovering energy. In recent years, there were two main ways to improve the sewage treatment processes, one is to improve the activated sludge process which derived many new processes already, the other one is to develop new technologies for the sewage wastewater treatment. The derived process of ASP can effectively improve the efficiency of sewage wastewater treatment, however resulted an even more energy demand generally and made the process become much more complex.

The new technologies being developed mainly based on biological treatment because of its efficiency and among them the anaerobic digestion (野池達也 et al., 2009) has become a focus due to its various advantages such as:

- ✓ High organic removal efficiency (normally > 85%) with no necessary to add nutrients such as nitrogen and phosphorus generally;
- ✓ Less energy consumed during processing meanwhile have the energy recovery potential by the produced methane biogas;
- ✓ Low generation of waste sludge with easy dehydration, which can be used as a high-quality fertilizer.

While there are some disadvantages of cause:

- ✧ The treated water still contains a certain COD/BOD amount which need the aerobic biological treatment for further step;

- ✧ Anaerobic bacteria breed slowly, strict requirements on environmental conditions, and sensitive to the poisons;
- ✧ There is a small amount of ammonia and hydrogen sulfide in the final product of anaerobic degradation which makes the effluent smelly.

2.2 Introduction of anaerobic membrane bio-reactor

The anaerobic membrane bio-reactor (AnMBR) integrates the anaerobic digestion process and the membrane technology (figure 2.2) so that created a new process which could provide with both the advantages of anaerobic digestion as well as the high efficiency of sludge-water separation due to the filtration by membranes. The wastewater is occurred the anaerobic fermentation by bacteria anaerobically which release methane gas as a byproduct and can be combusted to generate heat or electricity energy, then it is filtered and separated to make the permeate treated water and activated sludge apart by the membrane pores.

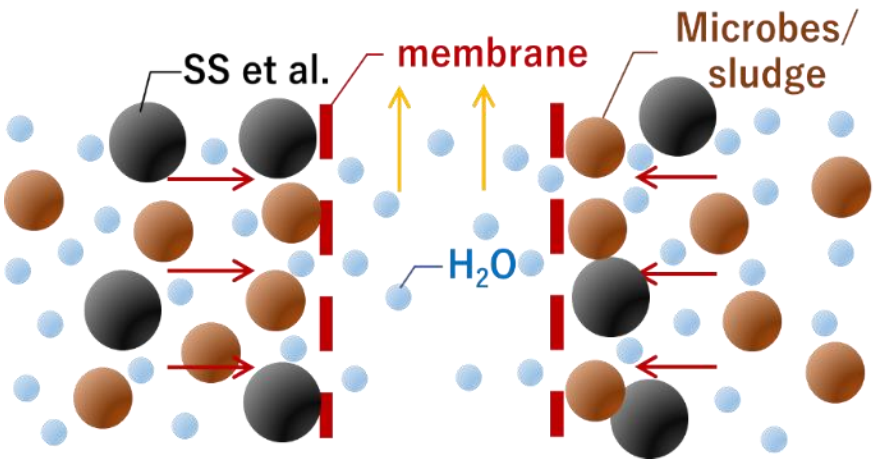


Fig. 2.2 Membrane filtration principle.

2.2.1 Types of membranes

The configuration of membrane was started as plat sheet and already being widely used in the world. Then hollow fiber membrane appeared and becoming more mainstream in recent years due to the characteristics such as: filling density of membrane area per unit volume was higher, easy for backwash controlling, simple in structure and low price. Some photos of flat sheet and hollow fiber membrane are shown in figure 2.3.

The membrane can also be classified by materials and pore size. So far, the most common materials for membrane is included with polyethylene (PE), polypropylene (PP), polytetrafluoroethylene (PTFE), polyvinylidene fluoride (PVDF), polyvinylchloride (PVC) and ceramic. In addition, the membrane filtration can be distinguish as Microfiltration (MF, 0.1~1μm or 0.1~5μm), Ultrafiltration (UF, 0.01~0.1μm), Nanofiltration (NF, 1~10nm), and Reverse Osmosis (RO, No pores or 0.1~1nm) by the different size of membrane pore (Yoon, 2015) for removing different size of ingredients in wastewater (figure 2.4).

2.2.2 Reactor configurations of the AnMBR

AnMBR were essentially implemented based on two configurations as be known: external/side-stream configuration and submerged/ immersed configuration.

Until submerged/immersed configuration reactor were commercialized, MBR relied on crossflow filtrations using mostly tubular membrane modules and some plate and frame membrane modules. It is called side-stream membrane process (side-stream AnMBR for combined with anaerobic reactors) because of the membrane module is set separately outside of reactor. The invitation of submerged/immersed configuration membrane filtration (submerged AnMBR for combined with anaerobic reactors) was on the purpose of saving capital and operational costs by directly placing membranes in mixed liquor without housings. After that, the submerged AnMBR also developed the external/side-stream configuration for the reactors can be separated as two part beneficial to the maintenance (external submerged AnMBR). The schematic diagram for those AnMBR is shown in figure 2.5.

Generally, side-stream AnMBR provides more direct hydrodynamic control of fouling, and offers the advantages of easier membrane replacement and high fluxes but at the expense of frequent membrane cleaning and high energy consumption (Le-Clech et al., 2006). Compared to side-stream AnMBR, submerged AnMBR directly places the membranes into the liquid and sludge. A suction pump is used to drag the permeate through the membranes. Several distinct advantages of submerged AnMBR are their much lower energy consumption, easier cleaning procedures, as well as the milder operating conditions and easier fouling control due to the lower tangential velocity (Lin et al., 2013). Then, the further step development for submerged AnMBR as the system constructed as external submerged AnMBR makes it inherited the advantages of both sides except for more energy is needed for the recycling pump between the two parts compare to the single submerged AnMBR (Mahboubi et al., 2016).

In addition, in order to combine a AnMBR system, any type of anaerobic reactor can be used for the anaerobic digestion process in principle together with the three main configurations. Normally, CSTR and UASB are widely used.

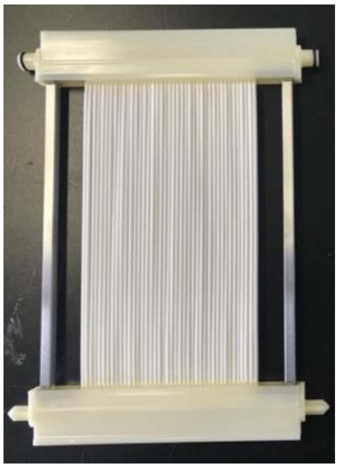


Fig. 2.3 Flat sheet (left) and hollow fiber (middle and right) membrane.

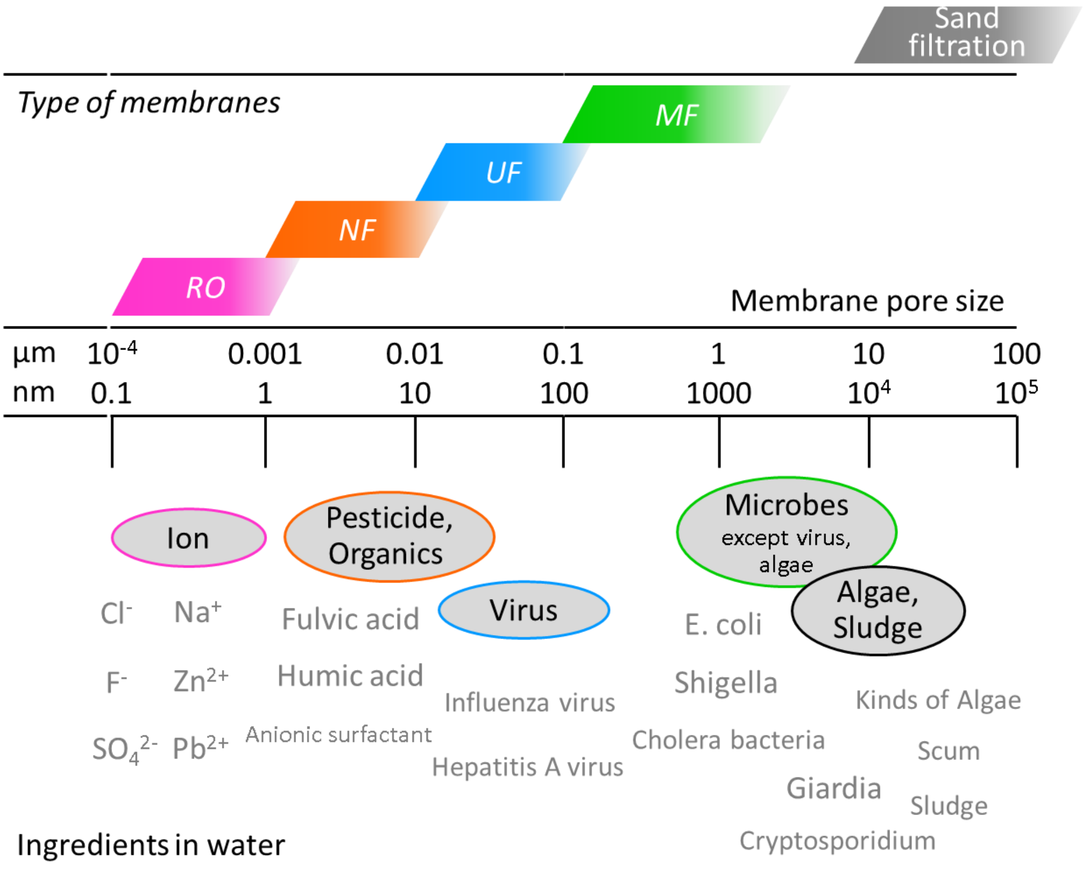


Fig. 2.4 Membrane filtration principle.

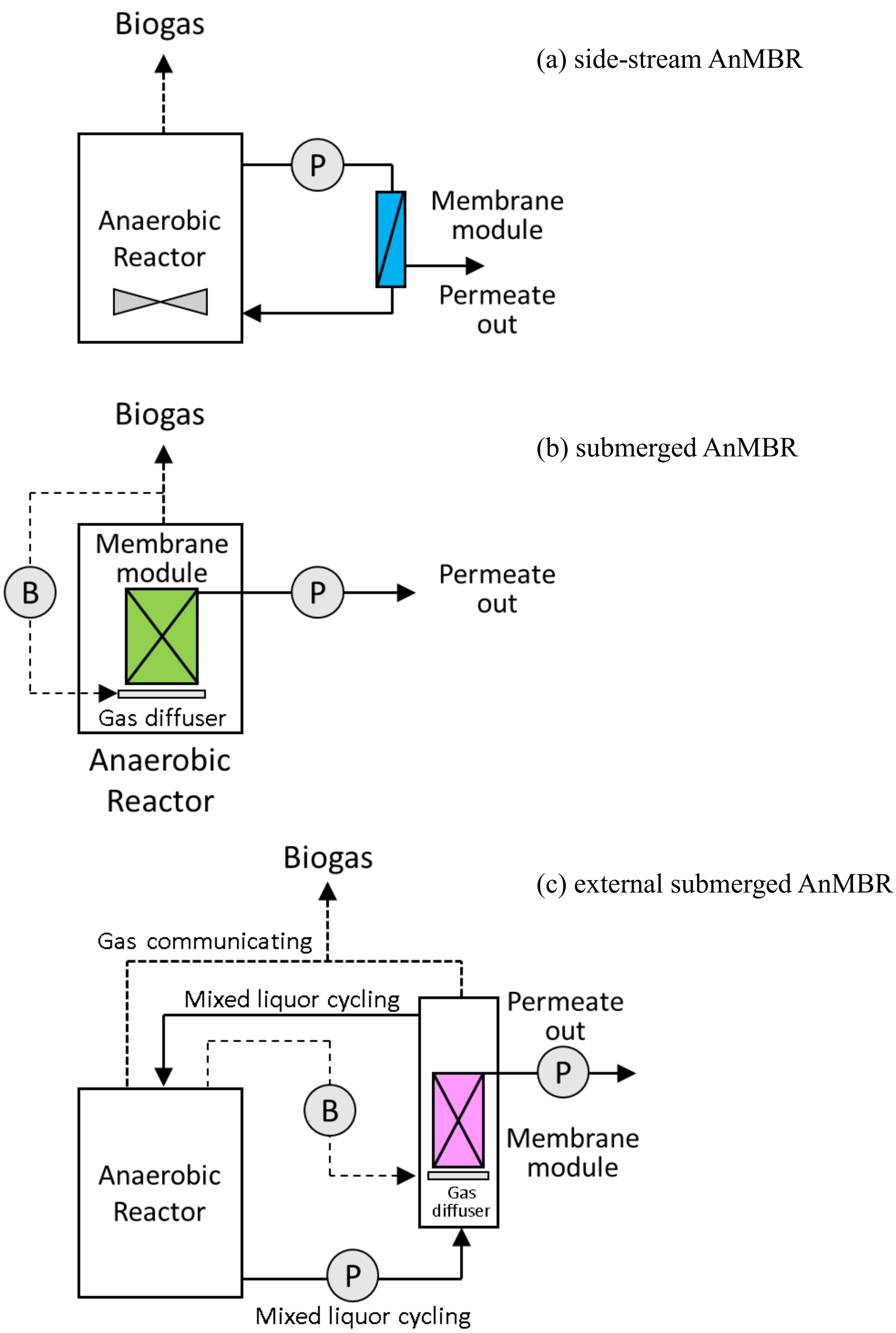


Fig. 2.5 Three kinds of main configurations of AnMBR.

2.2.3 Biodegradation mechanisms of anaerobic digestion

Anaerobic digestion normally refers to a biochemical conversion process, by which microorganisms convert chemical energy in biomass feedstock into a biogas with energy content in an oxygen-free environment (Balaman, 2018). The pathways of anaerobic digestion are normally considered as two phases consisted by four steps to be known as hydrolysis, acidogenesis, acetogenesis and methoanogenesis which is shown in figure 2.6. The four steps are carried out by distinct consortia of bacteria, namely fermentative bacteria, syntrophic acetogens, homoacetogens, hydrogenetrophic methanogens and aceticlastic methanogens (Batstone et al., 2002). In recent years, the anaerobic digestion process has been applied commercially with success in a multitude of situations and for a variety of biomass sources. The organic matters for biomass sources are always complex polymers which can be categorized as carbohydrates, proteins and fats, and their removal efficiencies are highly dependent on the anaerobic digestion ability due to the four key steps during the anaerobic digestion process. In addition, as CH_4 is the final product which released as a biogas of the anaerobic respiration, anaerobic digestion was also called as methane fermentation.

Because of AnMBR was combined by anaerobic digestion process and membrane filtration technology, the biodegradation mechanisms of anaerobic digestion are usually considered to be the same biodegradation mechanisms in AnMBR.

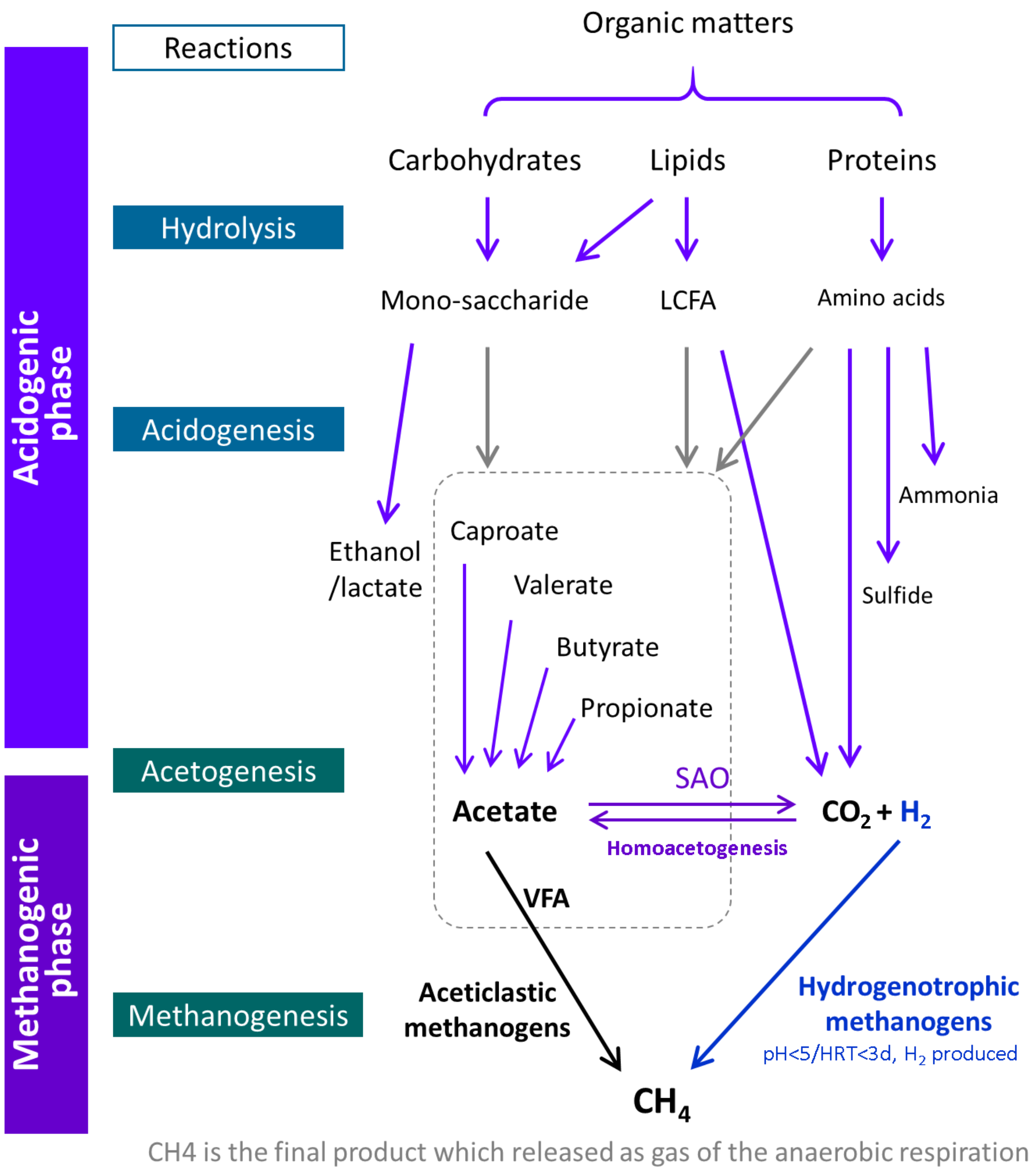


Fig. 2.6 Biodegradation pathways of anaerobic digestion.

2.2.4 Advantages and disadvantages of AnMBR

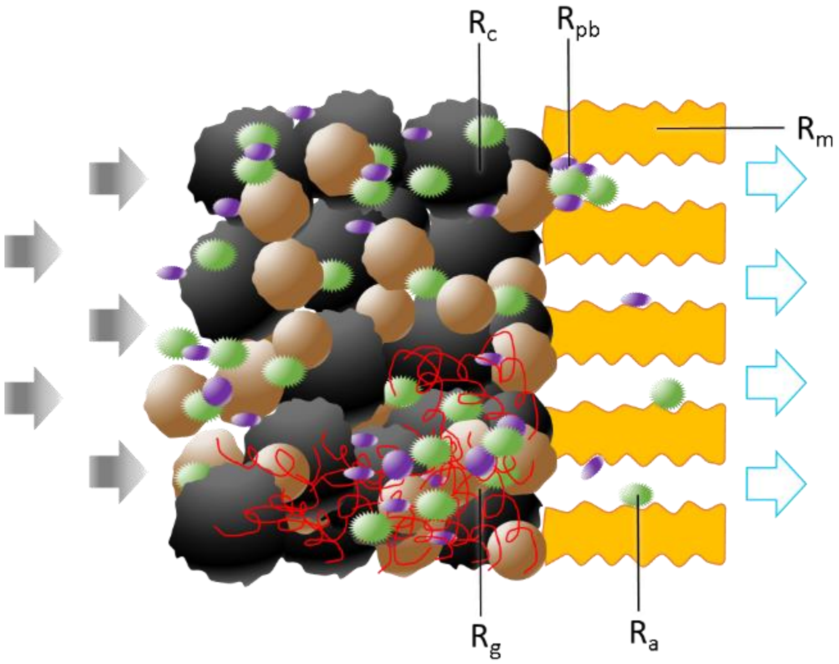
Usually, AnMBR is considered to be a sustainable alternative for the sewage wastewater treatment due to the energy generated by the biogas produced can achieve beyond the energy required for maintaining the process (Dvořák et al., 2016). So summarize the advantages of AnMBR:

- ✓ Efficient solid-liquid separation, good and stable effluent quality;
- ✓ The microorganisms can be completely trapped in the bioreactor, making the operation control more flexible and stable, due to the high-efficiency retention of the membrane;
- ✓ Space saving during the treatment process;
- ✓ High volume loading resulted by high concentration of microorganisms (1/3 of the standard conventional ASP and 1/5 of OD process);
- ✓ Conducive to the retention and growth for the microorganisms with slow growth speed;
- ✓ No final sedimentation tank is required;
- ✓ No management on sludge recycling makes it easy for maintenance;
- ✓ Almost no SS and Escherichia coli contain in effluent especially pore size smaller than 0.4μm;
- ✓ Easy to achieve process monitoring and automatic control;
- ✓ Generally, AnMBR operated in a high volume loading and low sludge loading and reduced the treatment cost for the waste sludge due to the low sludge yield and the easy dehydration property of anaerobic sludge.

However, as methanogens are obligate anaerobic bacteria, a strict anaerobic environment is required. Moreover, although the energy consumed for the filtration can achieve beyond the energy required for maintaining the process, the membrane fouling is still a problem. More energy may require for overcoming the resistance by the membrane itself as well as the resistance caused by kinds of fouling (figure 2.7). Therefore, a regular chemical cleaning is normally utilized for cleaning the foulant to reduce the resistance and extend the membrane life till the membrane cannot be recovered to a high filtration efficiency.

Thus, summarize the disadvantages of AnMBR:

- ✧ A strict anaerobic environment is required;
- ✧ Much energy is required for overcoming the resistance via filtration;
- ✧ A regular chemical cleaning is needed because of membrane fouling.



Rm = membrane resistance;

Ra = adsorption, biofouling; Rpb = pore plugging;

Rc = cake layer; Rg = gel layer.

Fig. 2.7 Membrane filtration resistance and membrane fouling.

2.3 Researches on AnMBR treating sewage wastewater

Up to now, AnMBR has been successfully applied in the field of industrial wastewater treatment and there are plenty of studies related to AnMBR focused on high organic strength wastewater or industrial wastewater while still lacking of development for treating the real sewage wastewater as the features of: large quantity, complicated composition, instable and low concentration of pollutant (Lei et al., 2018). However, despite that, there are some studies related to synthetic sewage wastewater or the low organic strength wastewater have been reported and obtained some achievements.

2.3.1 Organic pollutant removal

Usually, organic matters mainly indicated by chemical oxygen demand (COD) and biochemical oxygen demand (BOD), suspended solid (SS), total nitrogen (TN), ammonia (NH_4^+-N) and total phosphorus (TP) are the main concerns of common pollutants in the sewage wastewater and the treatment performances for these pollutants show a tendency to appear different fates in an AnMBR. The organic matters are always complex polymers which can be categorized as carbohydrates, proteins and fats, and their removal efficiencies are highly dependent on the anaerobic digestion ability due to the four key steps during the anaerobic digestion process. Normally, in the AnMBRs, SS could be removed to a large extent due to the filtration (MF and UF for wastewater treatment) by membrane module while TN, ammonia and TP is incapable of removal because of the mechanisms of regular anaerobic digestion process known so far.

Seib et al., 2016 operated four bench-scale AnMBRs at 10°C and 25°C separately by different AnMBR configurations (fluidized bed bioreactor and down floating filter) fed synthetic sewage wastewater first and then the real sewage wastewater and the organic removal efficiency exceeded 94% in the four AnMBRs. Watanabe et al., 2017 operated a submerged lab-scale AnMBR at temperature 25°C and decreased to 15°C later achieved COD removal efficiency ranged from 92% to even 98% though long HRT was utilized for the low temperature. Chen et al., 2017 compared two kinds of AnMBR (one is an external granular AnMBR and the other one is a submerged granular AnMBR) at the lab-scale fed synthetic sewage wastewater at room temperature 25°C and both AnMBR achieved higher than 91% for COD removal efficiency. Not just the lab-scale, A pilot-scale SAF-MBR fed by the primary settled sewage wastewater at 8~30°C with a total HRT as short as between 4.6 to 6.8h achieved an average COD removal more than 90% with 485 days continuous operation. Therefore, it was confirmed that a remarkable organic removal efficiency can be achieved by the AnMBR with different configuration or membranes reported in lab-scale fed synthetic sewage wastewater or the real sewage wastewater as well as the pilot-scale (table 2.1).

2.3.2 Sludge yield

Sludge in the bio-reactors is a combination of various microorganisms, solid-type organic matters, and even some inorganic matters (Rulkens, 2008). The low sludge yield is an advantage of the anaerobic wastewater treatment process (0.03~0.18 gMLVSS/gCODrem; Henze et al., 2008) compare with the aerobic wastewater treatment process (0.25~0.4 gMLVSS/gCODrem; Huang et al., 2001). While as a combination of anaerobic digestion and membrane separation technology, the performance for an AnMBR on the sludge yield is an important parameter as waste sludge treatment was reported as a major capital and operation consumption (occupied 65% of the whole operation cost) in a medium size WWTP applied ASP (Gray, 2017).

Theoretically, the sludge yield of AnMBR would be equal or higher than the simple anaerobic digestion process as the membrane separation limited the solids organic to get though the membrane which also count in the sludge yield especially in the situation of inadequate biodegradation. During the previous researches, Huang operated an external submerged AnMBR fed with domestic wastewater at temperature from 25°C to 30°C obtained a sludge yield value 0.125 gVSS/gCODrem. Later, an even smaller sludge yield as low as 0.06~0.09 gVSS/gCODrem was obtained by R. Chen et al., 2017 operated a lab-scale submerged AnMBR fed with synthetic sewage wastewater at room temperature 25°C. Furthermore, a pilot-scale staged anaerobic fluidized membrane bioreactor was applied to the domestic wastewater with low COD concentration (198~285 g/L) and a small sludge yield as around 0.05 gVSS/gCODrem was present by the long-term operation (Shin et al., 2014). Thus, it was confirmed that a small sludge yield can be obtained by the AnMBR with well bio-degradation performance in lab-scale as well as the pilot-scale fed by the domestic or sewage wastewaters (synthetic or real) (table 2.2).

2.3.3 Biogas yield and energy recovery

As mentioned above, sewage treatment though anaerobic fermentation release methane gas as a byproduct which could be combusted to generate heat or electricity energy (figure 2.8). Thus, it is considered to be a sustainable alternative for the sewage wastewater treatment anaerobically. The biogas produced during the AnMBR or other anaerobic process commonly refers to a mixture of gases mainly consists methane gas (CH_4), carbon dioxide (CO_2) and nitrogen gas (N_2) which produced by the biological degradation of organic matters in sewage wastewater under anaerobic environment conditions. Hydrogen gas (H_2) and hydrogen sulfide (H_2S) are sometimes also detected in the produced biogas (He et al., 2017; Li et al., 2015).

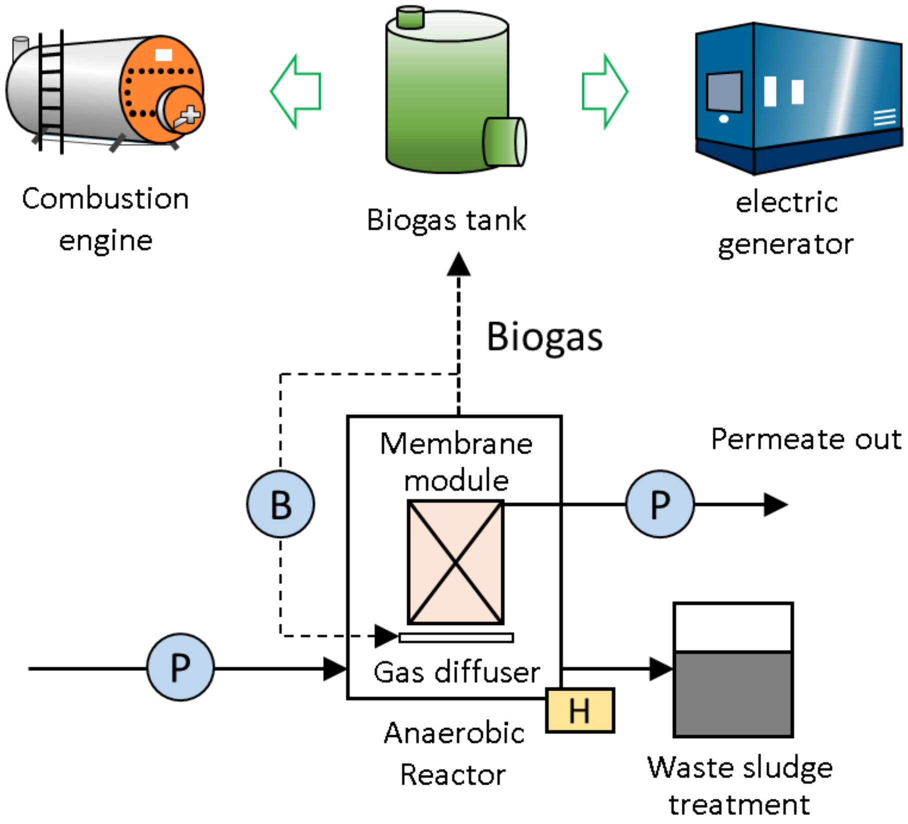
Researchers have focused on the biogas production and the methane gas yield by using AnMBRs treating the sewage wastewater. Methane yield of 0.133 L-CH_4/$gCOD_{rem}$ to

0.338 L-CH_4/$gCOD_{rem}$ have be obtained as different methane conversion rate in the studies listed in table 2.3. The results were included cases that treating the real sewage wastewater or real domestic wastewater. According to the results, the methane gas amount in the produced biogas was ranged from 60% to even as high as 90% normally which is hopeful to create a high efficiency for the energy conversion rate (R. Chen et al., 2017b; Gouveia et al., 2015a; Lin et al., 2013; Peña et al., 2019).

According to the concept of energy recovery by AnMBR shown in figure 2.8, the process can achieve 3 possible results:

- ✓ energy saving - the energy generated can be used back to the treatment process;
- ✓ energy neutral - the energy consumption for the sewage wastewater treatment is just covered by the energy generated from the biogas, thus, no more energy is needed from urban power network;
- ✓ energy positive - change the WWTP's role as an energy nets supplier.

A review study compared 11 pilot-scale AnMBRs treating sewage type wastewater and obtained a consequence that 6 of them achieved energy positive, 3 obtained energy saving (all less than 0.1 kW·h/m^3 during the process, far less than 0.27~0.60 kW·h/m^3 reported as the energy demand for traditional ASP treatment (Bodik and Kubaska, 2013)), 1 achieved energy neutral, and 1 required more energy due to the huge amount of cross flow gas for membrane fouling countermeasures during the operation (Lei et al., 2018).



E (consume) = E (pumps) + E (blower) + E (heater) + E (WS treatment) + E (electronic devices) +…

E (generate) = $\eta \times$ E (biogas)

η: conversion efficiency

① **E (consume)** > **E (generate)** Energy saving

② **E (consume)** = **E (generate)** Energy neutral

③ **E (consume)** < **E (generate)** Energy positive

Fig. 2.8 Concept of energy recovery by AnMBR.

2.3.4 Membrane fouling

Figure 2.7 has shown a membrane fouling concept caused by different resistances such as adsorption of sludge flocs and organic matters on the membrane surface formed cake or gel layers, adsorption or deposition of sludge flocs or other solid or dissolved matters to the membrane pore, and matters deposited and formed pore plugging (Le-Clech et al., 2006). The membrane fouling may result pore formed pathway impeded then cause the raising of TMP which more energy may require and the membrane would become unfiltered gradually for the treated water if the fouling reached to a certain extent (Ozgun et al., 2013). Therefore, in order to maintain the effectiveness of the membrane filtration, countermeasures like operation together with the cross flow gas or adding carriers (for example activated carbon) into the reactor to continuous scrape the membrane surface are usually used (Smith et al., 2012). Furthermore, a regular on-line chemical cleaning is normally utilized for cleaning the foulant to reduce the resistance and extend the membrane life till the membrane cannot be recovered to a high filtration efficiency (Stuckey, 2012). Due to the membrane fouling as a key factor for applying AnMBR to the sewage wastewater treatment (Yue et al., 2015), many achievements have been reported related in the AnMBR treating sewage or low-strength wastewater.

The achievement can be divided as effect of sewage substrate, operation conditions, characterization of sludge and microorganisms, bioactive metabolites, and then countermeasures.

(1) Sewage substrate. Substrate ingredients such as C/N ratio, COD/SO_4^{2-} ratio and specific organic matters (such as surfactants) have been reported lead out the membrane fouling via effected to different concentrations of extracellular polymeric substances (EPS) and soluble microbial products (SMP) (C. Chen et al.,

2018; Nie et al., 2017a, 2017b; Sarti et al., 2010). Besides, some inorganic matters such as metal ions and anions (CO_3^{2-}, SO_4^{2-}, PO_4^{3-}), was reported that can result membrane fouling (Wang et al., 2014). In addition, lower pH was reported that can lead to a low adherence and fouling propensity of EPS by compare of pH 6.3 and 8.3 in the experiments (Sweity et al., 2011).

(2) Operation conditions. Shorter HRT, bigger OLR and longer SRT induce a higher SMP production rate which result to an easier membrane fouling have been reported (Aquino et al., 2009; Aquino and Stuckey, 2004; R. Chen et al., 2017c; Win et al., 2016). Researches also reported that temperature can increase apparent sludge viscosity and induce a higher drag force to the membrane to result membrane fouling (Altmann and Ripperger, 1997). Moreover, it also has a significant effect on the microorganism communities which can affect the membrane fouling by different bioactive metabolites (Smith et al., 2015; Watanabe et al., 2017).

(3) Characterization of sludge and microorganisms. As it mentioned above, microorganism communities can produce different bioactive metabolites to affect the membrane fouling. In addition, MLSS has been reported as a factor of which can affect the membrane fouling (Dagnew et al., 2011).

(4) Bioactive metabolites. EPS and SMP were main metabolites as reported on membrane fouling foulant in AnMBR treating sewage wastewater (R. Chen et al., 2017a).

(5) Countermeasures. Except for those actions of operation with cross flow gas, adding of carriers and regular on-line chemical cleaning. There were also other countermeasures reported. For example, operation mood like suction

filtering/relax and suction filtering/backwashing are also used in long termed operation. Furthermore, a novel configuration of rotating membrane module was reported as an effective measure to extend the membrane life (Ruigómez et al., 2016). Afterwards, addition of flocculants was also reported have big benefits for membrane fouling through various effects (Deng et al., 2016; Díaz et al., 2014; Dong et al., 2015).

2.3.5 Degradation and influence of surfactant

Surfactants are organic compounds that are amphiphilic, meaning they contain both hydrophobic groups (their tails) and hydrophilic groups (their heads). Therefore, a surfactant contains both a water-insoluble (or oil-soluble) component and a water-soluble component (Myers, 2006). According to a report, about half of the world production of surfactants (estimated at 15 Mton/y) are soaps(Kosswig, 2000), which are rich in kinds of washes from kitchen sinks, baths or showers then discharged into the sewer systems.

Research on the effect of anionic surfactant inhibition on sewage treatment shows AnMBR was hence not suitable to dispose linear alkylbenzene sulfonate (LAS) containing sewage with higher concentration due to the results presented LAS can inhibit to the methanogen activity and can cause a higher membrane fouling rate as the microbial self-protection behaviour in coping with the LAS in sewage (Nie et al., 2017a). While on the research of degradation of non-ionic surfactant, it was found that alcohol ethoxylates (AE) could be efficiently degraded and converted into methane but it caused a higher membrane fouling rate because of the microbial self-protection behaviour by releasing more amounts of EPS and SMP (Nie et al., 2017b, 2017c).

2.3.6 Nutrients Removal by combined technologies

Since the mechanisms of regular anaerobic digestion process known so far is shown incapable of removal to the nutrients including total nitrogen, ammonia and total phosphorus, some technologies are required to implement to the treated water from AnMBR because of the important significance of nutrients removal which not only on purpose of meet high quality effluent requirements but also reduce the environmental issues such as eutrophication. However, the traditional methods for nutrients removal is normally not suit for the AnMBR treated water. For example, nitrification/denitrification process widely used so far in the ASP treatment require a much C-source or relatively high C/N ratio while AnMBR treated water has removed most of the COD amount already by the anaerobic digestion and combine nitrification/denitrification with AnMBR is also difficult by the process itself. Some other methods like ion exchange which is a classical chemical methods and the air stripping always have a big demand of energy. Compare with the traditional nitrogen removal processes mentioned above, a new biological process called anaerobic ammonium oxidation which also be known as anammox have be found and focused on as it's low emissions and energy saving (Yogev et al., 2017). Moreover, it can treat low C/N and high ammonium wastewater which is very suitable for treating the AnMBR effluent (Xing et al., 2015).

During the anammox process, the first step is nitritation which converted ammonium into nitrite, then the second step is the anammox reaction which convert ammonium and nitrite produce in first step into nitrogen gas. So the nitrogen was removed by nitrogen gas produced (figure 2.9). The reaction equations are shown below.

Nitritation (partial nitrification):

$$1.3NH_4^{+}+1.95O_2 \rightarrow 1.3NO_2^{-}+2.6H^{+}+1.3H_2O$$

Anammox reaction:

$$NH_4^+ + 1.3NO_2^- \rightarrow 0.26NO_3^- + 1.02N_2 + 2H_2O$$

Nitrogen removal (Nitritation + Anammox reaction):

$$NH_4^+ + 0.85O_2 \rightarrow 0.11NO_3^- + 0.445N_2 + 1.13H^+ + 1.43H_2O$$

For utilization, two stage and one stage anammox have been widely reported for treating different kinds of wastewaters. The process was considered to be strength on treating ammonia rich wastewaters especially with a high ammonia concentration or high nitrogen loading rate which have been reported in previous researches (He et al., 2016; Zhang et al., 2016). While there were also lab-scale one-stage anammox reactors reported by treating the low nitrogen concentration wastewater and achieved as high as around 80% removal efficiency of total nitrogen and 100% removal of ammonia nitrogen by a granular sludge CSTR as well as a carrier based reactor (Chen et al., 2019; R. Chen et al., 2018). In addition, there were also reported for anammox process applied in the mainstream of municipal sewage wastewater treatment process (Ali and Okabe, 2015; Cao et al., 2017).

In a previous study, a process combined anammox and hydroxyapatite (HAP) precipitation in an UASB expanded bed reactor for simultaneous nitrogen removal and phosphorus recovery was developed by applying specific Ca/P ratio and pH control (Ma et al., 2018). With a proper Ca/P ratio and pH control, the anammox reactor was transformed into an efficient process to simultaneously remove nitrogen and recover phosphorus and resulted a high phosphorus removal rate (0.14 ± 0.01 kg-P/m^3/d) as well as a stable high nitrogen removal efficiency (87.4 ± 2.9%) was achieved.

Therefore, combine AnMBR with anammox process is promising to generate a novel process for municipal sewage wastewater treatment by removal organics as well as the nutrients which could totally replace the current ASP treatment processes (figure 2.10).

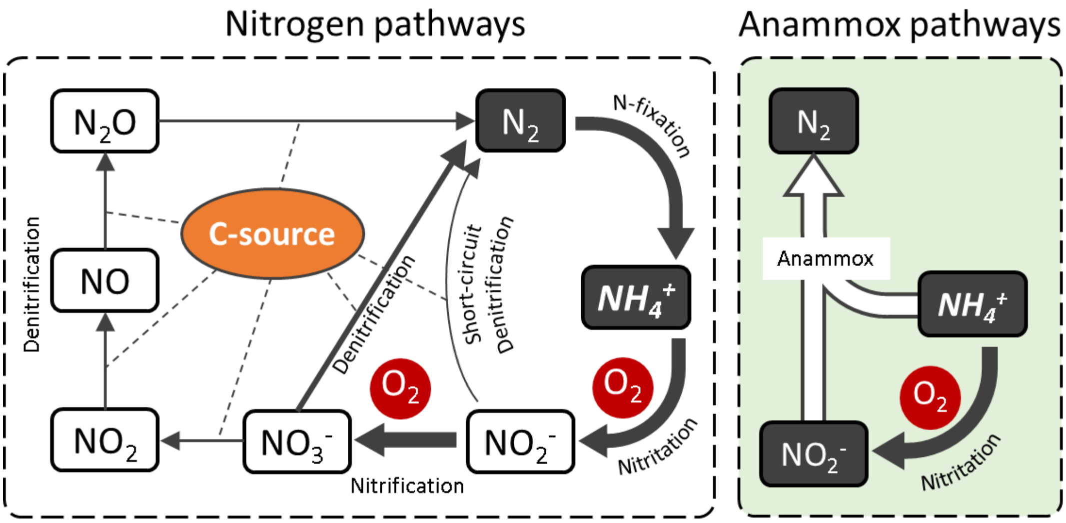


Fig. 2.9 Pathways of nitrogen and Anammox process.

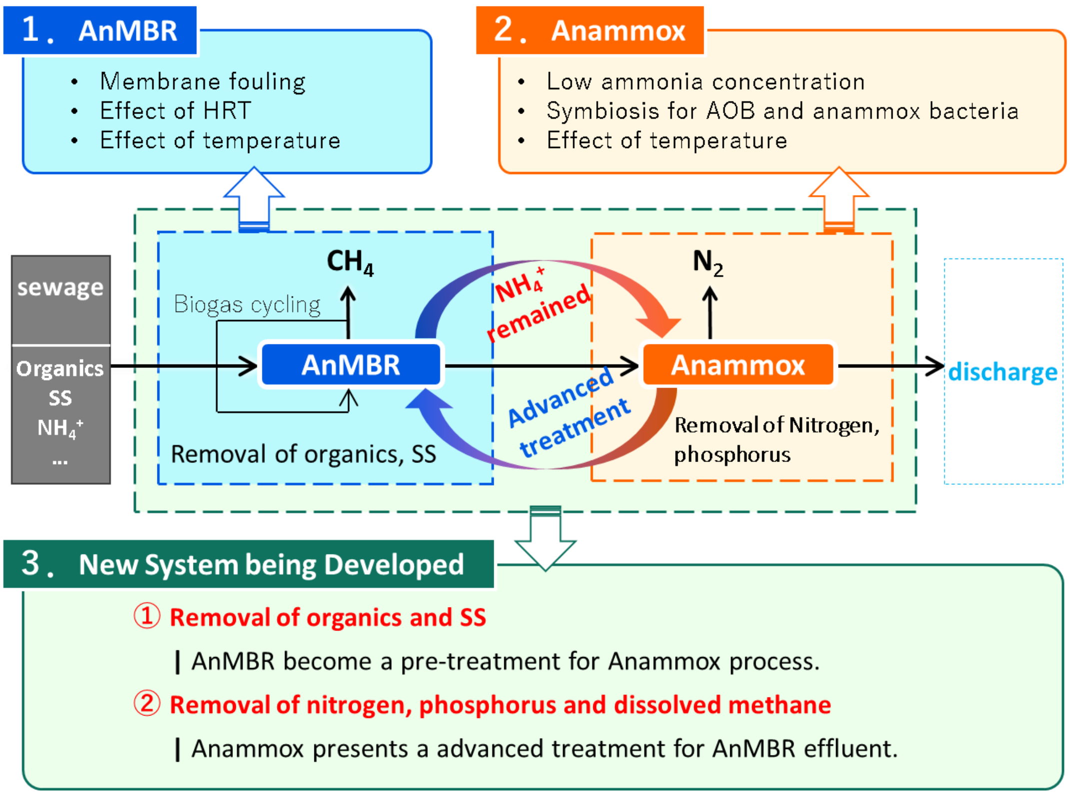


Fig. 2.10 Combined process by AnMBR and anammox process.

2.4 Application research on AnMBR treating real sewage

Application researches on the real sewage wastewater are listed as table 2.4 and table 2.5 for lab-scale and pilot-scale, respectively. The achievements have been introduced in the section 2.3 of this Chapter. While at the present stage, most of those researches based on treating the real sewage wastewater by AnMBR have the characteristic of focus on one-point which means, in other words, lack of comprehensive and systematic property. And so far, organic removal efficiency, membrane fouling and microbial communities are the popular research sites as reported relatively more. In addition, the reactors or system be used are normally very complex which also lead to a not-small energy consumption for operation or maintenance. Furthermore, some of the previous researches actually used the effluent of grit removal or primary sedimentation tank as the feeding substrate which means SS content may much lower than the raw sewage wastewater.

In this study, a new-designed simple structure AnMBR with a 20L reaction volume was applied in a wastewater treatment plant to start-up the process directly by the raw real sewage wastewater from the beginning gate of the WWTP even before the screen or the grit removal tank. A new-designed changeable submerged membrane module was set inside the AnMBR in the configuration of submerged AnMBR way. This study was in progressed under a serious topic including start-up the AnMBR by the real sewage wastewater, comparison of membrane pore size and the effect of HRT and temperature to form a systematically application researches based on the real sewage wastewater. The drawings of the new-designed AnMBR and the membrane modules are shown in the Appendix content (SUPPLEMENTARY DATA & FIGURES) of this thesis.

Table 2.1 The organic removal performance of AnMBRs treating sewage wastewater.

Anaerobic reactor (Volume/L)	Config.	Membrane	Water type	Tem. (°C)	HRT (h)	COD-in (mg/L)	COD-eff (mg/L)	CODre (%)	References
SAF (9)	ES	TM PVDF 0.1	RS	10~25	2.3	235~300	21~37	>86	(Yoo et al., 2013)
FBR (3.3)	ES	TM Ceramic 0.05	SS/RS	10/25	6~8	310~480	36~65	>94	(Seib et al., 2016b)
UAGB (4)	Sub.	HF PVDF 0.22	SS	20	12	330~370	29~32	91	(C. Chen et al., 2017)
CSTR (550)	ES	HF PVDF 0.04	RS	23	6.8	252±59	17~29	90	(Dong et al., 2015)
SAF (1700)	ES	HF PVDF 0.03	RS	8~30	4.5~6.8	198~285	14~28	93	(Shin et al., 2014)

Note: [Anaerobic reactor] SAF, staged anaerobic fluidized membrane bioreactor; FBR, fluidized bed bioreactor; UAGB, upflow anaerobic granular sludge blanket; CSTR, continuous stirred tank reactor; [Configuration] ES, external submerged; Sub., submerged; [Membrane] including membrane type, membrane material and pore size; TM, tubular membrane; HF, hollow fiber membrane; [Water type] RS, real sewage (municipal or domestic wastewater); SS, synthetic/man-made sewage or low organic strength wastewater.

Table 2.2 The sludge yield of AnMBRs reported.

Anaerobic reactor (Volume/L)	Config.	Water type	Tem. (°C)	HRT (h)	COD-in (mg/L)	CODre (%)	MLVSS (g/L)	Sludge yield (gVSS/gCODrem)	References
CMAC (6)	ES	RS	25~30	10	427±59	86	9.93	0.125	(Huang et al., 2011)
CSTR (6)	Sub.	SS	25	8~48	700±100	93	-	0.06~0.09	(R. Chen et al., 2017b)
CSTR (60)	ES	RS	35	2.2	396±101	87±7	4.7~20.1	0.07	(Mei et al., 2017)
UASB (284)	ES	RS	18±2	11.4	978±210	86	-	0.004	(Gouveia et al., 2015b)
SAF (1700)	ES	RS	8~30	4.5~6.8	198~285	93%	0.95~1.23	0.051	(Shin et al., 2014)

Note: [Anaerobic reactor] CMAC, completely mixed anaerobic reactor; CSTR, continuous stirred tank reactor; UASB, upflow anaerobic sludge blanket; SAF, staged anaerobic fluidized membrane bioreactor; [Configuration] ES, external submerged; Sub., submerged; [Water type] RS, real sewage (municipal or domestic wastewater); SS, synthetic/man-made sewage or low organic strength wastewater.

Table 2.3 The methane yield of AnMBRs treating sewage wastewater.

Anaerobic reactor (Volume/L)	Config.	Membrane	Water type	Tem. (°C)	HRT (h)	COD-in (mg/L)	Methane conversion rate(%)	Methane yield ($LCH_4/gCOD_{rem}$)	References
CSTR (6)	Sub.	FS PVDF 0.2	SS	25	8	700±100	96.5	0.338	(R. Chen et al., 2017b)
UAGB (4)	Side	HF PVDF 0.22	SS	20	12	330~370	45.3	0.161	(C. Chen et al., 2017)
UAGB (4)	Sub.	HF PVDF 0.22	SS	20	12	330~370	44.7	0.156	(C. Chen et al., 2017)
UAGB (3)	-	HF PVDF 0.22	SS	20	12	330~370	38.1	0.133	(C. Chen et al., 2017)
UASB (310)	ES	HF - 0.045	RS	6~30	10~13.4	892±271	67.1	0.235	(Gouveia et al., 2015a)

Note: [Anaerobic reactor] CSTR, continuous stirred tank reactor; UAGB, upflow anaerobic granular sludge blanket; UASB, upflow anaerobic sludge blanket; [Configuration] Sub., submerged; Side, side-stream AnMBR; ES, external submerged; [Membrane] including membrane type, membrane material and pore size; FS, flat sheet membrane; HF, hollow fiber membrane; [Water type] SS, synthetic/man-made sewage or low organic strength wastewater; RS, real sewage (municipal or domestic wastewater).

Table 2.4 Lab-scale AnMBRs treating the real sewage wastewater.

Anaerobic reactor /Volume(L)	Config.	Membrane	Tem. (°C)	HRT (h)	COD-in (mg/L)	COD-eff (mg/L)	CODre (%)	References
SAF/0.442	Sub.	HF - 0.1	30	10	342~527	40	84	(Yoo et al., 2012)
FBR/3.3	ES	TM Ceramic 0.05	10/25	6~8	310~480	36~65	>94	(Seib et al., 2016b)
CMAC/5	ES	FS - 0.45	25~30	10	427±59	60	86	(Huang et al., 2013)
CMAC/6	ES	FM PES 0.45	8~30	8-12	427±59	-	86	(Huang et al., 2011)
SAF/9	ES	TM PVDF 0.1	10~25	2.3	235~300	21~37	>86	(Yoo et al., 2013)
UASB/12.9	Sub.	TM PET 0.64	15~20	2.6	260±244	76±30	84	(An et al., 2009)
CSTR/50	Side	FS - 0.2	35	16	350~500	<30	98	(Kocadagistan and Topcu, 2007)

Note: [Anaerobic reactor] SAF, staged anaerobic fluidized membrane bioreactor; FBR, fluidized bed bioreactor; CMAC, completely mixed anaerobic reactor; UASB, upflow anaerobic sludge blanket; CSTR, continuous stirred tank reactor; [Configuration] Sub., submerged; ES, external submerged; Side, side-stream AnMBR; [Membrane] including membrane type, membrane material and pore size; HF, hollow fiber membrane; FS, flat sheet membrane; FM, frame membrane; TM, tubular membrane.

Table 2.5 Pilot-scale AnMBRs treating the real sewage wastewater.

Anaerobic reactor /Volume(L)	Config.	Membrane	Tem. (°C)	HRT (h)	COD-in (mg/L)	COD-eff (mg/L)	CODre (%)	References
UASB/310	ES	HF - 0.045	6~30	10~13.4	892±271	73~225	80~90	(Gouveia et al., 2015a)
-(CSTR)/350	ES	FS - 0.038	20/35	-	630±82	<80	90	(Martinez-Sosa et al., 2011)
CSTR/550	ES	HF PVDF 0.04	23	6.8	252±59	17~29	90	(Dong et al., 2015)
UASB/849	Side	TM - 100kDa	-	6	425±138	33±8	92	(Calderón et al., 2011)
UAGB/1500	Side	FS - 0.038	Environ. Tem.	16	197~553	-	86	(Martin-Garcia et al., 2011)
SAF/1700	ES	HF PVDF 0.03	8~30	4.5~6.8	198~285	14~28	93	(Shin et al., 2014)
-/2100	ES	HF - 0.05	33	6~21	445±95	77±33	90	(Giménez et al., 2011)

Note: [Anaerobic reactor] UASB, upflow anaerobic sludge blanket; CSTR, continuous stirred tank reactor; UAGB, upflow anaerobic granular sludge blanket; SAF, staged anaerobic fluidized membrane bioreactor; [Configuration] ES, external submerged; Side, side-stream AnMBR; [Membrane] including membrane type, membrane material and pore size; HF, hollow fiber membrane; FS, flat sheet membrane; TM, tubular membrane.

REFRENCES:

Ali, M., Okabe, S., 2015. Anammox-based technologies for nitrogen removal: Advances in process start-up and remaining issues. Chemosphere. https://doi.org/10.1016/j.chemosphere.2015.06.094

Alleman, J.E., 2005. The genesis and evolution of activated sludge technology. Indiana, USA 22.

Altmann, J., Ripperger, S., 1997. Particle deposition and layer formation at the crossflow microfiltration. J. Memb. Sci. https://doi.org/10.1016/S0376-7388(96)00235-9

An, Y., Wang, Z., Wu, Z., Yang, D., Zhou, Q., 2009. Characterization of membrane foulants in an anaerobic non-woven fabric membrane bioreactor for municipal wastewater treatmentAn, Y., Wang, Z., Wu, Z., Yang, D., & Zhou, Q. (2009). Characterization of membrane foulants in an anaerobic non-woven fabric memb. Chem. Eng. J. https://doi.org/10.1016/j.cej.2009.09.003

Angelakis, A.N., Koutsoyiannis, D., Tchobanoglous, G., 2005. Urban wastewater and stormwater technologies in ancient Greece. Water Res. https://doi.org/10.1016/j.watres.2004.08.033

Angelakis, A.N., Spyridakis, S. V, 1996. The Status of Water Resources in Minoan Times: A Preliminary Study, in: Angelakis, A.N., Issar, A.S. (Eds.), Diachronic Climatic Impacts on Water Resources. Springer Berlin Heidelberg, Berlin, Heidelberg, pp. 161–191.

Aquino, S.F., Gloria, R.M., Silva, S.Q., Chernicharo, C.A.L., 2009. Quantification of the inert chemical oxygen demand of raw wastewater and evaluation of soluble microbial product production in demo-scale upflow anaerobic sludge blanket reactors under different operational conditions. Water Environ. Res. 81, 608–616.

Aquino, S.F., Stuckey, D.C., 2004. The effect of organic and hydraulic shock loads on the production of soluble microbial products in anaerobic digesters. Water Environ. Res. 76, 2628–2636.

Balaman, S.Y., 2018. Decision-making for Biomass-based Production Chains: The Basic Concepts and Methodologies. Academic Press.

Batstone, D.J., Keller, J., Angelidaki, I., Kalyuzhnyi, S. V, Pavlostathis, S.G., Rozzi, A., Sanders, W.T.M., Siegrist, H., Vavilin, V.A., 2002. The IWA Anaerobic Digestion Model No 1 (ADM1). Water Sci. Technol. 45, 65–73. https://doi.org/10.2166/wst.2002.0292

Bodik, I., Kubaska, M., 2013. Energy and sustainability of operation of a wastewater treatment plant. Environ. Prot. Eng. 39, 15–24.

Calderón, K., Rodelas, B., Cabirol, N., González-López, J., Noyola, A., 2011. Analysis of microbial communities developed on the fouling layers of a membrane-coupled anaerobic bioreactor applied to wastewater treatment. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2011.01.007

Cao, Y., van Loosdrecht, M.C.M., Daigger, G.T., 2017. Mainstream partial nitritation–anammox in municipal wastewater treatment: status, bottlenecks, and further studies. Appl. Microbiol. Biotechnol. https://doi.org/10.1007/s00253-016-8058-7

Chen, C., Guo, W., Ngo, H.H., Chang, S.W., Duc Nguyen, D., Dan Nguyen, P., Bui, X.T., Wu, Y., 2017. Impact of reactor configurations on the performance of a granular anaerobic membrane bioreactor for municipal wastewater treatment. Int. Biodeterior. Biodegrad. https://doi.org/10.1016/j.ibiod.2017.03.021

Chen, C., Guo, W.S., Ngo, H.H., Chang, S.W., Nguyen, D.D., Zhang, J., Liang, S., Guo, J.B., Zhang, X.B., 2018. Effects of C/N ratio on the performance of a hybrid sponge-assisted aerobic moving bed-anaerobic granular membrane bioreactor for municipal wastewater treatment. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.09.062

Chen, C., Guo, W.S., Ngo, H.H., Liu, Y., Du, B., Wei, Q., Wei, D., Nguyen, D.D., Chang, S.W., 2017. Evaluation of a sponge assisted-granular anaerobic membrane bioreactor (SG-AnMBR) for municipal wastewater treatment. Renew. Energy 111, 620–627. https://doi.org/https://doi.org/10.1016/j.renene.2017.04.055

Chen, R., Ji, J., Chen, Y., Takemura, Y., Liu, Y., Kubota, K., Ma, H., Li, Y.-Y., 2019. Successful operation performance and syntrophic micro-granule in partial nitritation and anammox reactor treating low-strength ammonia wastewater. Water Res. 155, 288–299.

Chen, R., Nie, Y., Hu, Y., Miao, R., Utashiro, T., Li, Q., Xu, M., Li, Y.Y., 2017a. Fouling behaviour of soluble microbial products and extracellular polymeric substances in a submerged anaerobic membrane bioreactor treating low-strength wastewater at room temperature. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2017.02.046

Chen, R., Nie, Y., Ji, J., Utashiro, T., Li, Q., Komori, D., Li, Y.-Y., 2017b. Submerged anaerobic membrane bioreactor (SAnMBR) performance on sewage treatment: removal efficiencies, biogas production and membrane fouling. Water Sci. Technol. 76, 1308–1317. https://doi.org/10.2166/wst.2017.240

Chen, R., Nie, Y., Tanaka, N., Niu, Q., Li, Q., Li, Y.Y., 2017c. Enhanced methanogenic degradation of cellulose-containing sewage via fungi-methanogens syntrophic association in an anaerobic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.09.046

Chen, R., Takemura, Y., Liu, Y., Ji, J., Sakuma, S., Kubota, K., Ma, H., Li, Y.-Y., 2018. Using Partial Nitrification and Anammox To Remove Nitrogen from Low-Strength Wastewater by Co-immobilizing Biofilm inside a Moving Bed Bioreactor. ACS Sustain. Chem. Eng. 7, 1353–1361.

Cooper, P.F., 2007. Historical aspects of wastewater treatment. Decent. Sanit. reuse concepts, Syst. Implement.

Dagnew, M., Parker, W., Seto, P., Waldner, K., Hong, Y., Bayly, R., Cumin, J., 2011. Pilot testing of an AnMBR for municipal wastewater treatment. Proc. Water Environ. Fed. 2011, 4931–4941.

Deng, L., Guo, W., Ngo, H.H., Zhang, H., Wang, J., Li, J., Xia, S., Wu, Y., 2016. Biofouling and control approaches in membrane bioreactors. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.09.105

Díaz, H., Azócar, L., Torres, A., Lopes, S.I.C., Jeison, D., 2014. Use of flocculants for increasing permeate flux in anaerobic membrane bioreactors. Water Sci. Technol. https://doi.org/10.2166/wst.2014.153

Dong, Q., Parker, W., Dagnew, M., 2015. Impact of FeCl3 dosing on AnMBR treatment of municipal wastewater. Water Res. 80, 281–293. https://doi.org/10.1016/J.WATRES.2015.04.025

Dvořák, L., Gómez, M., Dolina, J., Černín, A., 2016. Anaerobic membrane bioreactors—a mini review with emphasis on industrial wastewater treatment: applications, limitations and perspectives. Desalin. Water Treat. https://doi.org/10.1080/19443994.2015.1100879

Ghneim., A., 2010. Wastewater reuse and management in the Middle East and North Africa : a case study of Jordan. Berlin : Universitätsverlag der TU Berlin.

Giménez, J.B., Robles, A., Carretero, L., Durán, F., Ruano, M. V, Gatti, M.N., Ribes, J., Ferrer, J., Seco, A., 2011. Experimental study of the anaerobic urban wastewater treatment in a submerged hollow-fibre membrane bioreactor at pilot scale. Bioresour. Technol. 102, 8799–8806. https://doi.org/https://doi.org/10.1016/j.biortech.2011.07.014

Gordon, L.D.B., 1851. A short description of the plans of Captain James Vetch for the sewerage of the metropolis.

Gouveia, J., Plaza, F., Garralon, G., Fdz-Polanco, F., Peña, M., 2015a. Long-term operation of a pilot scale anaerobic membrane bioreactor (AnMBR) for the treatment of municipal wastewater under psychrophilic conditions. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2015.03.002

Gouveia, J., Plaza, F., Garralon, G., Fdz-Polanco, F., Peña, M., 2015b. A novel configuration for an anaerobic submerged membrane bioreactor (AnSMBR). Bioresour. Technol.

Gray, N., 2017. Water technology. CRC Press.

He, C.S., He, P.P., Yang, H.Y., Li, L.L., Lin, Y., Mu, Y., Yu, H.Q., 2017. Impact of zero-valent iron nanoparticles on the activity of anaerobic granular sludge: From macroscopic to microcosmic investigation. Water Res. https://doi.org/10.1016/j.watres.2017.09.061

He, S., Zhang, Y., Niu, Q., Ma, H., Li, Y.Y., 2016. Operation stability and recovery performance in an Anammox EGSB reactor after pH shock. Ecol. Eng. https://doi.org/10.1016/j.ecoleng.2016.01.084

Henze, M., van Loosdrecht, M.C.M., Ekama, G.A., Brdjanovic, D., 2008. Biological Wastewater Treatment: Principles, Modelling and Design. https://doi.org/10.2166/9781780401867

Huang, X., Gui, P., Qian, Y., 2001. Effect of sludge retention time on microbial behaviour in a submerged membrane bioreactor. Process Biochem. https://doi.org/10.1016/S0032-9592(01)00135-2

Huang, Z., Ong, S.L., Ng, H.Y., 2013. Performance of submerged anaerobic membrane bioreactor at different SRTs for domestic wastewater treatment. J. Biotechnol. https://doi.org/10.1016/j.jbiotec.2013.01.001

Huang, Z., Ong, S.L., Ng, H.Y., 2011. Submerged anaerobic membrane bioreactor for low-strength wastewater treatment: Effect of HRT and SRT on treatment performance and membrane fouling. Water Res. https://doi.org/10.1016/j.watres.2010.08.035

Jenkins, D., Wanner, J., 2014. Activated sludge-100 years and counting. IWA publishing.

Khouri, N., Kalbermatten, J., Bartone, C., 1994. Reuse of Wastewater in Agriculture: A Guide for Planners Sanitation Report.

Kocadagistan, E., Topcu, N., 2007. Treatment investigation of the Erzurum City municipal wastewaters with anaerobic membrane bioreactors. Desalination. https://doi.org/10.1016/j.desal.2006.10.038

Kosswig, K., 2000. Surfactants. Ullmann’s Encycl. Ind. Chem., Major Reference Works. https://doi.org/doi:10.1002/14356007.a25_747

Le-Clech, P., Chen, V., Fane, T.A.G., 2006. Fouling in membrane bioreactors used in wastewater treatment. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2006.08.019

Lei, Z., Yang, S., Li, Y. you, Wen, W., Wang, X.C., Chen, R., 2018. Application of anaerobic membrane bioreactors to municipal wastewater treatment at ambient temperature: A review of achievements, challenges, and perspectives. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2018.07.050

Li, W., Niu, Q., Zhang, H., Tian, Z., Zhang, Y., Gao, Y., Li, Y.Y., Nishimura, O., Yang, M., 2015. UASB treatment of chemical synthesis-based pharmaceutical wastewater containing rich organic sulfur compounds and sulfate and associated microbial characteristics. Chem. Eng. J. https://doi.org/10.1016/j.cej.2014.08.085

Lin, H., Peng, W., Zhang, M., Chen, J., Hong, H., Zhang, Y., 2013. A review on anaerobic membrane bioreactors: Applications, membrane fouling and future perspectives. Desalination. https://doi.org/10.1016/j.desal.2013.01.019

Ma, H., Zhang, Y., Xue, Y., Li, Y.Y., 2018. A new process for simultaneous nitrogen removal and phosphorus recovery using an anammox expanded bed reactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2018.07.044

Mahboubi, A., Ylitervo, P., Doyen, W., De Wever, H., Taherzadeh, M.J., 2016. Reverse membrane bioreactor: Introduction to a new technology for biofuel production. Biotechnol. Adv. https://doi.org/10.1016/j.biotechadv.2016.05.009

Martin-Garcia, I., Monsalvo, V., Pidou, M., Le-Clech, P., Judd, S.J., McAdam, E.J., Jefferson, B., 2011. Impact of membrane configuration on fouling in anaerobic membrane bioreactors. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2011.07.042

Martinez-Sosa, D., Helmreich, B., Netter, T., Paris, S., Bischof, F., Horn, H., 2011. Anaerobic submerged membrane bioreactor (AnSMBR) for municipal wastewater treatment under mesophilic and psychrophilic temperature conditions. Bioresour.

Technol. 102, 10377–10385. https://doi.org/https://doi.org/10.1016/j.biortech.2011.09.012

Mei, X., Quek, P.J., Wang, Z., Ng, H.Y., 2017. Alkali-assisted membrane cleaning for fouling control of anaerobic ceramic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.02.052

Melosi, M. V, 2008. The sanitary city: Environmental services in urban America from colonial times to the present. University of Pittsburgh Pre.

Myers, D., 2006. Surfactant science and technology. Wiley Online Library.

Nie, Y., Kato, H., Sugo, T., Hojo, T., Tian, X., Li, Y.Y., 2017a. Effect of anionic surfactant inhibition on sewage treatment by a submerged anaerobic membrane bioreactor: Efficiency, sludge activity and methane recovery. Chem. Eng. J. https://doi.org/10.1016/j.cej.2017.01.022

Nie, Y., Niu, Q., Kato, H., Sugo, T., Tian, X., Li, Y.Y., 2017b. Efficient methanogenic degradation of alcohol ethoxylates and microbial community acclimation in treatment of municipal wastewater using a submerged anaerobic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.11.128

Nie, Y., Tian, X., Zhou, Z., Li, Y.Y., 2017c. Impact of food to microorganism ratio and alcohol ethoxylate dosage on methane production in treatment of low-strength wastewater by a submerged anaerobic membrane bioreactor. Front. Environ. Sci. Eng. https://doi.org/10.1007/s11783-017-0947-1

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. Sep. Purif. Technol. https://doi.org/10.1016/j.seppur.2013.06.036

Peña, M., do Nascimento, T., Gouveia, J., Escudero, J., Gómez, A., Letona, A., Arrieta, J., Fdz-Polanco, F., 2019. Anaerobic submerged membrane bioreactor (AnSMBR) treating municipal wastewater at ambient temperature: Operation and potential use for agricultural irrigation. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2019.03.019

Ruigómez, I., Vera, L., González, E., González, G., Rodríguez-Sevilla, J., 2016. A novel rotating HF membrane to control fouling on anaerobic membrane bioreactors treating wastewater. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2015.12.011

Rulkens, W., 2008. Sewage Sludge as a Biomass Resource for the Production of Energy: Overview and Assessment of the Various Options. Energy & Fuels 22, 9–15. https://doi.org/10.1021/ef700267m

Sarti, A., Pozzi, E., Chinalia, F.A., Ono, A., Foresti, E., 2010. Microbial processes and bacterial populations associated to anaerobic treatment of sulfate-rich wastewater. Process Biochem. https://doi.org/10.1016/j.procbio.2009.09.002

Seib, M.D., Berg, K.J., Zitomer, D.H., 2016a. Influent wastewater microbiota and temperature influence anaerobic membrane bioreactor microbial community. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.05.098

Seib, M.D., Berg, K.J., Zitomer, D.H., 2016b. Low energy anaerobic membrane bioreactor for municipal wastewater treatment. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2016.05.007

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). Bioresour. Technol. https://doi.org/10.1016/j.biortech.2014.02.060

Smith, A.L., Skerlos, S.J., Raskin, L., 2015. Anaerobic membrane bioreactor treatment of domestic wastewater at psychrophilic temperatures ranging from 15 C to 3 C. Environ. Sci. Water Res. Technol. 1, 56–64.

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. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2012.04.055

Stuckey, D.C., 2012. Recent developments in anaerobic membrane reactors. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2012.05.138

Sweity, A., Ying, W., Belfer, S., Oron, G., Herzberg, M., 2011. PH effects on the adherence and fouling propensity of extracellular polymeric substances in a membrane bioreactor. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2011.04.056

Tzanakakis, V.E., Paranychianaki, N. V., Angelakis, A.N., 2007. Soil as a wastewater treatment system: Historical development. Water Sci. Technol. Water Supply. https://doi.org/10.2166/ws.2007.008

Wang, Z., J., M., C., T., K., K., Q., W., X., H., 2014. Membrane Cleaning in Membrane Bioreactors : A Review. J. Memb. Sci.

Watanabe, R., Nie, Y., Wakahara, S., Komori, D., Li, Y.Y., 2017. Investigation on the response of anaerobic membrane bioreactor to temperature decrease from 25 °C to 10 °C in sewage treatment. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.07.001

Win, T.T., Kim, H., Cho, K., Song, K.G., Park, J., 2016. Monitoring the microbial community shift throughout the shock changes of hydraulic retention time in an anaerobic moving bed membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2015.11.085

Xing, B.S., Guo, Q., Yang, G.F., Zhang, J., Qin, T.Y., Li, P., Ni, W.M., Jin, R.C., 2015.

The influences of temperature, salt and calcium concentration on the performance of anaerobic ammonium oxidation (anammox) process. Chem. Eng. J. https://doi.org/10.1016/j.cej.2014.12.007

Yogev, U., Sowers, K.R., Mozes, N., Gross, A., 2017. Nitrogen and carbon balance in a novel near-zero water exchange saline recirculating aquaculture system. Aquaculture. https://doi.org/10.1016/j.aquaculture.2016.04.029

Yoo, R., Kim, J., McCarty, P.L., Bae, J., 2012. Anaerobic treatment of municipal wastewater with a staged anaerobic fluidized. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2012.06.028

Yoo, R.H., Kim, J.H., McCarty, P.L., Bae, J.H., 2013. Effect of temperature on the treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor. Water Sci. Technol. 69, 1145–1150. https://doi.org/10.2166/wst.2013.793

Yoon, S.-H., 2015. Membrane bioreactor processes: principles and applications. CRC press.

Yue, X., Koh, Y.K.K., Ng, H.Y., 2015. Effects of dissolved organic matters (DOMs) on membrane fouling in anaerobic ceramic membrane bioreactors (AnCMBRs) treating domestic wastewater. Water Res. 86, 96–107. https://doi.org/10.1016/J.WATRES.2015.07.038

Zhang, Y., He, S., Niu, Q., Qi, W., Li, Y.Y., 2016. Characterization of three types of inhibition and their recovery processes in an anammox UASB reactor. Biochem. Eng. J. https://doi.org/10.1016/j.bej.2016.01.022

野池達也，佐藤和明，安井英斉，李玉友，落修一, 2009. メタン発酵. 技報堂出版.

Chapter 3

Effect of membrane pore size on start-up and long-term operation performance

3.1 Introduction

In the last hundred years, Activated Sludge Process (ASP) has been developed into a mature process for treating sewage wastewater. However, during the ASP treatment, there are some issues presented, for example:

- ✧ a great deal of energy is needed for aeration requirement;
- ✧ a large amount of greenhouse gas is discharged;
- ✧ a big amount of waste sludge is produced.

On the other hands, sewage is the most abundant type of wastewater and a valuable resource containing water, nutrients and energy which is worthy of recovery and reuse. If recovery and reuse could be achieved in technology, it would be possible that sewages become net supplier of renewable resources, energy and reclaimed water (Khiewwijit et al., 2015; Ozgun et al., 2013). Consequently, development of appropriate technology that can convert sewage into high level renewable energy and high quality reclaimed water is very important.

The anaerobic process does not have those disadvantages which ASP have which mentioned above and has drawn considerable attention for its ability to convert chemically bound energy in the organic pollutants to useful energy namely biogas (Shizas and Bagley, 2004). While there are two points which become the main obstacles to applying anaerobic digestion directly to sewage treatment. Firstly, anaerobic sludge shows a trend of slowly grow, especially under the condition of low organic strength feeding. Secondly, it is hard to separate activated sludge and the treated water in traditional anaerobic digestion process. That appears even in the situation of treating a very big amount of wastewater. While the processes for treating the sewages, an issue always be faced is to dealing with the large amount of low organic strength wastewater.

The anaerobic membrane bio-reactor (AnMBR) integrates the anaerobic digestion process and the membrane technology so that created a new process which could provide with both the advantages of anaerobic digestion as well as the high efficiency of sludge-water separation due to the filtration by membranes. Usually, organic matters mainly indicated by chemical oxygen demand (COD) and biochemical oxygen demand (BOD), suspended solid (SS), total nitrogen (TN), ammonia (NH_4^+-N) and total phosphorus (TP) are the main concerns of common pollutants in sewage, and the treatment performances for these pollutants show a tendency to appear different fates in an anaerobic membrane reactor. The organic matters are always complex polymers which can be categorized as carbohydrates, fats and proteins, and their removal efficiencies are highly dependent on the anaerobic digestion ability due to the four key steps to be known as hydrolysis, acidogenesis, acetogenesis and methoanogenesis during the anaerobic digestion process. The four steps are carried out by distinct consortia of bacteria, namely fermentative bacteria, syntrophic acetogens, homoacetogens, hydrogenetrophic methanogens and aceticlastic methanogens (Batstone et al., 2002). Normally, in the AnMBRs, SS could be removed to a large extent due to the micro- filtration or ultra-filtration by membrane module while TN, ammonia and TP is incapable of removal because of the mechanisms of anaerobic digestion known so far.

Up to now, AnMBR has been successfully applied in the field of industrial wastewater treatment and there are plenty of studies related to AnMBR focused on high organic strength wastewater or industrial wastewater while still lacking of development for treating the real municipal sewage as the features of: large quantity, complicated composition, instable and low concentration of pollutant (Lei et al., 2018). However, despite that, there are some studies that related to the low organic strength wastewater or sewage based on man-made synthetic wastewater have be reported.

High efficiency of organic removal and methane conversion rate were obtained upon small sludge yield reported in the previous research (Chen et al., 2017b). Research on the effect of anionic surfactant inhibition on sewage treatment shows AnMBR was hence not suitable to dispose linear alkylbenzene sulfonate (LAS) containing sewage with higher concentration due to the results presented LAS can inhibit to the methanogen activity and can cause a higher membrane fouling rate as the microbial self-protection behaviour in coping with the LAS in sewage (Nie et al., 2017a). While on the research of degradation of non-ionic surfactant, it was found that alcohol ethoxylates (AE) could be efficiently degraded and converted into methane but it caused a higher membrane fouling rate because of the microbial self-protection behaviour by releasing more amounts of extracellular polymeric substances (EPS) and soluble microbial products (SMP) (Nie et al., 2017b, 2017c). A research that contributed to a better understanding of properties of EPS and SMP and their roles in membrane fouling in an AnMBR treating low strength sewage at room temperature was also reported (Chen et al., 2017a). Moreover, studies that related to the low organic strength wastewater or sewage based on synthetic wastewater also have been reported on the field of biogas energy recovery (Hasan et al., 2014; Song et al., 2018), effect of HRT and SRT (Huang et al., 2011), effect of operation temperature, biomass concentration (Barreto et al., 2017), and the fouling of membranes (Gao et al., 2010; Hong et al., 2014; Meng et al., 2017), but rarely have the researches focused on the membrane pore size.

Normally, in AnMBRs, SS could be removed to a large extent due to the filtration or by membrane module, and that has been proved by using submerged AnMBR treating two types of synthetic sewage with SS contained or not contained and evaluated the performance of AnMBR at 25 °C (Watanabe et al., 2016). Not just SS in the influent water, but also the microorganisms in the activated sludge, all kinds of solids can also be obstructed by the membrane filtration process (Guglielmi et al., 2010). Membranes' filtration process included Microfiltration (MF, 0.1~1μm or 0.1~5μm), Ultrafiltration (UF, 0.01~0.1μm), Nanofiltration (NF, 1~10nm), and Reverse Osmosis (RO, No pores or 0.1~1nm) depends on the pore size of the membrane itself (Yoon, 2015). MF/UF can be highly effective in eliminating bacteria and used widely in the wastewater treatment while NF/RO are more used for drinking water or surface water filtration.

UF pore size are also expressed as molecular weight cut off by the classification of membranes according to pore size (Yoon, 2015). It was found that many researches or cases used UF to treat wastewaters including the municipal sewage wastewater by the literature work. For example, Yoo et al., 2013 applied.1 μm pore size hollow fiber membranes in a lab-scale SAF-MBR system to treat the domestic wastewater. Mei et al., 2017 applied flatsheet ceramic membrane module with 0.08 μm pore size in two lab-scale AnCMBRs operated in parallel to investigate the fouling control by treating the real domestic wastewater. Yue et al., 2015 also used 0.08 μm pore size ceramic membrane to treating the domestic wastewater researched on membrane fouling effects. Sweity et al., 2011 studied on pH effects on the adherence and fouling propensity of EPS in a UF membrane bioreactor treating the long-strength wastewater (COD 418±123 mg/L in influent). Gouveia et al., 2015 even applied only 0.045 μm pore size UF in a pilot plant by AnSMBR to treating the municipal wastewater. While weather it is needed for such small pore size of UF membrane to complete the water-solid separation, it is a question. In addition, because of different pore size ranges can obstruct matters with different particle size but smaller pore size has a demand for more energy for pump suction in the same liquid condition be filtered, it is important to figure out which kind of pore size is more suitable for the sewage wastewater treatment by using AnMBRs in order to achieve more economical and reasonable operation.

In this chapter, two mini-pilot scale AnMBRs by different pore size membranes (one was 0.4μm pore size MF, the other one was 0.05μm pore size UF) was installed in the wastewater treatment plant. The performance of those two AnMBRs were evaluated by the effluent water quality, bio-gas production, the membrane's operation properties and the microbial community analysis to verify the feasibility for AnMBR start-up and treating the real sewage wastewater as well as seek for more suitable membrane pore size for the sewage wastewater treatment by AnMBRs on the purpose of achieving economical and reasonable operations.

3.2 Materials and Methods

3.2.1 Consist and operation of the mini-pilot AnMBR systems

Two AnMBRs were installed in Sen-En wastewater treatment plant (S-WWTP) located in Tagajo city so that the original municipal sewage wastewater could be pumped into the reactors. The raw municipal sewage wastewater was pumped continuously from the beginning of the S-WWTP into a 100 L sewage bucket together with a continues stirring

(US540-401) to keep the sewage fresh and there was over-flow pipeline setting to make the extra sewage flow back under gravity. Basic water quality index of the raw municipal sewage wastewater was shown in table 3.1.

Those 2 AnMBRs were set with 2 membranes for each reactor and named by M1 (MF, 0.4μm pore size) and M2 (UF, 0.05μm pore size). The two kinds of membrane are shown in figure 3.1. The influent of each AnMBR was taken from the same sewage bucket by a peristaltic pump (FP-100-1515). For each AnMBR system, the solid-liquid separation was permeated using a micro-filtration module (Mitsubishi Chemicals, Japan) by a peristaltic pump (FP-100-1515). The produced biogas was recycled by a diaphragm pump (APN-110KV-1, Iwaki, Japan) to scour the membranes' surface as a fouling control method via a gas diffuser set directly below the membrane module (Martin-Garcia et al., 2011). A digital pressure meter (AP-10S & AP-V85, Keyence, Japan) was installed between the membrane module and the permeate pump to measure the trans-membrane pressure (TMP) and the TMP data was recorded by a multi input data logger (NR-500 & NR-HA08, Keyence, Japan) connected to a computer and controlled by the installed software. Biogas production was measured by a wet gas meter. The operation temperature of 25°C was controlled by a water bath equipment (NTT-20S). The whole system's sketch is flowing the structure shown in figure 3.2.

Seed sludge was taken from the full-scale waste sludge treatment process inside the S-WWTP. In order to obtain a faster and better microbial acclimation, the 2 AnMBRs were fed by not just real sewages but also mixed with total COD concentration of 400 mg/L glucose and methanol from day 21 till day 37. The HRT for sewage was controlled as 24 hours in the beginning 37 days. Besides, one membrane was shut down in M2 for adjusting the total membrane area as the same level as M1 during the period of day 48 to day 90. The detail operation conditions are shown in table 3.2.

Table 3.1 Basic water quality index of the real sewage wastewater.

Parameter	Value	Unit
Sewage COD	400 ± 150	mg/L
Sewage BOD	180 ± 80	mg/L
Sewage TN	23 ± 7	mg/L
Sewage TP	10 ± 5	mg/L
Sewage TS	900 ± 100	mg/L
Sewage SS	200 ± 50	mg/L
pH	7.3 ± 0.3	
temperature	10~25	°C

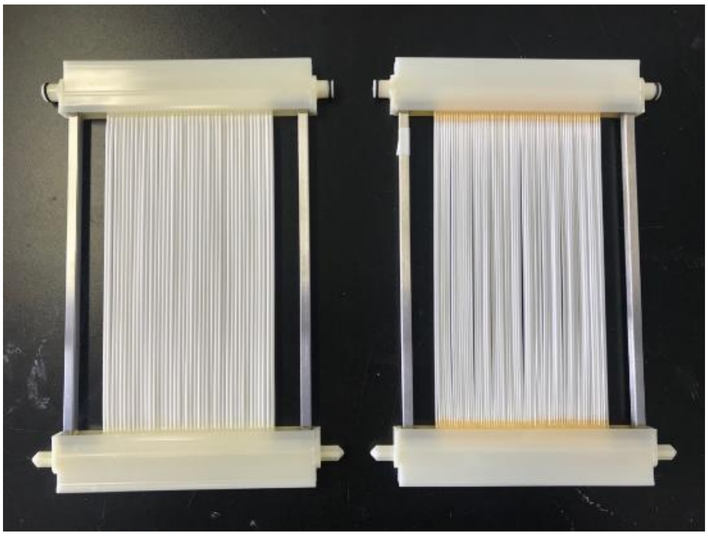


Fig. 3.1 Two kinds of membrane (pore size, left:0.4μm; right0.05μm).

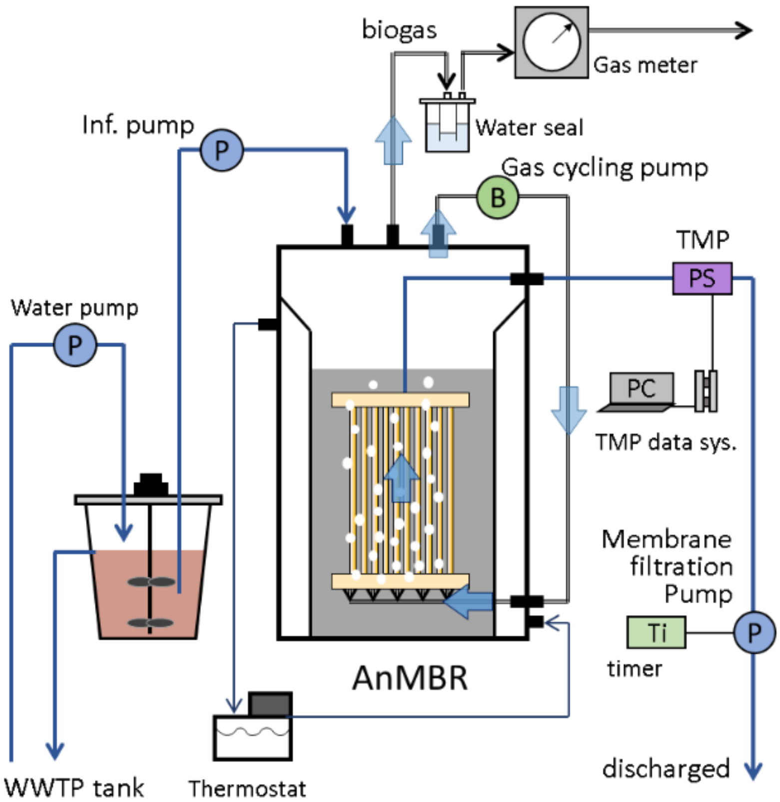


Fig. 3.2 The AnMBR system consist.

Table 3.2 The operation conditions of AnMBRs

Items	M1				M2				
Membrane	Hollow fiber, PVDF								
Membrane pore size (μm)	0.4 (MF)				0.05 (UF)				
Total membrane area (m^2)	0.146				0.270		0.135	0.270	
HRT (h)	24*	24	12	14.4	24*	24		14.4	12
FLUX (m/d)	0.137		0.274	0.228	0.074		0.148	0.123	0.148
Operated period (Day-)	7-37	38-48	49-84 136-150	85-135	7-37	38-47 90-108	48-90	109-118	119-170
Filtration mode (X min on / Y min off)	4 / 1			2 / 1	4 / 1			2 / 1	4 / 1
Reaction volume (L)	20								
Temperature (℃)	25.2 ± 0.2								
Gas circulation (L/min)	10		10-17		10				10-17

24*: microbial acclimation period, HRT 24 hours, sewage (day1-20), sewage + glucose + methanol (day 21-37).

3.2.2 Samples collection and analysis methods

Influent, effluent and mixed liquor samples were regularly taken in order to analysis water quality index and the sludge traits. The analysis of COD, BOD, SS, mixed liquor suspended solid (MLSS) and mixed liquor volatile suspended solid (MLVSS) were in according with standard methods (APHA, 2005). The proportion of CH4, CO2, and N2 in biogas produced was measured using a gas chromatograph (Shimadzu, GC-8A, Japan) equipped with a thermal conductivity detector. Dissolved methane in the effluent was determined using a headspace technique which has been described by former researchers (Watanabe et al., 2016). All the methane measurements of methane gas were normalized to the standard temperature and pressure (STP: 0 degree, 1atm). In daily operations, pH for influent and effluent and oxidation reduction potential (ORP) for the activated anaerobic sludge was measured by a pH meter (TOADKK, DM-32P, Japan) and ORP meter (TOADKK, RM-30P, Japan).

The removal efficiency of the pollutant calculated is listed below.

$$COD_{RE} = \frac{COD_{inf} - COD_{eff}}{COD_{inf}} \times 100\%$$

$$BOD_{RE} = \frac{BOD_{inf} - BOD_{eff}}{BOD_{inf}} \times 100\%$$

$$SS_{re} = \frac{SS_{inf} - SS_{eff}}{SS_{inf}} \times 100\%$$

where COD_{inf}, COD_{eff} and COD_{RE} are the COD of the influent, the effluent and the removal efficiency of COD, respectively; BOD_{inf}, BOD_{eff} and BOD_{RE} are the BOD of the influent, the effluent and the removal efficiency of BOD, respectively; SS_{inf}, SS_{eff} and SS_{RE} are the SS of the influent, the effluent and the removal efficiency of SS, respectively.

The calculate equation of sludge yield is:

$$Sludge\ \ yield = \frac{\Delta MLVSS}{COD_{rem}} = \frac{(MLVSS_2 - MLVSS_1)V}{t(COD_{inf} - COD_{eff})Q}$$

where COD_{inf}, COD_{eff} and COD_{rem} are the COD of the influent, the effluent and the removal of COD, respectively; $\Delta MLVSS$ is the variation of MLVSS; $MLVSS_2$ and $MLVSS_1$ represent MLVSS value in two different time; V is the reaction volume of AnMBR; Q is the sewage treatment capacity in a certain time.

The calculate equations of biogas production rate and biogas yield is:

$$biogas\ \ production\ \ rate = bpr = \frac{V_{gas}}{Q}$$

$$biogas\ \ yield = \frac{bpr}{COD_{rem}}$$

where V_{gas}, Q, bpr and COD_{rem} are the volume of biogas produced, sewage treatment capacity in a certain time, biogas production rate, and the removal of COD, respectively.

Microbial community structural analysis was carried out on the sludge of each of the series 1 and 2 AnMBR reactors, and the sludge collected during operation. The sludge (named as M0, M1_24, M1_12) which was seeded on the day of operation start ,47 (HRT = 24 hours) and 90 (HRT = 12 hours) days after the start of operation of series 1, and sludge in reactor on day 90 of series 2 (HRT = 24 hours) were collected.

3.2.3 Batch test

The batch test was carried out in 120 mL glass serum bottles using mixed liquor taken from the mini-pilot during HRT 6 hours' continuous operation as the test sludge. The mixed liquor was sampled by sealed sample sterilization bottles to keep the oxygen out. In each serum bottle, 49.0 mL raw mixed liquor and 0.5 mL $Na_2S{\cdot}9H_2O$ solution (250 mg/L as a final concentration in the vial) which was used as the reducing agent by injected into each bottle on purpose of obtain absolutely anaerobic condition was added with a series of sodium hypochlorite concentrations: 0.25, 0.5, 1, 3, 5, 7, 10 g/L. There was no further nutrient solution added due to it enriched 1000 to 2000 mg/L COD in the raw mixed liquor and the real sewage wastewater components used as the nutrient solution makes it more close to reflect out the real specific methanogenic activity (SMA). The total volume of the liquid was fixed to 50 mL by distilled water refilled, thus the headspace was 70 mL. The serum bottles were sealed with rubber stoppers and secured by aluminum crimp. Oxygen in headspace of the bottles was purged with nitrogen gas for 10 minutes. Distilled water or other solutions were also pre-treated by the nitrogen gas. The temperature was controlled around 35 degrees and biogas production and composition was measured every 3 ~ 5 hours according to the biogas volume and expressed as the value at standard condition. The blank sample was conducted in two replicates to ensure its reliability (Nie et al., 2017b).

3.2.4 Off-line membrane cleaning

The membranes were replaced on day 90 and the taken out membranes one was used for off-line membrane cleaning and the fouled matters analysis as it was reported that fouling fraction is widely used to provide a further understanding of the contribution of fouling layers (Ferrero et al., 2012; Kalboussi et al., 2017; Kola et al., 2014). The equipment and structure of the off-line membrane cleaning is shown in figure 3.3. The cleaning is divided as physical cleaning and chemical cleaning by following steps:

<u>Step 1. Wipe the outer surface of fouled membrane carefully by a sponge;</u>

<u>Step 2. Backwashing with 20L distilled water for 2 hours;</u>

<u>Step 3. Soaked in distilled water for another 22 hours;</u>

<u>Step 4. Backwashing with 20L sodium hypochlorite solution (7g/L) for 2</u>

hours;(Metzger et al., 2007; Wu et al., 2008)

Step 5. Soaked in sodium hypochlorite solution (7g/L) for another 22 hours;

Step 6. Backwashing with 20L critic acid (7g/L) for 2 hours;

Step 7. Soaked in critic acid (7g/L) for another 22 hours.

The whole process was controlled temperature at 25 degrees and after every single step the TMP-FLUX test was implemented. The International Union of Pure and Applied Chemistry defines fouling as: the process that results in a decrease in performance of a membrane, caused by the deposition of suspended or dissolved solids on the external membrane surface (surface fouling), on the membrane pores (blocking fouling), or within the membrane pores (blocking fouling as well) (Koros W J et al., 1996). When clean water is filtered, the membrane material is the only resistance caused (*Rm*). The flux is than called the clean water-flux. As a result of the accumulation of particles on the membrane through the filtration of water with a certain level of suspended solids, a cake layer will form on the membrane surface (*Rc*; particles). Therefore, it was found that gel layer rather (*Rg*; biofouling) than cake layer was more easily formed when soluble microbial products content in sludge suspension was relatively high (Hong et al., 2016). Resistance as a consequence of adsorption in or on the membrane is called biofouling (*Ra*). Then if the particles stacked to forming blocked membrane pores, it is called pore plugging (*Rpb*; scaling). The TMP-FLUX test after every step can present the resistance by cake layer (or gel layer), biofouling or scaling which were three main groups of pollutants that can be distinguished from membrane fouling.

3.2.5 Mini-module cleaning

The second taken out membrane was cut and picked out a part of 70 mm with a membrane area of 6 cm^2 for the mini-module chemical cleaning. The mini-module cleaning was proceeded by an experiment equipment shown in figure 3.4 by the following steps:

Mini-step 1. Backwashing with sodium hypochlorite solution (3g/L) for 2 hours;

Mini-step 2. Soaked in sodium hypochlorite solution (3g/L) for another 22 hours;

Mini-step 3. Backwashing with 20L critic acid (20g/L) for 2 hours;

Mini-step 4. Soaked in critic acid (20g/L) for another 22 hours.

The whole process was under room temperature and after every single step the TMP-FLUX test was implemented. SEM analysis was experimented before and after the mini-module cleaning by a SEM instrument (SU-1500, HITACHI, Japan).

3.2.6 TMP-FLUX test

TMP-FLUX test used the same equipment of off-line membrane cleaning or mini-module cleaning. A combination of TMP statistics was recorded by applied different FLUX ranged from 0.1 m/d to 0.5 m/d. The temperature was controlled along the tests.

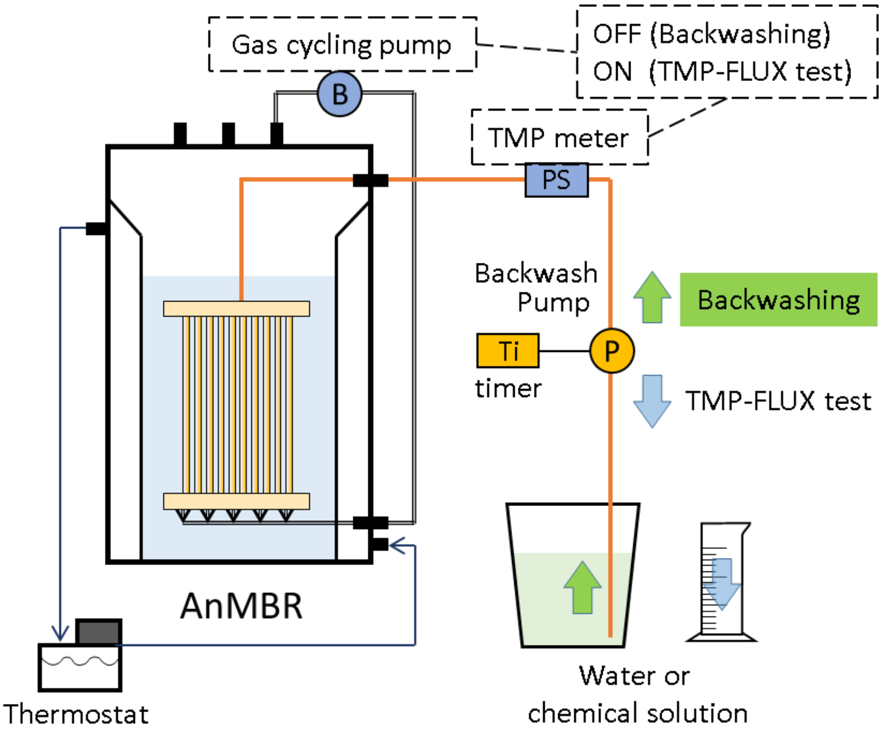


Fig. 3.3 Off-line membrane cleaning equipment.

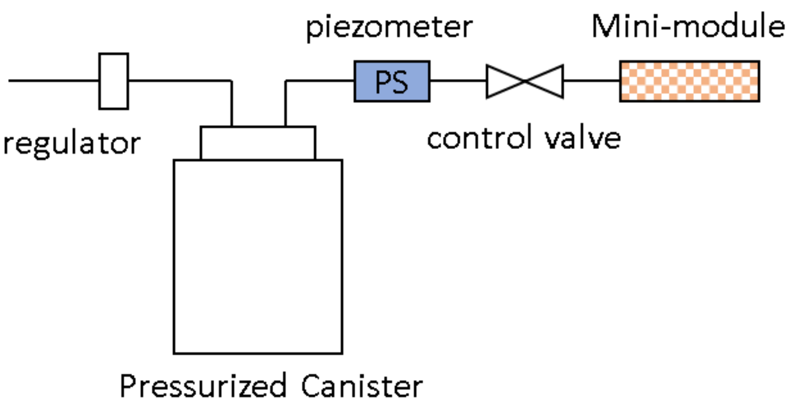


Fig. 3.4 Mini-module cleaning equipment.

3.2.7 16S rRNA gene analysis

DNA extraction from the collected sludge followed by the Manufacture's instruction of ISOIL for Beads Beating kit (Nippon Gene), except prolonging the incubation time to 2 hours. For PCR, 341F (5'- CCT AYG GGR BGC ASC AG -3') and 806R-mix (a mixture of 806R (5'- GGA CTA CHV GGG THT CTA AT -3') and 806R-P (5'- GGA CTA CCA GGG TAT CTA AG-3' with the ratio of 30:1) targeting the prokaryotic 16S rRNA gene were used. A library was prepared by a three-step PCR. And 25 amplification cycles consisted the PCR reactions (at 94˚C for 5 sec, 50˚C for 30 sec, and 68˚C for 10 sec), followed by a final extension step at 68˚C for 7 min by using C1000 Touch™ Thermal Cycler (Bio-Rad Laboratories Inc., Japan). After finishing the preparation of the library, MiSeq reagent Kit v3 (Illumina) was used to do the sequencing. Sequence analysis was performed with QIIME 1.8.0 software. For the Operational Taxonomy Unit (OTU) grouped with 97% as the threshold, assignment was performed using the Greengene database, and analysis of the proportion of 16S rRNA gene in prokaryote and analysis of diversity were performed.

3.3 Results and Discussion

3.3.1 Annual data collection of influent raw sewage

The continues long-term operated experiments recorded temperature and pH values of the influent manual every day and COD and BOD in influent was measured 2 or 3 samples per week. These results and data during the 600 days has been organized into figures 3.5. According to the collected data, the sewage temperature in influent was presented lower than 10 degrees in winter while a little bit higher than 25 degrees in summer. Raw sewage pH value was ranged from 6.9 to 7.5 in the seasons operation. COD and BOD in influent was ranged from 200 to 600 and 90 to 280, respectively. SS also measured several times every season and influent SS was in the range of about 100 to 250mg/L. The influent sewage parameters were instability during the long-term operation and even in the same day, that is one of the big difference between real sewage wastewater and the synthetic sewage wastewater.

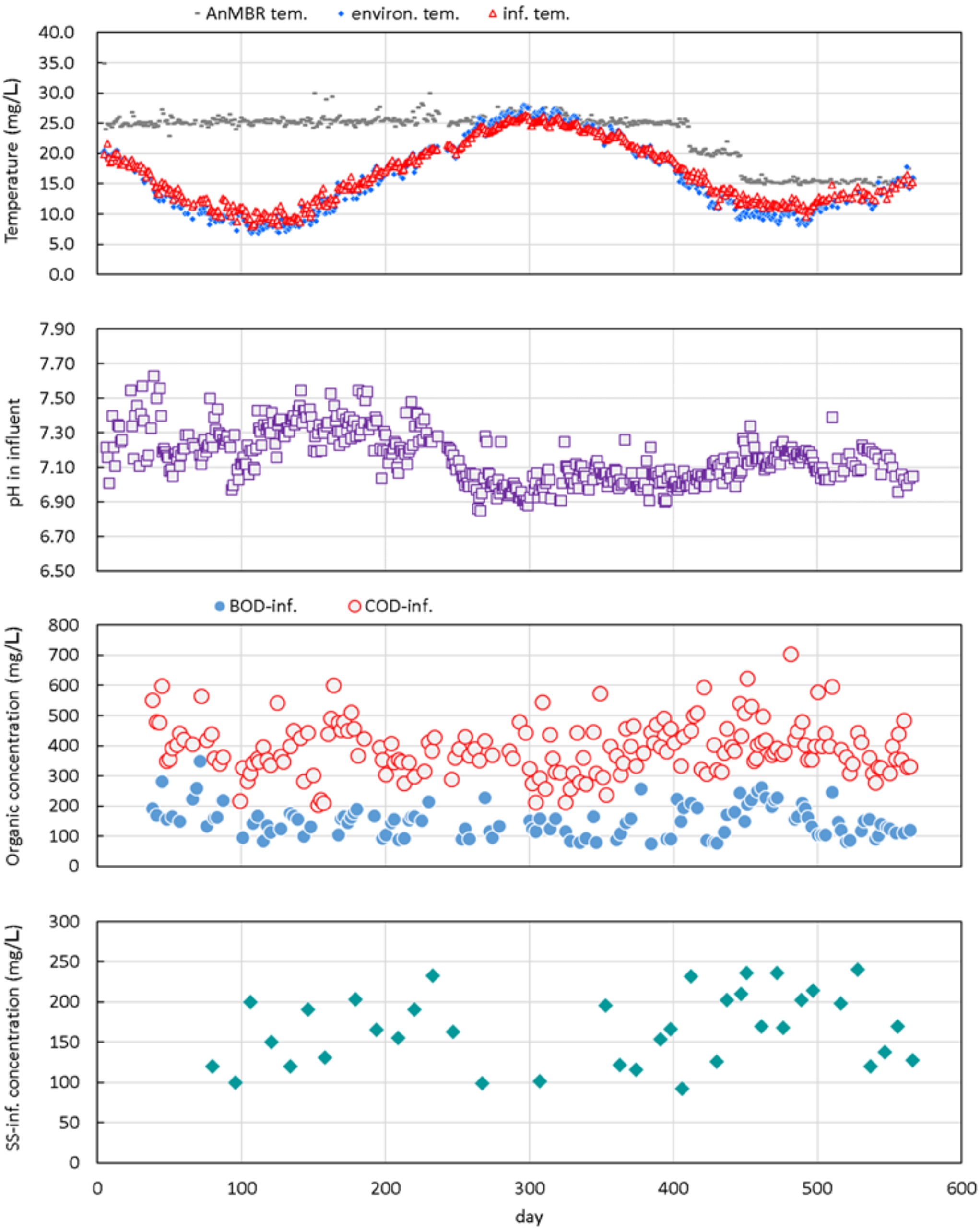


Fig. 3.5 Annual data collection results of influent raw sewage.

3.3.2 Pollutant removal efficiency

Figure 3.6 shows COD / BOD in influent and effluent and the removal efficiency. The figure shows that, for both AnMBRs, COD contained in effluent was decreased as time went on in the beginning period for microbial acclimation. As a result, the removal efficiency for COD and BOD also presented a trend of increasing and finally reached around 90% for COD and higher than 90% for BOD in both AnMBRs. It sent a signal that using the real sewage to start up AnMBRs has turn to a success. And a strategy of glucose and methanol additional with a total COD concentration as low as 400 mg/L, also made the progress achieved faster.

Completed the microbial acclimation, the removal efficiency for sewage was also presented well performance for AnMBRs through with HRT 24, 24.4 and 12 hours (COD around 90% and BOD higher than 90%). Most of the effluent COD were less than 50mg/L and effluent BOD were less than 15mg/L, which meant the organic matters were highly degraded by both AnMBRs. Compare with the studies based on synthetic sewage (Chen et al., 2017b; Watanabe et al., 2014), the treatment performance also closes to those used synthetic sewage. The results confirmed that AnMBR treating the real sewage can achieve the a high organic removal efficiency and qualified effluent with low COD and BOD concentration at room temperature 25°C.

According to the long-tern operated data, the average value for each phase was calculated out for both AnMBRs and listed in table 3.3. For both AnMBR, SS in influent and effluent was measured for several times in each HRT condition, the removal efficiency all achieved 100% since there was no SS determined in effluent. The result identifies with mechanism of membrane filtration due to the pore size of membrane was 0.4μm in M1 and 0.05μm in M2, both less than 0.45μm, the measurement filter diameter for SS used in standard method. And the comparison for COD / BOD removal efficiency, there was no big difference between M1 and M2, though the removal efficiency for M1 was higher than M2 a bit during HRT12 on COD and BOD, HRT14.4 on COD. The results were also adopted unanimously with previous studies treating synthetic sewage by 0.2μm pore size plate membrane module (Chen et al, 2017; Watanabe et al, 2015). That comes out a conclusion that the organic removal efficiency in sewage treatment or sewage purification performance has no effect with the pore size of the membrane if the pore size less than 0.4μm.

Table 3.3 The performance of the pollutant removal and the bio-gas production

	HRT	**COD**		**BOD**		**SS**	
MBR No.	/h	**EFF*** mg/L	**RE**** %	**EFF** mg/L	**RE** %	**EFF** mg/L	**RE** %
M1	24	53.6	88.9	16.1	91.4	0	100.0
	14.4	39.0	89.0	7.9	93.8	0	100.0
	12	42.1	90.0	11.9	93.9	0	100.0
M2	24	47.1	88.9	12	94.0	0	100.0
	14.4	42.8	88.9	7.6	92.8	0	100.0
	12	45.6	88.7	11.7	92.4	0	100.0

*EFF - pollutant concentration in the effluent;

**RE - removal efficiency;

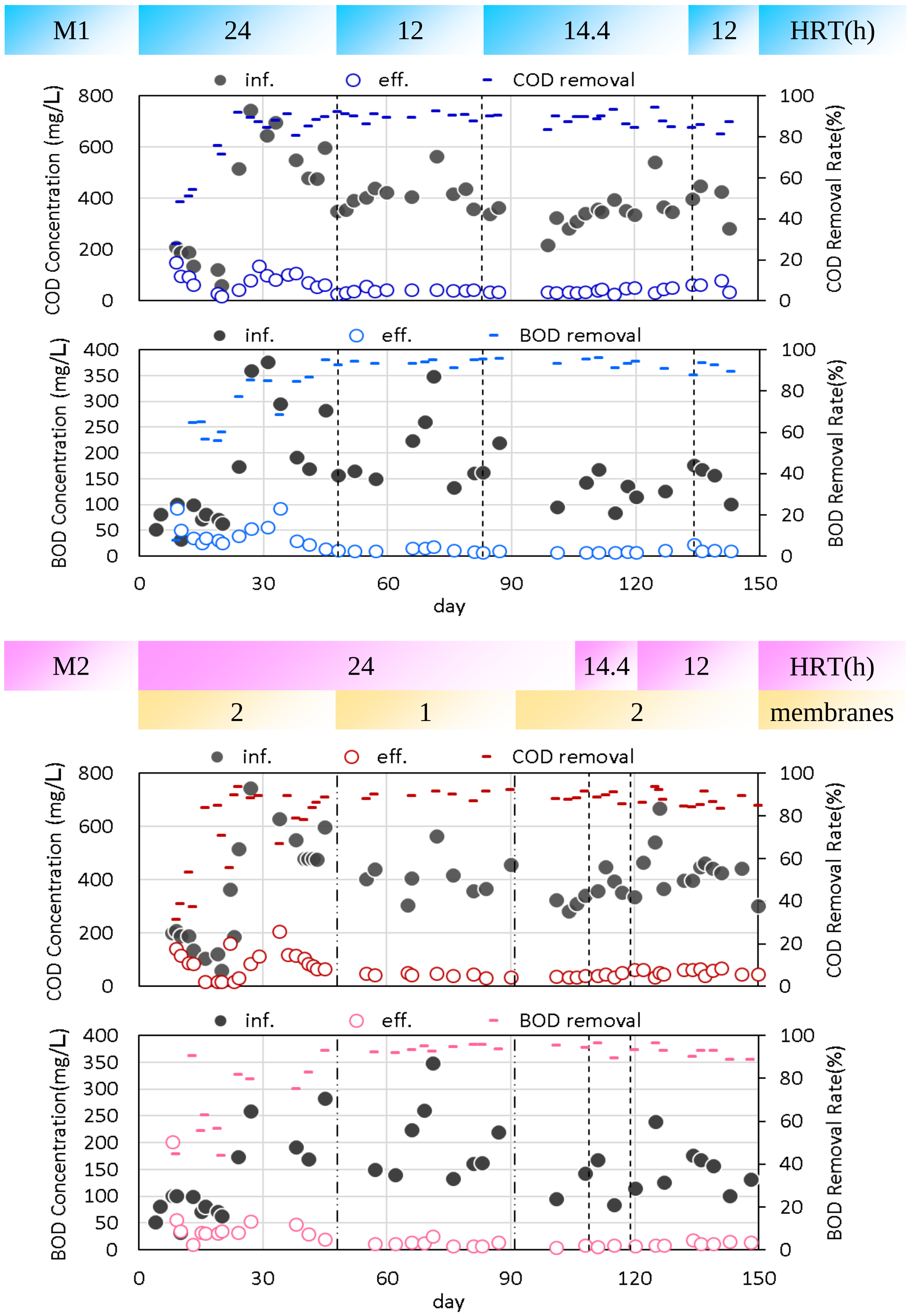


Fig. 3.6 COD and BOD of influent and effluent and the removal efficiency.

3.3.3 Biogas production effect

The biogas production and composition of CH_4, CO_2, N_2 in the biogas produced is shown in figure 3.7. In M1, daily biogas production rate (sludge yield) was ranged from 0.04 to 0.08 L-gas/L-water during the HRT24 condition after the microbial acclimation, then gradually increased as the number of days increased. The average daily biogas production rate is shown in figure 3.8 shows it clearly that the biogas production rate was achieved around 0.08 L-gas/L-water in HRT 14.4 and 12 hours. The biogas yield was calculated as 0.23 L-gas/g-COD_{rem} in M1 which was even higher for some cases treating high strength wastewater, for example, a biogas yield of 0.04 L-gas/g-COD_{rem} (HRT 12 hours) and 0.12 L-gas/g-COD_{rem} (HRT 24 hours) was achieved for treating pharmaceutical wastewater (COD 4250~5129mg/L) by AnMBR in 40°C (Chen et al., 2018). In M2, almost the same but a little higher biogas production was achieved compare with M1, 0.076 L-gas/L-water (24hours HRT), 0.094 L-gas/L-water (14.4hours HRT), 0.086 L-gas/L-water (12hours HRT) and the biogas yield was 0.22 L-gas/g-COD_{rem}, 0.27 L-gas/g-COD_{rem}, 0.25 L-gas/g-COD_{rem}. According to the gas composition of CH_4, CO_2, N_2 in the produced biogas (figure 3.7), an average of 75.10% methane content was obtained in M1 (ranged from 70.02%~77.58% in stable period) and almost the same result of 76.11% in M2 was obtained (ranged from 72.43%~79.63% in stable period). This result also matches with the previous study used synthetic sewage that achieved an up to 80% methane content in biogas produced. Normally, the CH_4 and CO_2 content in biogas produced by methane fermentation during anaerobic digestion was known as 60% and 40% by the chemical mechanisms and also had been reported by a lot of researchers in their own long-term experiments. While the higher CH_4 content and lower CO_2 content presented in cases of treating sewage is considered to be caused by the short HRT. Long

HRTs resulted a less influent and effluent so that less produced CO_2 dissolved in water. The same AnMBR for treating the synthetic sewage was also applied for thermophilic high-solids co-digestion of coffee processing wastewater, CH_4 and CO_2 content in biogas produced was around 60% and 40% respectively in HRT conditions of 50 days to 5 days. Since the sewage has the characters of massive volume and low organic strength, the HRTs always need to be as short as 6~12 hours to meet the treatment requirements. That resulted biogas dissolved in water and discharged with the treated water permeated as effluent then more biogas dissolved in the new water system in AnMBR consisted with the new influent sewage. So far in the results, it can tell us that CO_2 dissolved more than CH_4 mainly because of carbonates can provide alkalinity (Brandt et al., 2017) that happened to be needed in fermentation process. The massive volume sewage in short HRT also lead to nitrogen gas was detected 15 ~ 20% in effluent, nonetheless, resulted methane gas content was about 72 ~ 80% which provides a higher purity of methane gas that can be reused more efficiently in heat or power generation. Considering the methane content and the methane yield can be calculated out: 0.17 L-CH_4/g-COD_{rem} (M1, HRT12h) and 0.20 L-CH_4/g-COD_{rem}, higher than a study treated municipal wastewater (COD 1729±914mg/L) by a AnSMBR in HRT 8~10h, 8~33°C (Peña et al., 2019). The results showed that AnMBRs were performed well for treating low organic strength real sewage and the effect was even better than high strength on the aspect of biogas reuse or energy recovery potential.

The results indicated that AnMBR was performed well for treating low organic strength real sewage and with a good energy recovery potential. In addition, the performance of biogas production was not presented big difference between the two different pore size membranes.

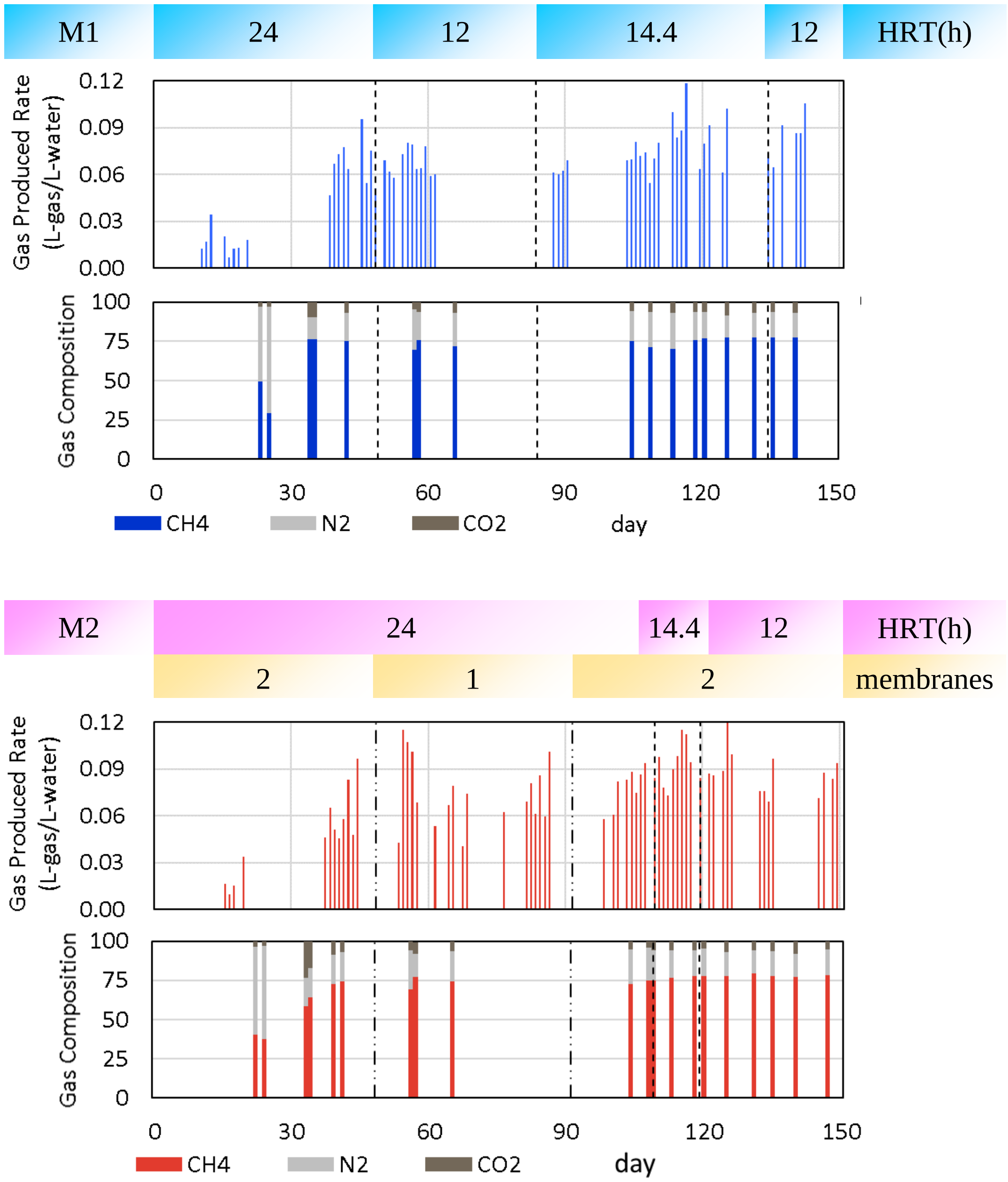


Fig. 3.7 Biogas production rate and the composition.

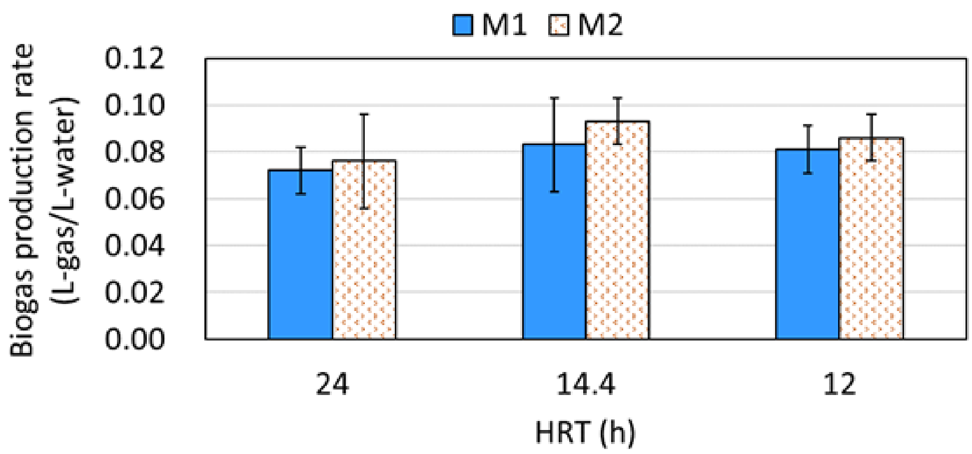


Fig. 3.8 Average pollutant removal efficiency and biogas production.

3.3.4 Filtration performance and the optimal filtering conditions

M1 was launched on FLUX 0.137 m/d at a HRT condition of 24 hours. Since transmembrane pressure (TMP) was not increased at all in the period of microbial acclimation and for another 11 days' operation by real sewage fed only under a HRT condition of 24 hours, the FLUX was raised up to 0.274 m/d by shortened HRT to 12h (Figure 3.9). Afterwards, the appearance of TMP increasing was brought out and the membrane fouling was found after operated for 13 days, then tried several times to stop and restart the membrane filtration but did not work efficaciously. Thereafter, the membranes still showed available as of the HRT was extend to 14.4h by the change of filtration mode to less strength permeate to 2 minutes permeate with every 1 minute relax. The membranes were replaced on day 90 because of TMP still increased obviously in HRT 14.4h. The new membranes were capable of work about a week later. TMP was not increased quickly during the started 2 weeks' operation, but suddenly was unable to control below 30kPa which was defined as a maximum operation for the membrane by the membrane producers.

In order to break out of the filtration impasse, before the FLUX was characterized as what it could be, a parameter of cross-flow gas velocity (CFGV) was considered to be redesigned. CFGV is decided (equation below) by the cycling of the biogas produced in the process as well as the cross-sectional area of the membrane module itself.

$$CFGV = \frac{Q_p}{A_m}$$

Where *CFGV* is the cross-flow gas velocity (m/h), Q_p is the operating capacity of the biogas cycling pumps (m^3/h), and A_m is the cross-sectional area of the membrane module (m^2).

Researches in the past barely had focus on CFGV for any type of discussion, though the biogas cycling by a gas diffusers was widely applied since it is known as a fouling control method in the bio-membrane reactors due to the shear force especially in the cross flow modules. TMP - FLUX tests result (figure 3.10) in different CFGV condition showed that FLUX was positively correlated with the CFGV condition (figure 3.11) had confirmed the shear force by the gas diffusers is useful in the cross flow membrane modules. TMP - FLUX tests also implemented in the conditions of different temperature and MLSS inside the AnMBR. A positive correlation (figure 3.12) was obtained between FLUX and temperature indicated that the operation temperature can also increase the filtration ability of membrane considered to be caused by the change of activated anaerobic sludge property. MLSS effect was less to the filtration in the TMP - FLUX test MLSS conditions when FLUX was below 0.3 m/d (figure 3.10) while a significant negative correlation was drawn out as figure 3.13 between FLUX and the condition of MLSS due to a bigger concentration of mixed liquor might be easier to block the membrane surface and increase the filtration resistance. Those results can provide a reference value for deciding the conditions when operating a AnMBR to the real sewage wastewaters. Thus, the optimal CFGV condition in this long-term experiment is considered as in the range of 80 m/h to 120 m/h because of the effect of CFGV was reduced when higher than 120 m/h even though more energy was needed for the gas diffuser pumps. Furthermore, the proven relations between FLUX and CFGV, temperature, MLSS also can provide a temporary measure if the membrane was fouled or more FLUX was needed since comparing with the cost and restart-up time it takes for membrane module reconstructed, changing the operation parameters was considered to be a better option.

The problem faced in M1 was solved by increasing the CFGV value. CFGV was increased from 70 m/h to 119 m/h on day 135 and FLUX was raised up to 0.274 m/d again by shortened HRT to 12 hours. According to the long-term operation figure 3.9, it is clear that stable low TMP operation for FLUX 0.274 m/d also achieved in M1 by increasing CFGV.

On the side of M2, TMP was remained at a very low status as FLUX was 0.074 m/d while only 6 days operated till the membrane fouled phenomenon presented since FLUX was raised to 0.148 m/d by disable one of the membranes. While during day 119 to day 134, the membrane filtration was achieved low level TMP operation though visible growth showed out since FLUX was raised to 0.148 again by shortened HRT into 12 hours since the CFGV also increased to 119 m/h the same as M1.

Compare with the two AnMBRs with different membrane modules, the FLUX was achieved as 0.274 m/d operation lasted for 2 weeks in M1 while just 0.148 m/d in M2 continued only 6 days in the same CFGV condition of 70 m/h. After increased the CFGV to 119 m/h, a low TMP was achieved in FLUX 0.274 m/d in M1 but M2 only obtained FLUX 0.148 m/d in the same condition. That indicated a bigger suction pressure is needed for the smaller pore size membrane to implement the filtering in the same FLUX under the same conditions. So that, much more energy is required to the filtration pumps for the smaller pore size membrane modules.

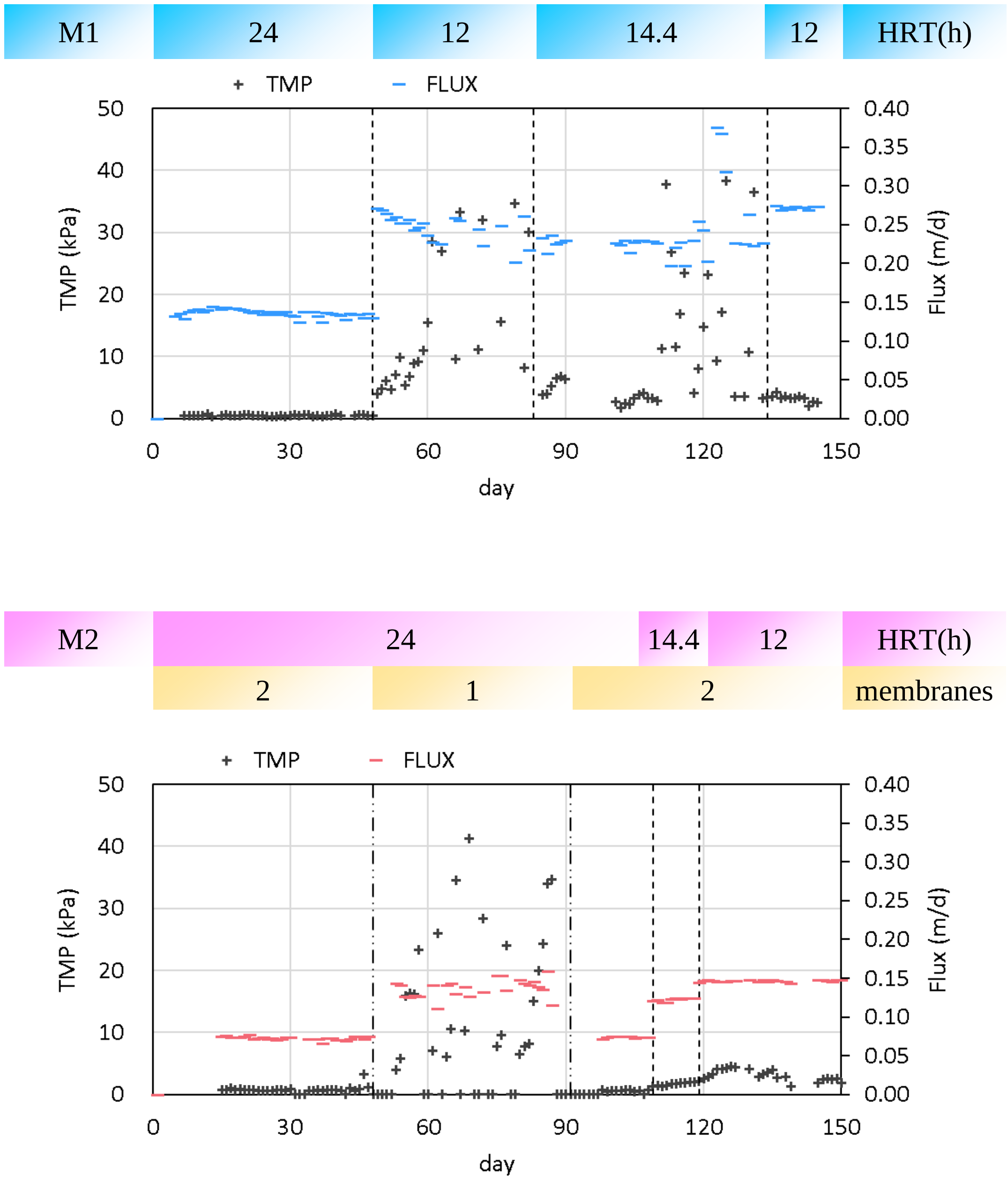


Fig. 3.9 TMP – FLUX record during the long-term operation.

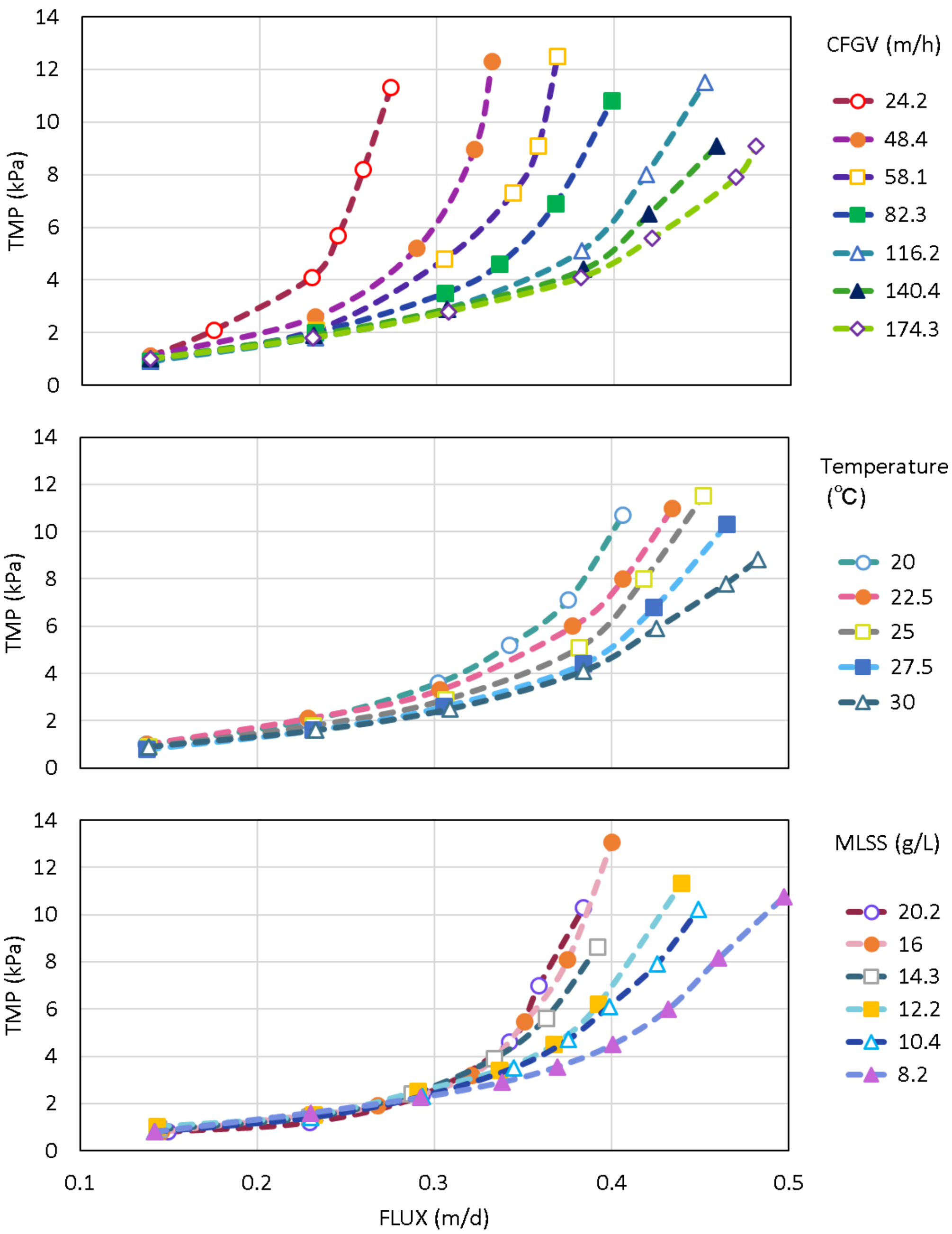


Fig. 3.10 TMP - FLUX tests in different CFGV, temperature and MLSS conditions.

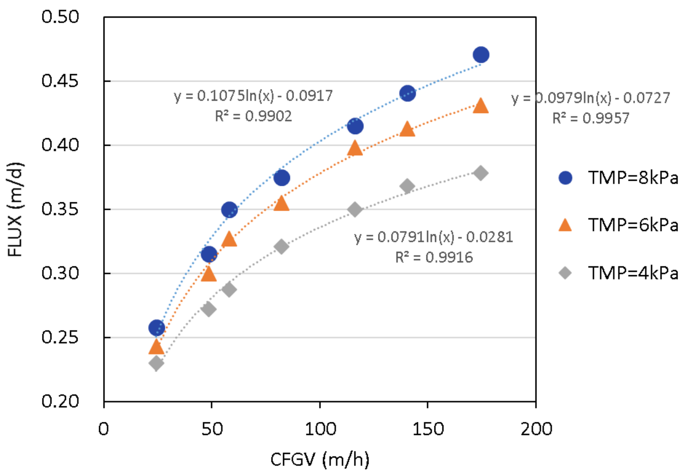


Fig. 3.11 The relations between FLUX and CFGV.

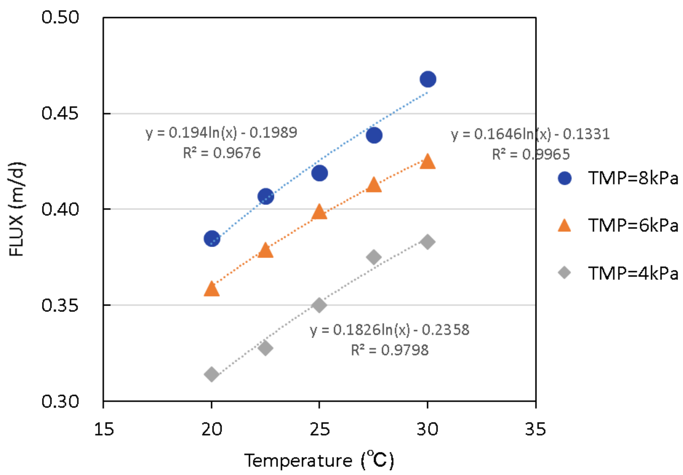


Fig. 3.12 The relations between FLUX and temperature.

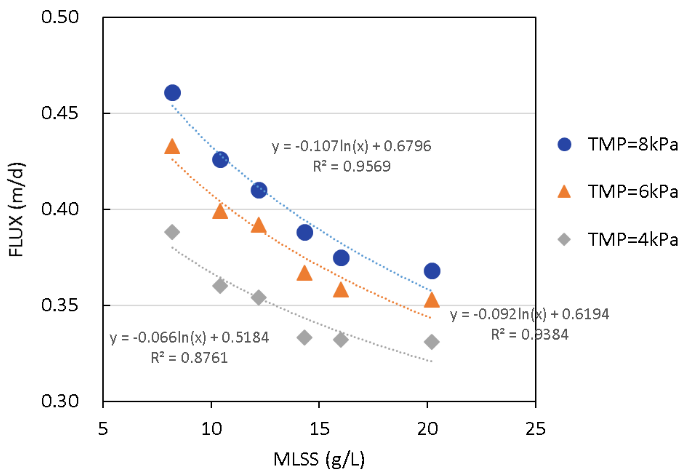


Fig. 3.13 The relations between FLUX and MLSS.

3.3.5 Reaction indexes and sludge character

Figure 3.14 shows pH and ORP for each AnMBR in the long-term experiment. For both AnMBRs, pH in effluent was able to be controlled mostly above 6.8 exactly more than the pH of methane deactivation of 6.5 in the methane fermentation process. On the other hand, ORP for the anaerobic sludge retained inside the AnMBRs was also determined in the daily operations. ORP under -300 is reported as a significant parameter to creating the anaerobic environment. According to the result, ORP in this study was mostly ranged from -300 to -350 mV in the two AnMBRs indicated that reactors could ensure a good anaerobic environment for the methane fermentation proceeding.

Figure 3.15 shows the properties of the mixed liquor. The MLSS was ranged from 6 ~ 10 g/L and MLVSS 5 ~ 8 g/L in M1. The same situation happened in M2 which MLSS 6 ~ 8.5 g/L and MLVSS 5 ~ 7 g/L. The MLSS/MLVSS ratio kept 0.82±0.1 in M1 and 0.84±0.2 in M2, which means few organic or inorganic solid matters were continuously accumulated in the mixed liquor. The average sludge yield in the long-term experiment was determined as 0.09 g-VSS/g-COD_{rem}, much lower than the sludge yield in a range of 0.25~0.4 g-VSS/g-COD_{rem} in a conventional aerobic activated sludge process (Huang et al., 2001). Thus AnMBRs are confirmed to be characterized by little production of excess sludge and small demand of COD for biomass multiplication in addition to a high efficiency of organic removal and biogas production compare with the previous research based on the synthetic sewage (Chen et al., 2017c).

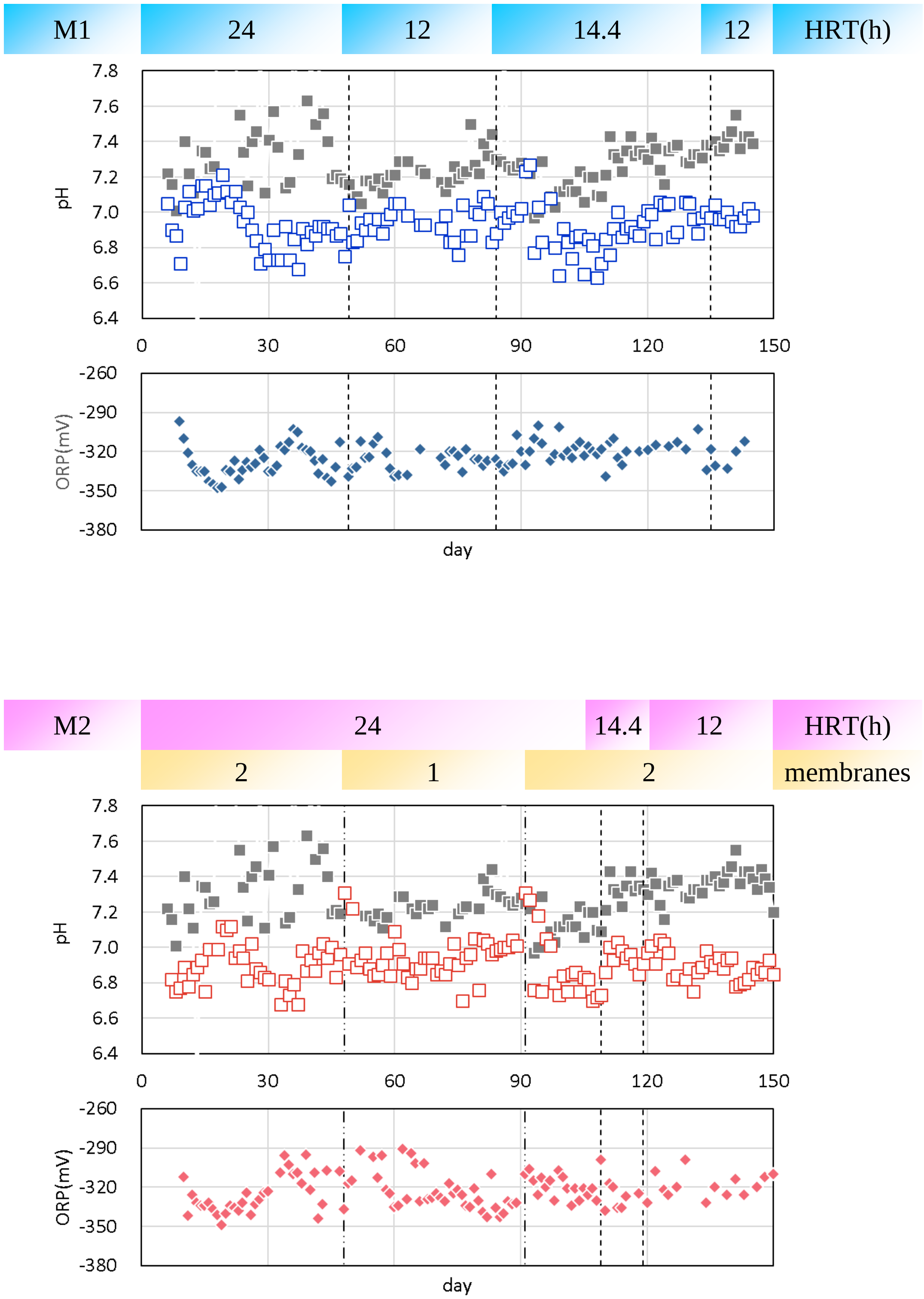


Fig. 3.14 Influent and effluent pH and ORP in AnMBRs.

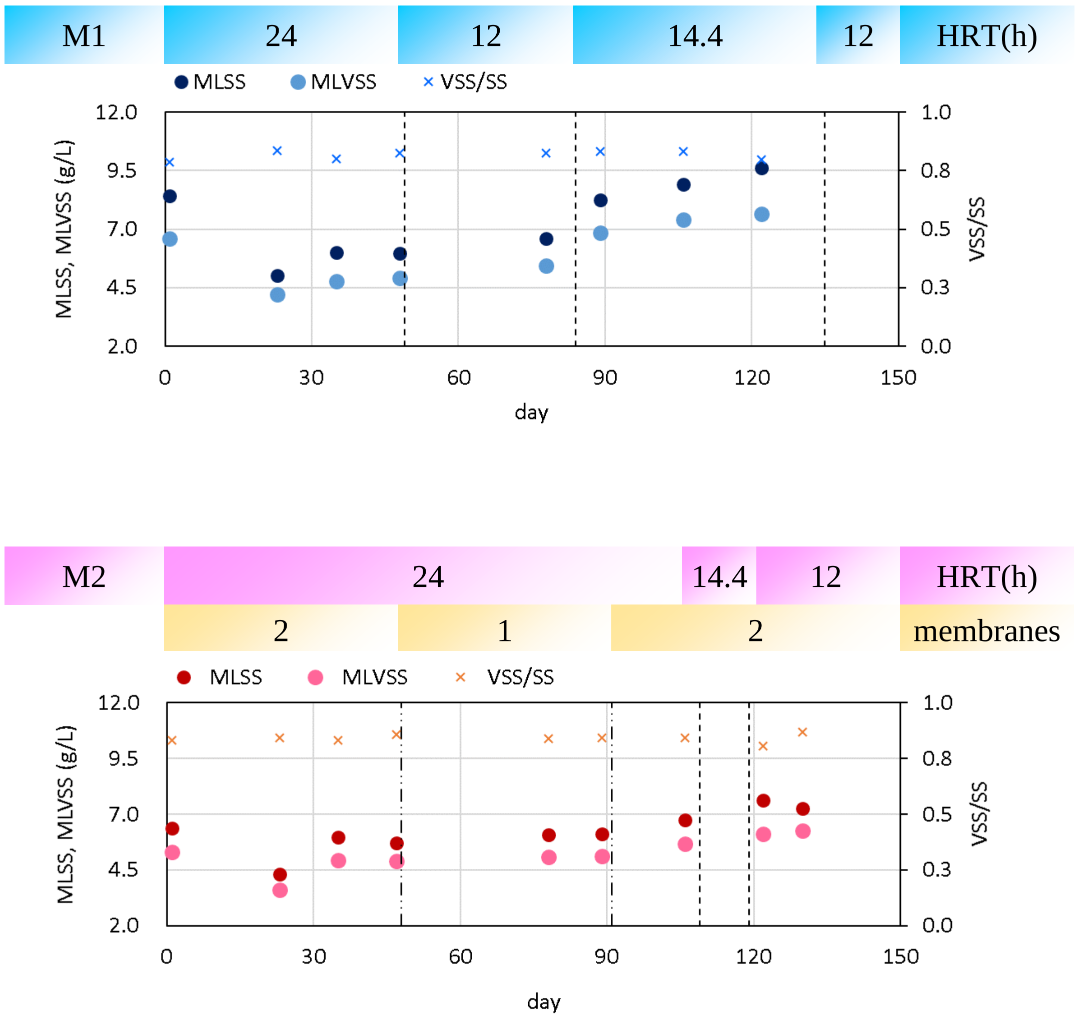


Fig. 3.15 MLSS/MLVSS for each AnMBR.

3.3.6 Batch test results

Figure 3.16 shows the methane gas production in the batch test implemented with different sodium hypochlorite concentration. The result is quite clear that the biogas production was close between the blank control and sodium hypochlorite concentrations under 1 g/L. Biogas production was apparently affected in the 3 g/L sodium hypochlorite solution serum bottle. Then, it was almost generated no biogas in the bottles that sodium hypochlorite solution concentration above 5 g/L.

Specific methanogenic activity (SMA) was calculated by the batch test results shown in figure 3.17. SMA shown a reduction as long as the sodium hypochlorite solution concentration increased. This result is different with previous studies that widely used sodium hypochlorite solution concentrations ranged from 0.25 to 7 g/L to implement the online cleaning in AnMBR (Kalboussi et al., 2017; Wu et al., 2008), which is considered to be caused by the differences of sludge properties in treating the sewage wastewaters with other anaerobic digestion sludge.

Combine with the results above, it can be draw out that sodium hypochlorite solution around or higher than 3g/L might cause microbial activity cannot be restored in a short time, so that less than 1 g/L is an acceptable concentration for the microorganisms in situation of treating the sewage wastewater. According to the online membrane cleaning steps, 1.0 L sodium hypochlorite solution pumped into a 20.0 L AnMBR resulted a 20 times dilution. So, sodium hypochlorite solution concentration used for backwashing in this study was determined as 7 g/L to ensure a high membrane cleaning performance during the backwashing via a relatively high concentration solution with result of only 0.35g/L sodium hypochlorite remained inside the AnMBR after the online membrane cleaning.

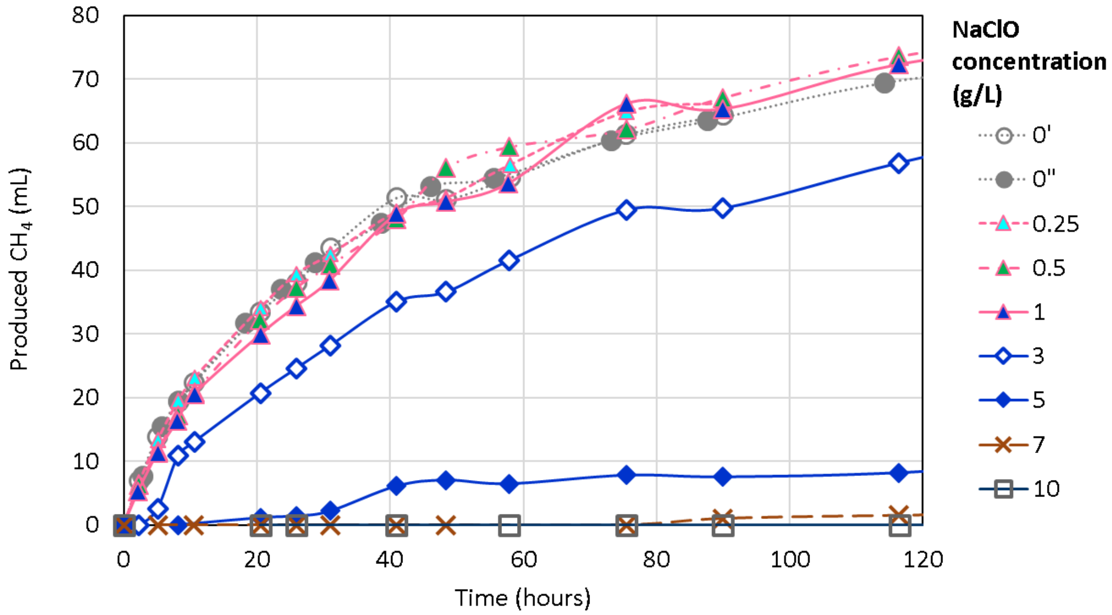


Fig. 3.16 Methane gas produced during the activity test.

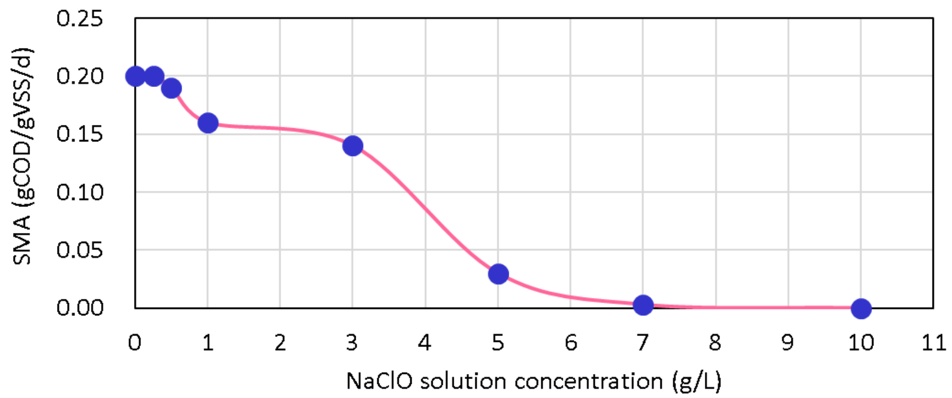


Fig.3.17 Specific methanogenic activity in different NaClO solution.

3.3.7 Membrane cleaning Analysis

The membrane presented macroscopic changes during each step of the membrane cleaning works (Figure 3.18 shows the photographs taken for compare with the un-used membrane). Then in figure 3.19, it shows the distilled water filtration experiment after each step of the membrane cleaning works. Compare with un-used membranes performance, figure 3.20 can be calculated by the slopes and shown as figure 3.19, it indicated that 46% fouled matters was easily wiped out by sponge on the outer surface of the membrane which is considered to be the surface fouling while it is hard to analysis the ratio of cake layer or gel layer based on available data, 7% was removed by the clean water backwash process which is considered as blocking fouling (pore plugging), then 37% was removed by the chemical cleaning process which also is considered as blocking fouling (biofouling). Among the chemical cleaning procedure, all of the pollutant was removed by sodium hypochlorite solution indicated that the blocking fouling formed in this experiment consisted only by organic matters as the sodium hypochlorite solution is used for removing organic matters and citric acid is used for removing inorganic matters. Finally, the off-line cleaning achieved about 90% removable from the fouled membrane.

Figure 3.21 shows the membrane restoration efficiency after each cleaning step compared with the un-used new membrane. The result presented that around 80% of restoration efficiency was achieved via the off-line membrane filtration and the rest 20% caused by the 10% un-removed fouled matter showed in figure 3.20 was considered to be the nonuniform transit through membrane pores during the backwashing. For that reason, the membrane restoration efficiency obtained a 100% as in mini-module cleaning work (figure 3.22). That was also verified by the SEM result shown in figure 3.23.

The results indicated that surface and blocking fouling was half and half caused mainly by the organics rather than inorganics in sewage treatment. Off-line membrane cleaning can achieve a high restoration efficiency as it obtained 80% restoration in this experiment and it presented a potential of 100% restoration according to the mini-module cleaning.

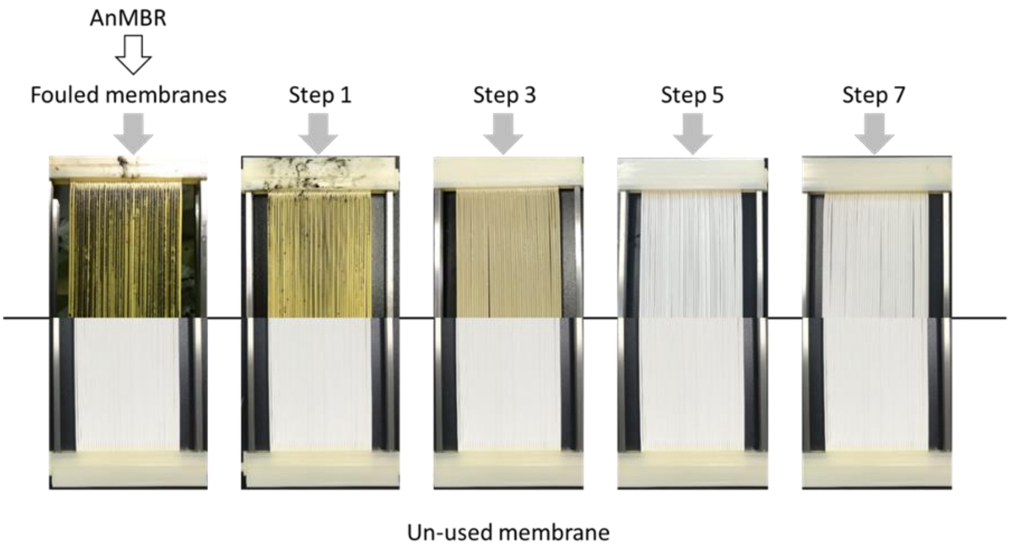


Fig. 3.18 Photographs taken after off-line membrane cleaning steps.

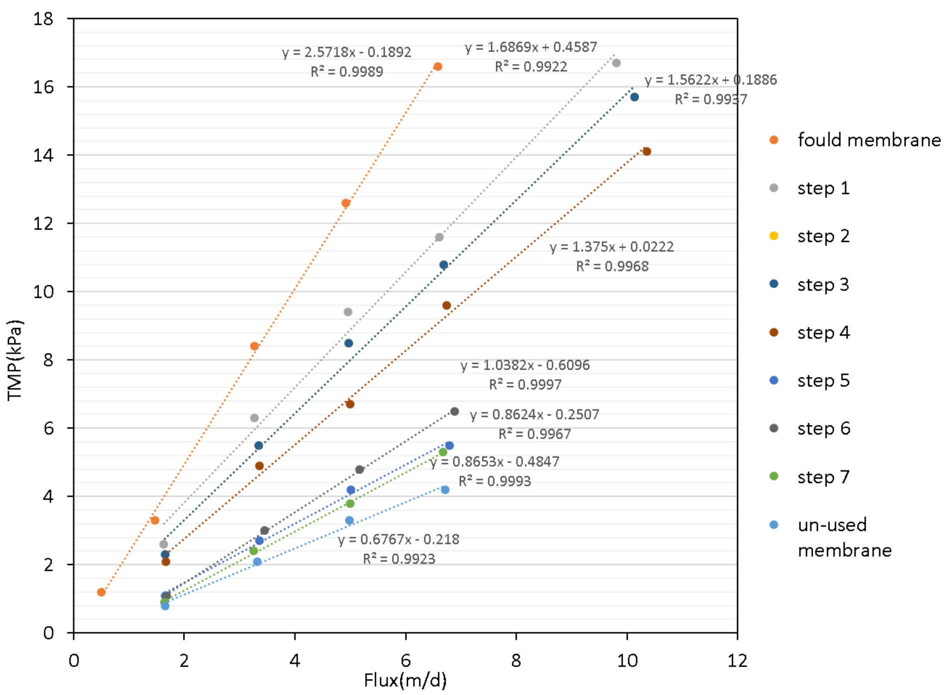


Fig. 3.19 Original TMP-FLUX data of off-line cleaning.

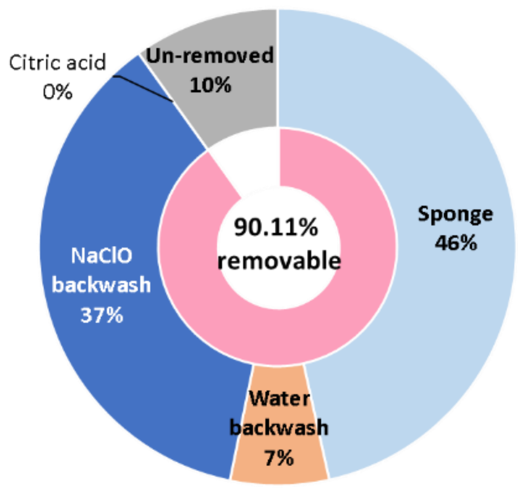


Fig. 3.20 Fouled removable analysis via membrane off-line cleaning.

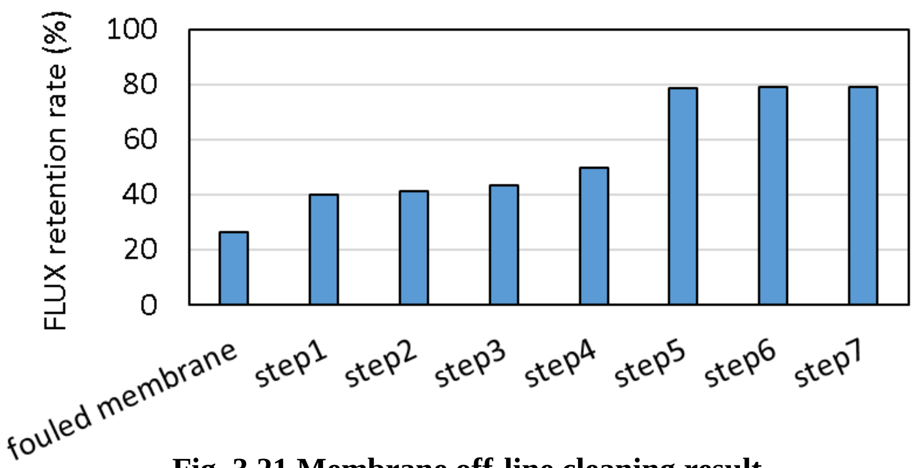


Fig. 3.21 Membrane off-line cleaning result.

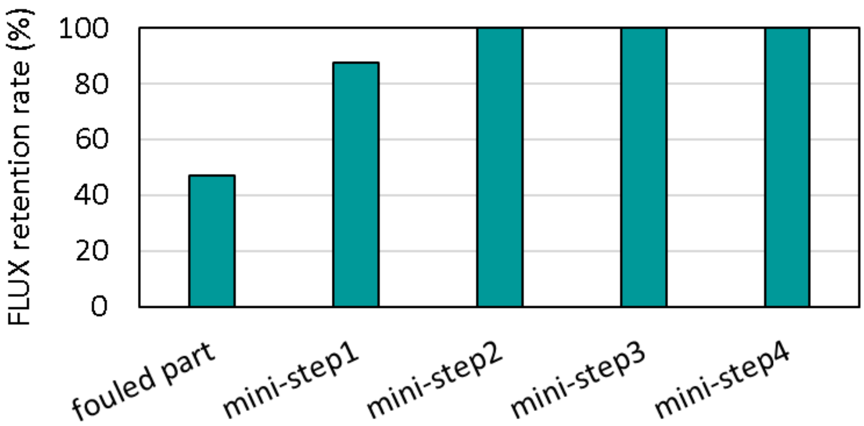
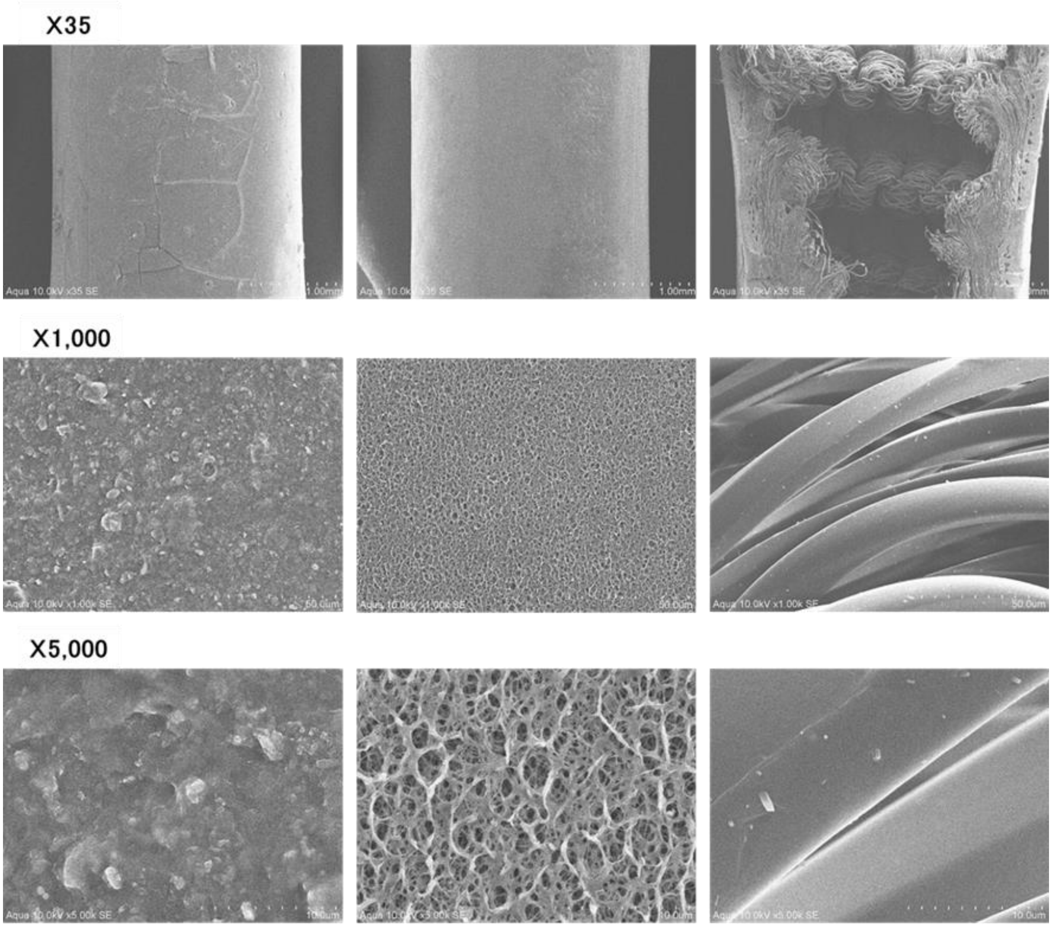


Fig. 3.22 Mini membrane module cleaning result.



Uncleaned membrane Outer surface | **Cleaned membrane Outer surface** | **Cleaned membrane Inner surface**

Fig. 3.23 SEM results of outer membrane surface as before and after cleaning.

3.3.8 Analysis of microbial structure

A) Microbial diversity in samples

The number of sequences obtained from each sample was about 60,000 to 80,000. The sampling depth was set to 60,000 in order to prevent variation due to sequencing depth when comparing samples. Table 3.4 shows the number of OTUs, the OTU estimator (Chao 1), and the diversity index (Shannon). All of the three index decreased after domestication, which suggested that a specific group adaptable to the AnMBR environment would be dominant with the stabilization of the treatment.

Table 3.4 Alpha_diversity of sludge sample in startup period

Sample	OTU	Chao1	Shannon
M0	2964	6066	7.865
M1_24	2375	4297	6.910
M1_12	2167	4008	6.185
M2_24	1879	3575	5.438

B) Characteristics and functions of methanogens adapted in startup period

Sequencing reads assigned as Archaea were 2381, 3403, 4184 and 2552, respectively in the four sludge samples. Figure 3.24 showed the major methanogen diversity in genera level of archaea. Seed sample contained the most kinds of methanogens comparing with the samples taken from AnMBR reactor, which suggested that specific methanogenic groups adapted by AnMBR environment. Those 8 genera consisted over 95% of the archaea sequencing reads in M1_24, M1_12, and M2_24 samples. *Methanoculleus*, which belonged to hydro-genotropic methanogens, was only detected in seed sample while presented absence during the long-term operation (Maria, 2017). *Methanosaeta* was the dominant genus of methanogens that use acetate as electron donors to produce methane in samples taken from reactor, and the result was similar with other studies treating sewage under anaerobic condition (Kong et al., 2018). *Methanobacterium* showed an 8.15% relative abundance of methanogens in M2, while only 1.32% presented in sample M1_24.

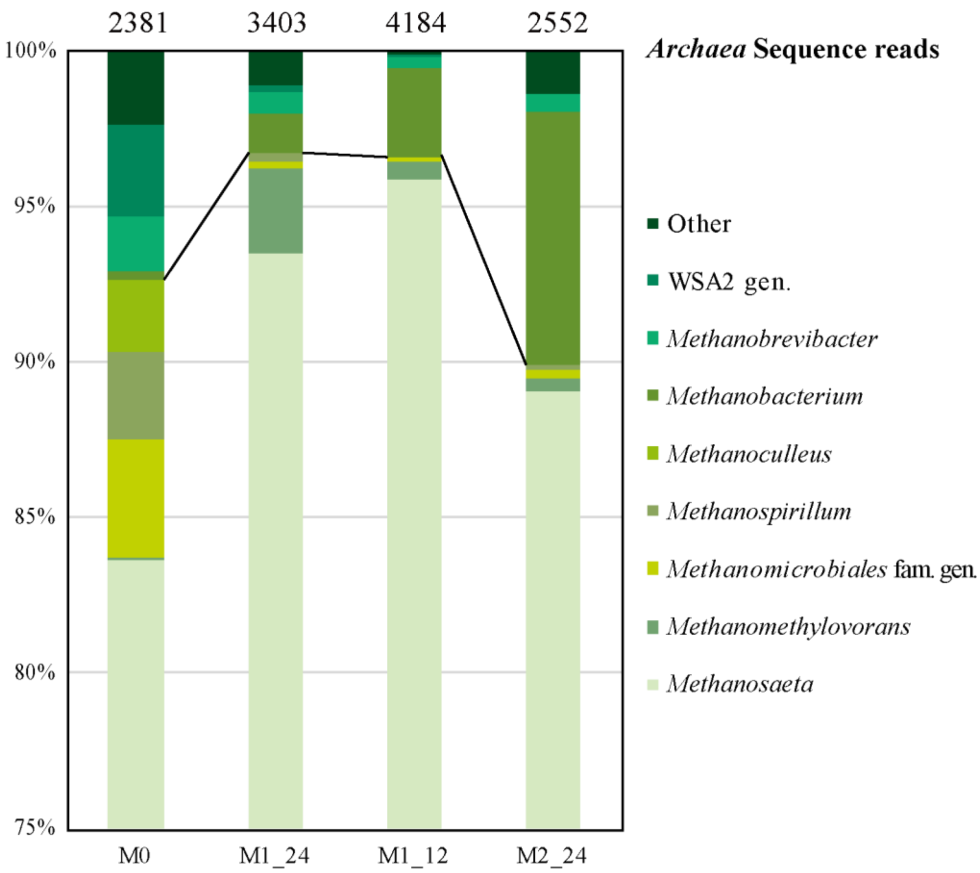


Fig. 3.24 Major methanogens in sludge samples

C) <u>Characteristics and functions of methanogens adapted in startup period</u>

It is known that the phyla *Chloroflexi*, *Firmicutes*, *Proteobacteria*, *Bacteroidetes*, *Actinobacteria*, *Synergistetes*, *Sirochachaetes*, which were known to be dominant in the general anaerobic digester. Those phyla were also dominant in the AnMBR.

Figure 3.25 showed dominant bacterial members in AnMBR reactor. The relative abundance of Order *Anaerolineales*, *Bacteroidales*, *Clostridiales* and *Lactobacillales* consisted over 65% in samples taken from AnMBR reactor. Among the Firmicutes, the groups belonging to the order *Lactobacillales* and *Clostridiales* are particularly dominant, suggesting that these groups play a role in acid production and acetic acid production. *Clostridiales* existed the most in the sample M2_24 than other samples, owning a 39.8% relative abundance of bacterial members. Order *Anaerolineales* (over 20% in bacterial members) belonged to Chloroflexi showed its most important position in M1 and M2 samples. Members of *Anaerolineales* are known connected to ferment sugars and they play an important role in degradation of a variety of carbohydrates in anaerobic digesters (Azman et al., 2017). Higher relative abundance of *Bacteroidales* indicated that it was the key player in the hydrolysis of xylan and cellulose (Azman et al., 2017).

The microbial community analysis results above reinforce that it is considered that the AnMBR can be successfully applied to the real sewage treatment process.

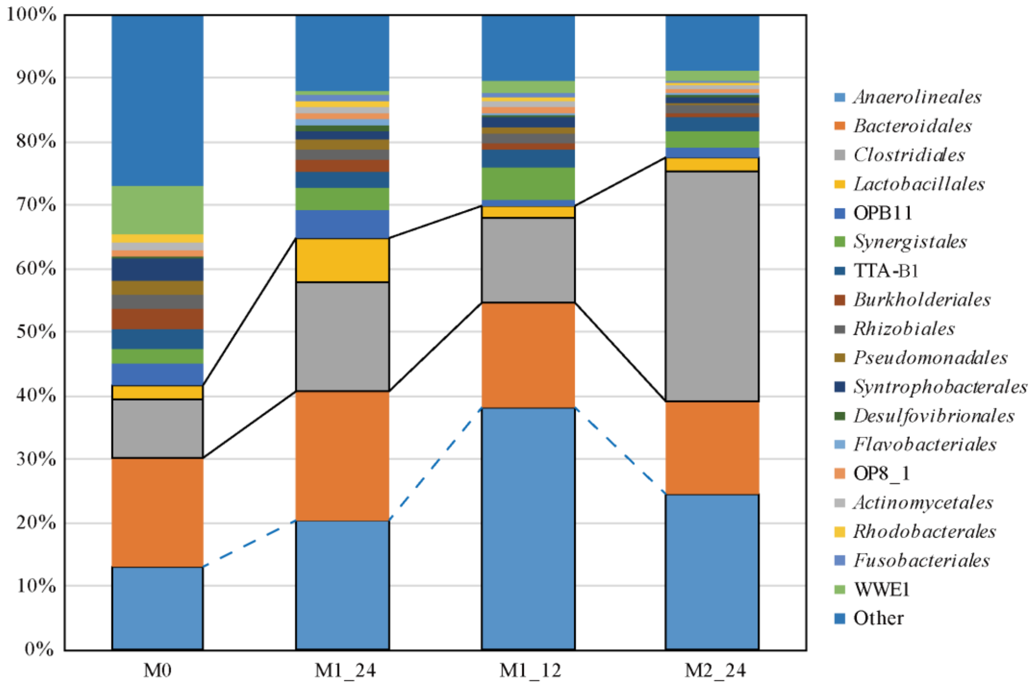


Fig. 3.25 Relative abundance of major bacterial members in sludge samples

3.3.9 Comprehensive comparison of MF and UF

A comprehensive comparison result is list in table 3.5 in aspects of organic removal, biogas produce performance, FLUX and the reaction index (MLSS, pH, ORP). The comparison result shown that there was almost no difference in organic removal efficiency, biogas production performance and the reaction indexes while the UF presented a much smaller FLUX as compared with MF in the same filtration conditions.

Table 3.5 Comprehensive comparison for MF / UF

Compare items	M1(MF)	M2(UF)
COD_{RE} (%)	89.3	88.8
BOD_{RE} (%)	93.0	93.1
SS_{RE} (%)	100	100
Biogas production rate (L-gas/L-water)	0.078	0.080
Methane content (%)	74.9	75.7
FLUX (m/d)	0.274	0.148
MLSS (g/L)	6~10	6~8.5
pH-eff	6.9 (6.7~7.1)	6.9 (6.7~7.1)
ORP (mV)	-324 (-300~-348)	-322 (-291~-349)

The distribution of particles in the AnMBRs is showed in figure 3.26 (above is M1/MF and below is M2/UF). According to the particle distribution results, M1 and M2 presented similar maximum particle size distribution as between 10 ~ 100 μm with the same percent revealed and even no particles was detected under 0.4 μm which indicated that there is no meaning for using 0.05 µm pore size (UF) to through the filtration process. Hence, it can be learned that UF (0.05 µm pore size) membrane cannot provide a better filtration effect comparing with the MF (0.4 µm pore size) membrane on AnMBR treating sewage wastewater.

Accordingly, sewage wastewater treated by AnMBR is considered to be recommended for using MF as the experiment results of this study shown almost no difference in organic removal efficiency, biogas production performance and the reaction indexes but the UF presented a much smaller FLUX with no better filtration effect can be provided in comparison with MF in the same filtration conditions.

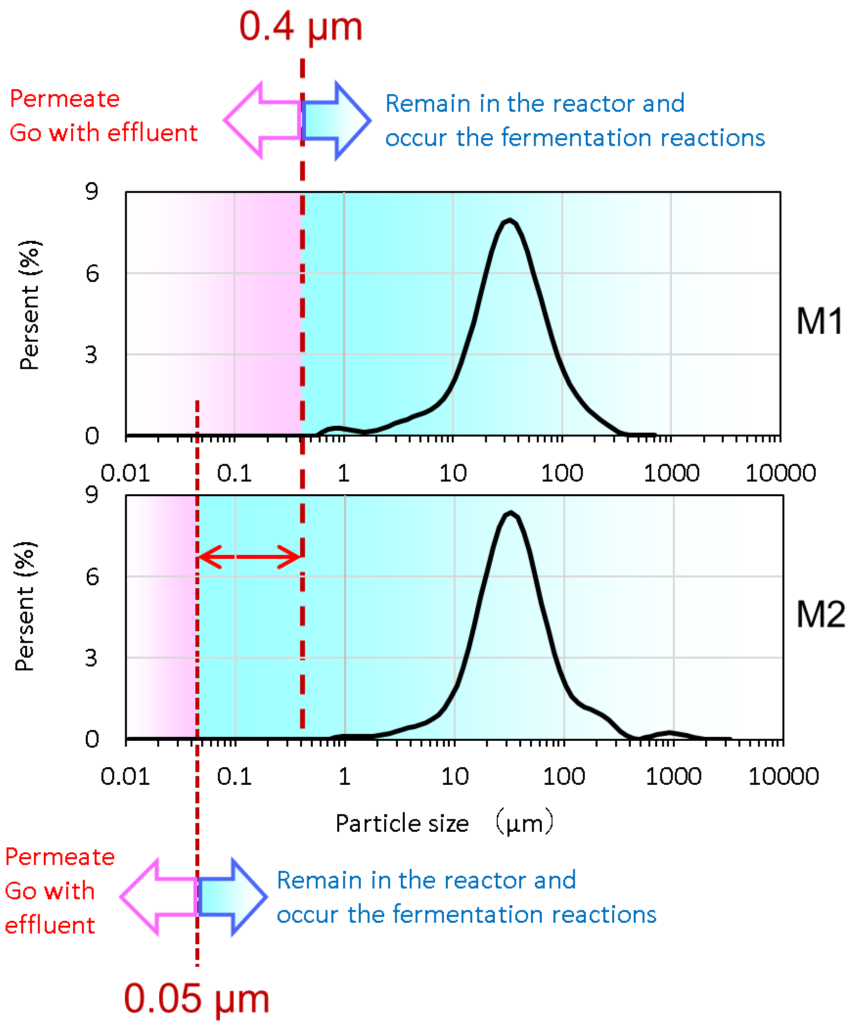


Fig. 3.26 Particle distribution for each AnMBR.

3.4 Conclusions

As installed two AnMBRs feeding by real sewage wastewater and operated for more than 150 days, the following conclusions can be obtained:

(1) The AnMBRs with different pore size membranes applied for treating the real sewage wastewater were started-up successfully and verified a good performance on organic pollutant removal (COD removal efficiency around 89%) with a great potential of energy recovery due to the methane yield was achieved 0.18 ~ 0.20 L-CH_4/g-COD_{rem} (dissolved methane was not included).

(2) Sodium hypochlorite solution used for online membrane cleaning should be less than 1 g/L as the final concentration in AnMBR on the purpose of protecting the microorganisms inside the reactor. Off-line membrane cleaning can achieve 80% potential of filtration ability recovery.

(3) Compare with the two different pore size membrane, the microfiltration membranes (0.4μm pore size used in this study) are relatively more appropriate for treating the sewage wastewater than the ultrafiltration membranes (0.05μm pore size used in this study) because of it can achieve the same treatment performance and biogas production with a relatively lower energy consumption for overcoming the suction pressure caused during the filtration process.

REFRENCES:

APHA, 2005. Standard Methods for the Examination of Water and Wastewater, 21st ed., America Water Works Association and Water Environment Federation, Wahington, DC, USA.

Azman, S., Khadem, A.F., Plugge, C.M., Stams, A.J.M., Bec, S., Zeeman, G., 2017. Effect of humic acid on anaerobic digestion of cellulose and xylan in completely stirred tank reactors: inhibitory effect, mitigation of the inhibition and the dynamics of the microbial communities. Appl. Microbiol. Biotechnol. 101, 889–901. https://doi.org/10.1007/s00253-016-8010-x

Barreto, C.M., Garcia, H.A., Hooijmans, C.M., Herrera, A., Brdjanovic, D., 2017. Assessing the performance of an MBR operated at high biomass concentrations. Int. Biodeterior. Biodegrad. https://doi.org/10.1016/j.ibiod.2016.10.006

Batstone, D.J., Keller, J., Angelidaki, I., Kalyuzhnyi, S. V, Pavlostathis, S.G., Rozzi, A., Sanders, W.T.M., Siegrist, H., Vavilin, V.A., 2002. The IWA Anaerobic Digestion Model No 1 (ADM1). Water Sci. Technol. 45, 65–73. https://doi.org/10.2166/wst.2002.0292

Brandt, M.J., Johnson, K.M., Elphinston, A.J., Ratnayaka, D.D., 2017. Chapter 7 - Chemistry, Microbiology and Biology of Water, in: Brandt, M.J., Johnson, K.M., Elphinston, A.J., Ratnayaka, D.D.B.T.-T.W.S. (Seventh E. (Eds.), . Butterworth-Heinemann, Boston, pp. 235–321. https://doi.org/https://doi.org/10.1016/B978-0-08-100025-0.00007-7

Chen, R., Nie, Y., Hu, Y., Miao, R., Utashiro, T., Li, Q., Xu, M., Li, Y.Y., 2017a. Fouling behaviour of soluble microbial products and extracellular polymeric substances in a submerged anaerobic membrane bioreactor treating low-strength wastewater at room temperature. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2017.02.046

Chen, R., Nie, Y., Ji, J., Utashiro, T., Li, Q., Komori, D., Li, Y.-Y., 2017b. Submerged anaerobic membrane bioreactor (SAnMBR) performance on sewage treatment: removal efficiencies, biogas production and membrane fouling. Water Sci. Technol. 76, 1308–1317. https://doi.org/10.2166/wst.2017.240

Chen, R., Nie, Y., Kato, H., Wu, J., Utashiro, T., Lu, J., Yue, S., Jiang, H., Zhang, L., Li, Y.Y., 2017c. Methanogenic degradation of toilet-paper cellulose upon sewage treatment in an anaerobic membrane bioreactor at room temperature. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.12.089

Chen, Z., Li, X., Hu, D., Cui, Y., Gu, F., Jia, F., Xiao, T., Su, H., Xu, J., Wang, H., Wu, P., Zhang, Y., Jiang, N., 2018. Performance and methane fermentation characteristics of a pilot scale anaerobic membrane bioreactor (AnMBR) for treating pharmaceutical wastewater containing m-cresol (MC) and iso-propyl alcohol (IPA). Chemosphere. https://doi.org/10.1016/j.chemosphere.2018.05.008

Ferrero, G., Rodríguez-Roda, I., Comas, J., 2012. Automatic control systems for submerged membrane bioreactors: A state-of-the-art review. Water Res. https://doi.org/10.1016/j.watres.2012.03.055

Gao, D.-W., Lee, Y.H., Wong, C.-Y., Yeh, D.H., Zhang, T., Tang, C.-Y.Y., Criddle, C.S., Wu, W.-M., 2010. Membrane fouling in an anaerobic membrane bioreactor: Differences in relative abundance of bacterial species in the membrane foulant layer and in suspension. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2010.08.031

Gouveia, J., Plaza, F., Garralon, G., Fdz-Polanco, F., Peña, M., 2015a. A novel configuration for an anaerobic submerged membrane bioreactor (AnSMBR). Bioresour. Technol.

Gouveia, J., Plaza, F., Garralon, G., Fdz-Polanco, F., Peña, M., 2015b. Long-term operation of a pilot scale anaerobic membrane bioreactor (AnMBR) for the treatment of municipal wastewater under psychrophilic conditions. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2015.03.002

Guglielmi, G., Chiarani, D., Saroj, D.P., Andreottola, G., 2010. Sludge filterability and dewaterability in a membrane bioreactor for municipal wastewater treatment. Desalination. https://doi.org/10.1016/j.desal.2009.06.074

Hasan, S.W., Elektorowicz, M., Oleszkiewicz, J.A., 2014. Start-up period investigation of pilot-scale submerged membrane electro-bioreactor (SMEBR) treating raw municipal wastewater. Chemosphere. https://doi.org/10.1016/j.chemosphere.2013.11.009

Hong, H., Lin, H., Mei, R., Zhou, X., Liao, B.Q., Zhao, L., 2016. Membrane fouling in a membrane bioreactor: A novel method for membrane surface morphology construction and its application in interaction energy assessment. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2016.06.006

Hong, H., Zhang, M., He, Y., Chen, J., Lin, H., 2014. Fouling mechanisms of gel layer in a submerged membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2014.05.063

Huang, X., Gui, P., Qian, Y., 2001. Effect of sludge retention time on microbial behaviour in a submerged membrane bioreactor. Process Biochem. https://doi.org/10.1016/S0032-9592(01)00135-2

Huang, Z., Ong, S.L., Ng, H.Y., 2011. Submerged anaerobic membrane bioreactor for low-strength wastewater treatment: Effect of HRT and SRT on treatment performance and membrane fouling. Water Res. https://doi.org/10.1016/j.watres.2010.08.035

Kalboussi, N., Rapaport, A., Bayen, T., Ben Amar, N., Ellouze, F., Harmand, J., 2017. Optimal control of a membrane filtration system. IFAC-PapersOnLine 50, 8704–8709. https://doi.org/https://doi.org/10.1016/j.ifacol.2017.08.1554

Khiewwijit, R., Temmink, H., Rijnaarts, H., Keesman, K.J., 2015. Energy and nutrient recovery for municipal wastewater treatment: How to design a feasible plant layout? Environ. Model. Softw. 68, 156–165. https://doi.org/https://doi.org/10.1016/j.envsoft.2015.02.011

Kola, A., Ye, Y., Le-Clech, P., Chen, V., 2014. Transverse vibration as novel membrane fouling mitigation strategy in anaerobic membrane bioreactor applications. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2013.12.078

Kong, Z., Li, L., Li, Y.Y., 2018. Characterization and variation of microbial community structure during the anaerobic treatment of N, N-dimethylformamide-containing wastewater by UASB with artificially mixed consortium. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2018.08.020

Koros W J, H, M.Y., T, S., 1996. Terminology for membranes and membrane processes (IUPAC Recommendations 1996). Pure Appl. Chem. https://doi.org/10.1351/pac199668071479

Lei, Z., Yang, S., Li, Y. you, Wen, W., Wang, X.C., Chen, R., 2018. Application of anaerobic membrane bioreactors to municipal wastewater treatment at ambient temperature: A review of achievements, challenges, and perspectives. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2018.07.050

Maria, F. Di, 2017. The Recovery of Energy and Materials Sludge : Internal Environment of Digester and Methanogenic Pathway, Food Bioconversion. Elsevier Inc. https://doi.org/10.1016/B978-0-12-811413-1/00003-6

Martin-Garcia, I., Monsalvo, V., Pidou, M., Le-Clech, P., Judd, S.J., McAdam, E.J., Jefferson, B., 2011. Impact of membrane configuration on fouling in anaerobic membrane bioreactors. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2011.07.042

Mei, X., Quek, P.J., Wang, Z., Ng, H.Y., 2017. Alkali-assisted membrane cleaning for fouling control of anaerobic ceramic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.02.052

Meng, F., Zhang, S., Oh, Y., Zhou, Z., Shin, H.-S., Chae, S.-R., 2017. Review Fouling in membrane bioreactors: An updated review. Water Res. https://doi.org/10.1016/J.WATRES.2017.02.006

Metzger, U., Le-Clech, P., Stuetz, R.M., Frimmel, F.H., Chen, V., 2007. Characterisation of polymeric fouling in membrane bioreactors and the effect of different filtration modes. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2007.06.016

Nie, Y., Kato, H., Sugo, T., Hojo, T., Tian, X., Li, Y.Y., 2017a. Effect of anionic surfactant inhibition on sewage treatment by a submerged anaerobic membrane bioreactor: Efficiency, sludge activity and methane recovery. Chem. Eng. J. https://doi.org/10.1016/j.cej.2017.01.022

Nie, Y., Niu, Q., Kato, H., Sugo, T., Tian, X., Li, Y.Y., 2017b. Efficient methanogenic degradation of alcohol ethoxylates and microbial community acclimation in treatment of municipal wastewater using a submerged anaerobic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.11.128

Nie, Y., Tian, X., Zhou, Z., Li, Y.Y., 2017c. Impact of food to microorganism ratio and alcohol ethoxylate dosage on methane production in treatment of low-strength wastewater by a submerged anaerobic membrane bioreactor. Front. Environ. Sci. Eng. https://doi.org/10.1007/s11783-017-0947-1

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. Sep. Purif. Technol. https://doi.org/10.1016/j.seppur.2013.06.036

Peña, M., do Nascimento, T., Gouveia, J., Escudero, J., Gómez, A., Letona, A., Arrieta, J., Fdz-Polanco, F., 2019. Anaerobic submerged membrane bioreactor (AnSMBR) treating municipal wastewater at ambient temperature: Operation and potential use for agricultural irrigation. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2019.03.019

Shizas, I., Bagley, D.M., 2004. Experimental Determination of Energy Content of Unknown Organics in Municipal Wastewater Streams. J. Energy Eng. https://doi.org/10.1061/(ASCE)0733-9402(2004)130:2(45)

Song, X., Luo, W., Hai, F.I., Price, W.E., Guo, W., Ngo, H.H., Nghiem, L.D., 2018. Resource recovery from wastewater by anaerobic membrane bioreactors: Opportunities and challenges. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2018.09.001

Sweity, A., Ying, W., Belfer, S., Oron, G., Herzberg, M., 2011. PH effects on the adherence and fouling propensity of extracellular polymeric substances in a membrane bioreactor. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2011.04.056

Watanabe, R., Nie, Y., Takahashi, S., Wakahara, S., Li, Y.Y., 2016. Efficient performance and the microbial community changes of submerged anaerobic membrane bioreactor in treatment of sewage containing cellulose suspended solid at 25 °C. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.05.049

Watanabe, R., Qiao, W., Norton, M., Wakahara, S., Li, Y.-Y., 2014. Recent Developments in Municipal Wastewater Treatment Using Anaerobic Membrane Bioreactor: A Review. J. Water Sustain. https://doi.org/10.11912/jws.4.2.101-122

Wu, J., Le-Clech, P., Stuetz, R.M., Fane, A.G., Chen, V., 2008. Effects of relaxation and backwashing conditions on fouling in membrane bioreactor. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2008.06.057

Yoo, R.H., Kim, J.H., McCarty, P.L., Bae, J.H., 2013. Effect of temperature on the treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor. Water Sci. Technol. 69, 1145–1150. https://doi.org/10.2166/wst.2013.793

Yoon, S.-H., 2015. Membrane bioreactor processes: principles and applications. CRC press.

Yue, X., Koh, Y.K.K., Ng, H.Y., 2015. Effects of dissolved organic matters (DOMs) on membrane fouling in anaerobic ceramic membrane bioreactors (AnCMBRs) treating domestic wastewater. Water Res. 86, 96–107. https://doi.org/10.1016/J.WATRES.2015.07.038

Chapter 4

Effect of HRT on treatment performance at room temperature

4.1 Introduction

Sewage wastewater is the most abundant type of wastewater in the world and as developed for more than 100 years, the Activated Sludge Process (ASP) has been evolved into a mature process for treating sewage wastewater. However, the issues such as huge energy cost for aeration requirement and big amount of waste sludge production is urgent needs to be addressed. On the other hands, researches on sewage treatment solutions are tend to not just purify kinds of wastewaters but also attempt to get the energy recovery of resources recycling (Zakkour et al., 2001). Technologies already be used for treating some kinds of industrial wastewaters or waste sludge and achieved the recovery or reuse of energy and resources have been reported (Le Corre et al., 2009; Pandey et al., 2016; Rulkens, 2008). If it could be possible for processes treating sewages as well, the wastewater treatment plants could also become net suppliers of energy, renewable resources and reclaimed water. It will play a very important role in the construction of sustainable social development.

The anaerobic membrane bio-reactor (AnMBR) integrates anaerobic digestion and membrane technology creating a new process which provided with both the advantages of anaerobic digestion which have the potential of energy recovery through the methane fermentation process as well as the high efficiency of sludge-water separation due to the filtration by membrane (Bai and Leow, 2002). Plus, it could also solve the problem that the slow growing of anaerobic sludge especially for treating low organic strength sewage to achieve a high organic strength retained inside the reactor by the membrane filtration (Watanabe, 2004). Furthermore, the high efficiency separation for sludge and treated water it provides is suitable for treating a big amount of water of which exactly is one of the characteristics of the sewage wastewater (Judd, 2010).

Up to now, AnMBR has been successfully applied in the field of industrial wastewater treatment and there are plenty of studies related to AnMBR focused on high organic strength wastewater or industrial wastewater while still lacking of development for treating the real municipal sewage as the features of: large quantity, complicated composition, instable and low concentration of pollutant (Lei et al., 2018). However, despite that, there are some studies that related to the low organic strength wastewater or sewage based on man-made synthetic wastewater have be reported. High efficiency of organic removal and methane conversion rate were obtained upon small sludge yield reported in the previous research (Chen et al., 2017b). Research on the effect of anionic surfactant inhibition on sewage treatment shows LAS can inhibit to the methanogen activity and can cause a higher membrane fouling rate as the microbial self-protection behaviour in coping with the LAS in sewage (Nie et al., 2017a)while the alcohol ethoxylates (AE) could be efficiently degraded and converted into methane though it caused a higher membrane fouling rate due to the microbial self-protection behaviour by releasing more amounts of extracellular polymeric substances (EPS) and soluble microbial products (SMP) (Nie et al., 2017b, 2017c). A research that contributed to a better understanding of properties of EPS and SMP and their roles in membrane fouling in an AnMBR treating low strength sewage at room temperature was also reported (Chen et al., 2017a). AnMBR was proved to be a suitable process to treat SS containing sewage by provide a influence of cellulose as suspended solid (SS) and evaluated the performance of submerged AnMBR at 25°C using two types of synthetic sewage with SS contained or not contained under the HRT ranged from 48 to 6 hours (Watanabe et al., 2016). Effect of HRT (12, 10, 8 hours) and SRT on treatment performance was also be investigated by a synthetic sewage treatment study (Huang et al., 2011).

Based on the results of successfully started 2 AnMBRs feeding with real sewage wastewater described in Chapter 3, a mini-pilot AnMBR was operated in the wastewater treatment plant at room temperature (25°C) and continued to dealing with the real sewage at operated HRTs ranged from 12, 8, 6 to 4 hours in this Chapter. The performance of sewages treated by AnMBR in each HRT condition was investigated in aspects on pollutant removal performance, gas yield, sludge yield, COD balance as well as the filtration performance of the membranes.

4.2. Materials and methods

4.2.1 Consist and operation of the mini-pilot

A 20L AnMBR with hollow fiber type membranes set inside was installed as a mini-pilot in Sen-En wastewater treatment plant (S-WWTP) located in Tagajo city of Miyagi prefecture mentioned in chapter 3. The raw municipal sewage wastewater was pumped continuously from the beginning of the S-WWTP into a 100 L sewage bucket together with a continues stirring (US540-401) to keep the sewage fresh and there was over-flow pipeline setting to make the extra sewage flow back under gravity. The influent of AnMBR was taken from the sewage bucket by a peristaltic pump (FP-100-1515). Basic water quality index of the raw municipal sewage wastewater was also mentioned in table 3.1 of chapter 3. Solid-liquid separation was permeated using a micro-filtration module

(Mitsubishi Chemicals, Japan) by a peristaltic pump (FP-100-1515). Membrane module used in this research was a hollow fiber type polyvinylidene difluoride (PVDF) membrane with the pore size of 0.4μm.

The produced biogas was recycled by a diaphragm pump (APN-110KV-1, Iwaki, Japan) to scour the membranes' surface as a fouling control via a gas diffuser at the bottom of the membrane module (Martin-Garcia et al., 2011). A digital pressure meter (AP-10S & AP-V85, Keyence, Japan) was installed between the membrane module and the permeate pump to measure the trans-membrane pressure (TMP) and the TMP data was recorded by a multi input data logger (NR-500 & NR-HA08, Keyence, Japan) connected to a computer and controlled by the installed software. Biogas production was measured by a wet gas meter. The operation temperature of 25 degrees was controlled by a water bath equipment (NTT-20S).

4.2.2 Operation conditions of the mini-pilot

Seed sludge for the AnMBR was taken from the full-scale waste sludge treatment

Table 4.1 Detail experiment conditions in different HRTs.

Operated period (Day-)	1-100	101-197	198-251	252-322
HRT (h)	12	6	8	4
OLR (g-COD/L/d)	0.67	1.18	1.52	2.05
FLUX (m/d)	0.27	0.23	0.17	0.35
membrane area (m^2)	0.146	0.345		
CFGV* (m/h)	70 & 119	116		116 & 174
Permeate mode	4mins on / 1min off			

CFGV* is the cross-flow gas velocity (m/h).

process inside the S-WWTP and operated for over 100days. In this experiment, the long-term operation of AnMBR fed by only the real sewage wastewater. The details of the operation conditions such as HRT are shown in table 4.1.

4.2.3 Samples collection and analysis methods

Influent, effluent and mixed liquor samples were regularly taken in order to analysis water quality index and the sludge traits. The analysis of COD, BOD, SS, mixed liquor suspended solid (MLSS) and mixed liquor volatile suspended solid (MLVSS) were in according with standard methods (APHA, 2005). The proportion of CH4, CO2, and N2 in biogas produced was measured using a gas chromatograph (Shimadzu, GC-8A, Japan) equipped with a thermal conductivity detector. H_2S in produced biogas was determined by a detector tube type gas measuring device (GASTEC, GV-100, Japan) with the standard method of the device itself. Dissolved methane in the effluent was determined using a headspace technique which has been described by former researchers (Watanabe et al., 2016). All the methane measurements of methane gas were normalized to the standard temperature and pressure (STP: 0 degree, 1atm). In daily operations, pH for influent and effluent and oxidation reduction potential (ORP) for the activated anaerobic sludge was measured by a pH meter (TOADKK, DM-32P, Japan) and ORP meter (TOADKK, RM-30P, Japan).

The removal efficiency of the pollutant calculated is listed below.

$$COD_{RE} = \frac{COD_{inf} - COD_{eff}}{COD_{inf}} \times 100\%$$

$$BOD_{RE} = \frac{BOD_{inf} - BOD_{eff}}{BOD_{inf}} \times 100\%$$

$$SS_{RE} = \frac{SS_{inf} - SS_{eff}}{SS_{inf}} \times 100\%$$

where COD_{inf}, COD_{eff} and COD_{RE} are the COD of the influent, the effluent and the removal efficiency of COD, respectively; BOD_{inf}, BOD_{eff} and BOD_{RE} are the BOD of the influent, the effluent and the removal efficiency of BOD, respectively; SS_{inf}, SS_{eff} and SS_{RE} are the SS of the influent, the effluent and the removal efficiency of SS, respectively.

The calculate equation of sludge yield is:

$$Sludge\ \ yield = \frac{\Delta MLVSS}{COD_{rem}} = \frac{(MLVSS_2 - MLVSS_1)V}{t\left(COD_{inf} - COD_{eff}\right)Q}$$

where COD_{inf}, COD_{eff} and COD_{rem} are the COD of the influent, the effluent and the removal of COD, respectively; $\Delta MLVSS$ is the variation of MLVSS; $MLVSS_2$ and $MLVSS_1$ represent MLVSS value in two different time; V is the reaction volume of AnMBR; Q is the sewage treatment capacity in a certain time.

The calculate equations of biogas production rate and biogas yield is:

$$bpr = \frac{V_{gas}}{Q}$$

$$biogas\ \ yield = \frac{bpr}{COD_{rem}}$$

$$CH_4\ \ yield = \frac{bpr}{COD_{rem}} \times CH_4\%$$

where V_{gas}, Q, bpr and COD_{rem} are the volume of biogas produced, sewage treatment capacity in a certain time, biogas production rate, and the removal of COD, respectively.

The COD balance was calculated for influent, permeate effluent, biogas produced (methane gas discharged), H2S in the biogas produced, methane gas dissolved in the effluent and sludge growth, all was converted into COD value.

4.2.4 Online membrane cleaning

In order to achieve stable membrane filtration and extend the membrane life, online membrane cleaning by chemicals backwashing was implemented for every 10 ~ 15 days' operation. An even shorter frequency of every one week was also applied during the HRT 4h condition because of the high FLUX and rapid TMP growth. Since the fouling matters were mainly consisted by the organic matters which was investigated in last chapter, the chemicals for online cleaning is only used by sodium hypochlorite solution. In order to determine the concentration of sodium hypochlorite solution used for backwashing without reducing the sludge activity, batch test with different concentration of sodium hypochlorite solution was done. The online backwashing cleaning process is following the steps:

Step 1. Turn off influent pump, effluent pump and biogas cycling pumps;

Step 2. Opposite the direction of the effluent pump rotation as the backwashing pump;

Step 3. Prepare 1.0 L sodium hypochlorite solution*;(Metzger et al., 2007; Wu et al., 2008)

Step 4. Start the backwashing pump with a timer of 1min on / 3 min off mood to pump sodium hypochlorite solution back in to the reactor;

Step 5. Control the backwashing pump speed with a TMP under than 5kPa and implement about 30 minutes for the online cleaning;

Step 6. Waite for 10 minutes after the backwashing finished;

Step 7. Turn on biogas cycling pumps;

Step 8. Opposite the direction of the backwashing pump rotation again to return the effluent pump;

Step 9. Turn on effluent pump to recover the water level inside the reactor as well as observe the filtration process;

Step 10. Turn on influent pump.

4.3 Results and discussion

4.3.1 Organic pollutant removal

Figure 4.1 and figure 4.2 shows COD and BOD as in influent, effluent and the removal efficiency during the long termed operation experiment. COD and BOD in effluent and removal efficiencies were stably low in each HRT conditions. In addition, pH in effluent and ORP was also obtained stable during the long-term operation which were shown in figure 4.3. The average COD, BOD and SS performance (influent / effluent and the removal efficiency) at different HRT conditions were calculated and listed as table 4.2. According to the result, the effluent COD was under 50mg/L and the effluent BOD was under 10mg/L in HRT condition of 12, 8 and 6 hours. Moreover, COD removal efficiency in HRT 12h ~ 6h was obtained around 89%, which was recognized as a high organic removal performance in a previous research applying combined anaerobic-anoxic-oxic municipal wastewater treatment process with the same average COD removal efficiency

Table 4.2 Average COD, BOD, SS performance in each HRT.

HRT (h)	12	8	6	4
COD-in (mg/L)	372.0	393.0	383.0	350.0
COD-eff (mg/L)	39.9	49.9	43.1	54.5
COD_{RE} (%)	89.3	87.3	88.7	84.4
BOD-in (mg/L)	139.1	127.9	150.6	127.6
BOD-eff (mg/L)	10.8	10.2	9.1	13.6
BOD_{RE} (%)	92.2	92.0	94.0	89.3
SS-eff (mg/L)	0	0	0	0
SS_{RE} (%)	100	100	100	100

of around 89% performed (Garuti et al., 1992). While in HRT 4h it was decreased to less than 85%, lower but still was considered as a high level organic removal efficiency presented in aerobic treatment processes (Yao et al., 2013). On the performance of BOD removal, the average removal efficiency in HRT 4h was achieved only 87.3%, lower than that in other HRT conditions of above 92%. While, the removal efficiency of SS was achieved 100% in all HRT conditions due to the 0.4μm pore size filtration.

Comparing the performance in this study with previous researches (table 4.3) fed with synthetic sewage wastewater with a COD concentration of 400~550mg/L treated under mesophilic condition (25~35°C) by micro-membrane AnMBRs, this study achieved good performance. Though the effluent COD was a little more than previous researches, the performance was not bad considering treating the real sewage wastewater. The effluent water quality was presented a little bit worse since the HRT was shortened to 4 hours while the discharged water was still better than some cases treated the synthetic sewage wastewater at HRT 6 hours (Watanabe et al., 2016).

Table 4.3 Comparison of the COD removal with other similar published reports.

Tem. (°C)	HRT (h)	COD-inf (mg/L)	COD-eff (mg/L)	References
35	3 ~ 24	460	27 ~ 48	(Hu and Stuckey, 2006)
30	24	500	20	(Gao et al., 2010)
25~30	8 ~ 10	550	17	(Huang et al., 2011)
25	12 ~ 48	470 ± 82	34 ± 21	(Watanabe et al., 2016)
25	6 ~ 24	400 ± 150	< 50	This study
25	4	400 ± 150	54.5	This study

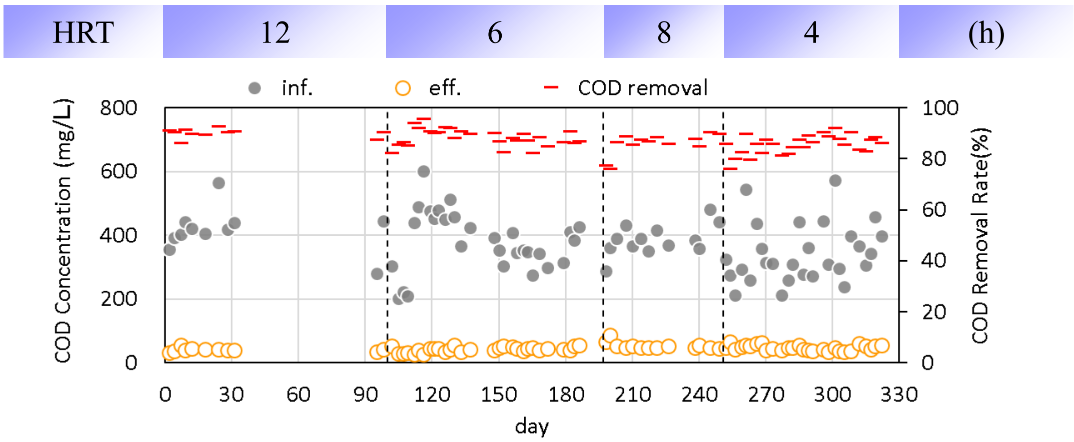


Fig. 4.1 COD in influent and effluent and removal efficiency during long-term operation.

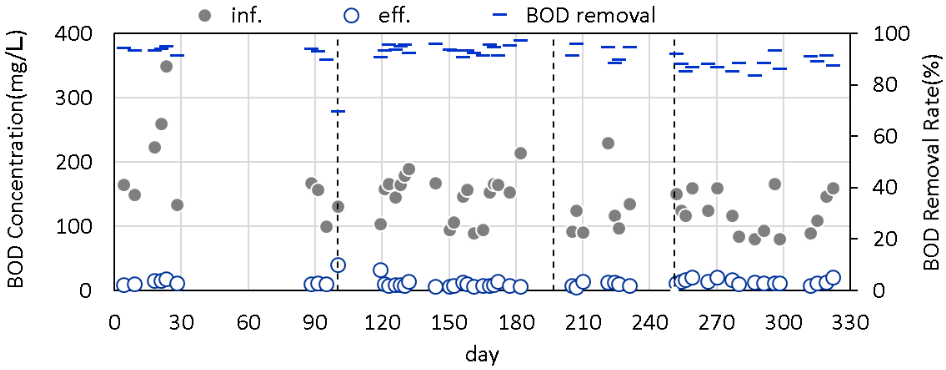


Fig. 4.2 BOD in influent and effluent and removal efficiency during long-term operation.

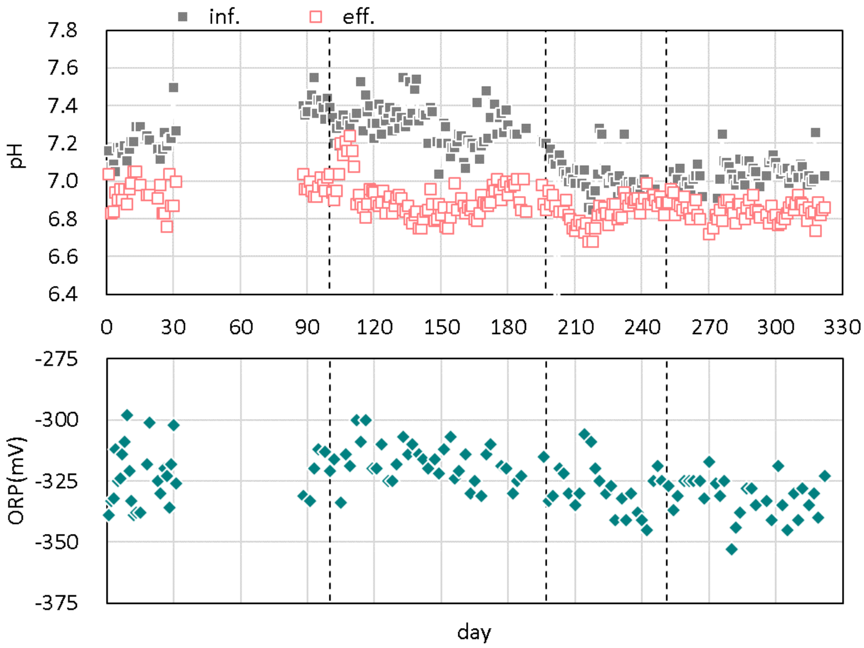


Fig. 4.3 pH and ORP during long-term operation.

4.3.2 Sludge yield

Figure 4.4 shows the mixed liquor concentration as MLSS and MLVSS during the long termed operation experiment. The MLSS was up to almost 20 g/L since the mixed liquor concentration was not controlled during HRT 6 ~ 12 hours for the purpose of investigate the membrane filtration performance in relatively high MLSS condition. Figure 4.5 shows the sludge yield calculation for each HRT condition followed the calculate equation mentioned in materials and methods section. The calculated sludge yield is obviously the slope in each calculation figures and shown in figure 4.6 by HRT as the x-coordinate. Overall, it was quite clear that sludge yield increased along with the HRT shortened especially in HRT 4 hours. In HRT 6 ~ 12 hours, the sludge yield was only 0.07 ~ 0.11 g-VSS/g-COD_{rem}, and the highest sludge yield as 0.22 g-VSS/g-COD_{rem}, presented in HRT 4h, was still much less than the sludge yield in a range of 0.25~0.4 g-VSS/g-COD_{rem} in a conventional aerobic activated sludge process (Huang et al., 2001).

Therefore, it can be draw out that the treatment performance was well on organics removal during HRT 6h to 12h in addition achieved a low sludge yield. While the removal efficiency decreased and sludge yield also respond quickly to a sharp increase since HRT was shortened to 4 hours.

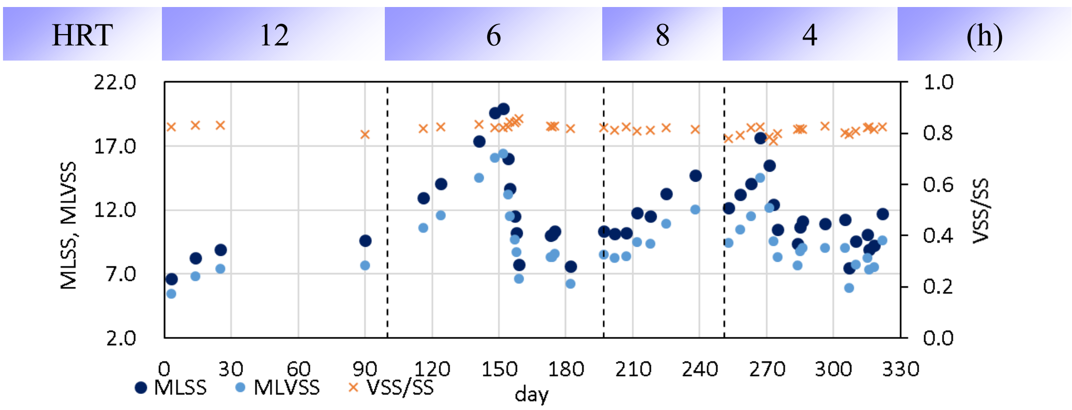


Fig. 4.4 MLSS and MLVSS during long-term operation.

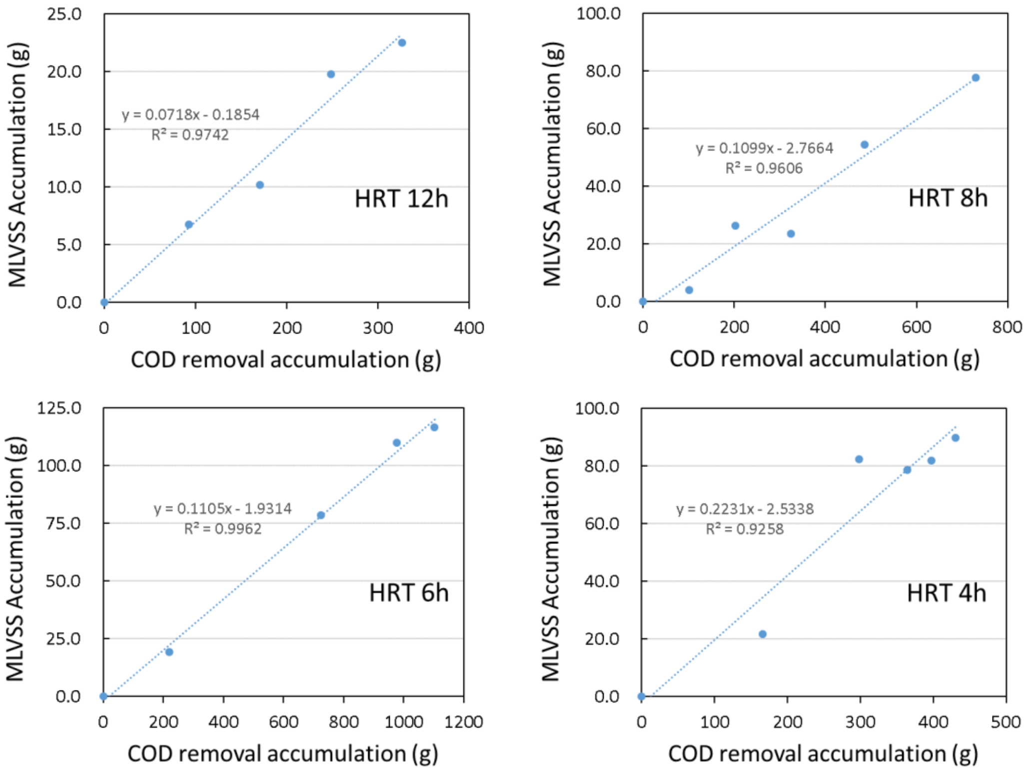


Fig. 4.5 MLSS accumulation by the COD_{rem} accumulation in each HRT.

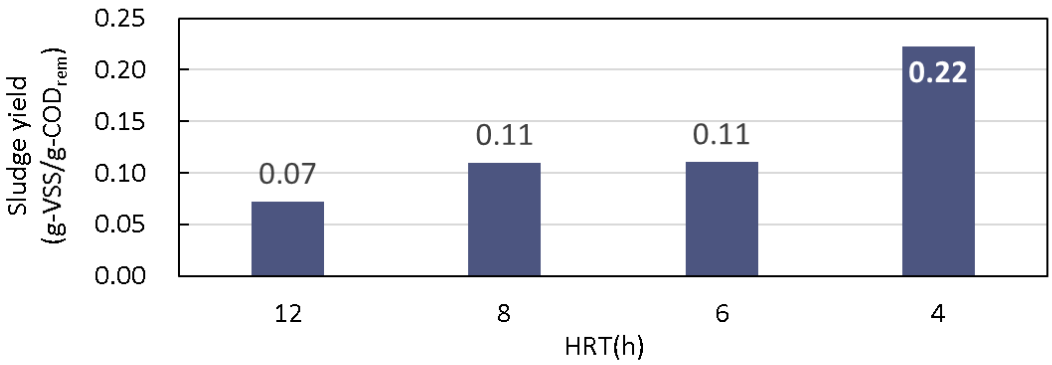


Fig. 4.6 Sludge yield in each HRT.

4.3.3 Biogas production

Figure 4.7 and figure 4.8 shows the biogas production rate and gas composition respectively during the long termed operation experiment and the highest biogas production was achieved as 0.12 L-gas/L-water during HRT 8h. Daily biogas production rate was not very stable thinkable due to some complicated reasons such as unstable organic content in the real sewage wastewater, unstable sewage wastewater temperature and the changeable of the environment temperature and barometric pressure although temperatures and pressures were considerations in the calculation for produced biogas to the standard conditions. On the other hand, the daily biogas composition was performed quiet stable even operated in different HRTs which indicated that methanogenic occurred relatively smooth during the overall experiment duration. H_2S gas content was shown as ppm unit in figure 4.9 which is useful for the calculation of COD balance.

Table 4.4 shows the average biogas production rate, gas composition, biogas yield and methane yield (CH_4 yield) in each HRT condition. Average biogas production rate achieved as high as 0.10 L-gas/L-water in HRT 12 and 8 hours and 0.09 L-gas/L-water in HRT 6h, higher than those achieved in chapter 3. While it was apparently decreased after HRT was shortened into 4 hours. In addition, the methane gas content was invariably obtained around 80% in the gas composition in each HRT condition as well as the nitrogen gas and carbon dioxide was existed of about 14% and 6%, respectively. In order to have a comparison with other previous researches, the parameter of biogas production rate could be expressed to biogas yield and methane yield by calculated combine with the influent water quality and the biogas composition data followed the calculation equations in materials and methods section and the obtained result is listed as the last two lines in table 4.4 in each HRT. Since the methane gas content in produced biogas was stable in different HRT conditions, a similar conclusion can be draw out that the biogas yield or methane yield decreased as long with the HRT shortened, just like the stepped down of biogas production rate performed.

Table 4.5 shows the comparison of methane yield with some published previous researches. It is not difficult to get that methanogens were performed well even by using the real sewage as feeding in HRT 6 ~ 24 hours. Though biogas yield was not so high in HRT 4h, it was also supposed to be an acceptable treatment plan considering the treatment capacity especially in some temporary situations when a requirement for dealing with a big amount of sewage wastewater was facing.

Thus, it is clear that applying submerged AnMBR to treating the real sewage wastewater even in the HRT condition as short as 6 hours, achievable of a high methanogenesis or biogas production performance have been confirmed.

Table 4.4 Average biogas performance in each HRT.

HRT (h)	12	8	6	4
Biogas production rate (L-gas/L-water)	0.10	0.10	0.09	0.06
CH_4 (%)	78.77	81.6	79.35	79.25
N_2 (%)	13.96	11.62	14.02	14.83
CO_2 (%)	7.27	6.78	6.63	5.92
Biogas yield (L-gas/g-COD_{rem})	0.30	0.28	0.25	0.20
Methane yield (L-CH_4/g-COD_{rem})	0.24	0.23	0.20	0.16

Table 4.5 Comparison of the CH_4 yield with other similar published reports.

Tem. (°C)	HRT (h)	Methane yield ($L\text{-}CH_4/g\text{-}COD_{rem}$)	AnMBR type	References
25	6 ~ 12	0.21 ~ 0.22	Submerged	(Ho and Sung, 2009)
28 ~ 33	8 ~ 12	0.21 ~ 0.29	Submerged	(Hongjun et al., 2011)
25~30	8 ~ 12	0.12 ~ 0.25	Side-stream	(Huang et al., 2011)
20	7 ~ 17	0.20	Side-stream	(Gouveia et al., 2015)
25	8 ~ 48	0.27 ~ 0.33	Submerged	(Chen et al., 2017c)
25	4	0.16	Submerged	This study
25	6 ~ 24	0.21 ~ 0.24	Submerged	This study

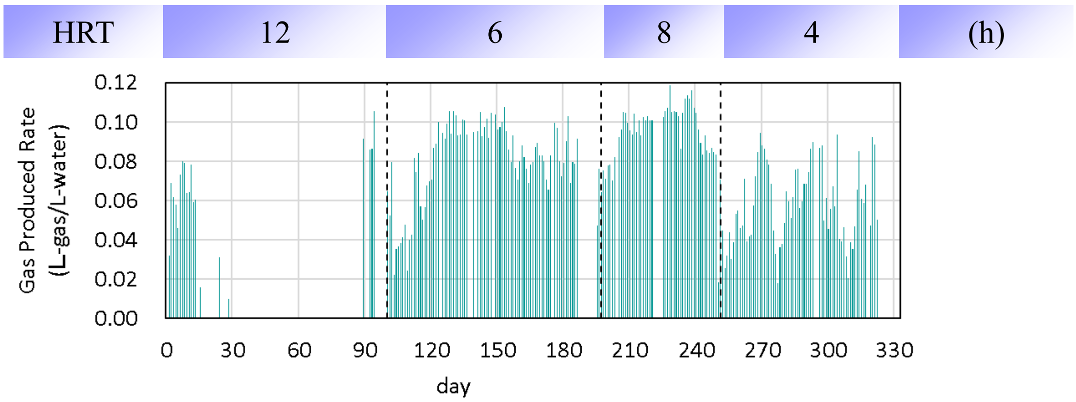


Fig. 4.7 Biogas production rate during long-term operation.

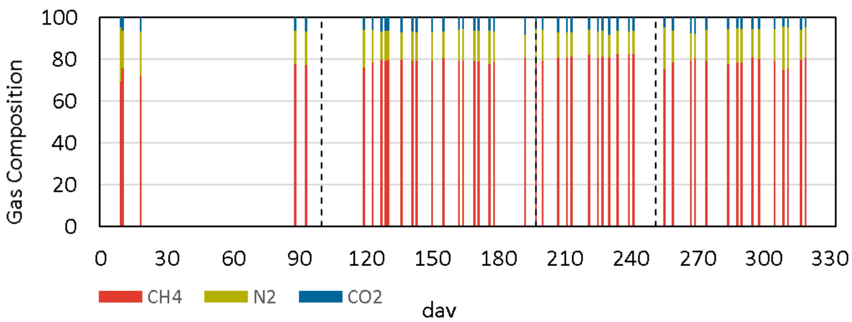


Fig. 4.8 Gas composition during long-term operation.

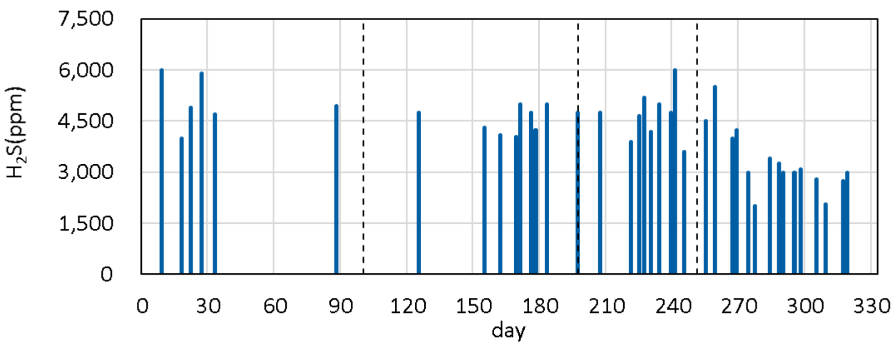


Fig. 4.9 H_2S gas produced during long-term operation.

4.3.4 COD balance analysis

Based on the results of effluent COD, sludge yield, biogas production, combine with the measured value of H_2S and dissolved methane, COD balance at different HRT conditions was calculated as shown in table 4.6 as absolute value of g-COD/d unit. It shown that COD converted to produced biogas in HRT 4h was even less than that in HRT 6h though COD content in influent sewage was increased per day which indicated that methanogenesis effect was not improved after HRT shortened into 4 hours from HRT 6h which also draw out that the micro-biological degradation was insufficient in HRT 4h condition.

Figure 4.10 shows the COD balance in percentage values. During HRT 6 to 12 hours, about 55 ~ 63% of COD in influent was transferred into methane gas by the anaerobic degradation indicating that the biogas-energy recovery efficiency could achieved around 60% in the HRT condition from 6 to 12 hours. After HRT was shortened into 4h, the biogas-energy recovery efficiency generated even as low as 41.6%. The reason is considered as the micro-biological degradation was insufficient in a short HRT condition as mentioned above. That analysis can also be verified by the percentages increased as the sludge growth and those remained in the effluent. The increase of sludge growth was caused by the organics especially SS cannot be biodegraded in time but existed inside of the reactor due to the membrane separation particularly since HRT was shortened into 4h. Table 4.7 shows the COD conversion to CH_4 in each HRT condition which was consisted as the amount in biogas and dissolved. The COD conversion rate to CH_4 shown a higher energy recovery potential if the dissolved methane could be gathered and as the same reason the COD conversion rate also decreased as the HRT shortened. In additional with those remained in the effluent, it can be learned that both soluble and insoluble organic matters were insufficient degraded in the short HRT condition. While between the soluble and insoluble biomass, insoluble biomass was more of un-degraded as a result of the percentages increased 2 times for sludge growth but less than 1.5 times for the organic remained in the effluent. However, despite the micro-biological degradation process by the anaerobic digestion, the biomass that stuck in the reactor also achieved removal thanks to the membrane filtration. Hence it shows that the membrane filtration did more contributions in the short HRT condition. Therefore, though micro-biological degradation and energy recovery efficiency was not performed well in HRT 4 hours, the purification effect of water quality was still shown a potential of practicable.

As a conclusion of COD balance result, it can be obtained that the reason for organic removal efficiency and biogas type energy recovery efficiency reduced in HRT 4h was considered to be the insufficient of the micro-biological degradation. Besides, HRT 4h condition was strained to operate the AnMBR in sewage purification owing to the membrane filtration.

Table 4.6 COD balance values (g-COD/d).

HRT (h)	12	8	6	4
COD-in	13.4	23.5	30.5	41.1
Biogas production	8.5	13	17.6	17.1
H_2S in biogas	0.0	0.1	0.1	0.1
Dissolved biogas	1.6	3.0	3.5	5.0
Sludge growth	1.1	2.9	3.9	10.2
COD-eff	1.4	3.2	3.4	6.4

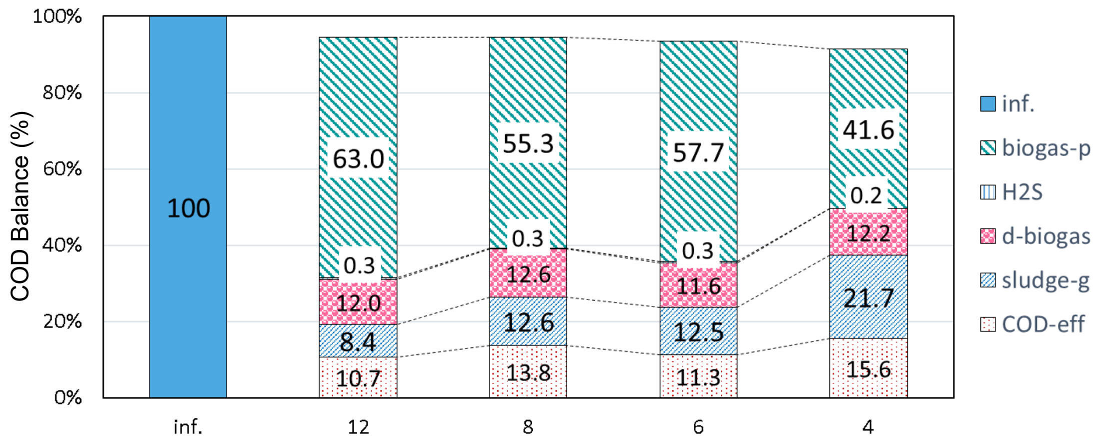


Fig. 4.10 COD balance (%).

Note:

inf.: COD in influent;

biogas-p: biogas produced (calculated by the discharged methane gas);

H2S: H_2S in the biogas produced;

Sludge-g: sludge growth;

COD-eff: COD in permeate effluent.

Table 4.7 COD conversion to CH_4 in each HRT.

HRT (h)	12	8	6	4
To biogas CH_4 (%)	63.0	55.3	57.7	41.6
To dissolved CH_4 (%)	12.0	12.6	11.6	12.2
Total (%)	75.0	67.9	69.3	53.8

4.3.5 Membrane performance

The TMP and FLUX data shown in figure 4.11 presents the membrane performance during the long-term operation. The CFGV was raised up to 119 m/h since day 80 to achieve the HRT 12h operation at a membrane area of 0.146 m^2, then it was set as 116 m/h to get close with 119 m/h which used in old membrane module as the new membrane module was updated with a total membrane area of 0.345 m^2.

On the basis of TMP-FLUX data from day 80 ~ 330, TMP was stabled at a very low value when FLUX was operated below 0.27 m/d (HRT 6~12h). While since FLUX was risen up to 0.35 m/d, the TMP was increased rapidly. Then for the sake of guarantee FLUX 0.35 m/d operation, the CFGV was upgraded to 174 m/h via increasing the biogas cycling. Whereas, the TMP still increased rapidly which the details are shown in figure 4.12. Furthermore, in short HRT conditions such as 4 hours, the concentration of the mixed liquor also must be controlled much stricter than HRT 6h or longer in order to maintain the filtration capacity of membranes. CFGV was returned to 116 m/h to figure out a possible of operation FLUX 0.35 m/d at the same CFGV as other HRTs since the treatment performance has been confirmed in HRT 4h. The growth rate of TMP in CFGV 116 m/h operation was presented 2 times than in CFGV 174 m/h operation and only lasted for 10 days for a continues period operation till the membrane had to stop due to the high TMP. In addition, no matter operated with high or low CFGV, even with the control of MLSS below than 11 g/L, the TMP still increased rapidly in FLUX 0.35 m/d and had to stop and clean the membrane during every short continuous operation period.

The results shown that applying the AnMBR directly into sewage wastewater treatment performed well on membrane filtration in FLUX condition below 0.27 m/d. While FLUX 0.35 m/d or even bigger is hard to operate due to the rapid increasing of TMP.

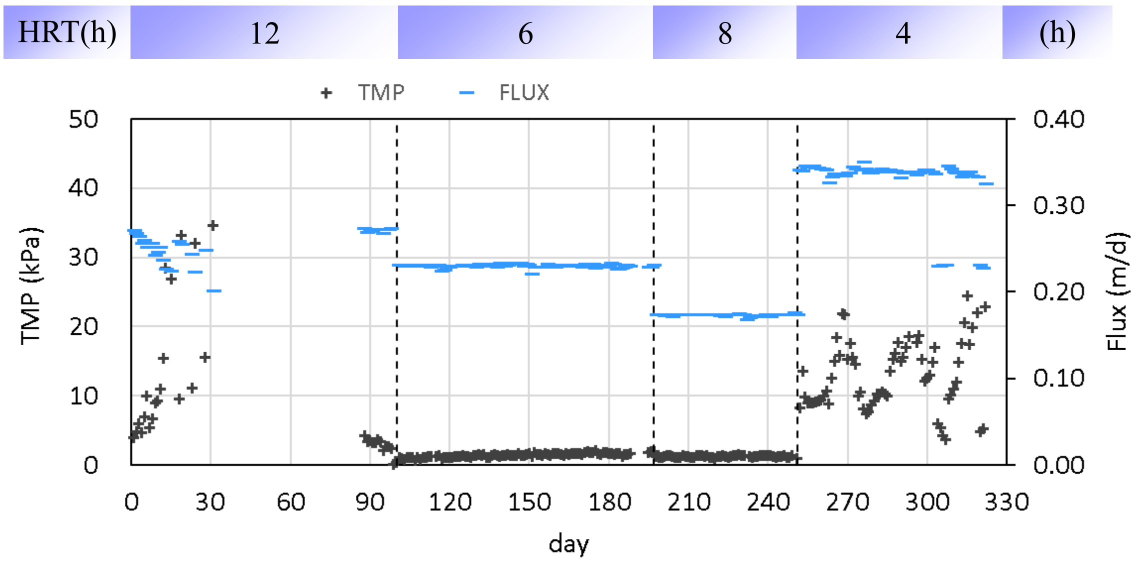


Fig. 4.11 TMP – FLUX record during long-term operation.

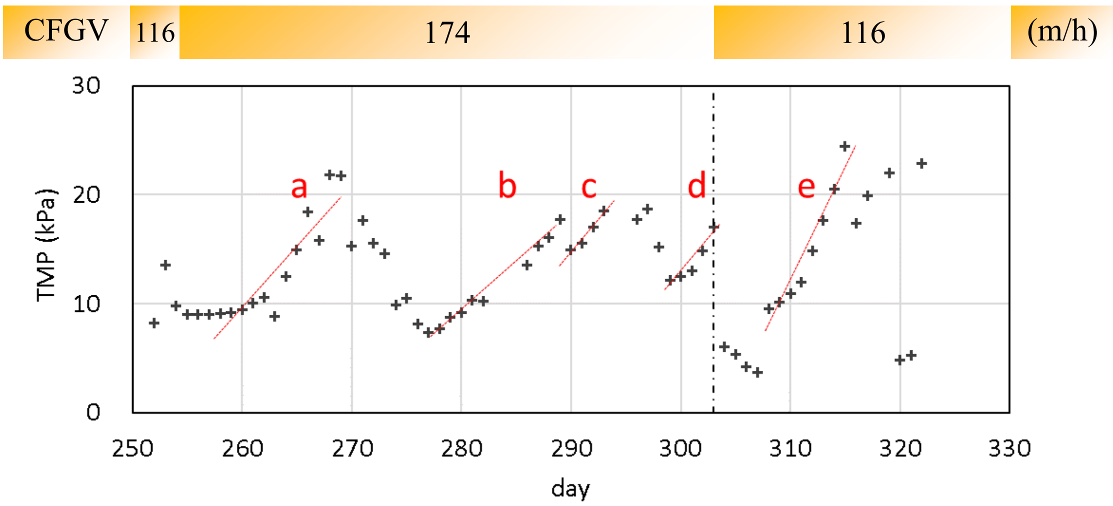


Fig. 4.12 TMP – FLUX record in HRT 4h.

Table 4.8 Slope for each fitted line in figure 4.12.

fitted line	a	b	c	d	e
slope	1.13	0.84	1.23	1.21	2.13

4.3.6 Comprehensive evaluation

A comprehensive comparison by the performances on aspect of organic removal (COD / BOD / SS), sludge character (MLSS and sludge yield), energy recovery potential (biogas production rate / methane content/biogas yield / methane yield), membrane filtration (FLUX and TMP growth), energy consumption (biogas cycling and temperature constant) as well as the operation conditions (sewage treatment capacity / pH in effluent / ORP) for different HRT conditions is shown in table 4.9. The weakness items were marked red color with bold and the incommensurable or non-differential items were marked in gray. As a result, HRT 4h presented the weakness compared with the other conditions on COD / BOD removal, sludge yield, biogas production rate (so did the biogas yield and methane yield), TMP growth, and cross-flow gas velocity while with strength on FLUX and sewage wastewater treatment capacity. Long HRTs, for example HRT 6h and 8h in this study, were shown a relatively low level of FLUX as well as the sewage wastewater treatment capacity which resulted an underutilization operation for the AnMBR system during treatment process.

As a result, compared the HRTs implemented at room temperature in this study, the suitable HRT was considered to be 6 hours.

Table 4.9 Comprehensive comparison in different HRTs.

HRT (h)	12	8	6	4
COD_{RE} (%)	89.3	87.3	88.7	**84.4**
BOD_{RE} (%)	92.2	92.0	94.0	**89.3**
SS_{RE} (%)	100	100	100	100
MLSS (g/L)	6.6~9.6	7.7~19.9	10.1~14.7	7.5~17.6
Sludge yield (g-VSS/g-COD_{rem})	0.07	0.11	0.11	**0.22**
Biogas production rate (L-gas/L-water)	0.10	0.10	0.09	**0.06**
Methane content	78.77	81.60	79.35	79.25
Biogas yield (L-gas/g-COD_{rem})	0.30	0.28	0.25	**0.20**
Methane yield (L-CH_4/g-COD_{rem})	0.24	0.23	0.20	**0.16**
FLUX (m/d)	0.25*	**0.17**	0.23	0.34
TMP growth (kPa/d)	- (low)	1.2	1.4	**16.3**
CFGV (m/h)	116	116	116	**174**
Temperature (m/h)	25	25	25	25
Treatment capacity (L/d)	**39.5**	59.8	79.6	117.3
pH-eff	6.94	6.84	6.83	6.84
ORP (mV)	-323	-328	-319	-332

*Note: different membrane module in HRT 12h.

4.4. Conclusions

As with the conditions of HRT ranged from 12 to 4 hours operated for almost one year, the results so far show that:

(1) Applying submerged AnMBR to treating the real sewage achieved high COD removal efficiency (89%) with sludge yield as low as 0.07 ~ 0.11 g-VSS/g-COD_{rem} in HRTs from 6 to 12 hours at 25 degrees.

(2) High energy recovery potential can be generated at 25 degrees as total COD conversion to CH_4 was obtained 68 ~ 75% during HRTs in 6 to 12 hours (methane gas yield: 0.20 ~ 0.24 L-gas/g-COD_{rem}).

(3) Organic loading rate was challenged as high as 2.05 g-COD/L/d in HRT 4 hours and obtained good performance of 84% COD removal efficiency with 0.22g-VSS/g-COD_{rem} of sludge yield.

REFRENCES:

APHA, 2005. Standard Methods for the Examination of Water and Wastewater, 21st ed., America Water Works Association and Water Environment Federation, Wahington, DC, USA.

Bai, R., Leow, H.F., 2002. Microfiltration of activated sludge wastewater—the effect of system operation parameters. Sep. Purif. Technol. 29, 189–198. https://doi.org/10.1016/S1383-5866(02)00075-8

Chen, R., Nie, Y., Hu, Y., Miao, R., Utashiro, T., Li, Q., Xu, M., Li, Y.Y., 2017a. Fouling behaviour of soluble microbial products and extracellular polymeric substances in a submerged anaerobic membrane bioreactor treating low-strength wastewater at room temperature. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2017.02.046

Chen, R., Nie, Y., Ji, J., Utashiro, T., Li, Q., Komori, D., Li, Y.-Y., 2017b. Submerged anaerobic membrane bioreactor (SAnMBR) performance on sewage treatment: removal efficiencies, biogas production and membrane fouling. Water Sci. Technol. 76, 1308–1317. https://doi.org/10.2166/wst.2017.240

Chen, R., Nie, Y., Tanaka, N., Niu, Q., Li, Q., Li, Y.Y., 2017c. Enhanced methanogenic degradation of cellulose-containing sewage via fungi-methanogens syntrophic association in an anaerobic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.09.046

Gao, D.-W., Lee, Y.H., Wong, C.-Y., Yeh, D.H., Zhang, T., Tang, C.-Y.Y., Criddle, C.S., Wu, W.-M., 2010. Membrane fouling in an anaerobic membrane bioreactor: Differences in relative abundance of bacterial species in the membrane foulant layer and in suspension. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2010.08.031

Garuti, G., Dohanyos, M., Tilche, A., 1992. Anaerobic-Aerobic Combined Process for the Treatment of Sewage with Nutrient Removal: The Ananox® Process. Water Sci. Technol. 25, 383–394. https://doi.org/10.2166/wst.1992.0170

Gouveia, J., Plaza, F., Garralon, G., Fdz-Polanco, F., Peña, M., 2015. Long-term operation of a pilot scale anaerobic membrane bioreactor (AnMBR) for the treatment of municipal wastewater under psychrophilic conditions. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2015.03.002

Ho, J., Sung, S., 2009. Effects of solid concentrations and cross-flow hydrodynamics on microfiltration of anaerobic sludge. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2009.08.047

Hongjun, L., Jianrong, C., Fangyuan, W., Linxian, D., Huachang, H., 2011. Feasibility evaluation of submerged anaerobic membrane bioreactor for municipal secondary wastewater treatment. Desalination. https://doi.org/10.1016/J.DESAL.2011.06.058

Hu, A.Y., Stuckey, D.C., 2006. Activated Carbon Addition to a Submerged Anaerobic Membrane Bioreactor: Effect on Performance, Transmembrane Pressure, and Flux. J. Environ. Eng. https://doi.org/10.1061/(asce)0733-9372(2007)133:1(73)

Huang, X., Gui, P., Qian, Y., 2001. Effect of sludge retention time on microbial behaviour in a submerged membrane bioreactor. Process Biochem. https://doi.org/10.1016/S0032-9592(01)00135-2

Huang, Z., Ong, S.L., Ng, H.Y., 2011. Submerged anaerobic membrane bioreactor for low-strength wastewater treatment: Effect of HRT and SRT on treatment performance and membrane fouling. Water Res.

https://doi.org/10.1016/j.watres.2010.08.035

Judd, S., 2010. The MBR book: principles and applications of membrane bioreactors for water and wastewater treatment. Elsevier.

Le Corre, K.S., Valsami-Jones, E., Hobbs, P., Parsons, S.A., 2009. Phosphorus recovery from wastewater by struvite crystallization: A review. Crit. Rev. Environ. Sci. Technol. 39, 433–477.

Lei, Z., Yang, S., Li, Y. you, Wen, W., Wang, X.C., Chen, R., 2018. Application of anaerobic membrane bioreactors to municipal wastewater treatment at ambient temperature: A review of achievements, challenges, and perspectives. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2018.07.050

Martin-Garcia, I., Monsalvo, V., Pidou, M., Le-Clech, P., Judd, S.J., McAdam, E.J., Jefferson, B., 2011. Impact of membrane configuration on fouling in anaerobic membrane bioreactors. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2011.07.042

Metzger, U., Le-Clech, P., Stuetz, R.M., Frimmel, F.H., Chen, V., 2007. Characterisation of polymeric fouling in membrane bioreactors and the effect of different filtration modes. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2007.06.016

Nie, Y., Kato, H., Sugo, T., Hojo, T., Tian, X., Li, Y.Y., 2017a. Effect of anionic surfactant inhibition on sewage treatment by a submerged anaerobic membrane bioreactor: Efficiency, sludge activity and methane recovery. Chem. Eng. J. https://doi.org/10.1016/j.cej.2017.01.022

Nie, Y., Niu, Q., Kato, H., Sugo, T., Tian, X., Li, Y.Y., 2017b. Efficient methanogenic degradation of alcohol ethoxylates and microbial community acclimation in treatment of municipal wastewater using a submerged anaerobic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.11.128

Nie, Y., Tian, X., Zhou, Z., Li, Y.Y., 2017c. Impact of food to microorganism ratio and alcohol ethoxylate dosage on methane production in treatment of low-strength wastewater by a submerged anaerobic membrane bioreactor. Front. Environ. Sci. Eng. https://doi.org/10.1007/s11783-017-0947-1

Pandey, P., Shinde, V.N., Deopurkar, R.L., Kale, S.P., Patil, S.A., Pant, D., 2016. Recent advances in the use of different substrates in microbial fuel cells toward wastewater treatment and simultaneous energy recovery. Appl. Energy 168, 706–723. https://doi.org/https://doi.org/10.1016/j.apenergy.2016.01.056

Rulkens, W., 2008. Sewage Sludge as a Biomass Resource for the Production of Energy: Overview and Assessment of the Various Options. Energy & Fuels 22, 9–15. https://doi.org/10.1021/ef700267m

Watanabe, A., 2004. Studies on membrane separation engineering for multi-component liquid food. Nippon Shokuhin Kagaku Kogaku Kaishi. https://doi.org/10.3136/nskkk.51.55

Watanabe, R., Nie, Y., Takahashi, S., Wakahara, S., Li, Y.Y., 2016. Efficient performance and the microbial community changes of submerged anaerobic membrane bioreactor in treatment of sewage containing cellulose suspended solid at 25 °C. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2016.05.049

Wu, J., Le-Clech, P., Stuetz, R.M., Fane, A.G., Chen, V., 2008. Effects of relaxation and backwashing conditions on fouling in membrane bioreactor. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2008.06.057

Yao, Y.C., Zhang, Q.L., Liu, Y., Liu, Z.P., 2013. Simultaneous removal of organic matter and nitrogen by a heterotrophic nitrifying-aerobic denitrifying bacterial strain in a membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2013.05.120

Zakkour, P.D., Gaterell, M.R., Griffin, P., Gochin, R.J., Lester, J.N., 2001. Anaerobic treatment of domestic wastewater in temperate climates: Treatment plant modelling with economic considerations. Water Res. https://doi.org/10.1016/S0043-1354(01)00145-2

Chapter 5

Effect of temperature on treatment performance of MF-MBR

5.1 Introduction

Recently, researches on wastewaters treatment solutions are tend to achieve recovery or recycling purpose of energy and resources during the process for pollutants removal (Zakkour et al., 2001). Technologies have already been used for treating some kinds of industrial wastewaters or waste sludge and achieved the recovery or reuse of energy and resources have been reported (Le Corre et al., 2009; Pandey et al., 2016; Rulkens, 2008) while it is still a bit backward for applying in sewage treatment. The anaerobic membrane bio-reactor (AnMBR) integrates anaerobic digestion and membrane technology creating a new process which provided with the potential of energy recovery through the methane fermentation process in anaerobic digestion as well as the high efficiency of sludge-water separation due to the filtration by membrane (Bai and Leow, 2002). It also has been successfully applied in dealing with industrial wastewater and is considered to be a trustworthy process (Lin et al., 2013). If the process could be possibly applied for treating sewages as well, the wastewater treatment plants could also become net suppliers of energy, renewable resources and reclaimed water which would make a big contribution for the construction of sustainable social development.

Some studies that related to the low organic strength wastewater or sewage based on man-made synthetic wastewater reported have confirmed that it is possible to be used for sewage treatment with a high efficiency of organic removal with less waste sludge produced as well as energy recovery from the produced biogas (Lei et al., 2018). A previous research based on synthetic sewage wastewater investigated the response of AnMBR to the temperature decreased from 25 to 10 on organic removal, membrane fouling and the microbial community while lack of the biogas production performance (Watanabe et al., 2017). Furthermore, it is also unknown that it could be able to obtain the same effect when dealing with the real sewage wastewater.

Based on the results of successfully started AnMBRs by the real sewage wastewater and decided the pore size of membrane described in Chapter 3 and investigated the effect of HRT in Chapter 4, the mini-pilot AnMBR was operated in the wastewater treatment plant at HRT condition of 6 hours and continued to dealing with the real sewage by operated temperatures from 25°C, 20°C then 15°C (HRT 6 to 24 hours) in this Chapter. Then the performance of sewages treated by AnMBR in each temperature condition and the different HRTs at low temperature condition was investigated in aspects on pollutant removal performance, gas yield, sludge yield, COD balance as well as the filtration performance of the membranes.

5.2. Materials and methods

5.2.1 Consist and operation of the mini-pilot

The same as mentioned in 4.2.1 of Chapter 4.

5.2.2 Operation conditions of the mini-pilot

In this experiment, the long-term operation of AnMBR fed by only the real sewage wastewater. The details of the operation conditions such as temperatures and HRTs are shown in table 5.1.

5.2.3 Samples collection and analysis methods

The same as mentioned in 4.2.3 of Chapter 4.

Table 5.1 Detail experiment conditions during long-term operation.

Operated period (Day-)	1-39	40-75	76-108	109-136	137-162	163-199
Temperature (°C)	25	20	15	15	15	15
HRT (h)	6	6	6	12	24	16
OLR (g-COD/L/d)	1.64	1.61	1.54	0.85	0.42	0.52
FLUX (m/d)	0.23	0.23	0.23	0.12	0.06	0.09
CFGV* (m/h)	116	116	116/174	116	116	116
Permeate mode	4mins on / 1min off					

CFGV* is the cross-flow gas velocity (m/h).

5.2.4 EPS and SMP detection

SMP and EPS were measured as those of carbohydrate and protein concentration. A mixed liquor sample with 20 mL was centrifuged for 15 minutes at 8000 rpm at 4°C and the supernatant was then filtered through a 0.45 μm filter. The obtained filtrate represented the SMP. EPS was obtained using a cation exchange resin (DOWEX R Marathon C, Na^+ form, Sigma-Aldrich, USA) extraction method. A mixed liquor sample with 20 mL was centrifuged for 15 minutes at 8000 rpm at 4°C and the sediments were re-suspended with a buffer solution (2 mmol/L Na_3PO_4, 4 mmol/L NaH_2PO_4, 9 mmol/L NaCl and 1 mmol/L KCl). Afterwards, resin (70 g/g-VSS) was added and mixed for 1 hour at 800 rpm. The mixture was first centrifuged for 10 mins at 8000 rpm and the obtained supernatant was then re-centrifuged for 10 mins at 8000 rpm. The finally obtained supernatant represented the EPS. The carbohydrate in SMP and EPS was determined using H_2SO_4/phenol oxidation and a colorimeter method, and the protein was measured using the Folin-Ciocalteu method. All analyses were conducted in two replicates.

5.2.5 Batch test for SMA

The batch test was carried out in 120 mL glass serum bottles using mixed liquor taken from the mini-pilot AnMBR during the stable operation period in each HRT or temperature condition as the test sludge. The mixed liquor was sampled by sealed sample sterilization bottles to keep the oxygen out. In each serum bottle, 40.0 mL raw mixed liquor was added with 0.5 mL $Na_2S{\cdot}9H_2O$ solution (250 mg/L as a final concentration in the vial) which was used as the reducing agent by injected into each bottle on purpose of obtain absolutely anaerobic condition. There was no further nutrient solution added due to it enriched 1000 to 2000 mg/L COD in the raw mixed liquor and the real sewage wastewater components used as the nutrient solution makes it more close to reflect out the real specific methanogenic activity (SMA). The total volume of the liquid was fixed to 50 mL by 9.5 mL distilled water refilled, thus the headspace in each serum bottles was 70 mL obtained. Then serum bottles were sealed with rubber stoppers and secured by aluminum crimp. Oxygen in headspace of the bottles was purged with nitrogen gas for 10 minutes. Distilled water or other solutions were also pre-treated by the nitrogen gas. The temperature was controlled around 35 degrees and the start timing was about 15 minutes later after discharged of the headspace gas expanded by heating. Then, biogas production and composition was measured every 3 ~ 5 hours according to the biogas volume and expressed as the value at standard condition (Nie et al., 2017b). The samples in each condition was conducted in two or three replicates to ensure its reliability.

5.3 Results and discussion on temperature effect

5.3.1 Organic pollutant removal

Figure 5.1 and figure 5.2 shows COD / BOD in influent and effluent and the removal efficiency. Figure 5.3 shows the average COD and BOD in effluent as well as the average removal efficiency for COD, BOD and SS at different temperature conditions. According to the result, the effluent COD was under 50 mg/L and the effluent BOD was under 20 mg/L in temperature 25°C and 20°C. In those two temperature conditions the average COD and BOD in effluent was about as low as 40 mg/L and 13 mg/L, respectively. But after the temperature was decreased to 15°C, COD and BOD in effluent was increased gradually and resulted the removal efficiency decreased day by day. The average COD and BOD in effluent was about 93 mg/L and 35 mg/L, respectively. Soluble COD in influent was measured as an average of 124 mg/L which equivalently that only around 25% of the soluble organic in the real sewage wastewater was biodegraded through the reactor. The removal efficiency of COD and BOD at temperature 15°C was only obtained 76.6% and 81.9%, respectively, even lower than that in HRT 4h at 25°C described in Chapter 4. And the around 80% removal efficiency was actually mainly contributed by the membrane filtration, considering the value of soluble COD in influent and COD in effluent. The above analysis shows that low temperature reduces the methanogenic activity and lead out the unsatisfactory performance for organic matters removal. As the previous study reported that the acetoclastic methanogen was more sensitive to the temperature than the hydrogenotrophic methanogen (Watanabe et al., 2017, 2014), it can be known that acetoclastic methanogen is the inhibitory factor in dealing with the real sewage wastewater in low temperature such as 15°C. In addition, pH and ORP was also obtained stable during the long-term operation which were shown in figure 5.4.

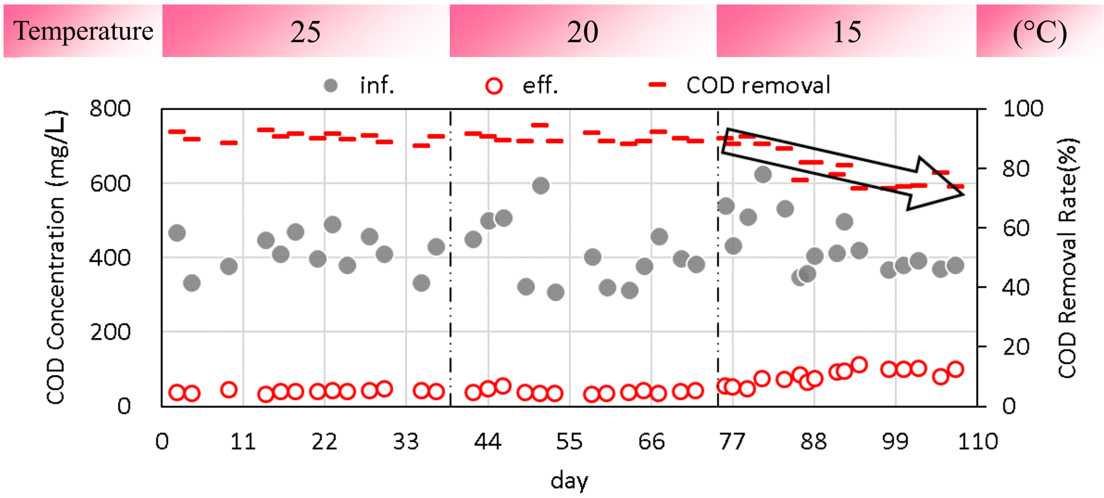


Fig. 5.1 COD in influent and effluent and the removal efficiency.

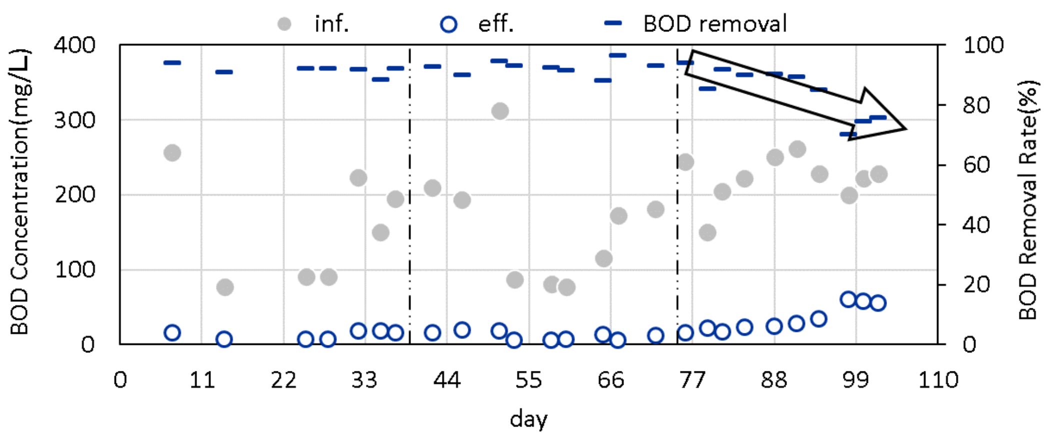


Fig. 5.2 BOD in influent and effluent and the removal efficiency.

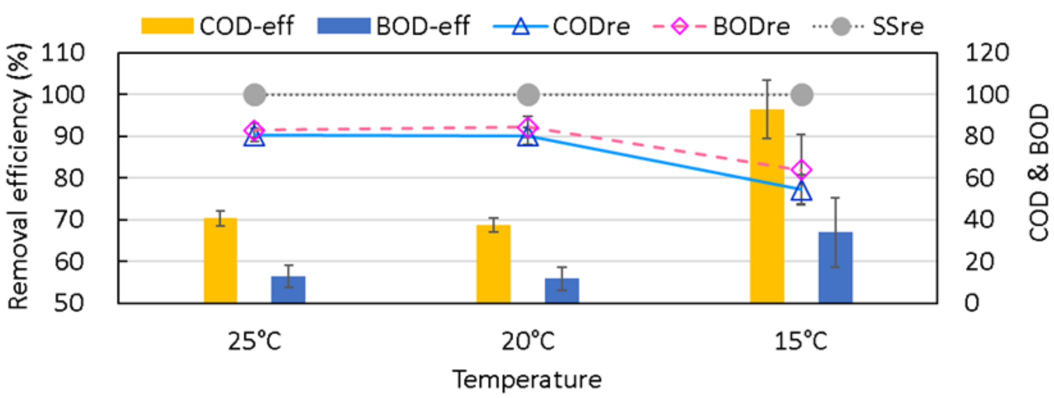


Fig. 5.3 Average COD, BOD, SS performance in different temperatures.

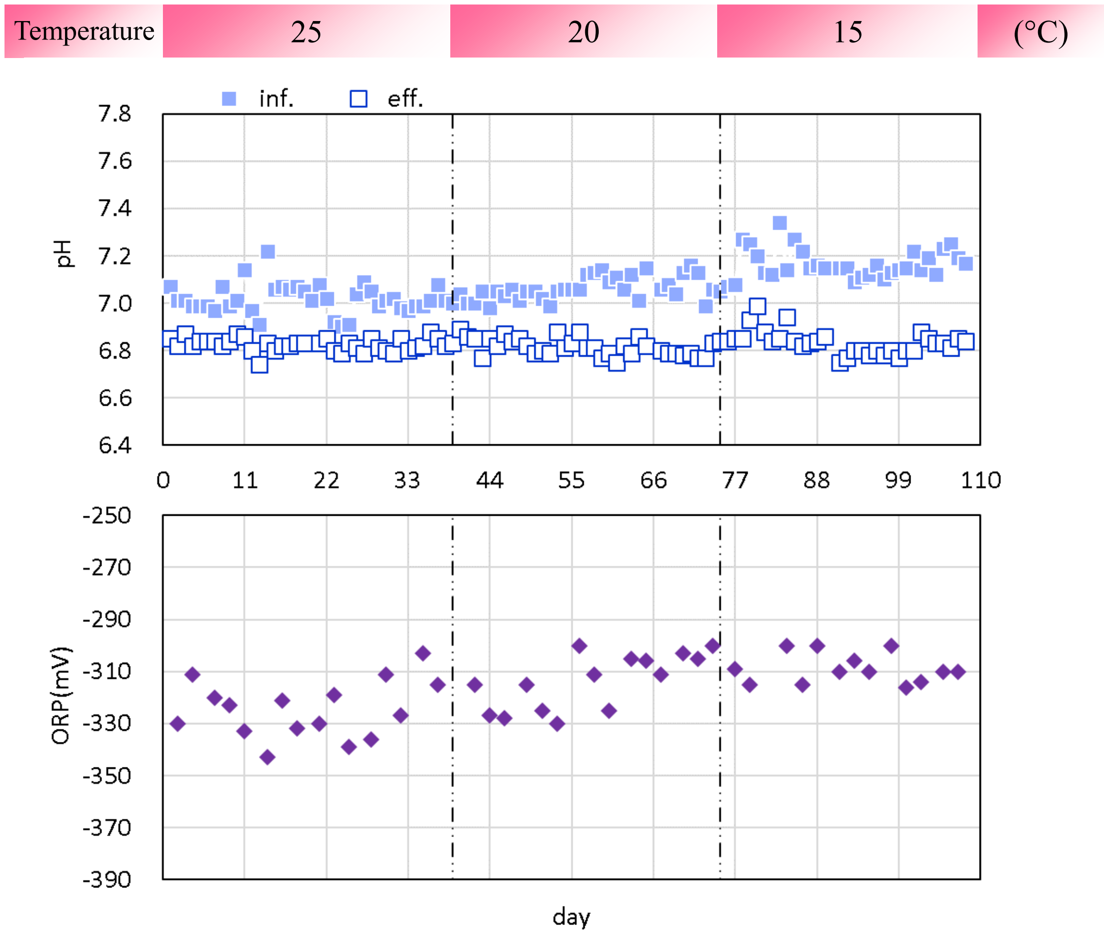


Fig. 5.4 pH and ORP during the long-term operation.

5.3.2 Sludge yield

Figure 5.5 shows the MLSS and MLVSS during the long-term operation experiment. The MLSS was strictly maintained in a range of 12 ~ 14.5 g/L and the MLVSS was in a range of 9.5 ~ 11.5 g/L at the conditions of temperature above 20°C. However, as it emerged the high operation pressure for TMP since temperature was decreased to 15°C, MLSS and MLVSS was controlled a bit lower in purpose of reduce the pressure during the membrane filtration process. Because of the controlling of MLSS and MLVSS, the mixed liquor inside the reactor was discharged diurnal due to a rapid growth rate of sludge in low temperature condition, which is known as the sludge yield (figure 5.6). In the case of continuous accumulation of SS/VS with a low biodegradation progress, the concentration of functional bacteria was diluted by the diurnal discharge of mixed liquor (Chen et al., 2017a). Thus, created a vicious circle and resulted, as one of the reasons, a worse biodegradation process.

According to the sludge yield at different temperature conditions calculated by figure 5.6 and shown in figure 5.7, it is obvious that the sludge yield increased along with the temperature decreased. The sludge yield in temperature 25°C was only 0.11 g-VSS/g-COD_{rem}, and increased to 0.20 g-VSS/g-COD_{rem} in temperature 20°C while then the biggest sludge yield of as high as 0.35 g-VSS/g-COD_{rem}, presented in the low temperature condition of 15°C, did not present any advantage comparing with the conventional aerobic activated sludge process which the sludge yield in a range of 0.25~0.4 g-VSS/g-COD_{rem} has been reported (Huang et al., 2001).

Therefore, it can draw a conclusion that the organics removal performance was able to remain as the temperature was above 20°C, but it become worse and the sludge yield presented sharp increase after the temperature was decreased to 15°C.

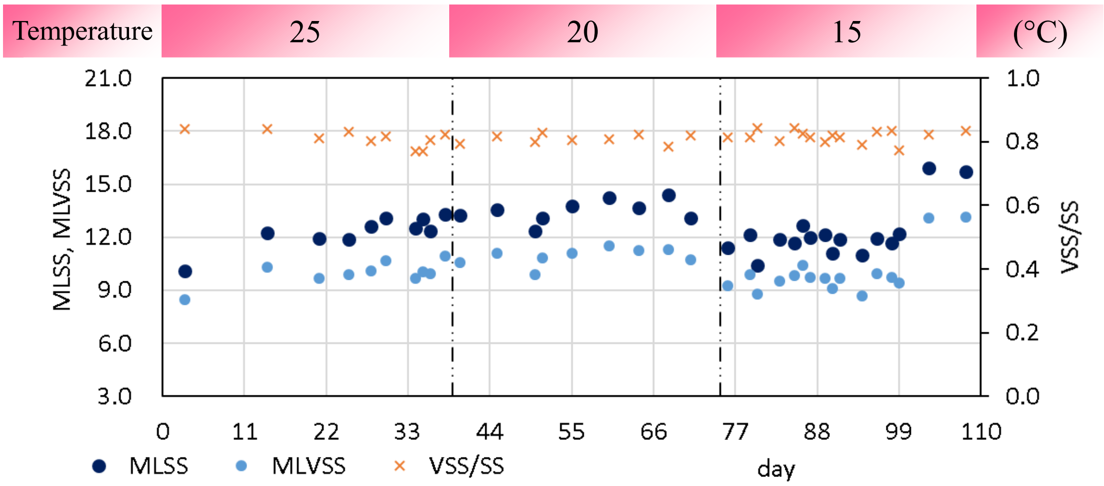


Fig. 5.5 MLSS and MLVSS during the long-term operation.

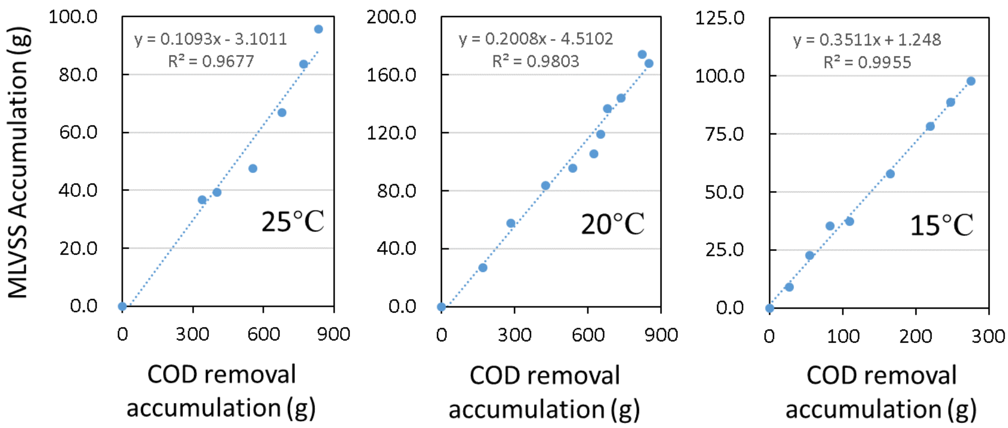


Fig. 5.6 Sludge yield calculation in each temperature condition.

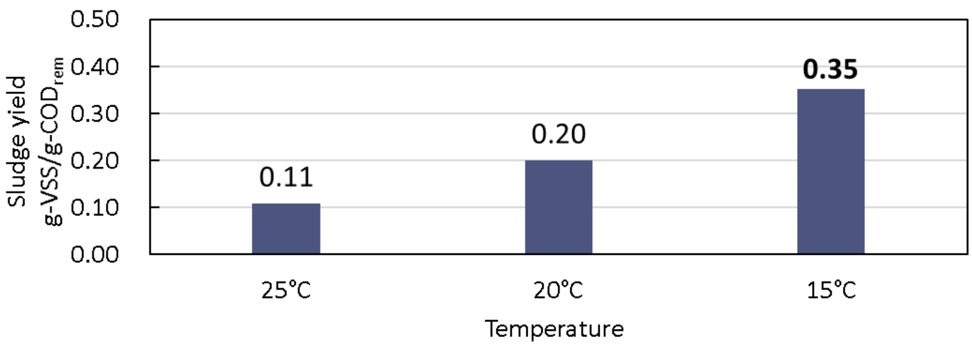


Fig. 5.7 Sludge yield in different temperature conditions.

5.3.3 Biogas production

Figure 5.8 and figure 5.9 shows the biogas production rate and gas composition during the long-term operation experiment. The daily biogas production rate was ranged from about 0.07 L-gas/L-water to 0.10 L-gas/L-water in temperature 25°C and 0.05 L-gas/L-water to 0.09 L-gas/L-water in temperature 20°C while declined significantly when the temperature was decreased to 15°C which only 0.02 ~ 0.04 L-gas/L-water biogas production rate was achieved. On the side of gas composition, methane gas content was stalely obtained around 80% when temperature was above 20°C while it was showed a significant day-to-day decline when decreased to low temperature as 15°C. The final daily methane gas content as operated for a month was only declined to 60% from 80% and it was considered to be not a decline end if the same condition operation continues as showed by the trend. Meanwhile, the nitrogen gas content was raised day by day. While as it is known that no nitrogen gas was produced from the anaerobic digestion process and the dissolved nitrogen gas in the sewage was the same theoretically. But poor biodegraded performance in low temperature condition resulted less methane gas produced as well as low biogas production rate obtained is considered to be the reason for the decline of methane gas content and growth of nitrogen gas content.

The average biogas production rate and biogas yield (including methane gas yield) in each temperature condition is shown in table 5.2 achieved average biogas production rate as high as 0.09 L-gas/L-water in temperature 25°C and 0.07 L-gas/L-water in temperature 20°C, basically the same level as those obtained in Chapter 4. Then it was dropped to only 0.03 L-gas/L-water after temperature was decreased to 15°C since the daily biogas production rate was declined day-to-day. In addition, the average methane gas content was declined to 67% for a month's operation and the CO_2 content also obtained less in temperature 15°C than the conditions of temperature above 20°C which bore out what was described above as of the poor biodegraded performance presented in low temperature condition.

Table 5.2 Average biogas performance in each temperature condition.

Temperature (°C)	25	20	15
Biogas production rate (L-gas/L-water)	0.09	0.07	0.03
CH_4 (%)	80.19	79.48	66.85
N_2 (%)	13.8	15.47	29.41
CO_2 (%)	6.01	5.05	3.74
Biogas yield (L-gas/g-COD_{rem})	0.23	0.20	0.09
Methane yield (L-CH_4/g-COD_{rem})	0.18	0.16	0.06

Biogas yield and methane yield in each temperature condition also calculated and showed in table 5.2. The decline of biogas yield in low temperature was more than that above 20°C temperature conditions because of less COD was removed by the low temperature condition and the decline of methane yield in low temperature was further more less as the lower methane gas content. Table 5.3 shows the comparison of the CH_4 yield with some previous researches published as well as Chapter 4. It is obviously that methanogenesis was performed well by feeding the real sewage wastewater in conditions of temperature above 20°C. But methanogenesis was performed worse in low temperature such as 15°C set in this long-term experiment.

The biogas performance showed that though the biogas performance drops as the temperature decrease, treating the real sewage in a condition of above 20°C can achieve a relatively high methanogenesis or biogas production performance but it is not ideal if the temperature be decreased as low as 15°C.

Table 5.3 Comparison of the CH_4 yield with other similar published reports.

Tem. (°C)	HRT (h)	CH_4 yield ($L\text{-}CH_4/g\text{-}COD_{rem}$)	AnMBR type	References
25	6 ~ 12	0.21 ~ 0.22	Submerged	(Ho and Sung, 2009)
20	7 ~ 17	0.20	Side-stream	(Gouveia et al., 2015)
25	8 ~ 48	0.27 ~ 0.33	Submerged	(Chen et al., 2017b)
25	4 ~ 24	0.16 ~ 0.24	Submerged	Chapter 4
20 ~ 25	6	**0.16 ~ 0.18**	Submerged	This study
15		**0.07**		

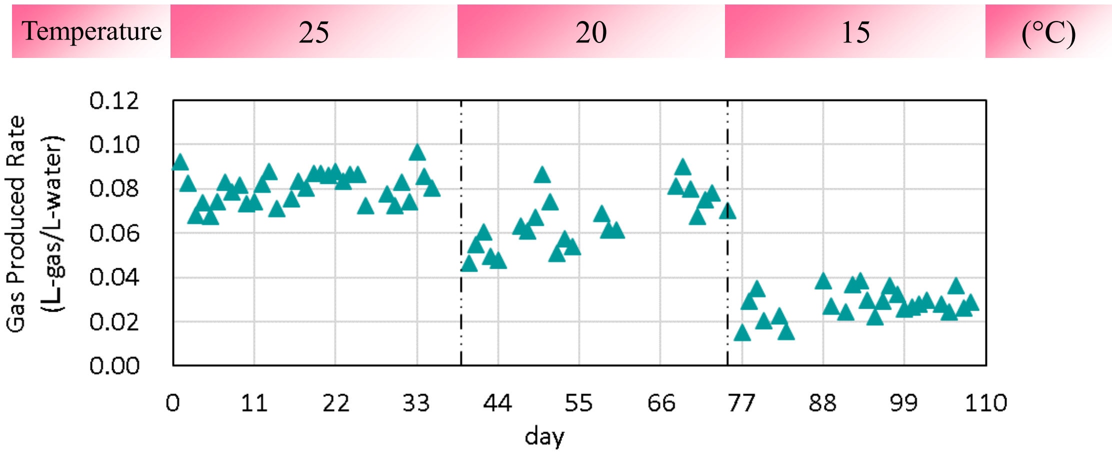


Fig. 5.8 Biogas production rate during the long-term operation.

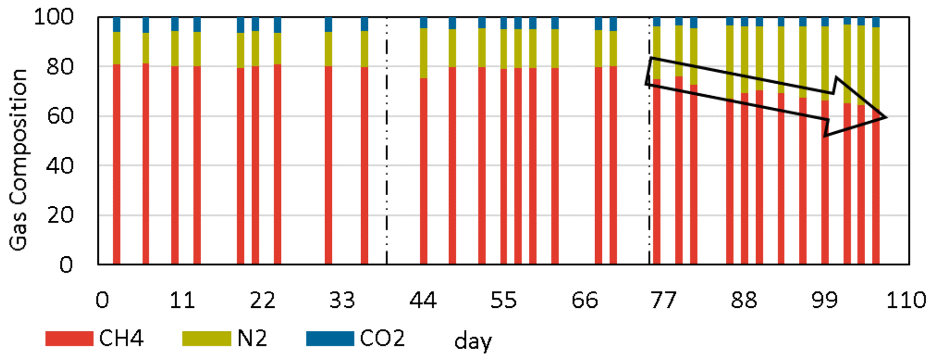


Fig. 5.9 Biogas composition during the long-term operation.

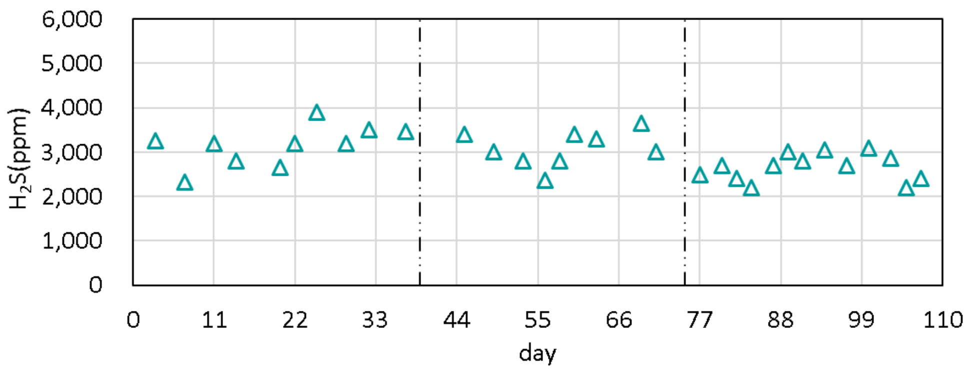


Fig. 5.10 H_2S concentration during the long-term operation.

5.3.4 COD balance analysis

Based on the results of effluent COD, sludge yield, biogas production, combine with the measured value of H_2S and dissolved methane, COD balance at different HRT conditions was calculated as shown in table 5.4 as absolute value of g-COD/d unit and figure 5.11 as percentage values. From the result, it is found that COD converted to produced biogas was decreased following the temperature decreased. From 25°C to 20°C condition, the decreased biogas amount was almost those of increased waste sludge amount and the same COD remained in effluent indicated out that during temperature 25°C operation the purification of sewage was mainly contributed by the methanogenesis though the anaerobic digestion thus the membrane filtration was used just as a sludge-water separation, but during temperature 20°C operation the purification process was contributed by a combined action from both the biodegradation as well as the membrane filtration. So, the membrane filtration can be used as a supporter if the biodegradation was performed poor when the temperature decreased. Then it was found that if the temperature continuous to drop to 15°C, the amount of sludge growth kept on increasing but the COD remained in effluent increased as well. This result showed that the support effects from membrane filtration was not enough for the micro-biological degradation decreased to treating the real sewage in HRT 6 hours if the temperature was set as low as 15°C.

In addition with the dissolved methane, the COD in influent conversion to CH_4 was more than 60% at temperature higher than 20°C by the anaerobic degradation indicating that the biogas-energy recovery efficiency could possibly achieved up to 60% (table 5.5). After the temperature was declined to 15°C, the COD in influent conversion to CH_4 generated sharp declines to only 34.4% as only 23.5% amount was in produced biogas and 14.7% was presented as dissolved methane. The reason was mentioned above as methanogenesis was performed worse in low temperature condition of 15°C.

As a conclusion of COD balance result, it can be obtained that a good performance on sewage treatment performance with a high biogas-energy recovery efficiency can be achieved if the reaction temperature condition for AnMBR is above 20°C. Moreover, it can draw out that the reason for organic removal efficiency and biogas type energy recovery efficiency reduced in low temperature of 15°C was showed to be the low activity of the micro-biological degradation. Besides, unlikely to the HRT 4h condition at temperature 25°C can achieve a struggle operation as the purification of sewage can be ensured described in Chapter 4, the condition for HRT 6h at temperature 15°C cannot be used to the real sewage treatment because of the poor performance of the sewage purification by the decreased methanogenesis activity at low temperature.

Table 5.4 COD balance values in different temperatures (g-COD/d).

Temperature (°C)	25	20	15
COD-in	32.3	31.4	30.7
Biogas production	19.1	15.0	6.0
H_2S in biogas	0.06	0.05	0.02
Dissolved biogas	3.5	4.4	4.5
Sludge growth	4.2	7.5	10.9
COD-eff	3.2	3.0	7.2

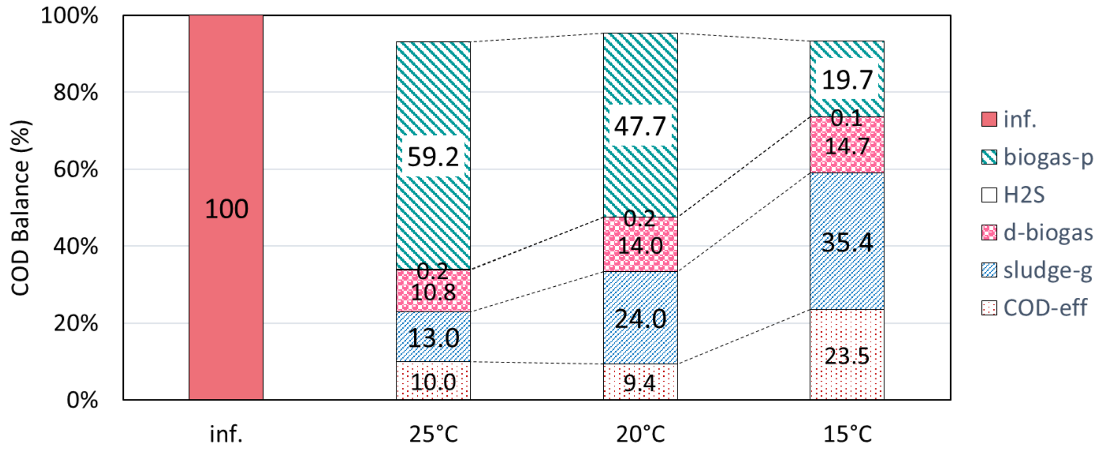


Fig. 5.11 COD balance in different temperatures (%).

P.S.

inf.: COD in influent;

biogas-p: biogas produced (calculated by the discharged methane gas);

H2S: H_2S in the biogas produced;

Sludge-g: sludge growth;

COD-eff: COD in permeate effluent.

Table 5.5 COD conversion to CH_4 in different temperatures.

Temperature (°C)	25	20	15
To biogas CH_4 (%)	59.2	47.7	19.7
To dissolved CH_4 (%)	10.8	14.0	14.7
Total (%)	70.0	61.7	34.4

5.3.5 SMA result

Figure 5.12 shows SMA and OLR (organic loading rate, calculated as the same unit of gCOD/gVSS/d with SMA and COD_{inf} and treatment capacity for the calculation was used the average values from the whole long termed operation in each condition) in different temperatures during the long-term operation. The calculation for SMA and OLR was following the equations below:

$$SMA = \frac{V_{gas}}{k \times Time_{bat} \times V_{bat} \times MLVSS}$$

where V_{gas} is the produced biogas integral (mL); k is a constant that used as 1g COD equals 0.35L CH_4 in standard condition; $Time_{bat}$ is the time integral for the batch test (d); V_{bat} is the mixed liquor volume that used for the batch test in each serum bottles (mL); then *MLVSS* represents mixed liquor volatile suspended solid value inside the mini-pilot reactor on the day of batch test implemented (g/L).

$$OLR = \frac{Q \times COD_{inf}}{V \times MLVSS}$$

where Q is the sewage wastewater treatment capacity per day (L); COD_{inf} is the COD contained in the influent (g/L); V is the reaction volume of the AnMBR (L); *MLVSS* represents mixed liquor volatile suspended solid value inside the mini-pilot reactor on the day of batch test implemented (g/L).

The result indicated that the SMA decreased as long with the decrease of operated temperature and the poor biodegraded performance presented in 15°C was also because of the low SMA caused by the low temperature operation. Moreover, the comparison between SMA and OLR in the same conditions shown that HRT 6h cannot be used as the operation condition during 15°C treatment due to the SMA was even less than OLR.

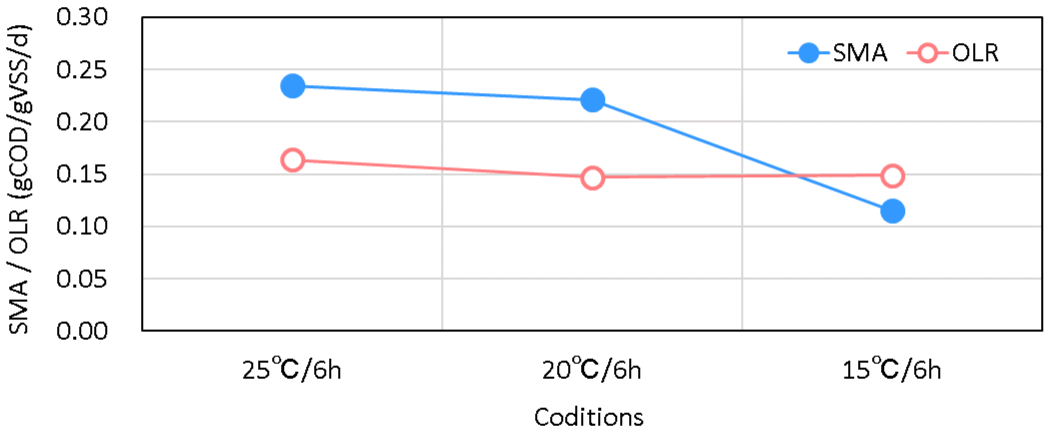


Fig. 5.12 SMA&OLR in different temperatures.

5.3.6 Membrane filtration and fouling risk

The TMP and FLUX recording data shown in figure 5.13 presents the membrane performance during the long-term operation. The CFGV was set as 116 m/h and the total membrane area was 0.345 m^2, which all were described in the method of experiment section. According to the TMP-FLUX result, TMP was stabled at a very low value when temperature was controlled at 25°C while it showed increasing after the temperature was decreased to 20°C even in the same FLUX condition. Then it showed rapidly increased as the temperature was decreased to 15°C and only obtained 2 ~ 3 days for every continuous operation period although the concentration of mixed liquor was lower than the conditions above 20°C. A comparison of the daily growth for each temperature condition was showed as figure 5.14. The figure shows that the slope of the TMP line during a stable operation period was increased apparently along with the decline of temperature condition. Furthermore, the slope increased much more in the situation of temperature 15°C than those above 20°C showed a rough operation in low temperature conditions. Then for the sake of guarantee low temperature operation, the CFGV was upgraded to 174 m/h via increasing the biogas cycling on day 93 during 15°C. Whereas, the average slope in each condition listed in table 5.6 shows that TMP increased rapidly in condition of 15°C with a bigger CGFV and that was still faster than the increases showed in 20°C even more energy was costed for the biogas cycling pumps.

The concentration of EPS and SMP shown in figure 5.15 presented that along with the temperature decreased, more EPS and SMP produced increased indicated a higher fouling risk should be taken in low temperature conditions. And the construction of protein or carbohydrate for EPS and SMP shown in figure 5.16 expresses that the reason for EPS or SMP increase since the temperature decrease was mainly caused by protein type EPS or protein type SMP because of the carbohydrate type of EPS and SMP was showed low increase by the decrease of temperature.

The results shown that applying AnMBR to the real sewage wastewater treatment, TMP and membrane fouling risk increases as the temperature decreases and it need more energy for biogas cycling pumps if the temperature is set as low as 15°C or even lower.

Table 5.6 Slope for each fitted line in figure 5.14.

Fitted line	25°C	20°C	15°C	15°C*
Average slope	0.2	1.1	11.9	1.5

Note: 15°C* is the condition of 15°C temperature with a CFGV 174 m/h.

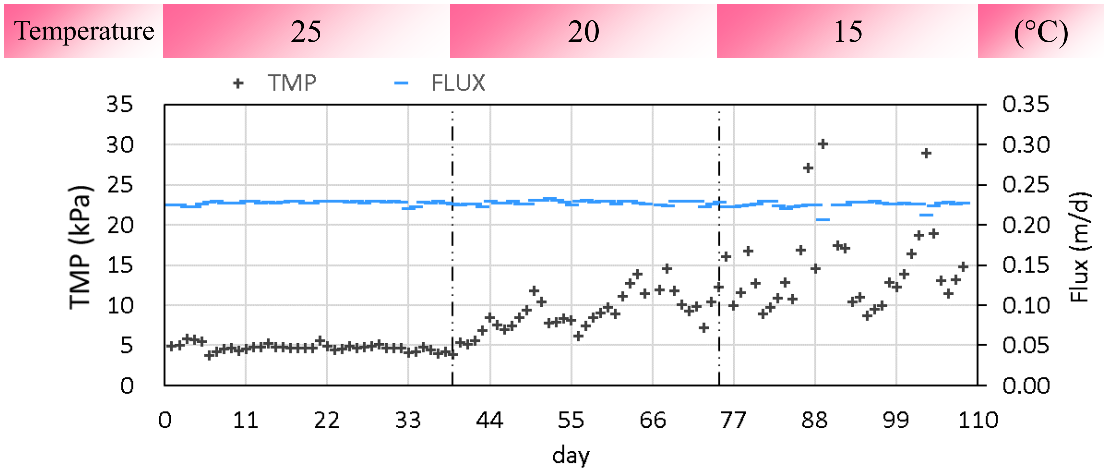


Fig. 5.13 TMP – FLUX record during the long-term operation.

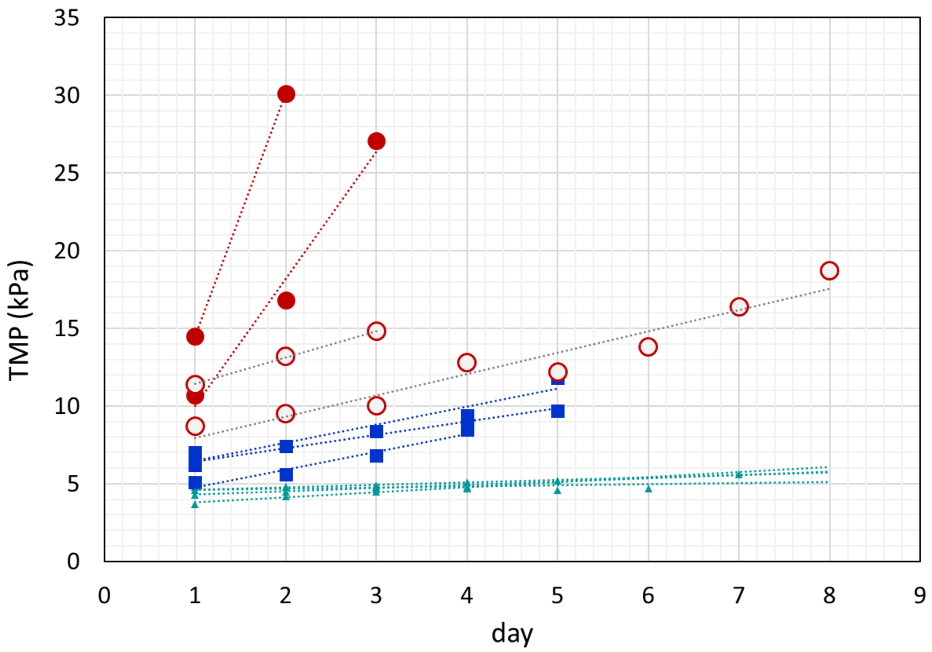


Fig. 5.14 TMP – FLUX comparison in different conditions.

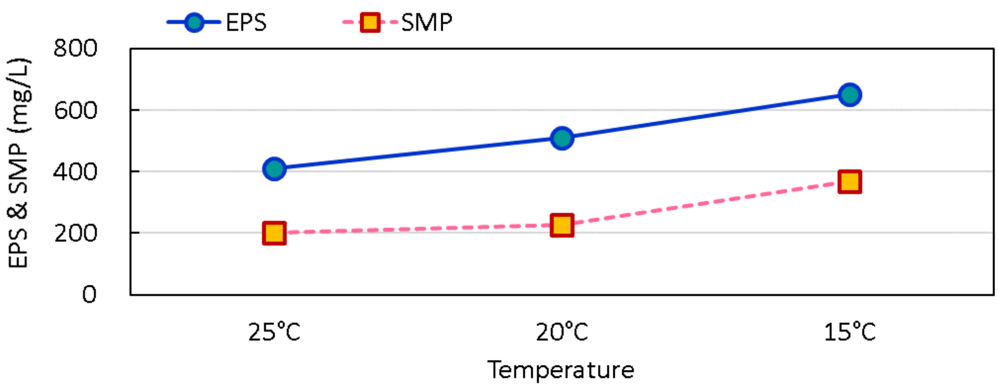


Fig. 5.15 EPS and SMP in different temperature conditions.

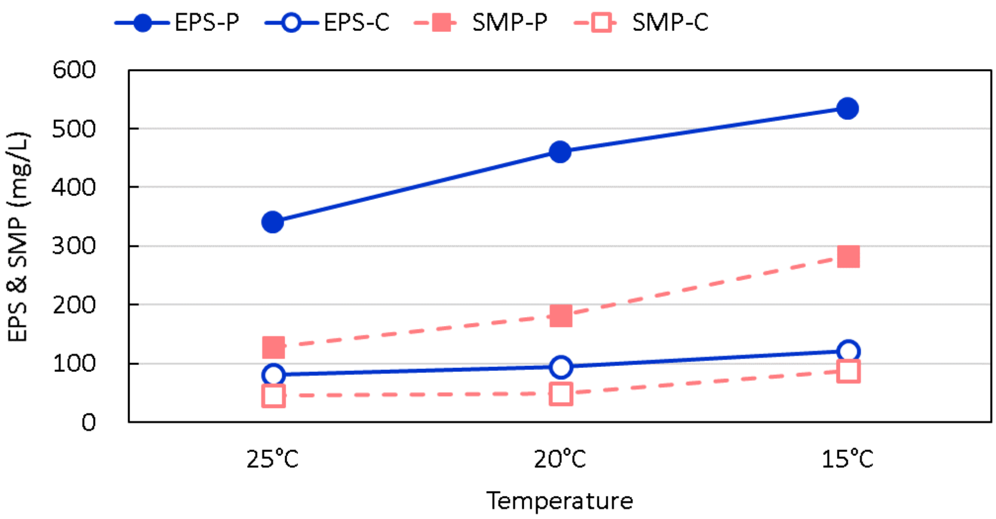


Fig. 5.16 Construction for EPS and SMP in different temperature conditions.

5.3.7 Section summarizes

As with the conditions of temperature decreased from 25°C to 15°C operated for 110 days, the conclusions can be obtained as:

(1) Applying AnMBR directly into real sewage wastewater treatment performed well in organic pollutant removal as well as energy recovery potential by biogas production at temperature above 20°C with a relatively low sludge yield.

(2) TMP as well as membrane fouling risk increases when the temperature decreases and more energy is needed for biogas cycling pumps if the temperature is set as low as 15°C or even lower.

(3) Low temperature of 15°C or even lower cannot be used for treating the real sewage wastewater in HRT 6 hours because of the poor performance of sewage purification and low biogas production performance due to the low methanogenesis activity in low temperature conditions. However, the temperature of the cities in the world showed a huge difference and the cities' temperature above 20 degrees in all season are rare which are principally tropical in distribution (Crowley, 2000; Huang et al., 2000). Therefore, it is necessary to seeking for a way to treating the sewage wastewater in low temperatures and this study implemented a continuous long termed operation experiment at 15°C.

5.4 Results and discussion at low temperature

5.4.1 Organic pollutant removal

Figure 5.17 shows COD in influent and effluent and the removal efficiency during long-term operation of HRT ranged from 6 to 24 hours at 15°C low temperature. The long-term operation result shown a trend of increase by extended HRT from 6 hours to 12 as well as 24 hours. The same situation was presented in the removal of BOD as shown in figure 5.18.

Figure 5.19 shows the average COD and BOD in effluent as well as the average removal efficiency for COD, BOD and SS at different HRTs at low temperature conditions. According to the result, the effluent COD and BOD was deceased after the HRT was extended to 12 hours and the COD was finally decreased to under 40mg/L and the effluent BOD was around 10mg/L in HRT 16 and 24 hours. Therefore, the removal efficiency of COD and BOD was recovered to above 90% in HRT 16 and 24 hours. The above analysis shows that at low temperature conditions, organic removal efficiency was increased and recovered by extending HRT condition. In addition, ORP value also obtained decreased and stabled in the long HRT condition due to the long-term operation pH and ORP shown in figure 5.20.

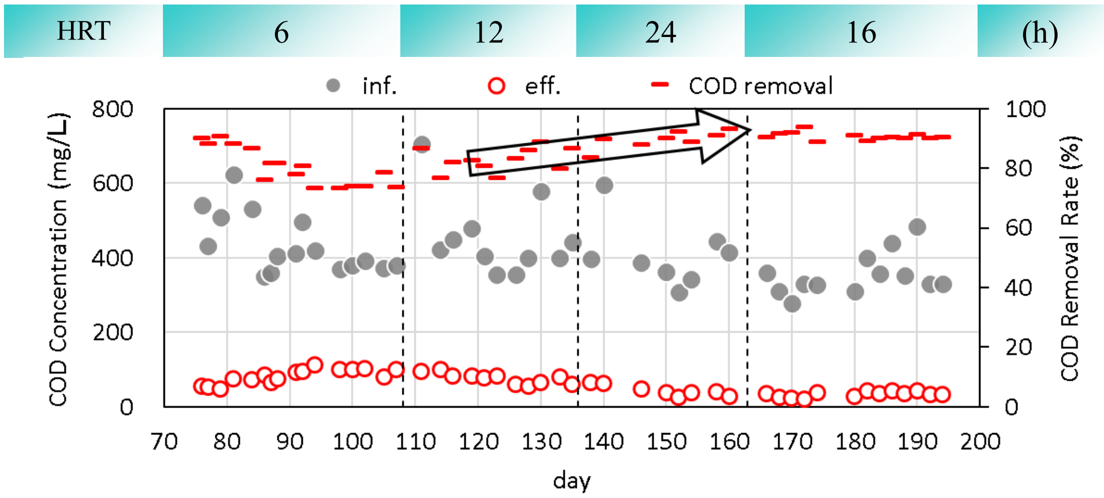


Fig. 5.17 COD in influent and effluent and the removal efficiency.

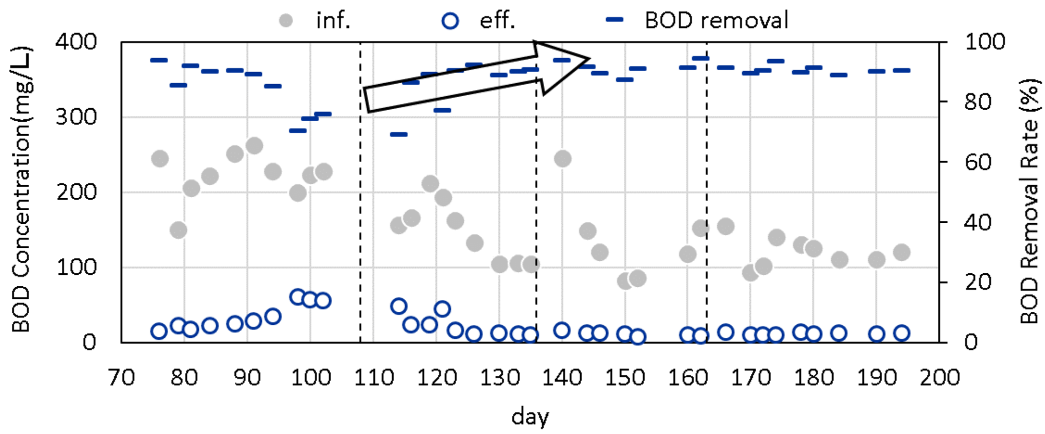


Fig. 5.18 BOD in influent and effluent and the removal efficiency.

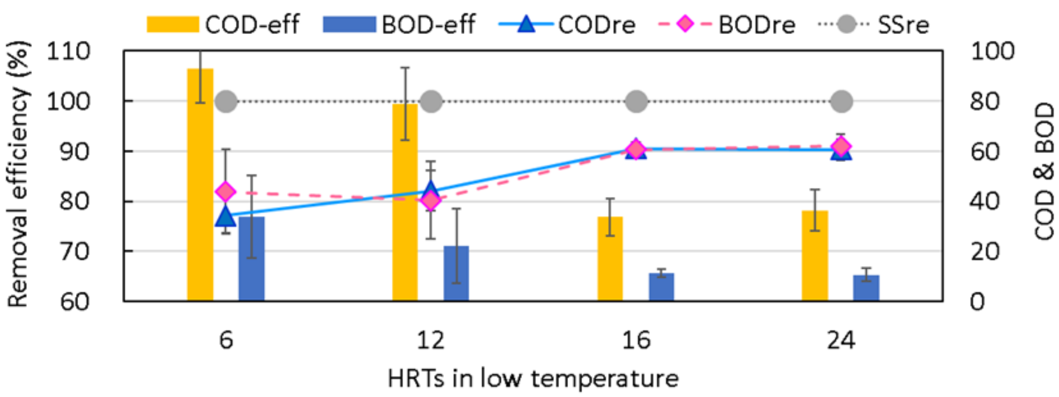


Fig. 5.19 Average COD, BOD, SS performance at low temperatures.

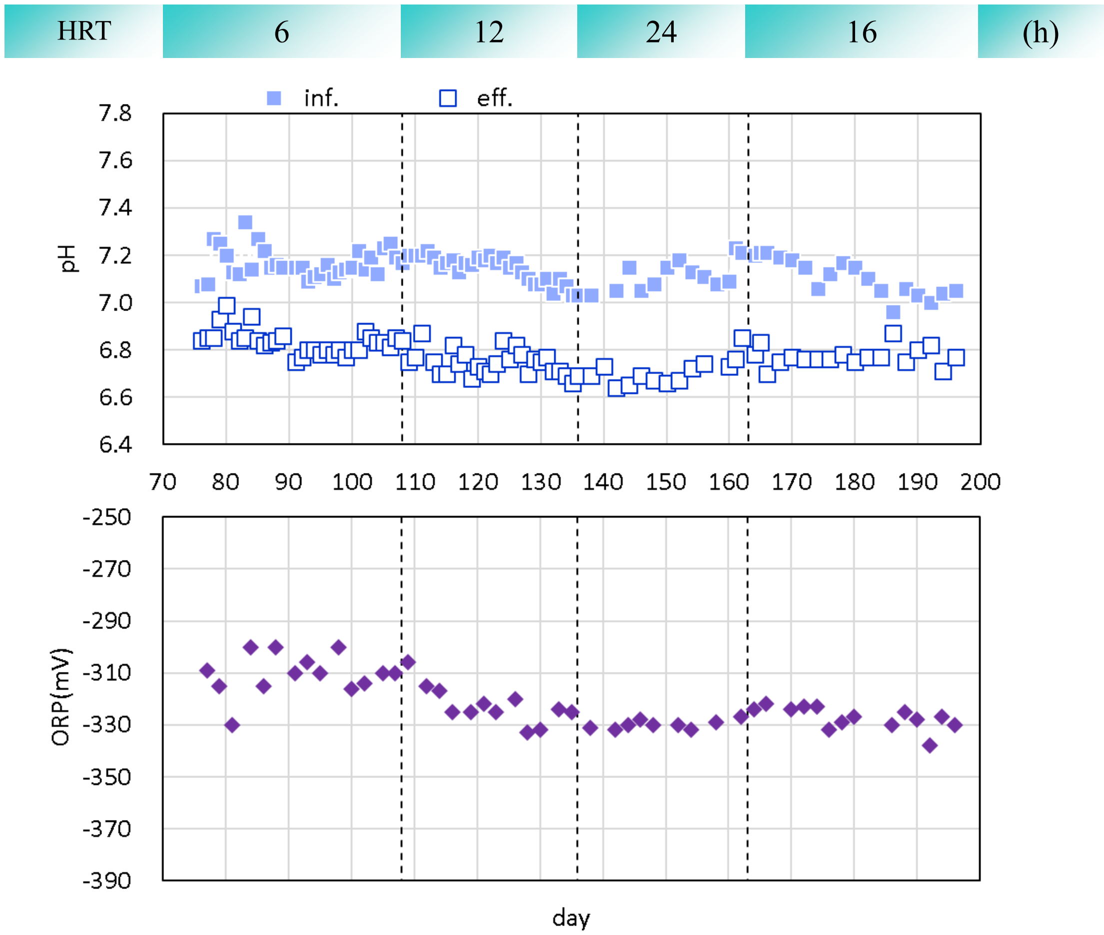


Fig. 5.20 pH and ORP during the long-term operation.

5.4.2 Sludge yield

Figure 5.21 shows the MLSS and MLVSS during the long-term operation experiment. The MLSS was maintained higher than HRT 6h / 15 ° C because of the poor methanogenesis performance. Then, mixed liquor concentration was decreased in HRT 24h due to a relatively longer time provided to the methane fermentation for organic matters accumulated in short HRTs by the membrane filtration process. During HRT 16h, the MLSS was controlled back to the range of 9 ~ 12 g/L and MLVSS 8 ~ 10 g/L.

Figure 5.22 shows the sludge yield calculation for each HRT condition followed the calculate equation mentioned in materials and methods section. The calculated sludge yield is presented as the slope in each calculation figures and shown in figure 5.23 by HRT as the x-coordinate in the low temperature conditions. It is obvious that the sludge yield decreased along with the HRT condition extending. Though during HRT 12h sludge yield was still relatively high as shown 0.21 g-VSS/g-COD_{rem}, it was decreased continuously since HRT was extended to 16 hours. Because of a relatively longer time had been provided to the methane fermentation for organic matters accumulated in short HRTs in HRT 24h and lead out the decrease of mixed liquor concentration, the sludge yield was obtained as a minus growing (-0.05 g-VSS/g-COD_{rem}).

Therefore, it can draw a conclusion that extending HRT is possible to achieve a high organic removal efficiency for treating the real sewage wastewater at low temperatures as well as obtain a relatively low sludge yield.

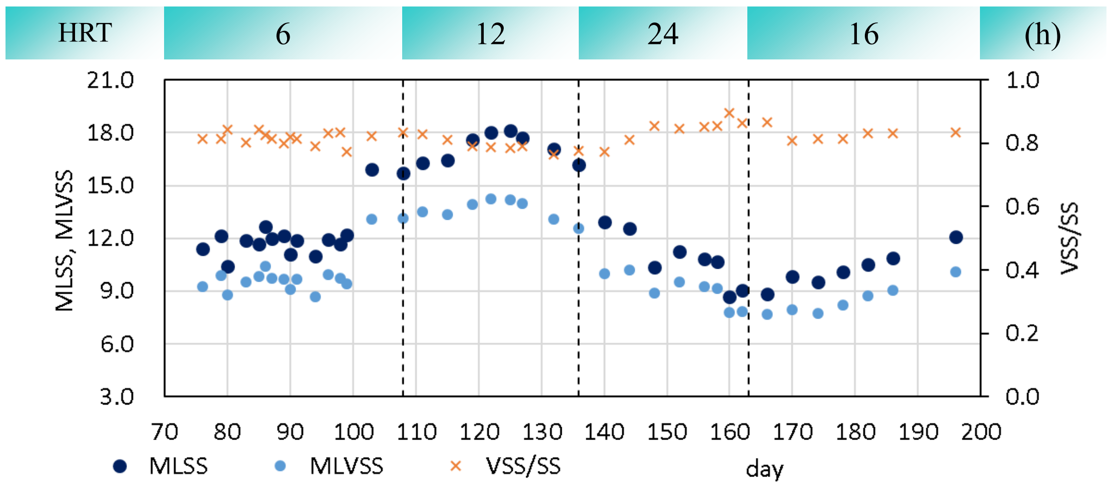


Fig. 5.21 MLSS and MLVSS during the long-term operation.

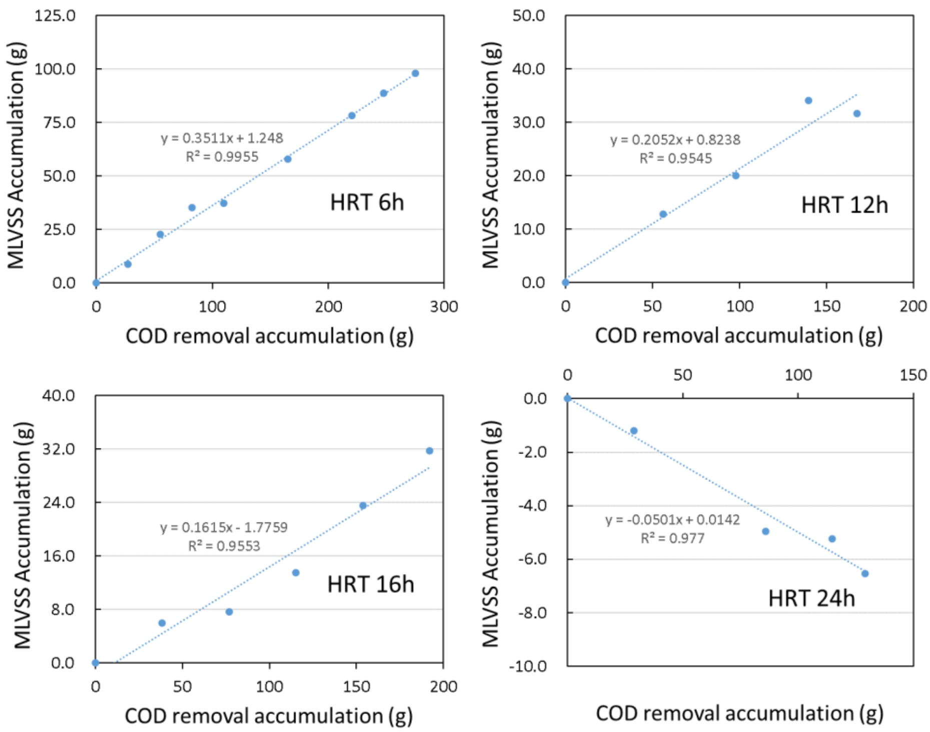


Fig. 5.22 Sludge yield calculation in different condition.

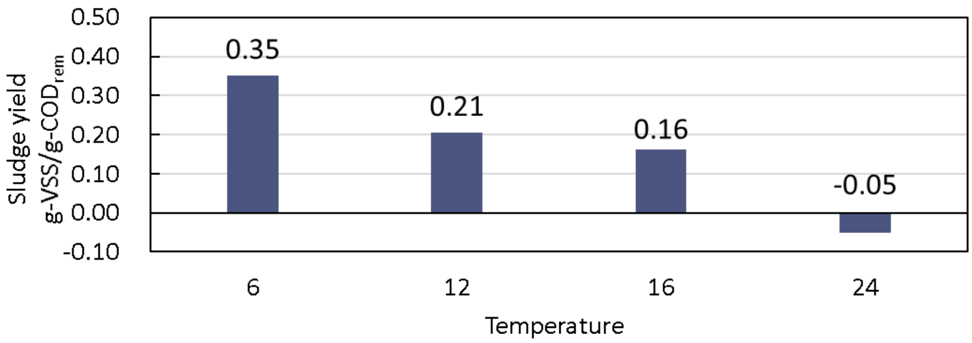


Fig. 5.23 Sludge yield in different HRTs at low temperature.

5.4.3 Biogas production

Figure 5.24 shows the biogas production rate during the long-term operation experiment. Daily biogas production rate was obtained stable generation in each HRT conditions and obvious growth by extending HRT condition. When HRT was adjusted from 24 hours to 16 hours, the biogas production rate was shown a rapid decrease because of the poor methanogenesis performed at low temperature and then returned to a level of more than 0.08 L-gas/L-water after about one week's operation. Methane gas content was also generated gradually increasing since HRT was adjusted to 12 hours due to the gas composition shown in figure 5.25.

The average biogas production rate and biogas yield (including methane gas yield) in each HRT condition at 15°C low temperature is shown in table 5.7. The result shown clearly that average biogas production has shown increase as the HRT extended. The highest average biogas production rate was achieved as high as 0.10 L-gas/L-water even at the low temperature in HRT condition of 24 hours and 0.09 L-gas/L-water in HRT 16h also performed well, the same level as those obtained in 25°C temperatures described in Chapter 4. In addition, the average methane gas content was restored to more than 80% in HRT 16 and 24 hours' operation and the N_2 content also obtained less compared with the short HRTs.

Biogas yield and methane yield in each temperature condition also has been listed in table 5.7. Biogas yield and methane yield also achieved increasing as long as the HRT extended due to the increasing of biogas production rate as well as methane gas content.

The biogas performance showed that in low temperature conditions, extending HRT condition can be an effective measure to achieving a better energy recovery potential by the improved biogas production performance.

Table 5.7 Average gas composition in each HRT condition at 15°C.

HRT (h)	6	12	16	24
Biogas production rate (L-gas/L-water)	0.03	0.06	0.09	0.10
CH_4 (%)	66.85	74.72	80.82	80.99
N_2 (%)	29.41	20.32	14.20	13.27
CO_2 (%)	3.74	4.96	4.98	5.74
Biogas yield (L-gas/g-COD_{rem})	0.09	0.19	0.29	0.31
Methane yield (L-CH_4/g-COD_{rem})	0.06	0.14	0.24	0.25

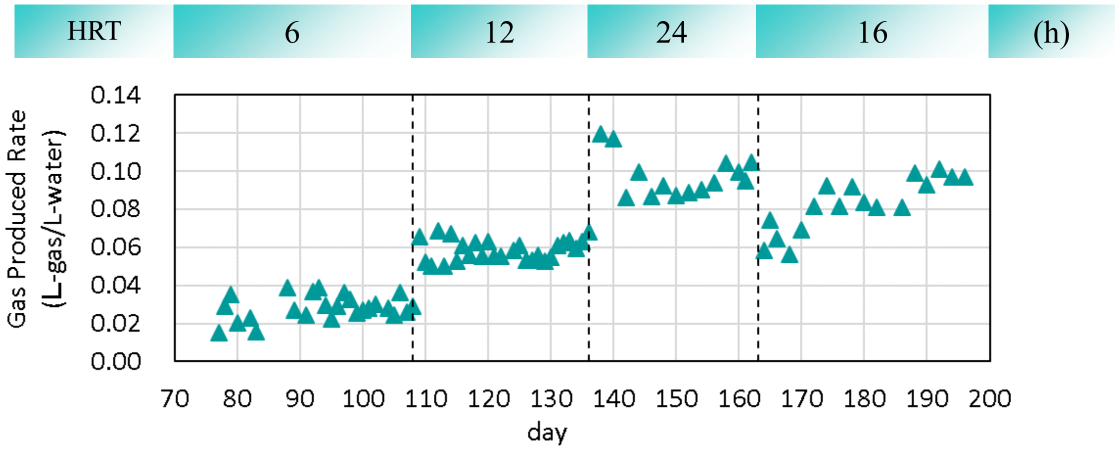


Fig. 5.24 Biogas production rate during the long-term operation.

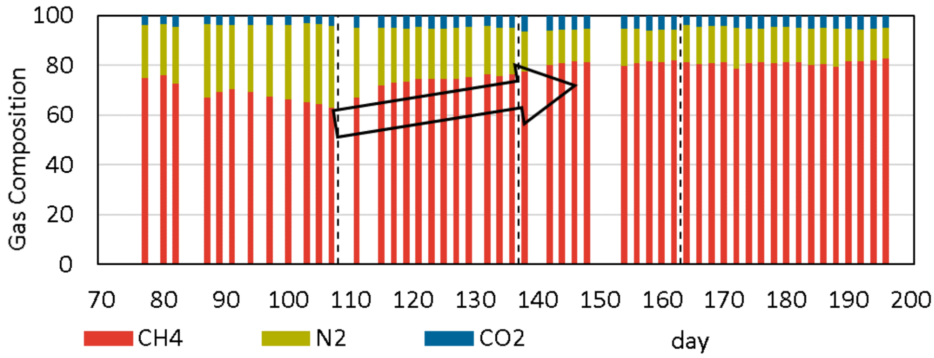


Fig. 5.25 Biogas composition during the long-term operation.

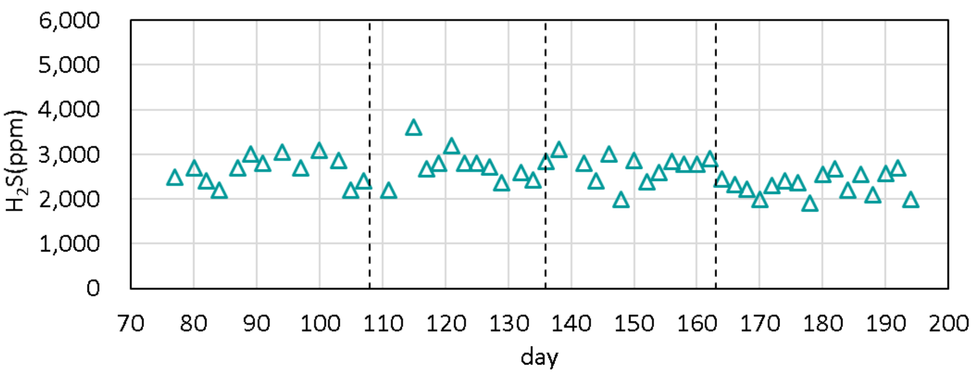


Fig. 5.26 H_2S concentration during the long-term operation.

5.4.4 COD balance analysis

COD balance at low temperature conditions in different HRTs was calculated and shown in table 5.8 as absolute value (g-COD/d) and figure 5.27 as percentage values. Compared with the HRT conditions of 6, 12 and 16 hours, it was found that COD amount in biogas produced was increased from the decrease of those COD amount of sludge growth and remained in effluent permeate. It was then obtained a further increase of COD amount as produced biogas due to the well biodegraded of solid organic matters as shown minus value of sludge yield in HRT 24 hours. Energy recovery potential could be generated well in HRT 16 and 24 hours as the COD amount was presented as 57.7% and 65.9%, respectively. Addition with the dissolved methane, the COD in influent conversion to CH_4 was achieved 70.1% in HRT 16h and even 83.4% in HRT 24h which indicated high energy recovery potential in low temperature conditions (table 5.9).

As a conclusion of COD balance result, it can be obtained that a good performance on sewage treatment performance with a high biogas-energy recovery efficiency can be achieved at low temperatures in relatively long HRT conditions.

Table 5.8 COD balance values in different HRTs at 15°C (g-COD/d).

HRT (h)	6	12	16	24
COD-in	30.7	15.5	10.3	7.0
Biogas production	6.0	5.9	6.0	4.6
H_2S in biogas	0.02	0.02	0.02	0.01
Dissolved biogas	4.5	2.1	1.3	1.3
Sludge growth	10.9	3.4	2.0	0.0
COD-eff	7.2	3.1	1.0	0.7

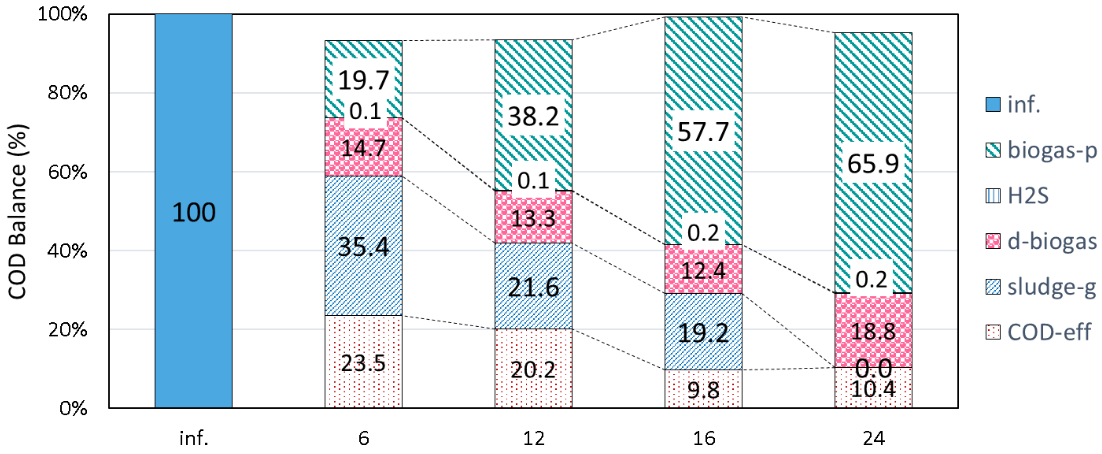


Fig. 5.27 COD balance in different HRTs at 15°C (%).

P.S.

inf.: COD in influent;

biogas-p: biogas produced (calculated by the discharged methane gas);

H2S: H_2S in the biogas produced;

Sludge-g: sludge growth;

COD-eff: COD in permeate effluent.

Table 5.9 COD conversion to CH_4 in different HRTs at 15°C.

HRT (h)	6	12	16	24
To biogas CH_4 (%)	19.7	38.2	57.7	65.9
To dissolved CH_4 (%)	14.7	13.3	12.4	17.5
Total (%)	34.4	51.5	70.1	83.4

5.4.5 SMA result at low temperatures

Figure 5.28 shows the SMA and OLR (calculated as the same unit of g-COD/g-VSS/d with SMA; COD_{inf} and treatment capacity for the calculation was used as the average values from the whole long termed operation in each condition) in different HRT conditions during the low temperatures operation. SMA value in 15°C was mostly shown around 0.10 g-COD/g-VSS/d except for less than 0.05 g-COD/g-VSS/d was obtained in the phase of HRT 12 hours. The reason was considered to be high-frequency discharge of mixed liquor during HRT 6h and then HRT 12h, which both had a high sludge yield, resulted the concentration of functional bacteria diluted (Chen et al., 2017a).

The result shown that both HRT 6h and 12h SMA was lower than OLR while SMA obtained higher than OLR in the longer HRTs which indicated that HRTs less 12 hours cannot be used as the operation condition during 15°C low temperature treatment. In order to achieve the low temperature operation, a longer HRT is required for decline the OLR less than low SMA in low temperatures.

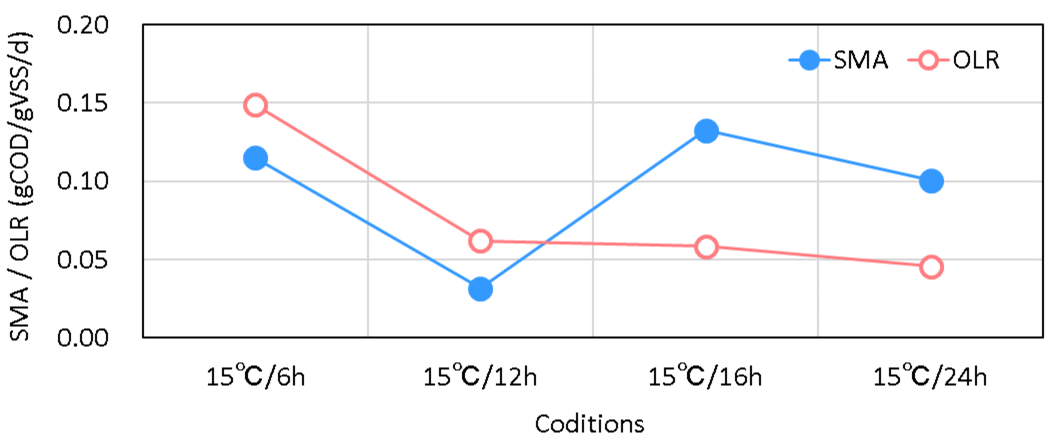


Fig. 5.28 SMA&OLR in different HRTs at low temperature operation.

5.4.6 Membrane performance

The TMP and FLUX recording data shown in figure 5.29 presents the membrane performance during the long-term operation at 15°C low temperature. The CFGV was declined to 116 m/h after HRT was extended to 12 hours or longer and TMP was stabled at a very low value in those conditions due to the smaller FLUX in long HRTs. This result shown that the membrane filtration was performed very well in the conditions of HRT 12 hours or longer even in the low temperature of 15°C.

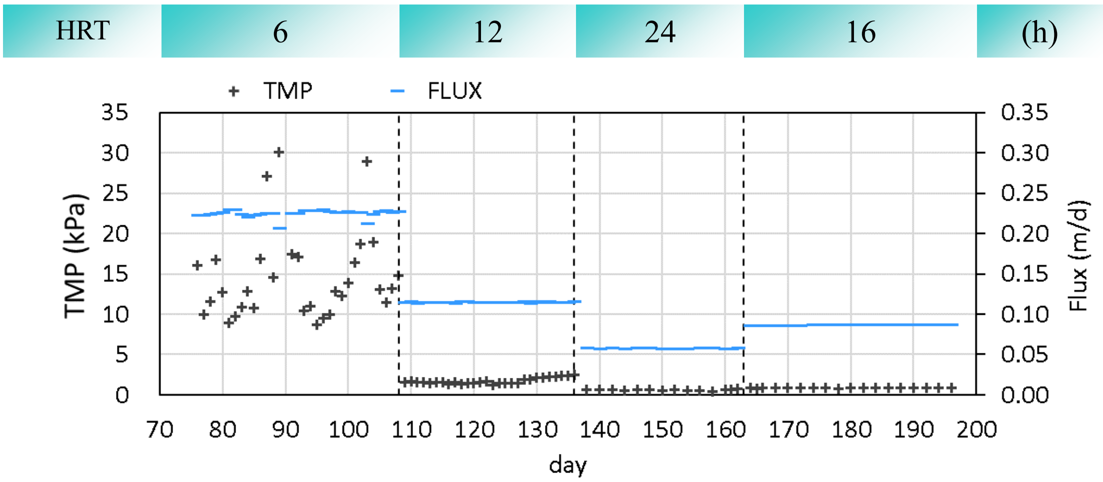


Fig. 5.29 TMP – FLUX record during the long-term operation.

5.4.7 Section summarizes

As operated at a low temperature of 15°C in different HRTs, the below conclusions can be obtained:

(1) Applying AnMBR directly into real sewage wastewater treatment at low temperatures could achieve well organic pollutant removal performance with a low sludge yield by extending HRT condition.

(2) A high energy recovery potential could be achieved by the generated biogas in HRT 16 or 24 hours at 15°C low temperature.

5.5 Comprehensive evaluation

A comprehensive comparison by the performances on aspect of organic removal (COD / BOD / SS), sludge character (MLSS and sludge yield), energy recovery potential (biogas production rate / methane content/biogas yield / methane yield / COD conversion to CH_4), membrane filtration (FLUX and TMP growth), energy consumption (biogas cycling and temperature constant) as well as the operation conditions (sewage treatment capacity / ORP) for different HRT conditions is shown in table 5.10. The weakness items were marked red color with bold and the incommensurable or non-differential items were marked in gray. In addition, the strength items at low temperature was marked blue color with underline.

As a result, HRT 6h at 15°C presented the weakness compared with the other conditions on organic removal, sludge yield, biogas production rate (so did the biogas yield and methane yield), methane content, COD conversion to CH_4, TMP growth, cross-flow gas velocity and ORP while with strength on FLUX and sewage wastewater treatment capacity. Among the HRTs implemented at 15°C low temperature, HRT 24h presented strengths on organic removal, sludge yield, biogas production rate (so did the biogas yield and methane yield), methane content, COD conversion to CH_4 and TMP growth while the sewage wastewater treatment capacity during the HRT 24 hours was obtained only 20L per day which also resulted an underutilization operation for the AnMBR system during treatment process. Because of organic removal efficiency was not reach the standards in HRT conditions of 6 hours and 12 hours, those conditions cannot be applied in the sewage wastewater treatment. Comparing with HRT 16 hours and 24 hours at 15° C low temperature, it was easily to found that the performances during these two HRT conditions was very close while can provide a bigger treatment capacity even in every aspect listed in table 5.10.

As a consequence, compared the HRTs implemented at low temperature of 15°C in this study, the suitable HRT was considered to be 16 hours.

Table 5.10 Comprehensive comparison for different HRTs.

Temperature (°C)	25	20	15	15	15	15
HRT (h)	6	6	6	12	16	24
COD_{RE} (%)	90.2	90.1	**77.2**	82.1	90.5	90.3
BOD_{RE} (%)	91.4	92.2	81.9	**80.2**	90.3	91.0
SS_{RE} (%)	100	100	100	100	100	100
MLSS (g/L)	11.9 ~13.1	12.5 ~14.5	10.5 ~12.5	15.7 ~18.1	9.0 ~12.9	8.8 ~12.1
Sludge yield (g-VSS/g-COD_{rem})	0.11	0.20	**0.35**	0.21	0.16	-0.05
Biogas production rate (L-gas/L-water)	0.09	0.07	**0.03**	0.06	0.09	0.10
Methane content	80.19	79.48	**66.85**	74.72	80.82	80.99
Biogas yield (L-gas/g-COD_{rem})	0.23	0.20	**0.09**	0.19	0.29	0.31
Methane yield (L-CH_4/g-COD_{rem})	0.18	0.16	**0.06**	0.14	0.24	0.25
COD conversion to CH_4 (%)	70.0	61.7	**34.4**	51.5	70.1	83.4
SMA-OLR	+	+	**negative**	**negative**	+	+
FLUX (m/d)	0.23	0.23	0.23	0.12	0.09	**0.06**
TMP growth (kPa/d)	0.2	1.1	**11.9**	0.1	0.0	0.0
CFGV (m/h)	116	116	**174**	116	116	116
Heater (°C)	**29.3**	24.3	18.2	17.4	16.3	16.4
Treatment capacity (L/d)	**78.7**	78.7	77.8	39.8	29.9	**20.0**
ORP (mV)	-325	-314	**-309**	-322	-327	-330

5.6. Conclusions

As with the conditions of temperature decreased from 25°C to 15°C operated for 110 days, the conclusions can be obtained as:

(1) Applying AnMBR directly into real sewage wastewater treatment performed well in organic pollutant removal (COD_{RE} around 90%) as well as energy recovery potential as methane yield 0.16~0.18 L-CH_4/g-COD_{rem} at temperature above 20°C with a low sludge yield under 0.20 g-VSS/g-COD_{rem} in OLR above 1.60 g-COD/L/d.

(2) Low temperature of 15°C cannot be used to treating the real sewage in short HRTs (< 12h) because of the poor performance due to the low methanogenesis activity in low temperature condition.

(3) TMP as well as membrane fouling risk increases when the temperature decreases and may lead out a more energy demand if the temperature is set as 15°C.

(4) Extending HRT to 16 or 24 hours can achieve good performance on organic pollutant removal (COD_{RE} 90%) with a low sludge yield (< 0.16 g-VSS/g-COD_{rem}) and a high energy recovery potential (0.25 L-CH_4/g-COD_{rem} methane yield) on treating real sewage wastewater as OLR up to 0.52 g-COD/L/d.

REFRENCES:

Bai, R., Leow, H.F., 2002. Microfiltration of activated sludge wastewater—the effect of system operation parameters. Sep. Purif. Technol. 29, 189–198. https://doi.org/10.1016/S1383-5866(02)00075-8

Chen, R., Nie, Y., Ji, J., Utashiro, T., Li, Q., Komori, D., Li, Y.-Y., 2017a. Submerged anaerobic membrane bioreactor (SAnMBR) performance on sewage treatment: removal efficiencies, biogas production and membrane fouling. Water Sci. Technol. 76, 1308–1317. https://doi.org/10.2166/wst.2017.240

Chen, R., Nie, Y., Tanaka, N., Niu, Q., Li, Q., Li, Y.Y., 2017b. Enhanced methanogenic degradation of cellulose-containing sewage via fungi-methanogens syntrophic association in an anaerobic membrane bioreactor. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.09.046

Crowley, T.J., 2000. Causes of climate change over the past 1000 years. Science (80-.). 289, 270–277.

Gouveia, J., Plaza, F., Garralon, G., Fdz-Polanco, F., Peña, M., 2015. Long-term operation of a pilot scale anaerobic membrane bioreactor (AnMBR) for the treatment of municipal wastewater under psychrophilic conditions. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2015.03.002

Ho, J., Sung, S., 2009. Effects of solid concentrations and cross-flow hydrodynamics on microfiltration of anaerobic sludge. J. Memb. Sci. https://doi.org/10.1016/j.memsci.2009.08.047

Huang, S., Pollack, H.N., Shen, P.-Y., 2000. Temperature trends over the past five centuries reconstructed from borehole temperatures. Nature 403, 756.

Huang, X., Gui, P., Qian, Y., 2001. Effect of sludge retention time on microbial behaviour in a submerged membrane bioreactor. Process Biochem. https://doi.org/10.1016/S0032-9592(01)00135-2

Le Corre, K.S., Valsami-Jones, E., Hobbs, P., Parsons, S.A., 2009. Phosphorus recovery from wastewater by struvite crystallization: A review. Crit. Rev. Environ. Sci. Technol. 39, 433–477.

Lei, Z., Yang, S., Li, Y. you, Wen, W., Wang, X.C., Chen, R., 2018. Application of anaerobic membrane bioreactors to municipal wastewater treatment at ambient temperature: A review of achievements, challenges, and perspectives. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2018.07.050

Lin, H., Peng, W., Zhang, M., Chen, J., Hong, H., Zhang, Y., 2013. A review on anaerobic membrane bioreactors: Applications, membrane fouling and future perspectives. Desalination. https://doi.org/10.1016/j.desal.2013.01.019

Pandey, P., Shinde, V.N., Deopurkar, R.L., Kale, S.P., Patil, S.A., Pant, D., 2016. Recent advances in the use of different substrates in microbial fuel cells toward wastewater treatment and simultaneous energy recovery. Appl. Energy 168, 706–723. https://doi.org/https://doi.org/10.1016/j.apenergy.2016.01.056

Rulkens, W., 2008. Sewage Sludge as a Biomass Resource for the Production of Energy: Overview and Assessment of the Various Options. Energy & Fuels 22, 9–15. https://doi.org/10.1021/ef700267m

Watanabe, R., Nie, Y., Wakahara, S., Komori, D., Li, Y.Y., 2017. Investigation on the response of anaerobic membrane bioreactor to temperature decrease from 25 °C to 10 °C in sewage treatment. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.07.001

Watanabe, R., Qiao, W., Norton, M., Wakahara, S., Li, Y.-Y., 2014. Recent Developments in Municipal Wastewater Treatment Using Anaerobic Membrane Bioreactor: A Review. J. Water Sustain. https://doi.org/10.11912/jws.4.2.101-122

Zakkour, P.D., Gaterell, M.R., Griffin, P., Gochin, R.J., Lester, J.N., 2001. Anaerobic treatment of domestic wastewater in temperate climates: Treatment plant modelling with economic considerations. Water Res. https://doi.org/10.1016/S0043-1354(01)00145-2

Chapter 6

Conclusions and perspectives

6.1 Conclusions

In this study, a series of studies was conducted on innovation of real sewage wastewater treatment by AnMBR and the conclusions can be summarized as following:

[Effect of membrane pore size on start-up and long-term operation performance]

(1) The AnMBRs with different pore size membranes applied for treating the real sewage wastewater were started-up successfully and verified a good performance on organic pollutant removal (COD removal efficiency around 89%) with a great potential of energy recovery due to the methane yield was achieved 0.18 ~ 0.20 L-CH_4/g-COD_{rem} (dissolved methane was not included).

(2) Sodium hypochlorite solution used for online membrane cleaning should be less than 1 g/L as the final concentration in AnMBR on the purpose of protecting the microorganisms inside the reactor. Off-line membrane cleaning can achieve 80% potential of filtration ability recovery.

(3) Compare with the two different pore size membrane, the microfiltration membranes (0.4μm pore size used in this study) are relatively more appropriate for treating the sewage wastewater than the ultrafiltration membranes (0.05μm pore size used in this study) because of it can achieve the same treatment performance and biogas production with a relatively lower energy consumption for overcoming the suction pressure caused during the filtration process.

[Effect of HRT on treatment performance at room temperature]

(4) Applying submerged AnMBR to treating the real sewage achieved high COD removal efficiency (89%) with sludge yield as low as 0.07 ~ 0.11 g-VSS/g-COD_{rem} in HRTs from 6 to 12 hours at 25 degrees.

(5) High energy recovery potential can be generated at 25 degrees as total COD conversion to CH_4 was obtained 68 ~ 75% during HRTs in 6 to 12 hours (methane gas yield: 0.20 ~ 0.24 L-gas/g-COD_{rem}).

(6) Organic loading rate was challenged as high as 2.05 g-COD/L/d in HRT 4 hours and obtained good performance of 84% COD removal efficiency with 0.22 g-VSS/g-COD_{rem} of sludge yield.

[Effect of temperature on treatment performance of MF-MBR]

(7) Applying AnMBR directly into real sewage wastewater treatment performed well in organic pollutant removal (COD_{RE} around 90%) as well as energy recovery potential as methane yield 0.16~0.18 L-CH_4/g-COD_{rem} at temperature above 20°C with a low sludge yield under 0.20 g-VSS/g-COD_{rem} in OLR above 1.6 g-COD/L/d.

(8) Low temperature of 15°C cannot be used to treating the real sewage in short HRTs (< 12h) because of the poor performance due to the low methanogenesis activity in low temperature condition.

(9) TMP as well as membrane fouling risk increases when the temperature decreases and may lead out a more energy demand if the temperature is set as 15°C.

(10) Extending HRT to 16 or 24 hours can achieve well organic pollutant removal performance (COD_{RE} 90%) with a low sludge yield (< 0.16 g-VSS/g-COD_{rem}) and a high energy recovery potential (0.25 L-CH_4/g-COD_{rem} methane yield) on treating real sewage wastewater as OLR up to 0.52 g-COD/L/d.

6.2 Perspectives

The conclusions are optimistic in this study which indicated that it is feasible to applying submerged AnMBR to the raw real sewage wastewater treatment especially when the temperature above 20 degrees, a relatively short HRT such as 6 hours have achieved stable operation. However, cities' temperature above 20 degrees in all season are rare in worldwide which are principally tropical in distribution (Crowley, 2000; Huang et al., 2000). The major cities are in the subtropics and even the cold zones. For those low temperature regions, longer HRTs may require in order to achieve a high organic removal performance with low sludge yield as well as generate the energy recovery. Though this study was not implemented operation temperature below 10 degrees, according to the previous research result, applying AnMBR to the extremely cold regions may need a very long HRT condition which decreased the sewage treatment capacity as result (Watanabe et al., 2017). Hence, properly heated is considered to be a better measurement if AnMBRs applied in the extremely cold regions.

Combining with development of other technologies AnMBR is hopeful to be applied in treating the municipal sewage wastewater achieving high removal efficiency on organic and nutrients with low waste sludge produced, and most important, realize the next generation sewage treatment process with characters of energy positive and resource recovery. The technologies can be combined to treating the AnMBR effluent are including but not limited to:

Anammox

The process was considered to be strength on treating ammonia rich wastewaters especially with a high ammonia concentration or high nitrogen loading rate which have been reported in previous researches (He et al., 2016; Zhang et al., 2016). While there were also lab-scale one-stage anammox reactors reported by treating the low nitrogen concentration wastewater and achieved as high as around 80% removal efficiency of total nitrogen and 100% removal of ammonia nitrogen by a granular sludge CSTR as well as a carrier based reactor (Chen et al., 2019; R. Chen et al., 2018). In addition, there were also reported for anammox process applied in the mainstream of municipal sewage wastewater treatment process (Ali and Okabe, 2015; Cao et al., 2017). Anammox process is very suitable to treat the AnMBR effluent due to the low C/N ratio which the details have been mentioned in the Literature review chapter of this thesis.

Chemical precipitation

Chemical precipitation is a process involves the addition of compounds of calcium, aluminum and iron. According to a report (Minnesota Pollution Control Agency, 2006), a dose of 1.0 mole of aluminum compound is sufficient per mole of phosphorus. Besides, aluminum compound makes it highly useable due to its less corrosive nature than ferric chloride. In terms of economic costs, the use of seawater as a dosing agent for magnesium salts can greatly reduce costs and the phosphorus recovery can even achieve 70% without adding other chemicals to adjust the pH value (Haifeng et al., 2007).

EBPR

Enhanced Biological Phosphorus Removal (EBPR) process utilized with the effect of polyphosphate accumulating organism (PAO) for phosphorus removal. The process is reported to be efficient in anaerobic rather than in aerobic (Tchobanoglous et al., 2003) and has a high potential of P removal ability (achieved 0.1mg/L of phosphorus in effluent) at modest cost and minimum sludge. The mechanism is described as phosphate is accumulated within the cells of PAOs in anaerobic environment and then biomass is separated from the wastewater, then phosphorus is recovered by phosphorus accumulating PAOs are processed later (Chapagain, 2016). However, it is not recommended to use directly as agricultural fertilizer because of the sludge produced in EBPR mostly contaminated with heavy metals, harmful pathogens and toxic substance which might interferes with the crops growth (Yuan et al., 2012).

Stirring, stripping and dissipation

A newly presented pre-anoxic MBR post-treatment was proved to be capable of consistently removing 80% of dissolved methane with peaks up to 95% via the anoxic stirring and the reported whole process was also presented achieved synergetic nitrogen removal (Silva-Teira et al., 2017). Another research implemented stripping and dissipation to two pilot-scale UASB with the results of achieved intermediate removal efficiencies of dissolved methane and sulfide were accomplished with the stripping technique (around 30% for methane and in the range of 40 to 60% for hydrogen sulfide, depending on the air injection rate applied), and very promising performance was obtained with the dissipation chamber technique, with removal efficiencies consistently above 60% being observed for dissolved methane and dissolved sulfide, even at low exhaustion rates. During the best operation condition, median removal efficiencies of as high as 73 and 97% were observed for dissolved methane and dissolved sulfide, respectively (Glória et al., 2016).

REFRENCES:

Chapagain, Y., 2016. Methods and Possibility for Recycling of Phosphorus from Sludge. Helsinki Metropolia University of Applied Sciences.

Crowley, T.J., 2000. Causes of climate change over the past 1000 years. Science (80-.). 289, 270–277.

Glória, R.M., Motta, T.M., Silva, P.V.O., Da Costa, P., Brandt, E.M.F., Souza, C.L., Chernicharo, C.A.L., 2016. Stripping and dissipation techniques for the removal of dissolved gases from anaerobic effluents. Brazilian J. Chem. Eng. 33, 713–721. https://doi.org/10.1590/0104-6632.20160334s20150291

Haifeng, F., Ying, H., Jinqing, L., 2007. Primly Methods and Common Process of Phosphorus Recovery from Wastewater. Chem. Eng. Equip. 2, 61-64,68. https://doi.org/10.3969/j.issn.1003-0735.2007.02.017

Huang, S., Pollack, H.N., Shen, P.-Y., 2000. Temperature trends over the past five centuries reconstructed from borehole temperatures. Nature 403, 756.

Minnesota Pollution Control Agency, 2006. Phosphorus Treatment and Removal Technologies.

Silva-Teira, A., Sánchez, A., Buntner, D., Rodríguez-Hernández, L., Garrido, J.M., 2017. Removal of dissolved methane and nitrogen from anaerobically treated effluents at low temperature by MBR post-treatment. Chem. Eng. J. https://doi.org/10.1016/j.cej.2017.06.047

Tchobanoglous, G., Burton, F.L., Stensel, H.D., 2003. Wastewater Engineering: Treatment and Reuse, 4th ed. ed. Boston : McGraw-Hill.

Watanabe, R., Nie, Y., Wakahara, S., Komori, D., Li, Y.Y., 2017. Investigation on the response of anaerobic membrane bioreactor to temperature decrease from 25 °C to 10 °C in sewage treatment. Bioresour. Technol. https://doi.org/10.1016/j.biortech.2017.07.001

Yuan, Z., Pratt, S., Batstone, D.J., 2012. Phosphorus recovery from wastewater through microbial processes. Curr. Opin. Biotechnol. 23, 878–883. https://doi.org/https://doi.org/10.1016/j.copbio.2012.08.001

APPENDIX

- ✧ **ABBREVIATIONS**
- ✧ **SUPPLEMENTARY DATA & FIGURES**

ABBREVIATIONS

AE	Alcohol Ethoxylates	mg/L
Anammox	Anaerobic ammonium oxidation	-
AnMBR	Anaerobic Membrane Bio-Reactor	-
AOB	Ammonium Oxidation Bacteria	-
AR	Aeration Rate	L/min
BOD	Biological Oxygen Demand	mg/L
BOD_{RE}	BOD Removal Efficiency	%
COD	Chemical Oxygen Demand	mg/L
COD_{RE}	COD Removal Efficiency	%
COD_{rem}	Removed COD	g, mg
CFGV	Cross Flow Gas Velocity	m/h
CSTR	Continuous Stirred Tank Reactor	-
DO	Dissolved Oxygen	mg/L
eff.	Effluent of the Reactor	-
EPS	Extracellular Polymeric Substances	mg/L
FISH	Fluorescence In Situ Hybridization	-
FLUX	permeate flow divided by the total membrane area	m/d
GCFV	Gas Cross Flow Velocity	m/h
HRT	Hydraulic Retention Time	hours
inf.	Influent Sewage	-
LAS	Linear Alkylbenzene Sulfonate	mg/L
MBR	Membrane Bio-Reactor	-
MLSS	Mixed Liquor Suspended Solids	g/L

MLVSS	Mixed Liquor Volatile Suspended Solids	g/L
OLR	Organic Loading Rate	gCOD/gVSS/d
ORP	Oxidation Reduction Potential	mV
SAnMBR	Submerged Anaerobic Membrane Bio-Reactor	-
SBR	Sequencing Batch Reactor	-
SMA	Specific Methanogenic Activity	gCOD/gVSS/d
SMP	Soluble Microbial Products	mg/L
SRT	Sludge Retention Time	days
SS	Suspended Solid	mg/L
SS_{RE}	SS Removal Efficiency	%
SV30	Sludge Volume / settling rate test	%
SVI	Sludge Volume Index	g/mL
TMP	Trans-Membrane Pressure	kPa
WWTW	Waste Water Treatment Works	-
WWTP	Waste Water Treatment Plants	-

SUPPLEMENTARY DATA & FIGURES

➪Drawing of the new designed AnMBR.

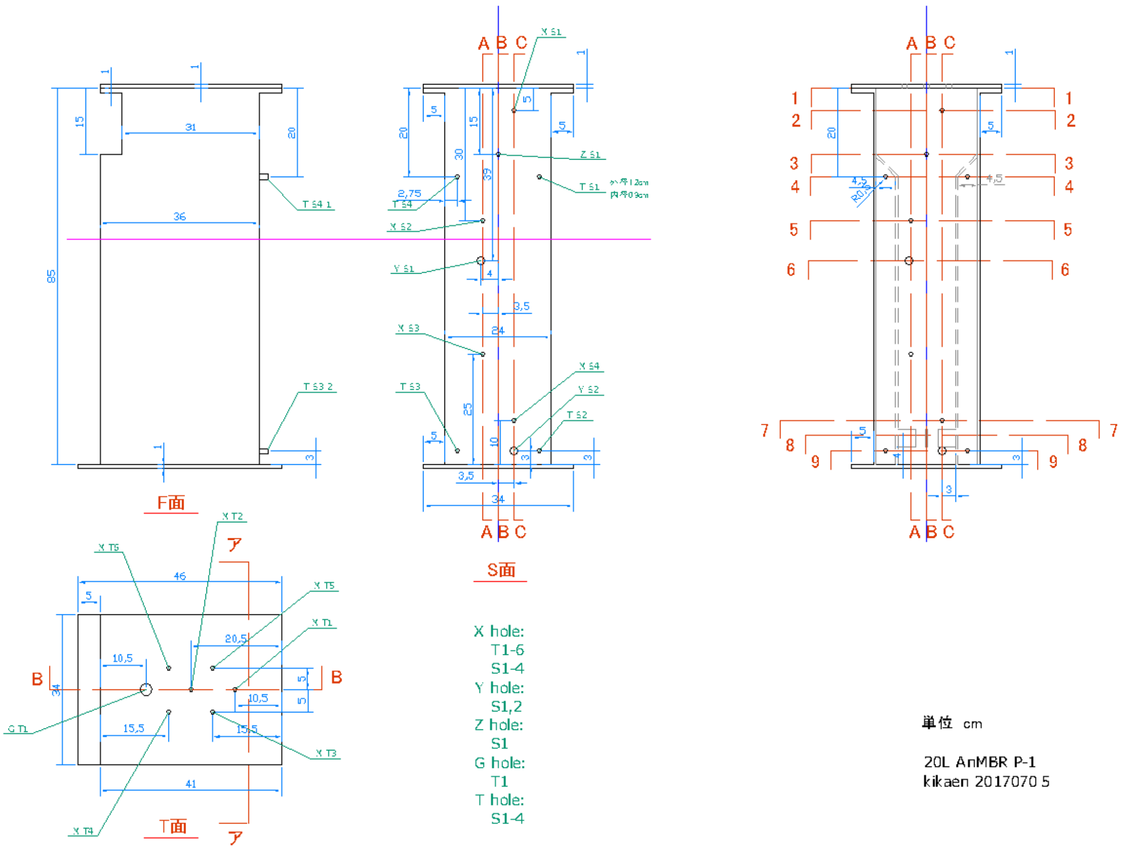


Fig. A.1 New-designed AnMBR integral drawing.

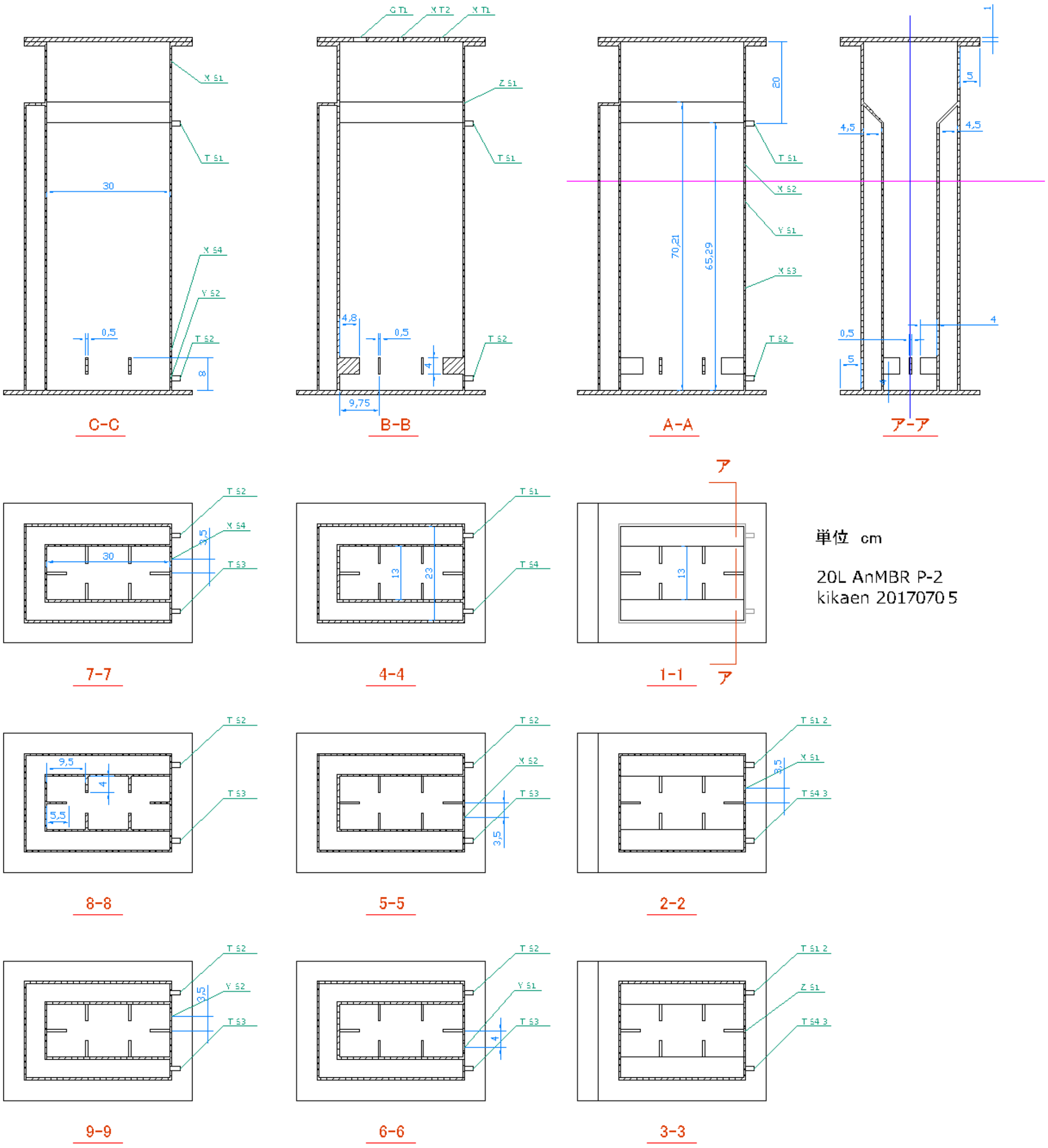


Fig. A.2 Details of the New-designed AnMBR transversal drawing.

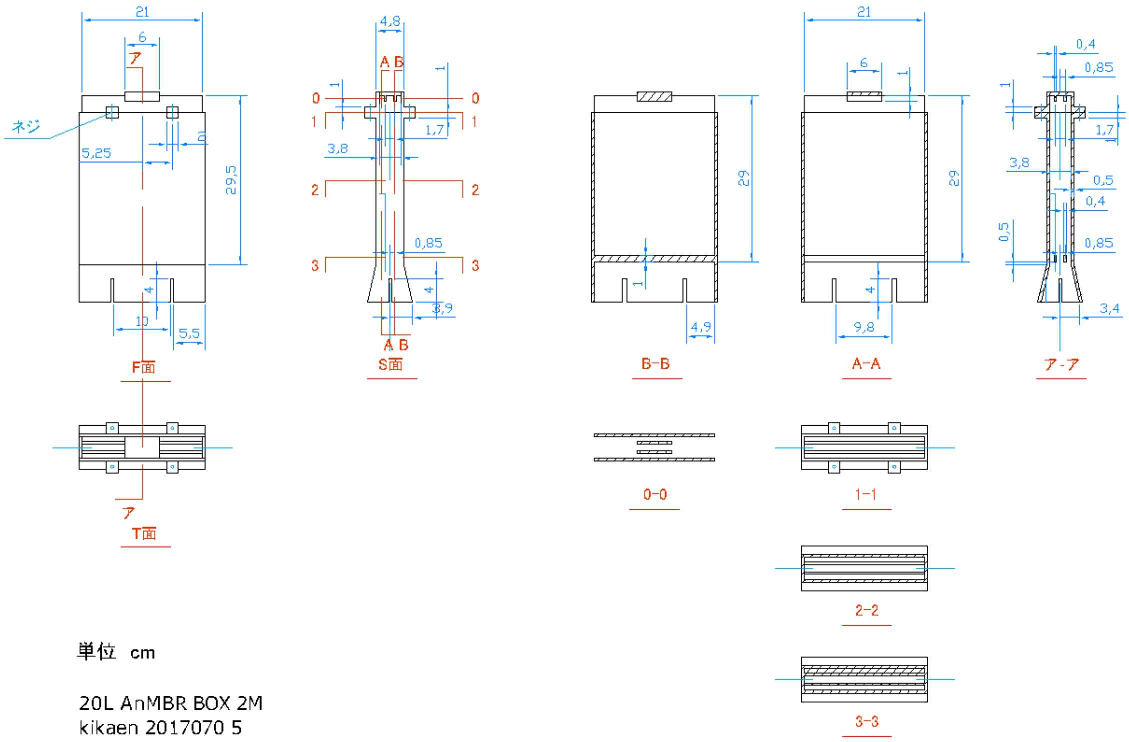


Fig. A.3 BOX of the membrane module.

➪Photos of the new designed AnMBR.

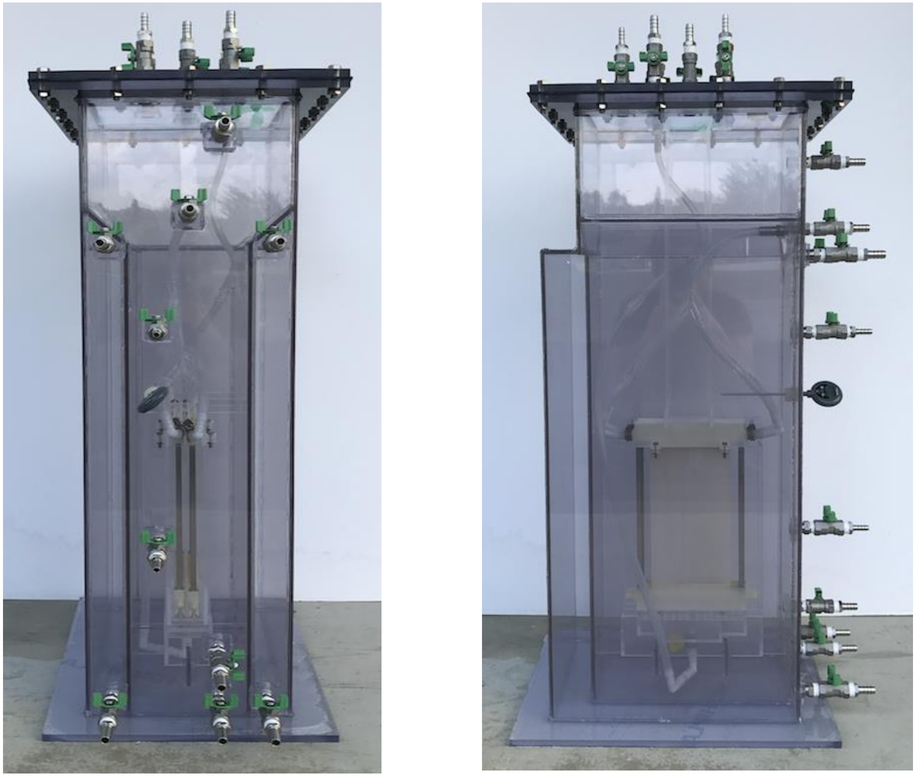


Fig. A.4 Photographs of the New-designed AnMBR with membrane module.

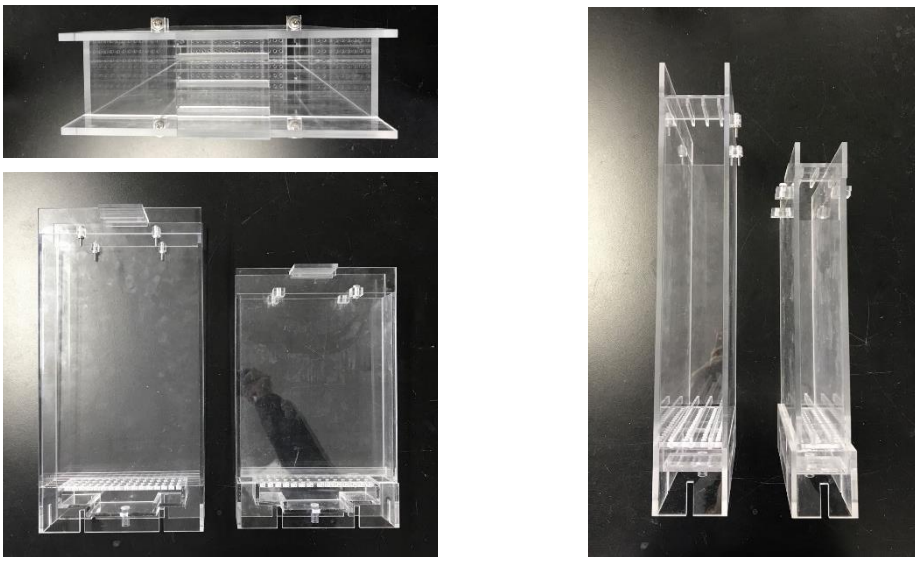


Fig. A.5 Photographs of the New-designed BOX of the membrane module.

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