CHANGE IN SLUDGE SETTLING AND FILTRATION PROPERTIES AND MEMBRANE FOULING TRENDS IN MBR ACTIVATED SLUDGE SYSTEMS OPERATED AT DIFFERENT SOLIDS AND HYDRAULIC RETENTION TIMES

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by

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The undersigned, appointed by the Dean of the Graduate School, have examined the thesis entitled

CHANGE IN SLUDGE SETTLING AND FILTRATION PROPERTIES AND MEMBRANE FOULING TRENDS IN MBR ACTIVATED SLUDGE SYSTEMS OPERATED AT DIFFERENT SOLIDS AND HYDRAULIC RETENTION TIMES

presented by Minghao Sun,

a candidate for the degree of Master of Science,

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DEDICATION

I dedicate this thesis to my beloved parents, whose moral encouragement and support helped me reaching my master's degree goal.

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Table of Contents

List of Illustrations

List of Abbreviations

SRT	Solids retention time
HRT	Hydraulic retention time
MBR	Membrane bioreactor
MLSS	Mixed liquor suspended solids
TMP	Trans-membrane pressure
CAS	Conventional activated sludge
WWTP	Wastewater treatment plant
DO	Dissolved oxygen
SS	Suspended solids
PVDF	Polyvinylidene fluoride
AO	Anoxic-oxic
A_2O	Anaerobic-anoxic-oxic
MABR	Membrane-aerated biofilm reactor

SMP	Soluble microbial products
OLR	Organic loading rate
EPS	Extracellular polymeric substances
LB-EPS	Loosely bound extracellular polymeric substances
SVI	Sludge volume index
DSVI	Diluted sludge volume index
SOUR	Specific oxygen uptake rate
TN	Total nitrogen
TTF	Time to filter

Abstract

The Membrane bioreactor (MBR) activated sludge process is increasingly used in wastewater treatment due to its excellence in solid-liquid separation, superior effluent quality, smaller bioreactor volume and foot print as compare to Conventional Activated Sludge (CAS) process. However, operational issues such as membrane fouling and sludge bulking affect its broad applications. As solids retention time (SRT) and hydraulic retention time (HRT) are the most important operating parameters in activated sludge systems, this research determined the effect of different SRTs (180 d, 90 d and 45 d) and HRTs (24 h, 12 h, and 6 h) on the change in sludge settling and filtration properties and membrane fouling trends while keeping the SRT/HRT ratio constant throughout the study period. The biomass concentrations increased from about 8,000 to 10,000 mg COD/L as SRT and HRT decreased proportionally. As SRT decreased to 45 d and HTR decreased to 6 h, significant sludge bulking and poor filtration with high Time to Filter (TTF) values were observed, largely due to the operation at low DO concentrations under high organic loading conditions. However, the system recovered in about 50 d after correction of low DO concentrations in the MBR. Due to the potential sludge bulking problems at

long SRT (180 d) operation, the results suggest the MBR operation at the SRTs of 45 to 90 d results in excellent sludge settling and filtration properties and effluent water quality.

1. Introduction

1.1 Membrane Bioreactor (MBR)

MBR activated sludge processes replace gravity-based sedimentation with membrane filtration to ensure high effluent water quality. Therefore, it is now widely used for municipal and industrial wastewater treatment (Beier et al. 2012, Fenu et al. 2010, Hoinkis et al. 2012). Because of its unique compact structure, which allows wastewater treatment at high mixed liquor suspended solids (MLSS) concentrations (e.g., 8-14 g/L), MBRs have been widely used for water reuse (Alturki et al. 2010, Atkinson 2006a). Although the CAS process is popularly used in municipal wastewater treatment worldwide, with the increase in water demand and more stringent regulatory requirements, more and more activated sludge wastewater treatment plants (WWTPs) are upgrading their facilities to improve treatment capacity for water reuse. However, due to the limited available space and volumetric organic loading rate in existing activated sludge systems, it is very challenging for the current WWTPs to meet the requirements. The MBR wastewater treatment process offers many advantages over the CAS process.

The average annual market growth rate is predicted to be 10.9% for MBR, which is significantly faster than other wastewater treatment technologies such as sequencing batch reactor (SBR) or biological aerated filters (BAF) (Judd 2008). This indicates that MBR plants will double every seven years for wastewater treatment. There are two MBR configurations, internal (submerged) and external (side-stream).

1.1.1 Internal (Submerged) MBR

The submerged membrane bioreactors (SMBR) are commonly used for wastewater treatment. In SMBR, the filtration elements are installed either in the main bioreactor or in a separate tank. The membranes used in SMBR operation can be either flat sheet or tubular or a combination of both, and may include a backwash system which will reduce the membrane fouling. Aeration is required to provide air scour to reduce membrane fouling. As the membranes are installed in the main reactor, membrane modules need to be removed from the vessel and transferred to an offline cleaning tank (Meng et al. 2008). Since the submerged system operates at a lower trans-membrane pressure (TMP) than an external system, it has a lower flux. However, it has the advantages of reduced fouling so less rigorous cleaning procedures are necessary as compared to the side-stream system (Churchouse 1997, Gander et al. 2000).

1.1.2 External (Side-stream) MBR

For a side-stream MBR system, the filtration units are installed externally to the main bioreactor. The biomass could be pumped directly through a number of membrane modules in series and back to the bioreactor, or the biomass is pumped to a bank of modules, from which a second pump circulates the biomass through the modules in series. Cleaning and soaking of the membranes can be undertaken in place with use of an installed cleaning tank, pump and pipework.

1.2 Advantages of MBR over Conventional Activated Sludge (CAS) Process

First, the MBRs require a much smaller footprint than CAS systems because the MLSS concentration in the MBRs are several times higher than that of CASs (Ben Aim and Semmens 2003, Chu et al. 2008, Huang et al. 2001), which means a higher applicable organic loading rate accompanied by higher biomass concentration (Falk et al. 2009, Fenu et al. 2010, Verrecht et al. 2010). Second, the MBR system can be more effective in simultaneous nitrification and denitrification, due to the maintenance of high organic loading rates and low Dissolved Oxygen (DO) concentrations (Baek and Pagilla 2008). Third, benefits of MBR operation yields high quality treated wastewater, easy control of SRT and HRT, and less sludge.

1.2.1 High Effluent Water Quality

The MBR process is capable of running at a longer SRT than a conventional activated sludge process, thus allowing the growth of slow-growing microorganisms, improving the removal of refractory organic compounds and making for a more robust system to load variations and toxic shocks. MBR process usually results in complete and stable nitrification owing to the retention of slow-growing nitrifying bacteria at a prolonged SRT (Davies et al. 1998, Li et al. 2006, Yoon et al. 2004).

Meanwhile, membrane process also acts as a barrier to separate suspended solids including microorganisms from water. The use of membranes can improve effluent water quality for water reuse since bacteria and suspended solids (SS) are larger than the membrane pore size. As a

result, membrane plays a role in disinfection to achieve up to 7 logs of inactivation of total coliforms (Hirani et al. 2010, Krauth and Staab 1993, Le-Clech 2010, Pollice et al. 2008, Rosenberger et al. 2002). So, the water produced from MBR process is almost free of pathogens and other microorganisms. The excellence in solid-liquid separation allows MBR operation at a very high MLSS concentration (up to approximately 20 g/L), which makes it efficient to deal with recalcitrant compounds. Therefore, MBRs have a superior effluent water quality than conventional activated sludge processes (Hirani et al. 2010, Krauth and Staab 1993, Le-Clech 2010, Pollice et al. 2008, Rosenberger et al. 2002).

1.2.2 Easy SRT and HRT Control

CAS processes cannot have a very long SRT, since long SRT will cause sludge bulking and high biomass concentrations, prohibiting the operation of gravity-based sedimentation in clarifiers. In contrast, in the MBR process, there is almost no sludge loss in the effluent, and the SRT can be controlled with more flexibility.

The MBR process allows for more flexible operation due to separate control of SRT and HRT. Unlike CAS operation, it does not need to consider sufficient long HRT for floc formation (Judd 2008, Khongnakorn et al. 2007, Teck et al. 2009). MBRs can be operated at a SRT as high as 100 days with the biomass concentration ranging from 10,000 to 50,000 mg/L(Muller et al. 1995). Nevertheless, the recent trend of MBR operation is to apply lower solids retention times (around 10–20 days), resulting in more manageable MLSS levels (10 to 15 g/L)(Le-Clech et al. 2006).

1.2.3 Less Sludge Wasting

One of the most competitive advantages of MBRs over traditional activated sludge systems is its long SRT operation, resulting in reduced sludge production (Gander et al. 2000). Sludge treatment and disposal is one of the major challenges in activated wastewater treatment plants and can represent up to 60% of their total operating costs (Canales et al. 1994, Wang et al. 2013). Therefore, effective reduction of sludge production is possible through MBR operation. Over the past decades, various sludge reduction technologies have been developed, which can be applied either in the sludge return line to promote sludge degradation, or in the sludge treatment processes to enhance aerobic or anaerobic digestion (Wang et al. 2013). Reducing sludge production in MBR wastewater treatment process is attracting extensive attention since it allows for decreased sludge production in the first place and therefore decreases the subsequent sludge management costs (Mahmood and Elliott 2006). In an MBR system operated at a long SRT, there is almost no sludge wastage.

1.3 MBR Operation

1.3.1 Organic and Nutrient Removal in the MBR

MBRs have become an important wastewater treatment process that is capable of transforming wastewater to high quality effluent suitable for various water recycling applications (Atkinson 2006b, Fane and Fane 2005). Due to recent technical innovation and drastic cost reduction in membrane materials, the high biomass concentrations at long SRTs are favorable for the

biodegradation of organic pollutants, resulting in high rate treatment systems.

In traditional MBRs intensive aeration is carried out to support microbial growth and control membrane fouling. This intensive aeration gives MBRs excellent removal capabilities when dealing with organic matter and ammonia nitrogen. However, the adverse effect of intensive aeration eliminates the anoxic conditions necessary for denitrification and results in poor total nitrogen removal in the MBR systems (Kim et al. 2008, Patel et al. 2005). Recent research efforts have been conducted to overcome this drawback and improve nitrogen removal with modified reactor configurations. For instance, the Anoxic/Oxic MBR removed COD, NH_4^+-N, and TN effectively with the average removal efficiencies of 96.4%, 99.1% and 75.8%, respectively (Kuang et al. 2012).

Biological nitrogen removal can efficiently remove organic nitrogen compounds to harmless nitrogen gas (N_2) and is generally more cost effective than physicochemical methods (Ahn 2006, Kim et al. 2008). By optimizing the ratio of sludge recirculation to the anoxic reactor, the biological nitrogen removal efficiency reached 90% in an MBR system consisting of two anoxic and aerobic reactors in series (Abegglen et al. 2008). In another study of the effect of MBR configuration on nitrogen removal, the A_2/O (Anaerobic/Anoxic/Oxic) MBR process achieved higher organic, total nitrogen and nitrate nitrogen removal efficiencies of 95%, 95% and 91%, respectively (Kim et al. 2008).

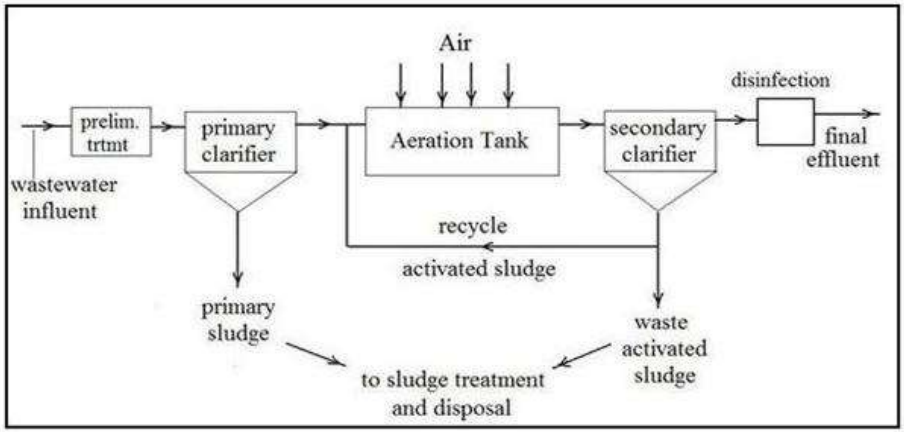


Figure1.1 Activated Sludge Wastewater Treatment Flow Diagram

The classical configuration of activated sludge process for nitrogen removal includes separate aerobic and anoxic zones arranged in an appropriate sequence to enable optimal performance. However, conventional biological nitrogen removal processes such as post-denitrification process have the disadvantage of requiring an external carbon source (Downing and Nerenberg 2008). Alternative processes using MBR and membrane-aerated biofilm reactor (MABR) techniques have the potential to overcome the disadvantage with simultaneous nitrification and denitrification. The characteristics of high biomass concentration in the MBR may allow simultaneous nitrification and denitrification because anoxic zone is formed in the inner side of biomass floc (Sarioglu et al. 2009).

Simultaneous nitrification and denitrification occur to accomplish biological nitrogen removal in a single sludge process. Compared to conventional biological nitrogen removal with nitrification and denitrification in two separate tanks, simultaneous nitrification and denitrification have advantages such as small footprint to save space and reduced construction costs (Bernat and

Wojnowska-Baryła 2007). Another advantage is a reduced demand or need for alkalinity chemicals (Andrade do Canto et al. 2008). Simultaneous nitrification and denitrification can take place inside the activated sludge flocs because of DO concentration gradients in the flocs. High DO concentrations at the exterior layer of flocs result in aerobic zone for autotrophic nitrification. Due to the limited DO diffusion and high oxygen consumption of nitrifiers, anoxic micro-zones develop in the inner rings of the floc, which favors the growth of heterotrophic denitrifiers to convert nitrates produced in exterior layers to nitrogen gas (Holman and Wareham 2005). Heterotrophic denitrifiers have ability to reduce nitrate under micro-aerobic conditions at the DO concentration of 0.8-2.0 mg/L (Bernat and Wojnowska-Baryła 2007). With possible co-respiration mechanism of aerobic denitrification, the heterotrophic denitrifiers can simultaneously use oxygen and nitrite/nitrate as electron acceptors. Furthermore, parallel channels of electron transport chains in microorganisms act to simultaneously transfer electron flows to denitrifying enzymes and oxygen-reducing enzymes (Huang and Tseng 2001). The high sludge concentrations in an MBR is not only beneficial to nutrient removal, but is also beneficial to the removal of micro-pollutants that tend to accumulate in the sludge, either due to their intrinsic hydrophobicity or via electrostatic interactions with the biomass (Sipma et al. 2010).

1.3.2 Effect of SRT on MBR Performance

SRT is one of the most important parameters affecting the biodegradation in activated sludge systems (Sipma et al. 2010). There have been many research efforts devoted to bioreactor

operating conditions and biomass characterization to achieve best performance (Delai Sun et al. 2007, Rosenberger et al. 2002, Tan et al. 2008). These include, but are not limited to, SRT (Ersu et al. 2010), sludge property characterization (Liang et al. 2010), and membrane fouling control (Menniti and Morgenroth 2010). SRT is the key design factor in activated sludge systems including MBRs. Although optimal operating conditions such as SRTs have yet to be clearly defined in MBRs, the membrane system can be operated at a much higher SRT than in the CAS system without affecting the biodegradation capacity (Pollice et al. 2008). Although the specific bacterial growth rates generally decreased as the SRTs increased, this did not affect organic degradation performance or effluent water quality with the help of high biomass concentrations (Pollice et al. 2008). Long SRTs also result in reduced sludge production in the MBRs. For instance, at a prolonged SRT of 300 d, the observed sludge yield and endogenous decay rate in the MBRs were 0.115 g VSS/g COD and 0.024 day^{-1}, respectively, half the reported lower values in the traditional CAS systems (Teck et al. 2009). Furthermore, since the concentration of soluble microbial products (SMP) that affect membrane fouling generally decreased with increasing SRT (Meng et al. 2009), it appears preferable to run MBRs at relatively long SRTs (e.g., SRT = 50 d) to control SMP concentration and improve organic and nutrient removal (Ersu et al. 2010).

Regarding the selection of SRT in MBR operation, in one study of the effect of SRT on the municipal wastewater treatment by pre-denitrification SMBR systems, the highest total nitrogen removal was achieved at an SRT of 33.3 days due to higher MLSS concentration and lower DO

concentrations in the mixed liquor recirculation flow (Tan et al. 2008). Studies also indicate that the importance of mixed liquor recirculation and DO control on nitrogen removal and membrane fouling control in pre-denitrification SMBR systems since higher aeration rate minimized membrane fouling while lower aeration rate improved total nitrogen removal (Tan et al. 2008). The positive effect of operation at a long SRT was also reported by Lesjean et al.(Lesjean et al. 2005), who found that the removal of pharmaceuticals increased with a sludge age of 26 days and inversely decreased when the sludge age was set at 8 days. Others, however, found that change in the SRT from 30 days to 10 days resulted in severe fouling (Zhang et al. 2006b). EPS is a big factor affecting membrane fouling, and a lower production rate can result from the reduced availability of easily biodegradable substrate at longer SRTs (Witzig et al. 2002) which leads a lower EPS concentration. Enhanced degradation of EPS at longer SRTs can be explained by a longer contact time between bacteria and these biopolymers (Mass é et al. 2006) More studies are needed to determine biomass characteristics, microbial activities and bioreactor configurations on nitrogen removal in the SMBR systems.

1.3.3 Effect of HRT on MBR Performance

Besides SRT, a relationship between HRT and biodegradation performance is expected as HRT determines the contact time between the pollutant and microorganisms (Sipma et al. 2010). Although MBR process has the capability of providing high removal efficiency of organic and nutrients with insensitivity to hydraulic fluctuation (Chang et al. 2002, Delai Sun et al. 2007, Rosenberger et al. 2002), reducing HRT leads to more severe membrane fouling (Chae et al.

2006a, Cho et al. 2005).

Since short HRT results in high OLR, HRT is a very important operating parameter in MBR systems, which correlated not only to the treatment efficiency but also to the characteristics of activated sludge (Meng et al. 2007). HRT may affect MBR performance through the release of EPS of bacterial origin. For instance, a low HRT could result in high MLSS concentration, sludge viscosity, and high EPS concentration (Meng et al. 2007). These factors resulted in more severe membrane fouling (Meng et al. 2009). Therefore, too low HRT may have a negative effect on membrane filtration, since the low HRT caused high OLR and potentially low DO concentrations which could cause excessive growth of filamentous bacteria in sludge. The growth of certain filamentous bacteria, such as *Sphaerotilus* and *Haliscomenobacer hydrssis*, is favored at relatively low DO concentrations(Liu and Liu 2006). Deficiency of DO is believed to be one of the major causes responsible for most filamentous growth in activated sludge process. Meanwhile, filamentous bacteria have high surface-to-volume ratio than non-filamentous bacteria which enable them to take up more nutrients as the bioreactor operates under high OLRs (Meng et al. 2007). Therefore, HRT has a great impact on the performance of MBR systems. Overall, HRT affects MLSS concentration, EPS, sludge viscosity and filamentous bacterial growth, which all have strong impact on membrane fouling.

1.3.4 Role of MLSS in MBR Operation

MLSS concentration may be linked to membrane fouling, sludge properties, effluent water quality, and so on in MBR systems, which not only affects the pollutant removal efficiencies, but

also affects the service life of membrane modules. However, SRT, HRT and SRT to HRT ratio are the most important parameters determining the biomass concentration and MBR performance.

The activated sludge concentration can be calculated by the following equation:

$$X = Y_H \frac{\Theta c}{\tau} \frac{1+f_D b_H \Theta_C}{1+b_H \Theta_C} (S_{so} - S_0) \quad (1.1)$$

Where

X = biomass concentration, mg/L

τ = hydraulic retention time, day

Θc = solid retention time, day

Y_H = intrinsic biomass yield, mg biomass COD/mg substrate COD

Sso = influent substrate concentration, mg/L

Ss = effluent substrate concentration, mg/L

b_H = biomass decay constant, day^{-1}

f_D = fraction of biomass forming biomass debris

Since the influent substrate concentration is a constant and the effluent substrate concentration is often negligible, while Y_H, b_H and f_D are constant for activated sludge in an MBR system, it is clear that the SRT and SRT to HRT ratio control the biomass concentration. As mentioned before, as SRT directly affect MBR performance and membrane fouling while HRT and MLSS can also affect MBR performance and membrane fouling, this thesis aimed to fix the SRT to

HRT ratio so that the MLSS can be kept at a relatively constant level in order to determine the effect of SRT and HRT on MBR performance.

1.4 MBR Fouling

1.4.1 Membrane Fouling Mechanisms

Membrane fouling is a major obstacle that hinders fast commercialization of MBR technology. the membrane fouling can be defined as the undesirable deposition and accumulation of microorganisms, colloids, solutes, and cell debris within/on membranes (Fenu et al. 2010) A very comprehensive review about fouling is available elsewhere (Chang et al. 2002, Sun et al. 2011), which is likely affected by sludge characteristics, operational parameters, membrane materials and feed-water characteristics. To date the fouling mechanism remains to be studied.

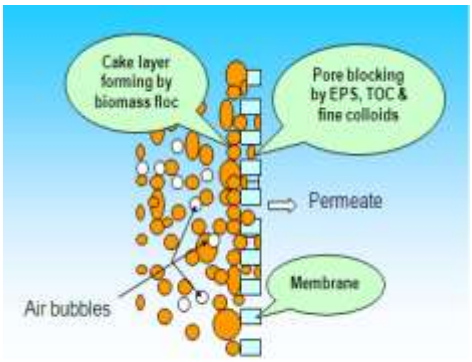


Figure 1.2 Mechanisms of Membrane Fouling

As a result of membrane fouling, there is a reduction of permeate flux or an increase of transmembrane pressure (TMP). Membrane fouling occurs due to the following mechanisms: (1) adsorption of solutes or colloids within/on membranes; (2) deposition of sludge flocs onto the

membrane surface; (3) formation of a cake layer on the membrane surface; (4) detachment of foulants attributed mainly to shear forces; (5) the spatial and temporal changes of the foulant composition after the long-term operation (e.g., the change of bacteria community and biopolymer components in the cake layer).

One of the fouling explanations is ‘cake layer’ theory. The resistance of the cake layer accounts for 95–98% of the total filtration resistances in membrane filtration (Ramesh et al. 2007). The permeability of the cake layer can be affected by flux, electrostatic interactions, and particle size. Several observations include the following (Petsev et al. 1993):

- If salts do not cause aggregation in the feed, the permeability of the cake layer sharply decreases with the increase in electrolyte concentration.
- The permeability of the cake layer sharply decreases with the increase in permeate flux because the increased flux results in a more compressed cake layer.
- The permeability of the cake layer increases with the surface potential of the particles due to the increase in the inter-particle repulsion. However, above a certain value of surface potential, a plateau value for the permeability is reached.
- The permeability of the cake layer passes through a minimum with the increase in the particle size.

A previous study (Rosenberger et al. 2006) showed that the non-settleable sludge fraction (soluble and colloidal material, i.e. polysaccharides, proteins, and organic colloids) were

impacted. The solutes in the sludge supernatant played a significant role in the initiation of cake layer formation (Bae and Tak 2005). Others have found the relative contributions of SS, colloids, and dissolved matter on membrane fouling were 24, 50, and 26%, respectively (Bouhabila et al. 2001). Hence, sludge deflocculation will cause an increase in small particles and soluble organic matter, which in turn lead to rapid decline of membrane permeability.

The overgrowth of filamentous bacteria in sludge could result in severe membrane fouling due to the formation of non-porous and thick cake layer (Meng et al. 2006a, Meng et al. 2006b). Since filamentous bacteria may produce more foulants (e.g. EPS) than did floc-forming bacteria(Choi et al. 2002), the overgrowth of filamentous bacteria can be considered as a fouling indicator.

1.4.2 Trans-membrane Pressure

Given the complex nature of the activated sludge, it is not surprising that the fouling behavior in MBRs is more complicated than that in most membrane applications. Generally, as shown in Figure 1.1, a three stage fouling history has been proposed (Cho and Fane 2002, Zhang et al. 2006a)

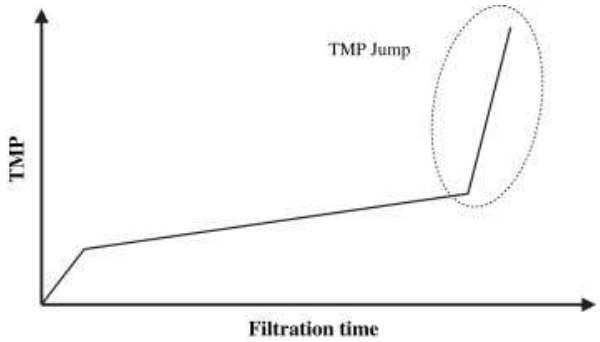


Figure 1.3 Schematic illustration of the occurrence of TMP jump in MBR operation.

Stage 1: an initial short-term rapid rise in TMP;

Stage 2: a long-term weak rise in TMP;

Stage 3: a sharp increase in $\frac{dTMP}{dt}$, also known as TMP jump.

The TMP jump (Figure 1.1) is believed to be the consequence of severe membrane fouling factors. Cho and Fane attributed the TMP jump to the changes in the local flux due to fouling eventually causing local fluxes to be higher than the critical flux (Cho and Fane 2002). Zhang et al. reported that the sudden jump was possibly not only due to the local flux effect, but also caused by sudden changes of the cake layer structure (Zhang et al. 2006b). Due to oxygen transfer limitation, the bacteria in the inner cake layer tend to die and release more extracellular polymeric substances (EPS). The sudden jump of TMP was closely related to the sudden increase in the concentration of EPS at the bottom of cake layer, which might be attributed to the bacterial decay in the inner of cake layer (Hwang et al. 2008).

The occurrence of the TMP jump also depends on operating conditions. For instance, an abrupt TMP jump of over 10 kPa was observed at 24 and 48 h for the fluxes of 30 and 20 L/(m^2 h), respectively (Pollice et al. 2005, Zhang et al. 2006b, a). However, there was no TMP jump during the 280 h operation at 10 L/ (m^2 h). The interactions between TMP jump and these operating parameters are very complex, and TMP jump occurs inevitably during long-term operation of MBRs. Thus, the overall goal of fouling control is to retard the occurrence of the TMP jump via operating the filtration below critical flux and through rigorous sludge characterization analysis.

1.4.3 Fouling due to EPS production

The EPS, produced by activated sludge, play an important role in cake layer formation on membrane surface thus causing bio-fouling (Ramesh et al. 2007). EPS composition in activated sludge also affects membrane fouling (Al-Halbouni et al. 2009). Generally, the specific cake layer resistance increased as the bound EPS level increased (Ahmed et al. 2007). While bound EPS can be fractionized into loosely bound EPS and tightly bound EPS, it was the loosely bound EPS that caused the fouling problem in MBRs (Ramesh et al. 2006). The loosely bound EPS was found to correlate with the performance of flocculation and sedimentation process as well as the dewaterbility of activated sludge (Li and Yang 2007). EPS in either bound or soluble form are currently considered as the predominant cause of membrane fouling in MBRs. The bound EPS consists of proteins, polysaccharides, nucleic acids, lipids, humic acids, etc., which are located at or outside the cell surface. Soluble EPS and SMP are generally considered the same. SMP can be defined as the pool of organic compounds that are released into solution from microbial metabolism (usually associated with biomass growth and decay) (Barker and Stuckey 1999).

Bound EPS is not only as a major sludge floc component keeping the floc in a three-dimensional matrix, but also as a key membrane foulant in MBR systems. As the bound EPS concentration increased, the specific cake resistance increased resulting in the rise of TMP (Ahmed et al. 2007, Chae et al. 2006b).

The EPS content is determined by the balance between microbial production and subsequent

degradation. It is suggested that EPS production is a result of bacterial response to changing environmental factors, including changes in the substrate concentration and stress conditions induced by shear and/or predation (Bossier and Verstraete 1996). Adverse environmental conditions may result in a higher EPS production due to switching on EPS production genes (Laspidou and Rittmann 2002). Therefore, it is not surprising that EPS production is closely related to microbial growth and substrate consumption rates, although there is still debate as to how SRT affects EPS production. Some studies indicated higher EPS concentration at longer SRTs (Badireddy et al. 2010, Ng and Hermanowicz 2005), while others observed lower concentrations at longer SRTs (Ahmed et al. 2007, Mass é et al. 2006).

Sludge deflocculation may lead to the release of EPS from sludge flocs into the mixed liquor(Morgan-Sagastume and Grant Allen 2005). Deflocculation refers to a dysfunction of the activated sludge process characterized by the formation of a very small sludge floc, or the absence of floc formation (Chae et al. 2006a). Deflocculation can be the result of operating conditions and environmental stresses such as shift in temperature, toxic compounds, metals, DO concentration, pH, substrate loading, and nutrient conditions (Li et al. 2008b). In the system with deflocculated sludge, the colloids and solutes in mixed liquor were the major contributors to membrane fouling (Meng and Yang 2007). Sludge bulking also impacts membrane fouling. In contrast, the impacts of bulking sludge on membrane fouling mainly resulted from the deposition of suspended solids onto the membrane surface. On the one hand, as the filamentous bacteria could deposit on the hollow fiber membrane and release the foulants on the membrane surface.

On the other hand, the filamentous bacteria can produce more biopolymers and lead to the increase of bound EPS in sludge flocs. Therefore, the severe cake fouling might be caused by the deposition of filamentous bacteria and bound EPS. Nevertheless, we still cannot conclude which one had a more significant influence on membrane fouling. So, further mechanistic research is needed to determine the impact of filamentous bacteria and bound EPS (Meng and Yang 2007).

1.4.4 MBR Fouling Control

Many anti-fouling strategies can be applied to MBR applications. They comprise, for instance, membrane backwashing, where permeate water is pumped back to the membrane, and flow through the pores to the feed channel, dislodging internal and external foulants. Backwashing with air is another common method where pressurized air in the permeate side of the membrane builds up and releases a significant pressure within a very short period of time. Membrane modules therefore need to be in a pressurized vessel coupled with a vent system. Air usually does not stay inside the membrane. If it did, the air would dry the membrane and a rewet step would be necessary, by pressurizing the feed side of the membrane. In addition, chemical cleaning may also be recommended, such as chemically enhanced backwash. The prevalent cleaning agents are sodium hypochlorite (NaClO) and citric acid. It is common for MBR suppliers to develop specific protocols containing information such as chemical concentrations and cleaning frequencies for chemical cleaning for individual facilities (Le-Clech et al. 2006).

1.5 Sludge Characterization

SMBR has been developed for municipal wastewater treatment in the last few decades to produce high quality water, reduce reactor sizes and minimize sludge production (Lesjean et al. 2004). However, SMBRs can be more complex to operate than the conventional activated sludge process because there is a lack of knowledge of sludge properties in the MBR and of their effects on membrane fouling.

1.5.1 Sludge Production

At long SRTs, mass balance on COD in the MBR suggests that around 90% of the influent COD is oxidized to carbon dioxide and MLSS concentration in the reactor is almost constant without sludge wastage (Yamamoto et al. 1989). In fact, sludge production is related to specific growth rate and MLSS concentration. If the MLSS concentration is relatively constant, the sludge settling and dewatering properties would be the most important factors.

1.5.2 Sludge Settling and Dewatering properties

In a study of the relationship between sludge properties and membrane fouling in MBRs, the dewatering ability and filterability of sludge in MBRs were evaluated by the measurements of specific resistance to filtration and sludge volume index (SVI) (Khongnakorn et al. 2007). The results indicated that the dewatering ability of sludge in the MBR was comparable to that in the CAS system. The capillary suction time of sludge in the MBR correlated with the amount of soluble microbial product (SMP) and associated filamentous bacteria. Hence, capillary suction

time couldbe a good indicator of membrane fouling potential (Pan et al. 2010). Within the SMP, the fraction of utilization associated products, due to their higher percentage of low molecular weight molecules, presented the highest specific cake resistance and appeared to have the highest fouling potential (Jiang et al. 2010).

1.5.3 Sludge Bulking

Sludge bulking, which is often caused by excessive growth of filamentous organisms in activated sludge, results in poor sludge settling, sludge loss from secondary clarifiers and deterioration of effluent water quality (Guo et al. 2012, Kappeler and Gujer 1994, Nielsen et al. 2009). Filamentous bulking has been found to have a strong influence on MBR fouling (Meng and Yang 2007). The overgrowth of filamentous bacteria leads to a sharp increase of bound EPS concentration and then induces the increase of sludge viscosity and sludge hydrophobicity (Chae et al. 2006a). In addition, the filamentous bacteria may release the foulants on the membrane surface. When the activated sludge flocculates poorly, the level of suspended solids of the supernatant will increase.

1.5.4 Sludge Filtration Property

There were many factors affecting membrane filtration property, such as MLSS concentration, which has been studied in the past 20 years. Membrane fouling is often considered to be caused by the deposition of particles on the membrane surfaces. However, MLSS concentrations between 2 and 24 g/L had little influence on filterability (Rosenberger and Kraume 2002). A

more recent study, however, indicated that there was little difference in filterability for the concentrations of MLSS ranging from 4 to 8 g/L, but there was a significant increase in critical flux when the MLSS concentration increased to 12 g/L (Le-Clech et al. 2003). So, a systematic method is needed to help determine the membrane filtration property.

The objective of this research was to systematically examine the effects of various sludge characteristics by using a submerged MBR operated at different SRT and HRT conditions to treat synthetic wastewater. The sludge in the MBR was characterized by measuring LB-EPS, MLSS, temperature, viscosity, TTF, and DSVI. Although SVI has been often used as an operating parameter to assess the membrane filterability for activated sludge (Roest et al. 2002), it is widely recognized that the deposition of particles on the membrane surface is fundamentally different from their settling by gravity. On the other hand, like DSVI, TTF has been also suggested as a filterability index to predict the fouling potential in wastewater MBR operations because of its simplicity for measurement (Rabie et al. 2001). TTF measures the effects of cake formation on sludge dewaterability only under static conditions. Nevertheless, neither TTF nor DSVI can be used individually to assess the MBR fouling potential because the accumulation of particles on the membrane surfaces involves different mechanisms from either the dewaterability under static conditions or settlability for gravity settling (Fan et al. 2006).

1.5.5 Sludge Activities

Organic matter and nutrients can be effectively removed in WWTPs using alternating

aerobic-anoxic processes such as two stage anoxic/aerobic process with flow recirculation, four-stage Bardenpho process, and simultaneous nitrification/ denitrification via intermittent aeration in a single biofilm reactor, sequencing batch reactor, or MBR (Danesh and Oleszkiewicz 1997, Grady 1999, Patel et al. 2005). Conventional nitrogen removal relies on sequential nitrification and denitrification by autotrophic and heterotrophic microorganisms, respectively.

The autotrophic and heterotrophic activities of activated sludge in the two MBRs during the study period were determined by batch extant respirometry with specific oxygen uptake rate (SOUR) measurement due to ammonia oxidation and acetate oxidation, respectively (Hu et al. 2002).

1.6 Research Objectives

Since SRT and HRT are the key operating parameters in active sludge bioreactor operation, it is important and necessary to study and compare the effect of SRT and HRT on sludge properties and membrane performance which are of significance to wastewater treatment. Currently, little performance evaluation work has been done to systematically change SRT and HRT values while maintaining a constant SRT to HRT ratio.

The main objectives of this study were in the following:

- To determine the effect of SRT and HRT on organic and nitrogen removal in MBR operation

- To evaluate the effect of SRT and HRT on settling and filtration property of activated sludge in MBRs
- To determine the effect of SRT and HRT on membrane fouling

2 Materials and Methods

2.1 Bioreactor Setup and Operation

Two bioreactors were set up for MBR performance evaluation. One served as a control system (Figure 2.1) operated at a constant SRT and HRT. The other MBR (Figure 2.2) was operated at different HRTs and SRTs but with constant SRT/HRT ratio so that the biomass concentrations in the MBR were maintained relatively constant throughout the study period.

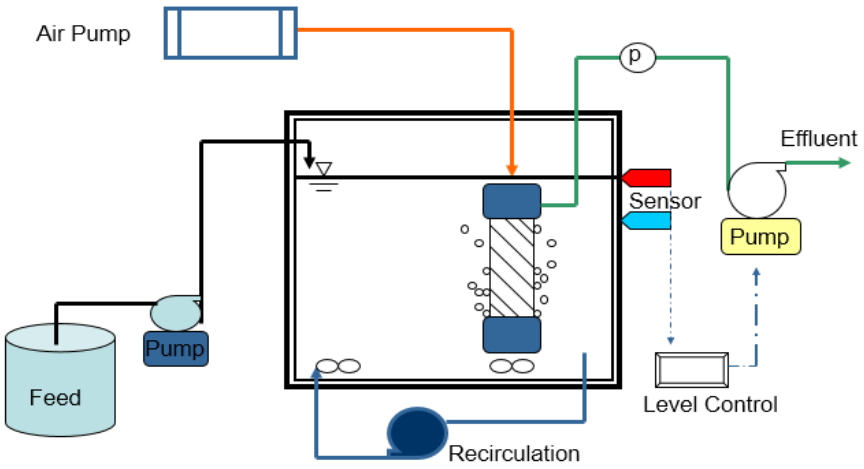


Figure 2.1 A schematic of a lab-scale submerged MBR with 180 d STR and 1 d HRT. P stands for a transmembrane pressure device.

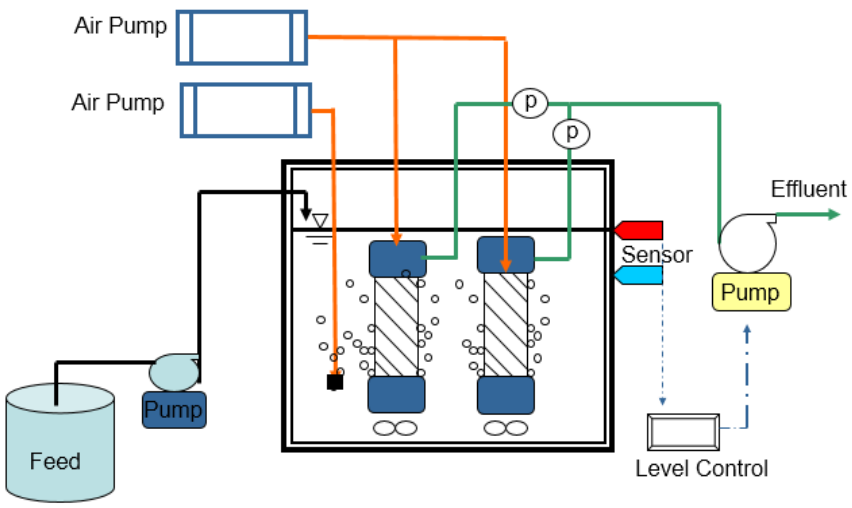


Figure 2.2 A schematic of a lab-scale MBR operated with different STRs and HRTs (Phase Ⅰ (SRT=180 d, HRT=1 d), Phase Ⅱ (SRT=90 d, HRT=12 h), PhaseⅢ(SRT=45 d, HRT=6 h)). P stands for a transmembrane pressure device

Both bench-scale MBRs were equipped with the ZeeWeed hollow fiber membrane module (GE Water & Process Technologies, Trevose, PA). The membrane module was made of PVDF with a nominal pore size of 0.1 μm and an effective surface area of 0.047 m^2. The MBRs had a total effective reactor volume of 7.2 L and was run under aerobic conditions. The upper and lower water level sensors (Cole-Palmer, Vernon Hills, Illinois) were applied to maintain a relatively constant mixed liquor volume in the MBRs. The volume difference between the upper and lower water level was less than 5% of the total mixed liquor volume in the MBRs. The sensor is designed to activate an onboard solid-state relay when the sensor detects a change of water level. When the water level reaches the upper limit because of continuous feeding, the upper level sensor triggers the operation of a permeate pump. When the water level reaches the lower limit, the lower level sensor assures pump shut-down. In this study, a suction peristaltic pump after the

membrane module acted as the permeate pump to produce a relative vacuum for permeate collection. An online digital pressure gauge (Cole Palmer) was installed to measure the TMP. The speed of permeate pump was set at a permeate flow rate higher than the influent flow rate so that the permeate pump was intermittently turned on and off by the upper and lower water level sensors, respectively, to keep the total mixed liquor volume relatively constant. An air pump supplied compressed air to the built-in orifices at the bottom of each membrane module at a constant air flow rate of 6 L/min for each membrane module to support aerobic biodegradation and control membrane fouling.

With a target HRT, the MBR was fed continuously with synthetic wastewater containing nonfat dry milk powder as a primary organic carbon source at a COD concentration of approximately 500 mg/L. Other nutrient of the synthetic wastewater included 51.7 mg/L total nitrogen (TN), 30 mg/L NH_4^+-N, and 6 mg/L PO_4^3-P. The micronutrients in the feed solution contained the following: 44 mg/L $MgSO_4$, 14 mg/L $CaCl_2 \cdot 2H_2O$, 2 mg/L $FeCl_2 \cdot 4H_2O$, 3.4 mg/L $MnSO_4 \cdot H_2O$, 1.2 mg/L $(NH_4)_6Mo_7O_{24} \cdot 4H_2O$, 0.8 mg/L $CuSO_4$, and 1.8 mg/L $Zn(NO_3)_2 \cdot 4H_2O$. The prepared synthetic wastewater was prepared and stored at room temperature (24 ± 2 ℃) in a covered 100 L plastic bin.

The inoculation sludge was taken from the aerobic tank from the Columbia Wastewater Treatment Plant (Columbia, MO), which has a treatment capacity of 20 million gal per day using conventional activated sludge process. A total of 24 L of sludge for each tank was acclimated to

the synthetic wastewater for three days and concentrated via sedimentation before the sludge was transferred to the MBRs. The starting MLSS concentration in each MBR was approximately 6,000 mg COD/L. During the start-up period of this study, there was no sludge wasted until the biomass COD concentration increased to about 8,000 mg COD/L. Afterward, sludge was wasted daily with the target SRT of 180 days to maintain relatively constant biomass concentrations in both MBRs. Initially, both MBRs were operated at the SRT of 180 d and HRT of 1d. One of the MBRs was then run at the fixed SRT/HRT ratio of 180 while varying SRT from 90 to 45 d and HRT from 12 h to 6 h, respectively. One more membrane module was installed in the tank in order to run the MBR at the HRT of 6 h.

2.2 Sludge Settling and Filtration properties

The sludge volume index (SVI), has become the standard measure of the settling property of activated sludge. It is defined as the volume in mL occupied by 1 g activated sludge after settling the aerated mixed liquor for 30 min (Lee et al. 1983). Due to the high biomass concentrations of MBRs, which leads poor settling, diluted SVI (DSVI) was used. The DSVI analysis was conducted by first diluting the sludge sample with wastewater effluent until the settled volume after 30 min was 250 ml/L or less (APHA 1998). In this research, the sludge was diluted by 3-5times before MLSS and DSVI were measured according to the Standard Methods.

Time to Filter (TTF) represents the filtration property of active sludge. TTF was determined by using a 90-mm Buchner funnel and filter papers (P5 with a pore size of 1 μm, Cat. No.: 09-801B,

Fisher Scientific). After pouring 200 mL mixed liquor directly from MBRs, the time required to obtain 100 mL of filtrate was measured at the vacuum pressure of 51 kPa (or 7.4 psi).

Sludge viscosity is another important factor for MBR system which has a strong relationship with membrane fouling and sludge bulking issues (Sweity et al. 2011). Activated sludge viscosity was measured at different periods in this research, using a rotational viscometer (Cole Parmer, P/N: 98965- 43).

2.3. Microbial Activities from the MBR operated at different SRTs and HRTs

Autotrophic and heterotrophic activities of microorganisms in the MBRs were determined through the specific oxygen uptake rate (SOUR) measurement (Hu et al. 2002). Aliquots (120 mL) of the sludge collected from each bioreactor that were operated at a defined SRT and HRT were poured into two 50 mL respirometric bottles followed by aeration with pure oxygen. The respirometric bottles were tightly capped with no headspace afterwards. At a predetermined time, an aliquot of substrate (with a final concentration of 10 mg N/L NH_4^+-N or 20 mg/L COD in acetate) was added using a 10 μL glass syringe. A decrease in the DO level in the respirometric vessel was measured by a DO probe (YSI model 5300A, Yellow Springs, OH) and continuously monitored at 4 Hz by an interfaced personal computer. The oxygen uptake rate was calculated based on a linear regression analysis because a zero-order reaction was observed for a long period of time. SOUR was calculated by dividing OUR by biomass concentration of each sample. All SOUR experiments were carried out in at least duplicate.

2.4. Live/Dead Fluorescent Staining and Microscopic Analysis

The activated sludge samples were subjected to live/dead analysis after fluorescent staining using the LIVE/DEAD® BacLight^{TM} bacterial viability kit (Invitrogen Co., Carlsbad, CA), according to the work reported elsewhere (Hu et al. 2003). A laser-scanning confocal microscope (Zeiss LSM 510 META) was used for fluorescence imaging of bacterial cells. Meanwhile, a bright microscope was regularly used for to determine the change in sludge properties such as floc size and morphology.

2.5. Membrane Fouling Monitoring and EPS Analysis

The TMP and permeate flux of each membrane module were closely monitored throughout the study period, during which time the membrane flux was maintained relatively constant by adjusting the speed of the permeate pump. When the TMP increased dramatically in a short period of time and the TMP level exceeded 45.5 kPa, the membrane module was taken out of the MBR for physical cleaning by rinsing with distilled water for 0.5 h before it was submerged in the mixed liquor in the MBR.

The biomass concentration and influent and effluent water quality parameters, such as ammonia-N, nitrate-N, nitrite-N, orthophosphate-P, and COD concentrations were determined following the standard methods (APHA 2005). Since loosely bound EPS is directly related to membrane fouling in activated sludge operation (Li and Yang 2007), it was monitored throughout the study according to the method described elsewhere (Hwang et al. 2007, Jorand et

al. 1994, Zhang et al. 1998) with minor modification. Briefly, aliquots (50 mL) of MLSS from the MBRs were centrifuged at 8,200 g for 5 min to separate the soluble EPS from bound EPS (Teck et al. 2009). The supernatant was discarded and pellets resuspended in a 50 mL solution containing 8.5% sodium chloride and 0.22% formaldehyde. The suspension was held in an ice bath and sonicated at a power output of 20 W for 30 min. After centrifugation at 12,000 g and 4 ℃ for 30 minutes, the loosely bound EPS concentration in the supernatant was analyzed for total polysaccharides and proteins, while the sum of the total polysaccharides and proteins was reported as the loosely bound EPS. Polysaccharides were determined by the phenol-sulfuric acid method with glucose as a standard (Dubois et al. 1956). Proteins were quantified by a modified Micro Lowry method using a total protein assay kit (Sigma-Aldrich, Product Code TP0300 and L 3540) containing standards of Bull Serum Albumin for calibration.

2.6. Biomass and Chemical Analysis

The biomass concentration in the MBRs and influent and effluent water quality parameters, such as ammonia-N, nitrate-N, nitrite-N, orthophosphate-P, and COD concentrations were determined following the standard methods (APHA 2005).

3 Results and Discussion

3.1 MBR Performance

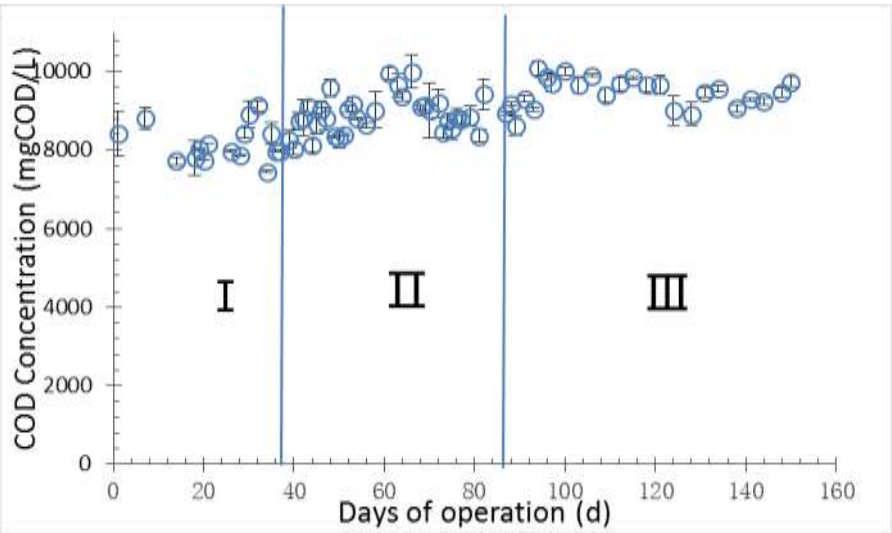


Figure 3.1. Biomass concentration in the MBR run at different SRTs and HRTs. The error bars represent the data range of duplicate samples. Phases Ⅰ, Ⅱ, Ⅲ represent the operating periods at different SRTs or HRTs: Phase Ⅰ (SRT=180 d, HRT=1 d), Phase Ⅱ (SRT=90 d, HRT=12 h), PhaseⅢ(SRT=45 d, HRT=6 h).

At the target SRTs of 180 days, 90 days and 45 days for Phases I, II, and III, respectively, by controlling the amount of sludge wasted, the HRTs were maintained at 1 day, 12 h and 6 h, respectively. Because a constant SRT/HRT ratio was maintained throughout the study period, the biomass concentrations ranged from 8,000 to 10,000 mg COD/L (Figure 1). Based on Equation 3.1, the observe biomass yield increases as SRT decreases because SRT has mainly effect on the observe yield).

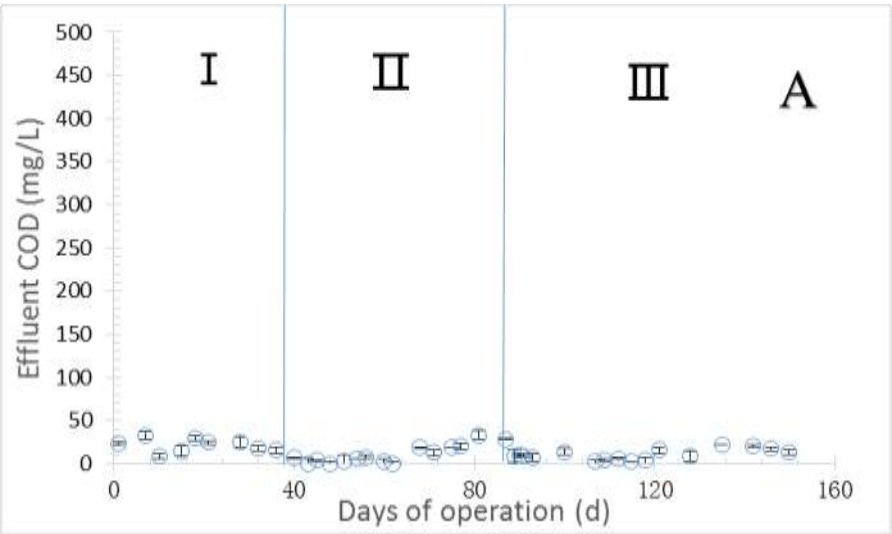
$$Y_{obs} = Y_H \frac{1+f_D b_H \Theta_C}{1+b_H \Theta_C} \quad (3.1)$$

where Y_H = intrinsic yield, mg/L, b_H = first-order biomass decay constant, day^{-1}, f_D = specific

growth rate, day^{-1}, Θc = solid retention time, day

As aresult, the biomass concentrations increased from an average of 7,697 ± 2,033 mg/L in Phase I to 8,889 ± 478 mg/L in Phase II and 9,473 ± 375 mg/L in Phase III.

The effluent COD data (Figure 3.2) demonstrated effluent COD were 13 ± 9 mg/L, with more than 97.4 % of influent COD removed due to long SRT operation along with the excellence in membrane filtration in MBR operation.



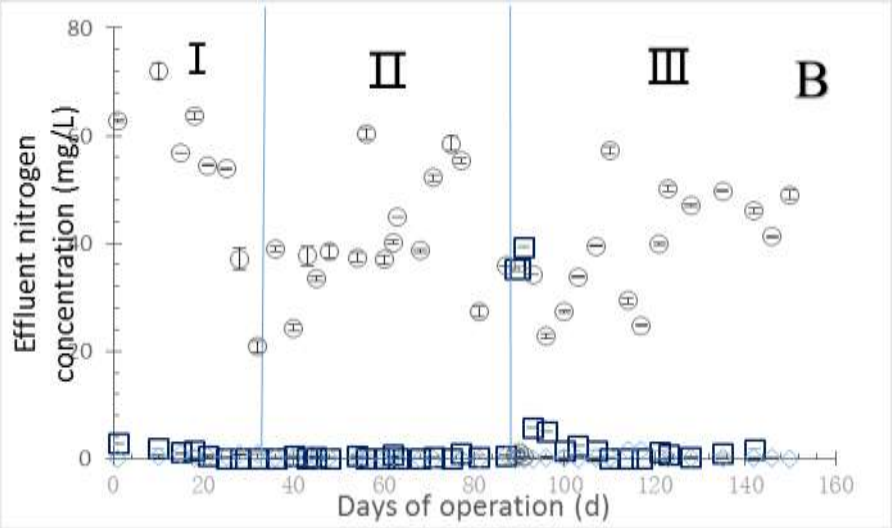


Figure 3.2. Change in the effluent COD concentration(A) and effluent concentrations of NH_4^+-N (□), NO_2^--N (◇) and NO_3^- -N (○) in the MBR systems(B) in phase Ⅰ, Ⅱ, Ⅲ. The error bars represent the data range of duplicate samples. Phases Ⅰ, Ⅱ, Ⅲ represent the operating periods at different SRTs or HRTs: Phase Ⅰ (SRT=180 d, HRT=1 d), Phase Ⅱ (SRT=90 d, HRT=12 h), PhaseⅢ(SRT=45 d, HRT=6 h).

A similar trend was also observed with respect to the concentrations of effluent nitrogen species (Figure 3.2). However, at the beginning of Phase III, due to the decrease in HRT from 12 h to 6 h, the influent flow rate doubled while the MBR was operated at a constant aeration rate, resulting in a decrease in DO concentration from 2 mg/L to 0.2 mg/L. As a result, the nitrate-N concentration dropped to almost 0, and the effluent ammonia-N concentration was almost as high as that of the influent, which was around 40 mg/L.

Three days after the MBR was run at low DO concentrations, another air pump was added to increase the DO concentration of the mixed liquor in the range of 2.0 to 3.0 mg/L, and the

effluent water quality recovered in 10 days. The ammonia-N concentration droped to 5.8 mg/L just in one day while nitrate-N concentrations increased to the level as before.

3.2 Change in Sludge Settling and Filtration Properties

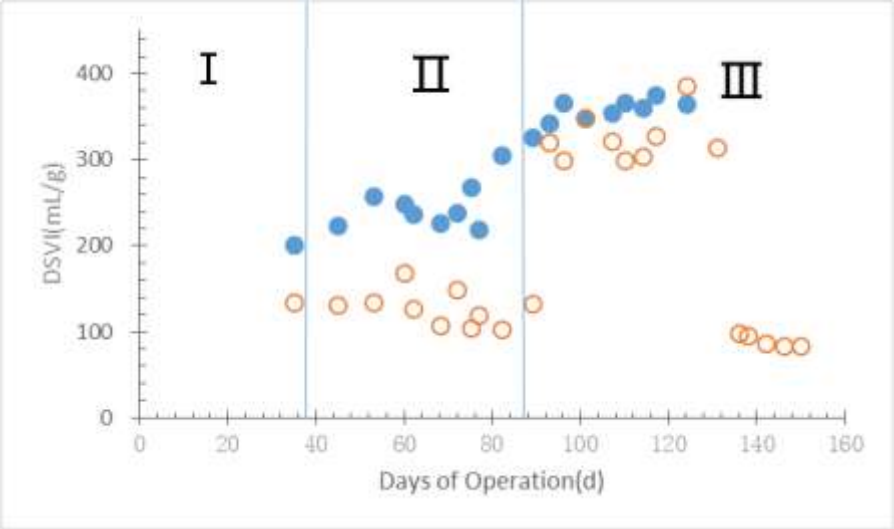


Figure 3.3. Changes in DSVI in the control MBR (SRT=180 d, HRT=1 d) (●) and the MBR operated at different SRTs and HRTs (○) in phases Ⅰ, Ⅱ, and Ⅲ as described earlier.

As shown in Figure 3.3, the DSVI values of the sludge from the control MBR continued to increase, which was attributed to the growth of filamentous bacteria (details described later). Due to the long SRT and low organic loading rate, MBR operation usully results in the growth of filamentous bacteria (Li et al. 2008a, Martins et al. 2004). For comparison, in the MBR with variable SRTs and HRTs, DSVI values were low in Phases I and II as there was barely filamentous bacteria (Figure 3.3) with the DO concentrations of the mixed liquor ranged from 2.0 to 3.0 mg/L. However, DSVI changed a lot at the beginning of Phase III because of the dramatic increase in filamentous bacteria which was likely caused by the low DO conditions,

although it lasted for only 3 days.

Even though the DO concentration was maintained to 2.0 to 3.0 mg/L, DSVI did not decrease promptly. In fact, it took 44 d (starting from day 131) before a significant decrease in DSVI was observed. A very short HRT under constant aeration conditions would lead to a low DO concentration in the MBR, which could help the growth of filamentous bacteria thus resultingin poor sludge settling.

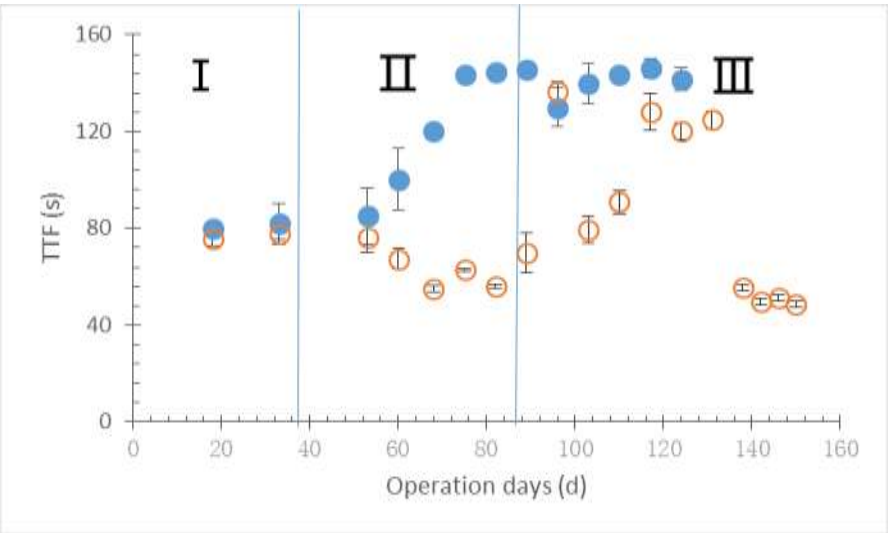


Figure 3.4. Time to filter for the sludge in the control-MBR (●) with fixed SRT and HRT and in the MBR run at different SRTs and HRTs (○) in phases Ⅰ, Ⅱ, and Ⅲ as described earlier. The error bars represent the data range of duplicate samples.

Another important sludge property is Time to Filter, and the less TTF represents the stronger filtration ability of sludge. Remarkably, the trend of change in TTF (Figure 3.4) was similar to that of DSVI, which was linked to the growth of filamentous bacteria and its effect on sludge settling and fouling property. For instance, in the control MBR, like DSVI, the TTF value

gradually increased with time as filamentous bacteria continued to grow (details described below). The growth of filamentous bacteria may facilitate the formation of cake-layer on the surface of membrane, which causes membrane fouling and results in the increase of TTF.

There were similarities between the DSVI profile and TTF profile in the MBR run at different SRTs and HRTs. However, there was difference between the increase of TTF and the growth of filamentous bacteria population or increase in DSVI during Phase III, indicating that not only the amount of filamentous bacteria but other factors may affect TTF measurement. Even though a significant growth of filamentous bacteria was observed (M-2, Figure 3.5), it might require some time for the filamentous bacteria to grow on the surface of membrane. As the filamentous bacteria was washed out on day 138, TTF decreased as low as before.

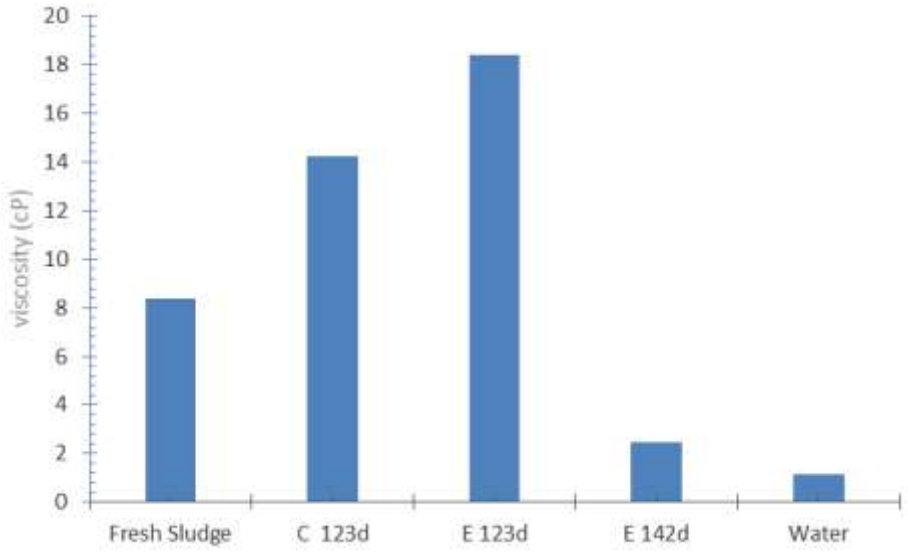


Figure 3.5. Change in sludge viscosity at 20 ℃ from different sludge sources with different filamentous bacteria population. C 123d was the sample taken from the control MBR on day 123, E 123d and E 142d were the samples taken from the MBR run at different SRTs and HRTs on days 123 and 142, respectively.

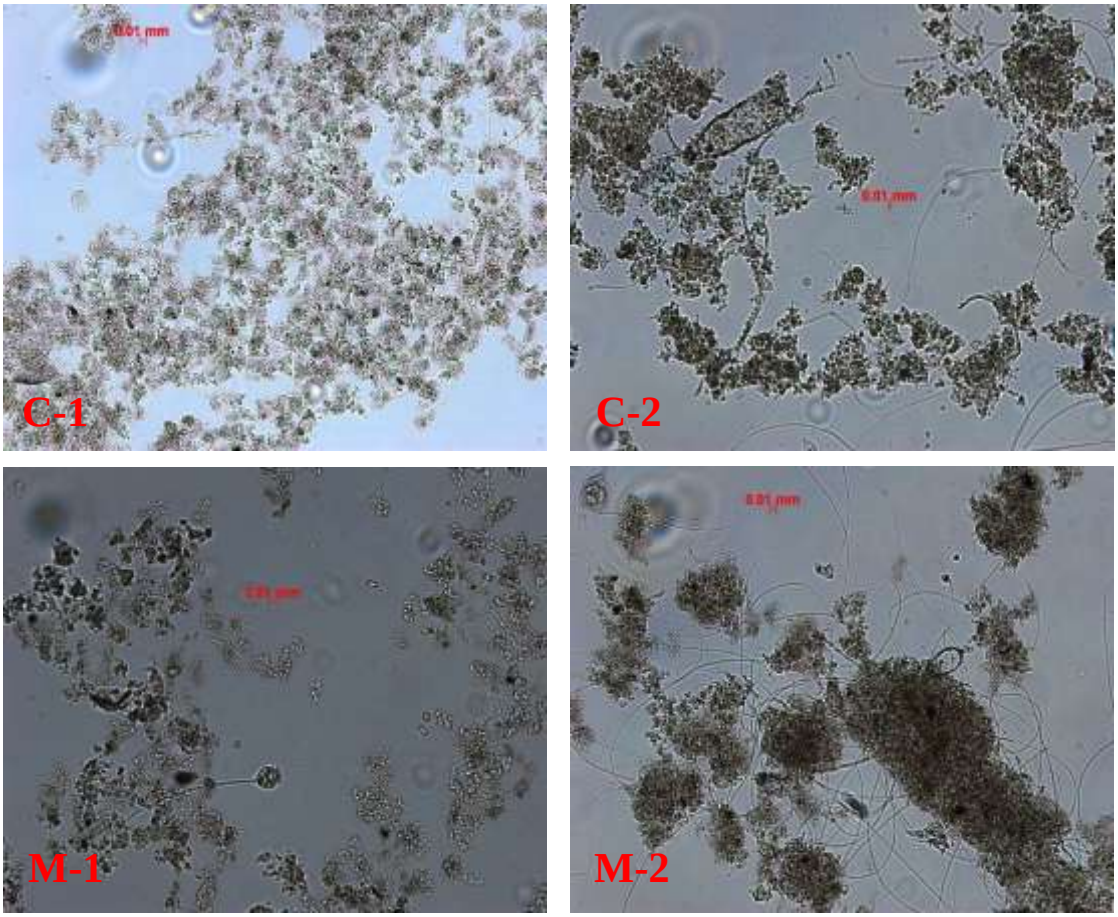
Viscosity was another important factor to track sludge properties in the MBRs. The increase in viscosity was linked to poor membrane permeability and high membrane resistance which resulted in worse membrane fouling (Chae et al. 2006b, Li et al. 2007, Trussell et al. 2007). Similar results were obtained in this study as viscosities for C 123d and E 123d samples with the sludge containing filamentous bacteria were much higher than that of E 142 sample where sludge had almost no filamentous bacteria. As the result, viscosity decreased as the filamentous bacteria was washed out after DO correction.

3.3 Change in Sludge Morphology

Since the sludge properties changed a lot because of the low DO concentration after shortening HRT to 6 h, light microscopy and fluorescent micrscopy were used to determine floc size and mophorology.

Light microscopic images were taken at the end each phase from both MBRs. For the sludge from the control tank, sludge samples C-1 and C-2 were taken on day 30 and day 90, respectively. It appeared that after a long-term operation, C-2 had more filamentous bacteria than C-1. For the MBR with variable SRTs or HRTs, M-1 was taken at the end of Phase I (on day 30, SRT = 180 d, HRT = 1d), there was almost no filamentous bacteria initially. As the SRT and

HRT decreased to 90 d and 12 in Phase II, sludge sample M-2 (taken on day 90), there appeared to be more filamentous bacteria and the flocs were bigger than in M-1. In M-3, due to low HRT (6 h) operation and low DO concentrations in the mixed liquor for three days, there were a dramatic increase in filamentous bacterial population. Only after about 50 d of operation at normal DO concentrations (2-3 mg/L), the sludge sample M-4 taken from Phase III shows the normal sludge properties with little growth of filamentous bacteria (Figure 3.6).



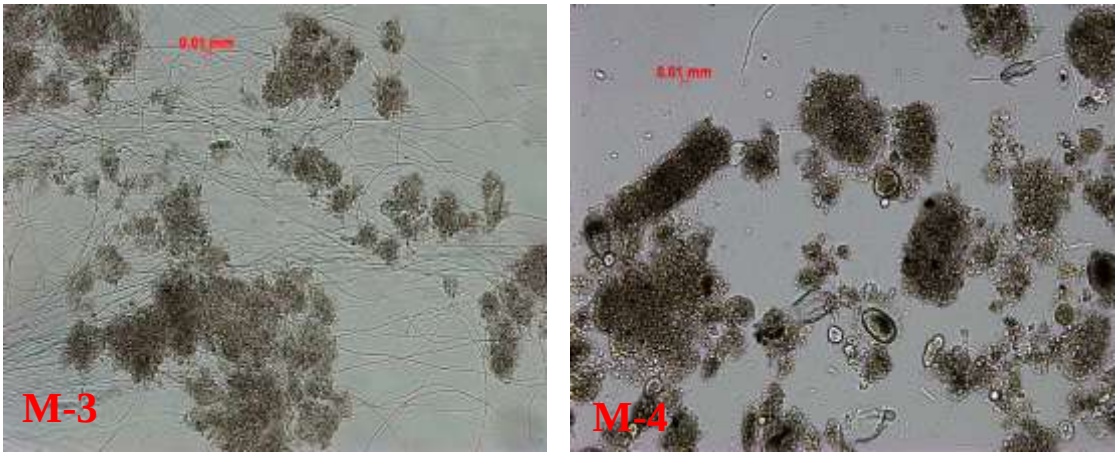
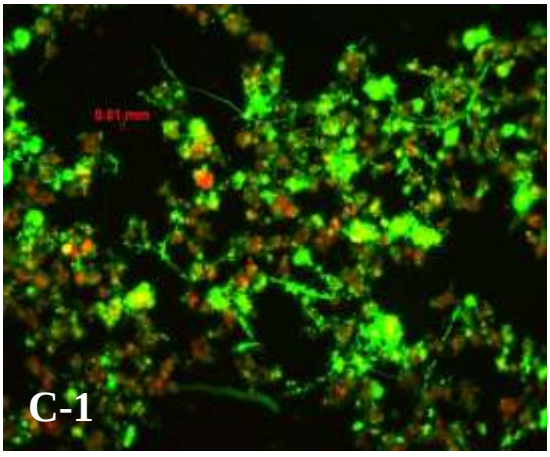


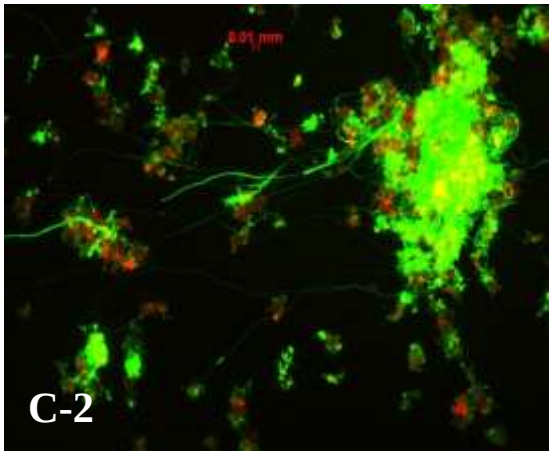
Figure 3.6. Light microscopic images of the sludge from the control MBR with fixed SRT/HRT and the MBR with different SRTs and HRTs.

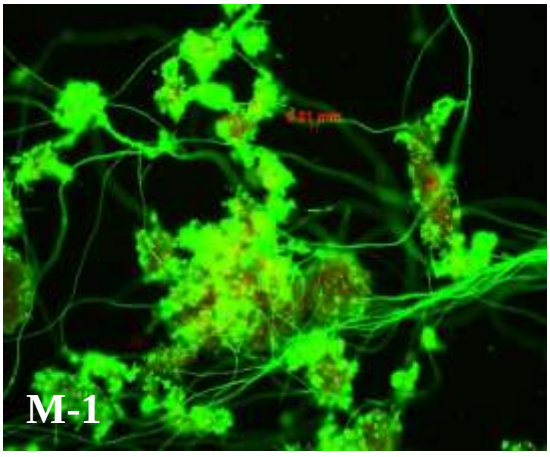
The activated sludge samples from both MBRs were further subjected to live/dead analysis after fluorescent staining.

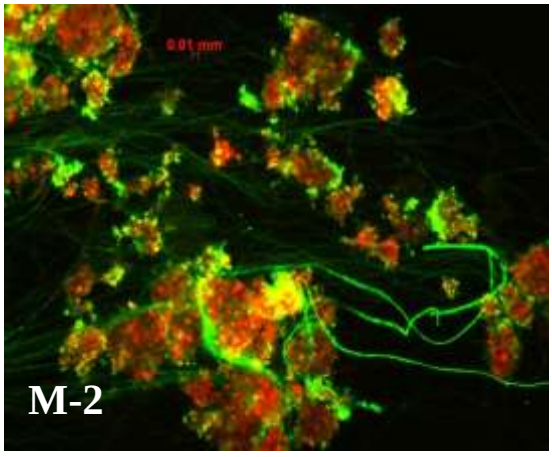
Pictures of sludge samples C-1 and C-2 from the control MBR were taken on day 87 and day 117, respectively. Consistent with that of light microscopy, there was little change in live to dead bacteria ratio by visulization. However, the population of filamentous bacteria appeared to increse with operating time.

Fluorescent images of sludge samples M-1, M-2, M-3 and M-4 from the MBR with different STRs and HRTs were taken on days 87, 117, 131, and 142, respectively.


C-1


C-2


M-1


M-2

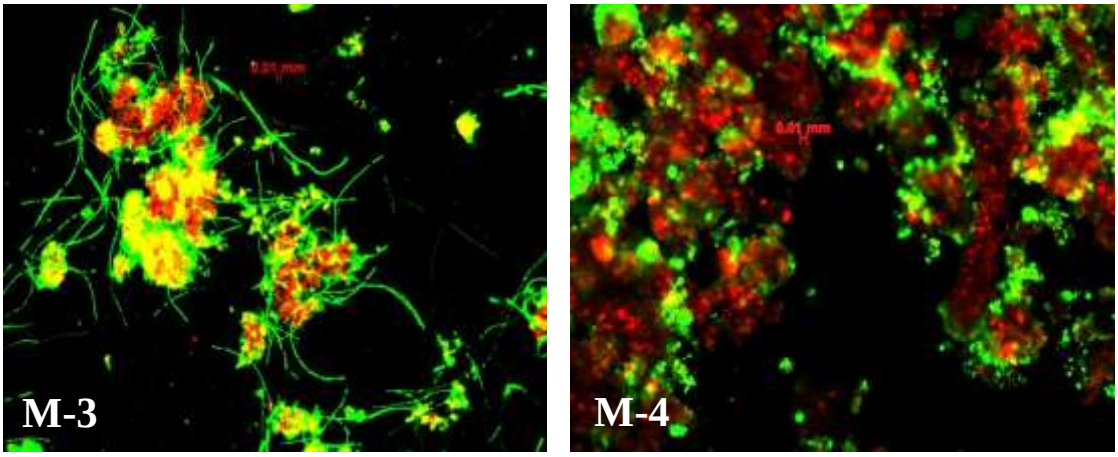


Figure 3.7. Fluorescent images of sludge samples taken for the control MBR (C-1, and C-2) with fixed SRT and HRT and the other MBR run at different SRTs and HRTs (M-1 to M-4). Dead cells are shown in red and live cells are shown in green after live/ dead staining.

Compared to the sludge sample M-1, more dead cells of M-2 to M-4 were observed as cultivation continued due to long SRT operation. Furthermore, fimamentous bacterial growth became significant in M-2 and M-3 samples. Due to the low DO concentration and high organic loading rate at the beginning of Phase III operation, there was a huge increase in filamentous bacteria as shown in M-2. Meanwhile, M-2 showed that almost all filamentous bacteria were alive compared to floc-forming bacteris, which suggests that filamentous bacteria have a stronger ability than other bacteria to survive well under low DO condtitions.

With a prolonged period of aeration to maintain the DO from 2 to 3 mg/L, sludge sample M-3 showed that the amount of filamentous bacteria decreased. After a total of about 50 d of vigourous aeration, there was almost no growth of filamentous bacteria in M-4, as was confirmed with low DSVI values (Figure 3.3).

3.4 Membrane Performance

Figure 3.8 describes the change in TMP and permeates flux of individual membrane modules in the MBR.

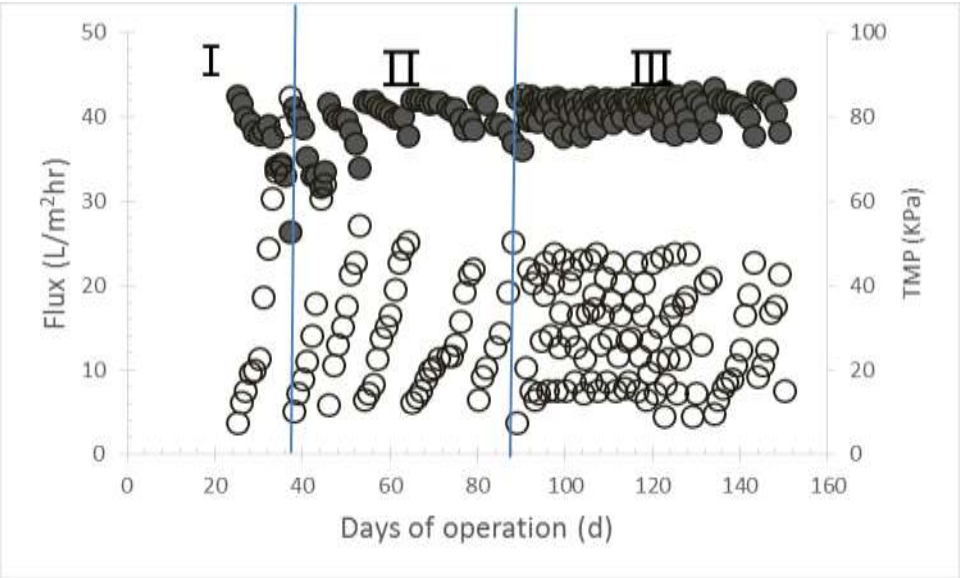


Figure 3.8. Change in the flux (●) and TMP (○) in the MBR run at different SRTs and HRTs in phases Ⅰ, Ⅱ, and Ⅲ as described earlier.

During Phases I and II, the membrane wash frequence was from 8 to 10 days. At the end of Phase II operation, the membrane wash interval increased to 14 days, as also evident from the low TTF data (Figure 3.4) and low viscosity (Figure 3.5) in the same period.

However, when HRT decreased further from 12 h to 6 h in Phase III operation, the membrane module had a poor performance due to fouling (details shown in Figure S1). The membrane

module had to be washed every two days since it was so easily blocked. The serious fouling issue was not only due to the doubled influent flow rate, but also due to the increase in filamentous bacterial population due to low DO conditions that lasted for three days. Only after about 50 d of vigorious aeration (from day 134 onward), the membrane module performance improved with less wash frequence needed.

3.5 Change in the EPS Concentration

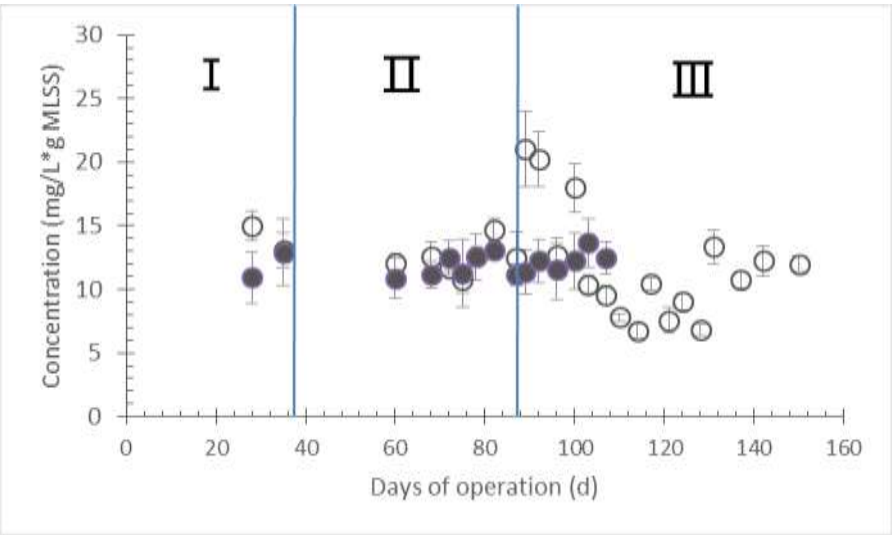


Figure 3.9. EPS concentration in the control-MBR (●) with fixed SRT/HRT and in the MBR run at different SRTs and HRTs (○) systems run at different SRTs and HRTs in phases Ⅰ, Ⅱ, and Ⅲ as described earlier. The error bars represent the data range of triplicate samples.

Compare to that of the control MBR, the EPS concentrations in the MBR run in Phases I and II were at the same level. However, at the beginning of Phase III operation under low DO conditions, the EPS concentration increased significantly (Figure 3.9). Nevertheless, the EPS concentration decreased after the correction of DO in the MBR run at the SRT of 45 d and HRT

of 6 h. After about 50 d of vigorious aeration, the EPS concentration remained low.

Even though the LB-EPS concentration remained unchanged at different SRTs and HRTs, the individual EPS component such as protein and polysaccharide had different respronses. As shown in Figure S3, polysaccharide concentration decreased as the SRT and HRT decreased. The change in protein concentration appeared to be oppsite with less significance (Figure S3). Possibly, longer SRTs resulted in biodegradation of proteins and lower EPS related protein production as they can serve as a food source for bacterial growth (Obayashi and Gaudy Jr 1973), especially at low organic loading rates.

Interestingly, the increase in protein fraction of EPS was accompanied by severe sludge bulking at the beginning of Phase III due to the operation under low DO conditions. Proteins are more likely to be involved in electrostatic bounds with multivalent ions because of the high content of amino acids and thus may play a more important role in the bioflocculation process than polysaccharides (Laspidou and Rittmann 2002).

3.6. Microbial Activities in the MBRs

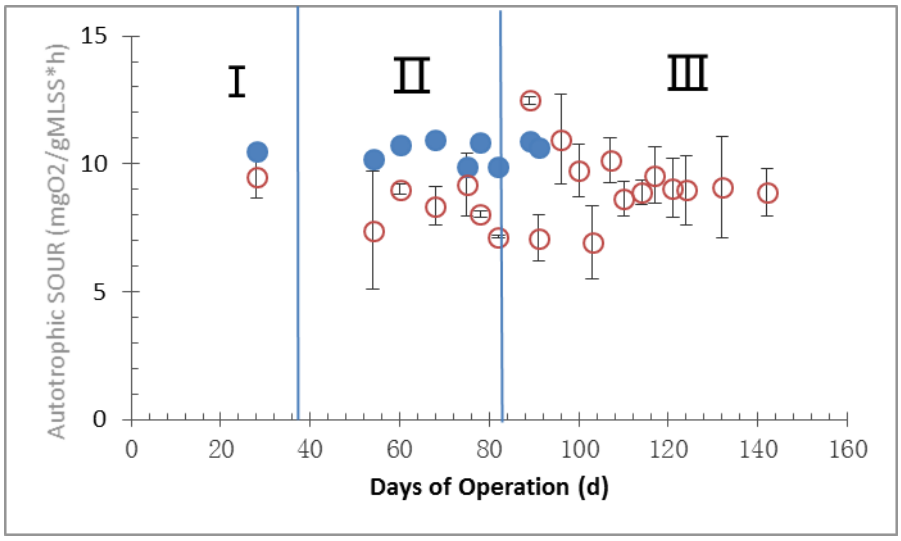


Figure 3.10. Autotrophic SOUR for the control-MBR (●) with fixed SRT and HRT and in the MBR run at different SRTs and HRTs (○) systems run at different SRTs and HRTs in phases Ⅰ, Ⅱ, and Ⅲ as described earlier. The error bars represent the data range of duplicate samples.

With the average autotrophic SOURs for the control MBR and the MBR run at different SRTs and HRTs were 10.50 ± 0.41 and 8.95 ± 1.30 $mgO_2/g/h$,ewspectively. There was no significant difference of autotrophic activitiy between the MBRs run at different SRTs and HRTs. Because SOUR was calculated by dividing the total biomass and nitrifying bacteria only represented a few percent of total biomass, the insignificant change in autotrophic SOUR is expected. It also suggests that the shorter HRT or the higher organic loading rate has little effects on autotrophic growth as long as SRT is long to keep the nitrifiers in the MBR.

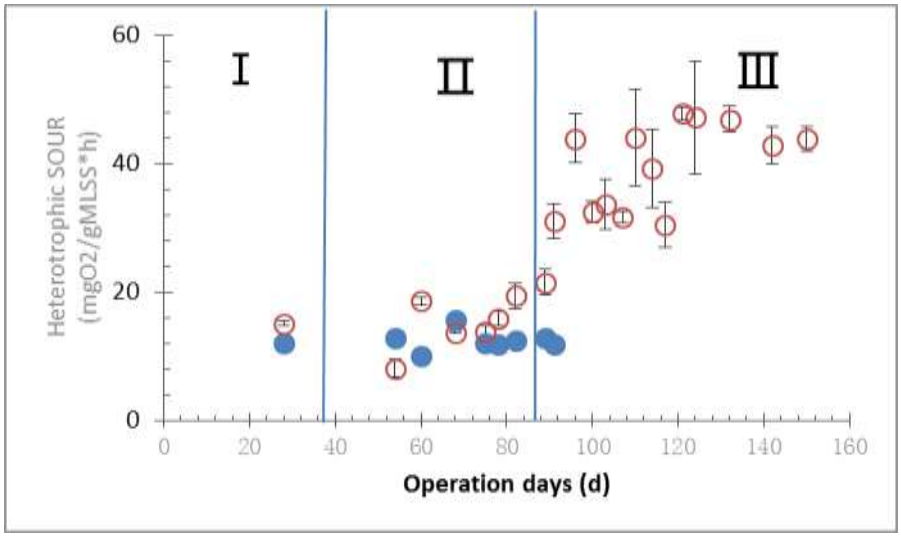


Figure 3.11. Heterotrophic SOUR for the control-MBR (●) with fixed SRT and HRT and in the MBR (○)run at different SRTs and HRTs in phases Ⅰ, Ⅱ, and Ⅲ as described earlier. The error bars represent the data range of duplicate samples.

On the other hand, the SOUR of heterotrophs increased as the SRT decreased (Figure 3.11). It is well known that the active biomass fraction decreas as SRT increases (Equation 1.1). Therefore, at relativelty constant biomass concentrations, the results of heterotrophic SOUR are consistent with prediction.

4. Conclusions

This research determined the sludge settling and filtration properties at constant SRT/HRT ratio while varying SRT and HRT proportionally. The sludge settling parameter DSVI appeared to have a good relationship with sludge filtration paramter TTF. As SRT decreased to 45 d and HTR decreased to 6 h, significant sludge bulking and poor filtration were observed, largely due to the excess growth of filamentous bacteria at low DO concentrations under high organic loading conditions. Lower HRT resulted in faster membrane fouling due to higher influent flow rate. However, the system recovered in about 50 d after the correction of low DO concentrations in the mixed liquor. Due to the potential sludge bulking problems at long SRT (180 d) operation, the results suggest the MBR operation at the SRTs of 45 to 90 d results in excellent sludge settling/filtration properties and effluent water quality.

5 Future Study

The mechanism of EPS production and its effect on membrane fouling are still not very clear. Furthermore, little is known about the flocculation ability of specific proteins and polysaccharides, which are involved in membrane fouling. Future study ia needed to characterize proteins and polysaccharides of EPS on a molecular basis. This would also give more detailed insight in the mechanisms involved in the bioflocculation and biofouling process.

Molecular biology work can be used in MBR study to determine the species of filamentous bacteria since different filamentous bacteria may have different effect on sludge settling and filtration properties. This study only evaluated one operating condition (at a fixed SRT/HRT ratio of 180) by changing SRT and HRT proportionally. More research is needed to determine the effect of SRT and HRT (with broad ranges) on sludge properties and overall MBR performance evaluation.

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Appendix:

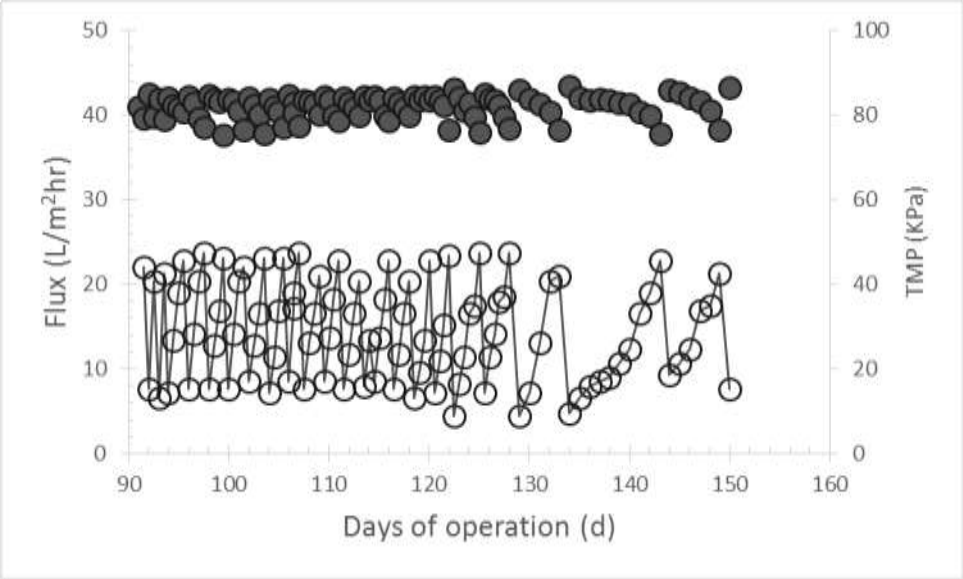

Figure S1. Changes in the Flux (●) and TMP (○) for third period in the MBR systems run at45 d SRT and 6 h HRTs.

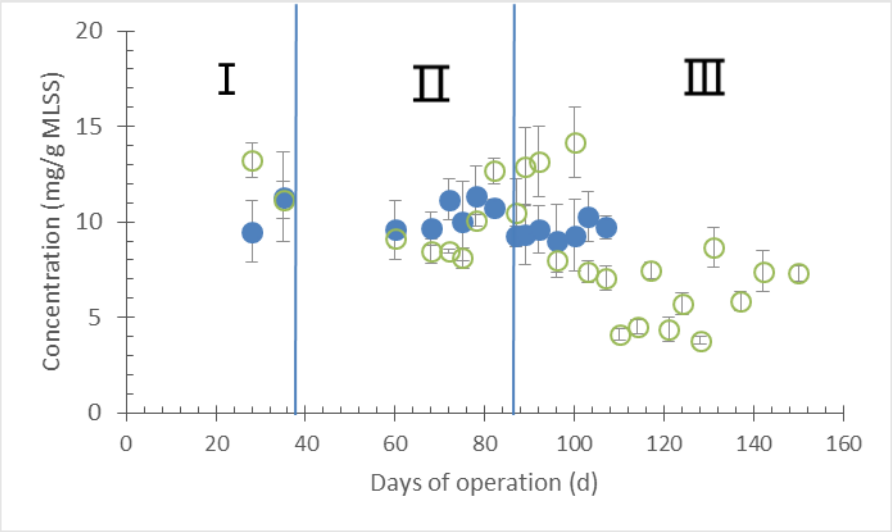

Figure S2. Changes in the polysaccharide concentration (●) for EPS in the control-MBR with fixed SRT and HRT and Polysaccharide concentration for EPS in the MBR run at different SRTs and HRTs (○). The error bars represent the data range of triplicate samples.

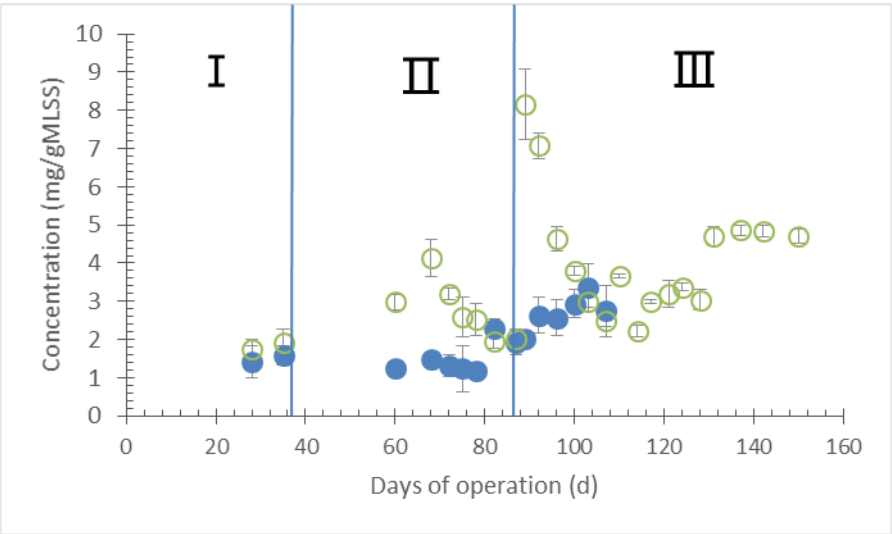

Figure S3. Changes in the protein concentration (●) for EPS in the control-MBR with fixed SRT and HRT and Protein concentration for EPS the MBR run at different SRTs and HRTs (○). The error bars represent the data range of triplicate samples.