Enhancement of wastewater treatment under low carbon/nitrogen ratio by using submerged membrane electro-bioreactor (SMEBR)

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Abstract

Enhancement of wastewater treatment under low carbon/nitrogen ratio by using submerged membrane electro-bioreactor (SMEBR)

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The submerged membrane electro-bioreactor (SMEBR) system has been proven to be effective for wastewater treatment, especially for nutrient removal. In this study, the SMEBR treatment efficiency was compared to conventional MBR treatment applied to the same municipal wastewater. The study was carried out in two side by side continuous flow reactors with the volume 14 liters over 6 months in lab. The changes in sludge quality, effluent quality and operational condition were recorded for both reactors while different C/N ratios (from 3 to 1) had been applied. The results proved that under an adequate dissolved oxygen and current density, the use of SMEBR under the low C/N ratio could improve total nitrogen and phosphorus removal by 30% (more than 50% of TN removal) and more than 99% respectively compared to MBR. Meanwhile, under the low C/N ratio, research has proven the SMEBR's high tolerance and fast recovery from the shock loading condition, helped balancing pH in the sludge, reduced membrane fouling and improved sludge dewatering properties. The results from this study show the possibility of the application of SMEBR to a variety of municipal wastewater.

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Dedication

I dedicate this thesis to my family and my girlfriend who will become to my wife after my graduation. Their caring, support and encouragement during my study and research was the driving force in helping me achieve my goals and fulfill the requirements of my degree.

Abbreviations

AMBR	Aeration Membrane Bioreactor	
C/N	Carbon/nitrogen ratio	
COD	Chemical oxygen demand	
COD/N	Chemical oxygen demand/nitrogen ratio	
DC	Direct current input	
DO	Dissolved oxygen	
EC	Electrical conductivity	
EMBR	Extractive Membrane Bioreactor	
F/M	Food to microorganisms (mass) ratio	
HRT	Hydraulic residence time	
MBR	Membrane bioreactor	
MLSS	Mixed-liquor suspended solids	
MLVSS	Mixed-liquor volatile suspended solids	
NH ₃ ⁻ -N	Ammonia nitrogen as nitrogen	
NO ₃ ⁻ -N	Nitrate nitrogen as nitrogen	
ORP	Oxidation reduction potential	
PO4 ³⁻	Orthophosphorus (Reactive phosphorus)	
SLSMBR	Solid/Liquid Separation Membrane Bioreactor	
SMEBR	Submerged membrane electro-bioreactor	
SMP	Soluble microbial products	
SRT	Sludge residence time	
TKN	Total Kjeldahl nitrogen	
TMP	Transmembrane pressure	
TN	Total nitrogen	
TSS	Total suspended solid	

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Chapter I

Introduction

1.1 Background

Water pollution is one of the major environmental problems in emerging countries with limited water resources. These countries include China, India, and those from the Middle East and Africa. However, due to several limitations (e.g. lack of technical and financial support, space for treatment facilities, etc.), effective wastewater treatment is difficult to achieve. Meanwhile, the effects of chemicals released into the environment have become more serious. At present, there is a need for adequate treatment methods that can reduce many adverse effects on the environment (e.g. eutrophication and red tide) of the rapidly increasing amounts of nutrient-loaded wastewater (Yang et al., 2010). Therefore, nutrient removal is one of the most important factors in designing wastewater treatment systems.

Nutrient removal in modern wastewater treatment is designed as a tertiary treatment process. Various types of treatment units and methods are available, such as flocculation, adsorption, and biological methods (Zhang et al., 2014). Each of these methods has benefits and limitations, which should be considered during the designing process.

The membrane bioreactor (MBR) process is a successful wastewater treatment method widely used throughout the world in conducting high quality COD removal. However, MBR is less efficient in nutrient removal. Meanwhile, the fouling problem is one major challenge in

MBR application (Bani-Melhem et al., 2010 and 2011; Ibeid et al., 2013; Hasan et al., 2012). Wastewater facilities consist of separate units for phosphorus removal and ammonia conversion to nitrates due to nitrification in oxidation conditions. Some facilities have built additional compartments for converting nutrients into gaseous nitrogen due to denitrification in anoxic conditions. However, successful denitrification can only take place when there is adequate C/N ratio, which means that an additional carbon source input should be required for the treatment of wastewater with low C/N ratio.

1.2 Motivation

There is variety of municipal wastewater characteristics while certain municipalities have diluted wastewater with a low C/N ratio. Usually, such ratio is not adequate for biological nitrogen removal. Since previous studies (Ibeid, 2011) reported the possibility of simultaneous removal of nutrients by a novel hybrid submerged membrane electro-bioreactor (SMEBR) using wastewater with a conventional range of C/N ratio (5:1 to10:1), it was necessity to investigate the SMEBR's response to other types of municipal wastewaters (i.e. different C/N). For highest C/N ratio almost complete removals of nitrogen phosphorous and carbon was observed (Ibeid, 2011), which were much better than results from MBR (working side by side with SMEBR). However, many municipalities with combine sewage system have low C/N ratio in wastewater to treat. In spite that the biological processes are affected by low C/N ratio, assessment of the efficiency of SMEBR at low C/N ratio compared to conventional MBR was required,

1.3 Objective

The main objective of this study is to explore the effectiveness of using the SMEBR system in removing nutrients from municipal wastewater, under the specific condition of a low organic carbon input. The particular objectives are as follows:

i) to find effective nitrogen removal under various C/N ratios, particularly a low carbon source input;

ii) to find the capability of phosphorus removal by SMEBR in relation to conditions required for an effective nitrogen removal; and

iii) to suggest operation conditions based on results for effective treatment, which include the control of C/N ratio (as the control of COD), pH, dissolved oxygen (DO), oxidation reduction potential (ORP), total suspended solid (TSS), electrical conductivity (EC) and current density (CD).

Chapter II

Literature review

Compared with MBR process, improvement in handing wastewater treatment has been shown in SMEBR; thus, this literature review discusses the processes that take place simultaneously in SMEBR, namely activated sludge, membrane filtration and electrokinetics. Previous studies have examined the design components and experimental purposes of the SMEBR with special emphasis on its operational condition under conventional C/N ratio.

2.1 Activated sludge process

Activated sludge system is designed to degrade organic compounds, the majority of which can be converted into CO₂ and water through the biological process. (Mackenzie et al., 2012). For the generation and reproduction of microorganisms in the sludge, a carbon source must be provided to serve as the food of the microorganisms. This carbonate organic compound is defined as the substrate, and its measurement is presented in BOD or COD (Zhang et al., 2014).

Apart from the input of the substrate, the effectiveness of the activated sludge system can also be affected by other conditions, such as temperature, pH, and salinity. (Buntner et al., 2013; Krzeminski et al., 2012). Microorganisms experience four phases in their lifecycle, which are lag, exponential growth, stationary, and death (Vesilind et al., 2010). The microorganisms' growth requires high aeration supply and good control of operational conditions. Under the suitable operational conditions, the activated sludge could provide a reliable treatment result, although some limitations and problems can arise. In such cases, solving them requires the application of additional treatments. To control the concentration of sludge, the settling and returning sludge processes have to be applied, and these require extra treatment units (Mackenzie et al., 2012). Moreover, to remove nitrogen compounds as a result of the biological process, the anaerobic-aerobic (activated sludge) process should be applied, which also requires additional operation units (Cho et al., 2005; Kyu-Hong et al., 2003; Christian et al., 2002). Some studies has shown the possibility of nitrogen removal in one reactor (Wang et al., 2008; Udert et al., 2008; Chiu et al., 2007; Wu et al., 2007), but no phosphorus removal. Nitrogen and phosphorus removal are conducting by different microorganisms; hence, with phosphorus removal, the biological treatment requires additional treatment units (Chiu et al., 2007; Wu et al., 2007), so that making the treatment reporcess become more complicate.

2.2 MBR process

As an improved technology of biological treatment, the membrane bioreactor process provides a simpler treatment. In operation, the membrane module successfully substitutes for secondary clarifiers. With smaller facilities, the system provides very reliable treatment results (Buntner et al., 2013; Li et al., 2013; Krzeminski et al., 2012). The classification of different types of MBR processes are based on the type of systems in the market, such as aeration membrane bioreactor (AMBR), extractive membrane bioreactor (EMBR), and solid/liquid separation membrane bioreactor (SLSMBR) (Li et al., 2013).

Applying MBR has several benefits, including the following: the system generates less waste sludge, minimizes the size of the treatment facility, and simplifies manipulation compared with the regular activated sludge treatment (Li et al., 2013; Krzeminski et al., 2012). However, MBR also has limitations and problems that need to be improved, such as the fact that it cannot provide reliable treatment effluent quality with the requirements of nutrient removal. Another major problem is fouling, which is related to the membrane filtration unit itself and the sludge quality (Bugge et al., 2013; Krzeminski et al., 2012; Ibeid, 2011). The MBR operational conditions are affected by fouling, especially the suction rate because sludge particles can block the pore of the membrane module (Bugge et al., 2013; Krzeminski et al., 2012). Membranes with the fouling problem reduce the efficiency of treatment as well as changes sludge volume and properties. Backwash should be frequently applied when the sludge has a high concentration, and the lifecycle of the membrane module are reduced in this case (Ibeid et al., 2013; Krzeminski et al., 2012).

Given that the efficient removal of COD through conventional MBR is related to an adequate biomass growth, sufficient dissolved oxygen is required in the bioreactor, which is similar to activated sludge process (Bugge et al., 2013; Chang et al., 2003). However, phosphorus removal requires a different treatment, and the conventional MBR system is not capable of its removal (Choi et al, 2009; Yoo et al., 1999). In this case, nutrient removal by MBR requires the inclusion of additional units (denitrification) and processes (coagulation) for nitrogen and phosphorus removal (Kim et al., 2010).

2.3 SMEBR

The SMEBR is a newly developed, latest generation MBR method. SMEBR applies an electrical field and combines membrane filtration with the activated sludge biological process (Bani-Melhem et al., 2011; Elektorowicz et al., 2009; Hasan 2011; Ibeid, 2011). Thus, the SMEBR system includes membrane filtration, electrokinetic process, and biological treatment in one reactor vessel (Bani-Melhem et al., 2011). Thus, through the SMEBR, the requirements of minimum space and high quality effluent (i.e., removal of COD, phosphorus, and ammonia removal) can be satisfied (Elektorowicz et al., 2009). Moreover, the development of the SMEBR allows for the creation of a hybrid system, where almost all carbon, nitrogen, and phosphorus can be removed in a single vessel (Bani-Melhem et al., 2011; Elektorowicz et al., 2009; Hasan, 2011; Ibeid, 2011).

2.3.1 Biological treatment

In activated sludge, carbon removal occurs through the mechanism of biodegradation, that is, the use of microorganisms to digest and degrade organic compounds (Guo et al., 2009; Kim et al., 2011; Tyagi et al., 1996). The compounds in wastewater have different biodegradation rates (Sims et al., 1999), and these are important factors to consider in designing treatment units and determining residence times. Separate operation units must be built to enhance the growth of dedicated microorganisms (Kim et al., 2011; Zhang et al., 2014; Vazquez et al., 2006). Many operational features are required, such as sufficient input of oxygen and food supplies (adequate F/M), a good control of temperature and pH and an adequate hydraulic/solid residence time

(HRT and SRT), to successfully run an activated sludge process with biodegradation (Vazquez et al., 2006; Guo et al., 2009). Compared with other wastewater treatment methods, biological treatment is a cost-effective option, and provides good treatment results with respect to carbon removal (Tyagi et al., 1996; Kim et al., 2011). However, the microorganisms are very sensitive to variations in operational conditions; thus, the system might not be able to adjust to sudden changes of conditions, such as variations in aeration or carbon inputs. The system also requires longer recovery time after the occurrence of shock loadings (Tyagi et al., 1996). Moreover, in some circumstances (e.g. low organic carbon supply or inadequate dissolved oxygen in the sludge), the microorganisms cease their activities, resulting in lack of treatment (Kim et al., 2011).

Furthermore, nutrient removal can be done by conducting nitrification and denitrification in the biological process (Cho et al., 2005; Kyu-Hong et al., 2003; Christian et al., 2002). Normally, two individual reactors are established for nitrogen removal: in the reactor with aerobic condition, the aerobic autotrophic nitrifiers transfer ammonia to nitrite and finally to nitrate. Then, in another reactor under anaerobic or anoxic conditions, nitrate transfer to nitrogen gas by heterotrophic denitrifying bacteria (He et al., 2009; Kim et al., 2011). The possibility of complete treatment in one reactor has been proved in some studies, but a good control of operation condition is required (Wang et al., 2008; Udert et al., 2008; Chiu et al., 2007; Wu et al., 2007). Biological treatment provides a cost effective solution of nitrogen removal, however, nitrification process requires relatively high oxygen consumption, and denitrification process is limited by the amount of organic carbon source input that is always deficient in the wastewaters (Guo et al., 2009). Hence, the application of biological treatment is limited. Nevertheless, it is expected that by combining other two treatments membrane filtration and electrokinetics) in SMEBR, the impact from the variation of `biological treatment results can be minimized.

2.3.2 Membrane filtration

As a newly developed technology, many types of membranes are used in the MBRs applied throughout the world. The mechanisms of each type of membrane are slightly different, which include micro-filtration, ultra-filtration, and nano-filtration modules. The identification depends on the utilization of the membrane (Chen et al., 2013). Membrane filtration depends on the pore size and surface area of the membrane being used, and these features are designed to fit various treatment requirements (Li et al., 2013). The cylindrical hollow fiber ultra-filtration membrane has been used in previous studies of the SMEBR (Bani-Melhem et al., 2011; Ibeid, 2011).

The major problem in applying membranes to the activated sludge is fouling. Then, a backwash must be applied after a certain period (a few days to months) of operation (Ibeid et al., 2013; Krzeminski et al., 2012). Fouling is caused by sludge particles on membrane surface, which is the precipitation of the dissolved materials inside the pores, and on the membrane surface. The pore blocking caused by fouling could reduce the rate of membrane filtration. (Cho et al., 1999; Crozes et al., 1997; Sharif, 2011). When the submerged membrane bioreactor works, the aeration supply creates turbulent flow to suspend sludge particles. However, the flow at the bottom of the reactor is slower than that on the surface, making it difficult for the sludge

to avoid settling. As a result, the bottom part of the submerged membrane can experience fouling (Kim et al., 2007). Once the foulants on the membrane exceed the critical value, the water suction rate by membrane system is dramatically reduced, and the sludge volume cannot maintain the balance because of the different flow rates (Kim et al., 2007).

The reduction of suction rate (flux decline) is subdivided into adsorption, concentration polarization, and reversible and irreversible fouling (Choi et al., 2005). The previous filtration experiments demonstrated that the membrane suction rate declines faster with increasing food concentration and membrane pore size and with decreasing tangential flow (Choi et al., 2005). According to Ibeid et al. (2013), the SMEBR can effectively reduce the fouling rate when applying in wastewater with high or low concentration of protein.

2.3.1 Electrokinetic process

Electro-coagulation is one of the electrokinetic processes applied in many different cases of wastewater treatment. The purpose of electro-coagulation is to destabilize suspended, emulsified, or dissolved contaminants in an aqueous medium by applying DC electrical current into the medium (Kobya et al., 2003). The mechanisms involved in electro-coagulation include coagulation, adsorption, precipitation and flotation (Kobya et al., 2003), which are mostly physical treatments. When the anode is made of perforated aluminum sheet, as suggested in a previous research (Ibeid et al., 2013; Ibeid, 2011), the Al³⁺ is released from anode by electrocoagulation process during the operation. The route of reactions has presented as follows (Ibeid, 2011):

 $Al^{3+} + OH^{-} \rightarrow Monomeric species (e.g. Al(OH)^{2+}, Al(OH)_{2}^{1+}, Al(OH)_{4}^{-}) \rightarrow Polymeric species$

(e.g.
$$Al_8(OH)_{20}^{4+}$$
, $Al_{13}(OH)_{34}^{5+}$)

The released Al^{3+} compounds causes the flocculation of organic sludge particles by reducing the absolute value of zeta potential to a certain level, where the Van der Waal forces are greater than the repulsive forces between the particles with negative charges (Ni'am et al., 2007; Larue et al., 2003). In this case, the settling rate of sludge is improved. The electro-coagulation process is capable of treating wastewater with different compounds. According to Rajeshwar et al., (1994), the benefits of applying an electrochemical process to wastewater treatment include environmental compatibility, versatility, energy efficiency, safety, selectivity, amenability to automation, and cost effectiveness. Furthermore, the application of electro-coagulation needs lower requirement of retention time and no chemical coagulants (Kobya et al., 2006), reduces the salt and ion content in the sludge (Mollah et al., 2001; Chen, 2004), generates flocs with bigger size and density (Larue et al., 2003). However, its usage has been limited by the power supply and relatively higher costs in some cases (Kobya et al., 2003). Moreover, the operation requires professional qualified staff that capable of running the system (Ibeid et al., 2013). By combining electrokinetic process in the conventional MBR system, the SMEBR proposed in the current study is expected to provide improved effluent quality with higher efficiency and lower requirements (Ibeid et al., 2013).

In conclusion, by combining an electrokinetic process, biodegradation, and membrane filtration in one reactor as SMEBR, effluent quality was effectively improved under regular

operational conditions. This finding has also been proven by several previous experiments under adequate organic carbon inputs (Elektorowicz et al., 2009; Ibeid, 2011; Wei et al., 2009).

2.4 Regular design of SMEBR

Previous studies have offered several options for designing an SMEBR system. The design relates to the consideration of the reactor size, flow rate, sludge or hydraulic residence time as well as many other conditions. The major purpose of the design is to generate an electrical field, and then perform biodegradation, electro-coagulation, and membrane filtration in one reactor. As the complete mix reactor, the SMEBR design has two zones, namely, Zone I and Zone II, which are established using two electrode units (Bani-Melhem et al., 2011). Zone I is located between the anode and the cathode, and provides electro-coagulation and biodegradation, whereas Zone II is located between the cathode and the membrane module, and provides membrane filtration and biodegradation.

Meanwhile, the materials of the anode and the cathode must be carefully considered because they affect the treatment result. Aluminum and stainless steel are viable options (Ibeid et al., 2013; Ibeid, 2011). Apart from the materials, aeration is also important. In designing the system, the aeration unit should be equally placed in both Zones I and II (Bani-Melhem et al., 2011), so that the treatment is not affected by unequal distribution of the aeration.

In this study, a cylindrical reactor was used, and the SMEBR system was designed as two immersed circular perforated electrodes in accordance with a previous work (Bani-Melhem et al., 2011). The design consisted of a continuous flow laboratory size system, and the membrane unit was placed at the center of the two electrodes (Figure 1).



Figure 1: Design of SMEBR as complete mix reactor (Bani-Melhem et al., 2011)

2.5 Mechanisms of nutrient removal

2.5.1 Nitrification and denitrification

In biological treatment, the nitrogen removal is done by conducting nitrification and denitrification process (Chiu et al., 2007; Wu et al., 2007). The nitrification process transfer ammonia nitrogen, which is common source of nitrogen in wastewater, to nitrate nitrogen. The reactions have presented as follows (Mackenzie et al., 2012):

The route of nitrification is

 $\rm NH_4^+ \rightarrow \rm NO_2^- \rightarrow \rm NO_3^-$

The formula of nitrification reaction is:

$$2 \text{ NH}_4^+ + 3 \text{ O}_2 \rightarrow 2 \text{ NO}_2^- + 2 \text{ H}_2\text{O} + 4 \text{ H}^+$$

$$2 \operatorname{NO}_2^- + \operatorname{O}_2 \rightarrow 2 \operatorname{NO}_3^-$$

$$NO_2^- + H_2O \rightarrow NO_3^- + 2H^+ + 2e^-$$

The denitrification, however, transfer nitrate nitrogen to nitrogen gases.

The route of denitrification is

 $NO_3^- \rightarrow NO_2^- \rightarrow NO + N_2O \rightarrow N_2$ (gas)

The equation can be expressed as:

 $2NO_3^- + 4H^+ + 4e^- \rightarrow 2NO_2^- + 2H_2O$

$$2\mathrm{NO}_2^- + 4\mathrm{H}^+ + 2\mathrm{e}^- \rightarrow 2\mathrm{NO} + 2\mathrm{H}_2\mathrm{O}$$

 $2NO+2H^++2e^- \rightarrow N_2O+H_2O$

 $N_2O+2H^++2e^- {\rightarrow} N_2+H_2O$

So, the overall route of nitrification/denitrification process is:

$$NH_4^+ \rightarrow NO_2^- \rightarrow NO_3^- \rightarrow NO_2^- \rightarrow NO + N_2O \rightarrow N_2$$
 (gas)

2.5.2 Phosphorus removal

In SMEBR, most of phosphorus removal can be done by electrokinetic process. When the DC electrical field was applied in SMEBR, Al³⁺ (coagulation agents) was generated due to the electrooxidation of the aluminum anode. (Mollah et al., 2004; Hasan et al., 2014). The released Al³⁺ will react with phosphorus compounds, especially orthophosphorus, the common source of phosphorus.

The reactions of phosphorus removal are presented as follows (Mackenzie et al., 2012):

$$3Al^{3+} + 2PO_4^{3-} + 3H_2O \rightarrow (AlOH)_3 (PO_4)_2 + 3H^+$$
$$Al^{3+} + PO_4^{3-} \rightarrow AlPO_4$$
$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+$$

Therefore, PO_4^{3-} was either precipitated into $(AIOH)_3(PO_4)_2$ and $AIPO_4$ or adsorbed by the produced strong adsorption agent $AI(OH)_3$ (Wei et al., 2009).

Since the mechanisms of phosphorus treatment are different from nitrogen removal, many types of biological treatment plants require additional treatment for phosphorus. The treatments like sequential batch reactors have already acquired the nitrogen removal in one reactor, but no phosphorus can be removed without additional treatments (Wang et al., 2008; Udert et al., 2008; Chiu et al., 2007; Wu et al., 2007).

2.6 Benefits of using the SMEBR

The SMEBR combines membrane filtration and activated sludge with an electrokinetic process, resulting in the generation of high-quality effluent and reduction of membrane fouling (Bani-Melhem et al., 2010; Bani-Melhem et al., 2011; Ibeid et al., 2013; Hasan et al., 2012). Compared with the conventional MBR system, the SMEBR system can provide better treatment results and simple control of the membrane unit (Bani-Melhem et al., 2011; Hasan et al., 2012). Furthermore, although conventional MBR can effectively remove COD, the nutrient removal process requires extra treatments. By contrast, SMEBR can provide a wider range of treatments,

including COD removal and elimination of nutrients, such as nitrogen and phosphorus compounds (Bani-Melhem et al., 2011; Hasan et al., 2012 and 2014; Ibeid, 2011).

Moreover, compared with regular treatments for nutrient removal similar to the MBR system, the size of the SMEBR is smaller because only a single reactor is required. Thus, the SMEBR can be used in places with limited spaces. Nevertheless, the minimized procedure can help to reduce the complications involved in system manipulation and maintenance (Elektorowicz et al., 2009).

Further, in improving membrane module, according to the previous studies, the SMEBR can reduce fouling rate by up to 16.3% without backwashing the membrane module. This capability improves filterability, provides more reliable effluent quality from membrane filtration, and extends the lifecycle of the membrane module (Bani-Melhem et al., 2010). The use of SMEBR also reduces transmembrane pressure (TMP) by as much as 5.8 times (Ibeid et al., 2013). Moreover, in the research done by Hasan, et al. (2012; Hasan, 2011), the result of TMP reduction was improved as much as 8 times in SMEBR application. By conducting SMEBR, the removal of soluble microbial products (SMP), which is the organic material that responsible for membrane fouling, can reach 80% (Ibeid et al., 2010; Ibeid, 2011).

2.7 Impact of low C/N ratio in wastewater treatment

Organic carbon input is the necessary food for microorganisms in the sludge (Zhang et al., 2014), and low food supply decreases the survival and reproduction rate of these microorganisms. The reduced amount of microorganisms leads to a lower rate of nitrogen

removal (Hu et al., 2014). The treatment result of the regular biological processe is affected by low C/N ratio, and that is why there is a need to introduce system improvements.

The condition of low C/N ratio is always found in residential wastewater (Hu et al., 2014), because of the low amount of carbon sources coming from residential activities. In comparison, other nutrients from residential activities, such as nitrogen and phosphorus compounds, are relatively higher.

When biological treatments (e.g., activated sludge processes) are no longer effective, one of the possible solutions is to add a carbon source, such as glucose. Although this can help to solve the problem, the extra chemical input is not cost effective and environmentally friendly. Hence, the current study hopes that the SMEBR can provide a better solution under this condition without additional chemical inputs (Ibeid, 2011).

As mentioned earlier, the current study focuses on the treatment with low organic carbon input and constant nutrient input condition (low C/N). The experimental equipment set up and operational conditions are mostly same as in previous experiment (Hasan et al., 2014) except C/N ratio.

Chapter III

Experimental work

The study was conducted for approximately 6 months, including the preparation and experiment periods. The study was conducted in three stages. A different C/N ratio was applied in each stage. The other characteristics of the synthetic wastewater were kept similar. The nutrient removal rate in each stage was assessed in relation to the C/N ratio.

The measurements in the daily observations included pH, ammonia nitrogen, nitrate nitrogen, dissolved oxygen, oxidation reduction potential (ORP), electrical conductivity, and current input. Total nitrogen measurement was performed every 3 or 4 days, and weekly measurements included those for COD and reactive phosphorus. Several suspended solid tests were implemented during the experiment to maintain the balance of the initial sludge and observe the changes within MLSS.

The main purpose of this study was to determine the efficiency of nutrient (nitrogen and phosphorous) removal under different C/N ratios. Nitrogen and phosphorus were added in synthetic wastewater under the forms of ammonia sulfate and potassium phosphate compounds, respectively. The amount of input compounds remained the same throughout the experiment. Hence, the comparison was accomplished. Nitrogen removal was particularly tested as ammonia-nitrogen (NH₃⁻), nitrate-nitrogen (NO₃⁻), total Kjehldahl nitrogen (TKN), and total nitrogen (TN). Meanwhile, phosphorous removal was analyzed as orthophosphorus (PO₄³⁻).

3.1 Synthetic wastewater preparation

3.1.1 Introduction

Laboratory-scale treatments designed for synthetic wastewater treatment were applied. The sources of nutrient utilized to prepare synthetic wastewater are shown in Appendix A1. Considering that the focus of the study is on nutrient removal in various C/N conditions, synthetic wastewater with an adequate C/N ratio was prepared every 2 to 4 days. To keep the characteristics of synthetic wastewater unchangeable, a quality check was conducted during the daily observations. The containers were cleaned after the previous synthetic wastewater when it was exhausted.

3.1.2 Source of nutrients

Two major nutrient components have been utilized in most studies on wastewater treatment. These two components are nitrogen and phosphorus. In this study, two types of sources were added into synthetic wastewater to function as the abovementioned two components.

3.1.2.1 Source of nitrogen

Ammonia nitrogen is a common nitrogen source that exists in wastewater, and its abundance has made it the major source of nitrogen. In this study, the source of nitrogen input was ammonia sulfate, which is a form of ammonia nitrogen. The major form of nitrogen compounds in the influent and reactor was ammonia nitrogen as well. Direct observation of ammonia nitrogen measurement in both influent and effluent shows the level of nitrification in the SMEBR and comparative MBR systems.

Thirty grams of ammonium sulfate was dissolved in 160 L of water in the preparation of synthetic wastewater. Thus, a total nitrogen concentration of 45 mg/L was maintained as the influent nitrogen concentration. The calculation was similar to the calculation of the C/N ratio in Section 3.2.1.

3.1.2.2 Source of phosphorus

In each stage, 6.16 g of potassium phosphate (KH₂PO₄) was added into 160 L of synthetic wastewater to maintain a concentration of approximately 20 mg/L PO_4^{3-} . The average orthophosphorus concentration in the synthetic wastewater was approximately 17 mg/L PO_4^{3-} to 21 mg/L PO_4^{3-} (Tab. 1).

	Minimum	Maximum	Average
Stage	concentration	concentration	concentration
	mg/L PO4 ³⁻	mg/L PO4 ³⁻	mg/L PO4 ³⁻
1	19.40	23.00	21.20
2	15.75	18.90	17.10
3	17.80	20.10	19.11

Table 1: Measured orthophosphorus input in each stage

The input was initially assumed to be 20 mg/L PO_4^{3-} on the average. The variation in phosphorous in the influent could be due to the incomplete dissolution of phosphorus sources in water during the preparation of synthetic wastewater. However, the differences were not large; thus, their impact is insignificant.

3.1.3 Source of organic carbon

At the beginning of the experiment (stage one), the researchers considered different sources of carbon (sodium acetate, peptone, and acetic acid), However, sodium acetate contains sodium, which releases metallic ions in the reactor and affects the electrokinetic process; acetic acid and peptone can affect the operational conditions, such as pH value, in the reactor. Subsequently, their usage would become limited. For simplicity, low potential impact, and cost effectiveness, glucose was selected as a carbon source.

3.2 Experimental setup

SMEBR method is an innovative design for wastewater treatment; it involves the control of biological processes through electrokinetics combined with membrane filtration. The experiment was conducted in two reactors: a target SMEBR (submerged membrane electrobioreactor) and a conventional submerged membrane bioreactor (MBR) as a comparative reactor. Both reactors had an effective volume of approximately 14 L, which is related to the control of hydraulic and solid residence time (HRT and SRT, respectively). Both reactors employed the same membrane module at the center. The membrane (Microza), which was

approximately 30 cm long, was designed by Asahi Kasei Company (Japan) for small-scale experimental use. The pore size of the membrane was $0.01 \ \mu m$ to allow for microfiltration.

The same initial conditions, including the quality and amount of wastewater input (nutrient input, carbon input and flow rate) and sludge (TSS, pH, volume), were applied in both reactors. As required for conventional MBRs, a sufficient amount of dissolved oxygen (DO) was maintained in the MBR to provide the best condition for the growth of aerobic microorganisms in the activated sludge. Different amounts of DO were supplied to SMEBR because this system contains different types of microorganisms (aerobic, anoxic, etc.). In such a case, aeration was adjusted to achieve ideal conditions for aerobic, nitrifying, and denitrifying bacteria in the same vessel. The decision on such adjustment depended on the previous measurements in the same stage, and the expected results in the subsequent measurements. The results from SMEBR were compared with the results from MBR to assess the degree of treatment improvement.

3.2.1 Calculation of C/N and COD/N ratio

3.2.1.1 C/N ratio

Given that the only changed initial operational condition in this study is the C/N ratio, the calculation and control of this ratio were prioritized in each stage. The nitrogen input from synthetic wastewater, which was 187.5mg of ammonium sulfate/L, was kept constant. The carbon input from synthetic wastewater was reduced in each stage as shown in Appendix A1, and the theoretical glucose concentration was calculated as shown in Table 2.
Stage	Glucose concentration g/L	Glucose deduction at each stage input
1	0.4	initial
2	0.26	35% of initial
3	0.12	70% of initial

Table 2: Theoretical glucose input concentration in each stage and the percentage

The C/N ratio in each stage was presented in table 3. The calculated C/N ratio is the average value, with a slight variation in carbon and nitrogen input.

Stage	Glucose g/180 L	Glucose g/L	Carbon g/L	Nitrogen	C/N
	water			g - N / L	ratio
1	64	0.36	0.14	0.045	3:1
2	41.6	0.23	0.09	0.045	2:1
3	19.2	0.11	0.04	0.045	1:1

Table 3: Experimental glucose and nitrogen input concentrations and C/N ratio

3.2.1.2 COD/N ratio

The COD/N ratio, which is shown in Table 4, was calculated with the value of the average measured COD and measured total nitrogen in synthetic wastewater.

Table 4. Concentration of average COD, total mitrogen and COD/IN ratio
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Stage	Average COD mg/L	Average Total Nitrogen mg/L	COD/N ratio
1	426.5	45	9:1
2	248.6	45	6:1
3	121.2	45	3:1

Table 4 shows that the COD/N ratio in each stage follows the trend of 9:6:3, which is similar to the trend of 3:2:1 exhibited by the C/N ratio. Such similarity verifies the accuracy of C/N ratio calculation.

3.2.2 Activated sludge

At the beginning of the study, 9 L of activated sludge from a wastewater treatment plant in St Hyacinthe, QC, was prepared. The concentration of MLSS in the original sludge was approximately 7000 mg/L to 8000 mg/L. The sludge was diluted in the first stage of the experiment to obtain 3000 mg MLSS/L to 4000 mg MLSS/L. In the second and third stages, the sludge was diluted more because the sludge from MBR in the previous stage was used as new sludge. With the same sludge concentration in both reactors, a comparison could still be performed in each individual stage. Before starting the experiment, two days of aeration was maintained in both reactors until the sludge maintained its brown color.

3.2.3 Synthetic wastewater

Considering that the aim of this experiment is to examine nutrient removal under a low organic carbon input, the synthetic wastewater that was applied as wastewater influent included both the carbon and nutrient sources. Two types of nutrients, namely, nitrogen and phosphorus, were regarded as the major nutrient components. In the experiment, specific amounts of nitrogen and phosphorus sources were prepared as presented in the discussion of materials in Section 3.1.2. These amounts were kept constant in all three stages of the experiment. However, the amount of the carbon source input was changed in each stage. The input amount during each stage was constant (Section 3.1.3).

3.2.4 Comparative reactor (MBR)

The capacity of the container in MBR was 20 L, and 14 L of sludge was reserved in the reactor to prevent or minimize spilling when the pumping system is not working properly. The air diffusers were placed at the bottom of the reactor to supply aeration. Three air diffusers were utilized: two pore stone air diffusers and a chain-formed diffuser. The ultrafiltration membrane module was placed at the center of the reactor. The design of MBR has presented in Figure 3.

3.2.5 Design of the SMEBR system

The design of the SMEBR system is similar to previous designs (Bani Melhem et al., 2011; Ibeid, 2011; Hasan, 2011). The system includes two cylindrical perforated electrodes (aluminum anode and stainless steel cathode) and a DC power supply (controlled by a timer) that provides intermittent current. The same type of membrane module for MBR was placed at the center of the reactor to provide filtration and pump out the effluent.

The SMEBR system functioned as a complete mix reactor with submerged electrodes and membranes. After adequate residence time, the treated water was continuously filtered out through the membrane. The measurements of remaining nutrient in effluent provided direct evidence of treatment improvement by SMEBR application.



Figure 2: The design of SMEBR

Electrodes

As mentioned in the previously, the SMEBR's electrical system included an anode (perforated aluminum sheet) and a cathode (stainless steel mesh). They are the center cylindrical units shown in Figures 2 and 3. Two modules were combined by wood stick and plastic ties, which was insulation materials, so that interruption can be minimized during the experiment.

The selection of the anode's material is important because the electrokinetic process would release aluminum materials from the anode to the sludge; the released material would then be involved in the biological reaction to improve the treatment result, especially for phosphorus removal. The stainless steel, however, would be unaffected. Moreover, to allow the sludge and synthetic wastewater to flow through the electrical field, the anode unit was not only larger than the cathode unit so that at least 5 cm distance can be maintained, but also contained holes similar to those of a net. The diameter of the anode unit was approximately 20 cm, and the submerged area was 24 to 25 cm high when the sludge was 14 L. Assuming that 40% of the plate has holes, the total submerged area is 60% of the total area. Information on the design details is presented in Table 5.

Volume of	Height of anode	Diameter of the	Percentage of	Effective area of
SMEBR	(submerged in	anode (cm)	openings	anode (cm ²)

sludge) (cm)

(liter)

Table 5: Desig	n of the	SMEBR	(anode)
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14	24 to 25	20	40%	900 to 950

3.2.6 Overview of the experimental system

The setup of the experimental system is shown in Figures 4.



Design of Experiment system

Figure 3: Setting up of the experimental system

A 3D illustration of the system is presented in figure 4.



Figure 4: Top view of the experimental system in 3D

A picture of operation units has been presented in Figure A2 (Appendix A6)

3.3 Operational conditions

Several operational conditions were applied to conduct the experiment and generate the expected results. The measurements were conducted through a rigorous protocol.

3.1.1 SRT and HRT

The sludge amount and concentration in the reactor were controlled by SRT. SRT is related to sludge properties, such as growth rate, decay rate, and type of microorganisms. In this study, SRT was set to 20 days. A large SRT provides additional time for biomass generation under a low organic carbon input. A specific amount of sludge was removed from both reactors every day to keep SRT approximately equal to 20 days. The amount should be related to the volume of the sludge on a particular day. However, to perform a reasonable comparison, when the sludge volume in one reactor exhibits even a slight difference, the amount of removed sludge does not change unless the sludge volume in the reactor changes significantly because of fouling or accidents. The sludge removed from MBR, which was mainly organic sludge, was stored and prepared for use in the next stage; however, the sludge from SMEBR was dumped because it contained many inorganic compounds.

HRT was approximately 14 h. This HRT value was set based on previous experiments as well, where the acceptable HRT was between 6 h to 15 h. Given that the sludge had a low organic carbon input, a long HRT would generate positive electrokinetic effects and improve the treatment (Hasan et al., 2014).

3.1.2 Daily maintenances

Except daily measurements, some maintenance of the experimental system has to be conducted in order to keep the treatment system running appropriately.

3.1.2.1 Current control in SMEBR

The current and voltage in the SMEBR system should be carefully maintained with an electrical timer and a DC power supply. A change in current would not be allowed unless adjustment is conducted for experimental purposes. In this study, the current automatically

switched in the electrokinetic process to maintain the level of current support. The voltage was adjusted through the DC power supply.

The optimal electrical exposure mode applied in this study was 20 min off and 5 min on, similar to previous experiments (Ibeid et al., 2013; Ibeid, 2011). An electrical timer was utilized to automatically switch the current on and off.

3.1.2.2 Membrane cleaning

To minimize the effect of fouling, the membrane in both reactors was cleaned daily (except in the fouling tests). During cleaning, the membrane module was removed from the reactor and washed with tap water for a few minutes to remove the sludge cake particles attached to the membrane surface (Bani Melhem et al., 2011). In cases where the membrane's suction capacity decreased obviously, diluted bleach solution was utilized to remove small organic particles in the pores of the membrane module.

3.1.3 Designed operational conditions

Information on the operational conditions is presented in Table 6.

Operational conditions	SMEBR	MBR
Initial sludge concentration mg/L	3000 - 4000 mg/L	3000 - 4000 mg/L
Sludge volume (suggested) L	14	14
Daily take-out sludge / 14 L sludge	700 mL	700 mL
Aeration supply (dissolved oxygen mg/L) ①	0 to 4 mg/L	5 to 9 mg/L
Hydraulic residence time (HRT)	14 hours	14 hours
Sludge residence time (SRT)	15-20 days	15-20 days
Current supply (5 min on, 20 min off)	1 – 1.1 A	-

Table 6: Proposed operational conditions

① The aeration supply would be adjusted according to the measured results.

3.4 Measurements

3.4.1 Measurement options and methods

Two types of measurements, namely, daily and non-frequent, were conducted. Daily measurements were conducted to measure the reactor's state and check the operational conditions that could be easily changed by the reaction environment. The measurements included those for ammonia nitrogen, nitrite nitrogen, DO, pH, ORP, and electrical conductivity. Non-frequent measurements were also conducted to measure the treatment results and operational conditions but not daily because the results are relatively stable. Non-frequent

measurements were conducted every 3 to 7 days and depended on the measurement objective. Non-frequent measurements included those for orthophosphorus, COD, total nitrogen, and total suspended solids.

Two measurement methods, namely, TNT Hach and electrode test methods, were utilized. The TNT Hach test involves measuring the collected sample by chemical kits to obtain an accurate result; however, the test is relatively expensive. Meanwhile, the electrode test requires calibration to adjust the test's accuracy but is suitable for multiple tests as well as costless after purchasing the electrode. The measurements conducted with TNT Hach tests included those for ammonia nitrogen, nitrate nitrogen, orthophosphorus, total nitrogen, TKN, and COD. The measurements conducted with the electrode test included those for DO, ORP, electrical conductivity, pH, ammonia nitrogen (stages 2 and 3), and nitrate nitrogen (stages 2 and 3).

3.4.2 pH

pH measurement was implemented to control one of the operational conditions. The measurement was performed at the beginning of each daily observation through electrode tests (by pH probe, product of Denver industrial). A sample of approximately 40 mL obtained from each reactor's effluent was subjected to pH measurement; approximately 10 min was spent waiting for a stable result. At least three reading were conducted in each measurement to guarantee the accuracy.

Without drastic changes in the operational conditions, the changes in pH are smooth and gradual. A large change in pH measurement indicates an operation problem. Additional measurements and adjustments should then be conducted to solve the problem.

3.4.3 Chemical oxygen demand (COD)

COD is a common indicator of the presence of organic compounds in a water sample, and its removal is one of the major considerations in wastewater treatment. In this study, COD removal was measured to establish adequate control of operational conditions and prove the treatment's efficiency.

To measure COD in both effluent and synthetic wastewater, water samples were collected from the treatment effluent and synthetic wastewater container. For a reasonable comparison, the synthetic wastewater sample was obtained at the same position, which is approximately 10 cm below the water surface.

COD analysis was conducted with TNT Hach measurement (TNT 822, 20–1500 mg COD/L). Weekly measurements were conducted, and an additional measurement was conducted when COD results were needed.

3.4.4 Electrical conductivity

Electrical conductivity measurement indicates the total ionized constituents in water and is related to the amount of cations or anions (Pescod, 1992). Heavy metal is one of the major compounds that provide high electrical conductivity in wastewater. Heavy metal is highly toxic,

particularly to microorganisms in sludge. Its control and removal are related to the treatment efficiency of biological methods and thus make the measurement important in any biological process.

The daily measurements in this study included electrical conductivity tests in both SMEBR and MBR. The inflow of electrical conductivity was also measured daily as a standard. Although heavy metal was not applied in the synthetic wastewater, the synthetic wastewater itself still contained conductivity. Meanwhile, the electrokinetic process in SMEBR precipitates the compounds with electrical charges. Thus, an improvement is expected when SMEBR is utilized as an electrical conductivity reducer. Under a low carbon source input condition, it could even help improve the activity of microorganisms in the activated sludge; in turn, this phenomenon enhances treatment efficiency.

In this study, electrical conductivity measurement was conducted through an electrode test (IntelliCAL[™] CDC401 Standard Conductivity Electrode, HACH) with an HQD meter for the measurement of electrical conductivity. At least three reading were conducted to guarantee the accuracy of the measurement.

3.4.5 Dissolved oxygen (DO)

DO measurement is one of the most important factor in this experiment because it is directly related to the result of nutrient removal. Thus, in nitrogen removal, the measured DO level can be the indicator of nitrification and denitrification control condition in the reactor.

In this study, DO measurement was conducted through an electrode test (IntelliCAL[™] LDO101 Standard Luminescent Dissolved Oxygen electrode) with an HQD meter. The measurements included the DO reading for at least one complete round (5 min current on and 20 min current off). In current on phase, 1 minutes between each reading, and in current off phase, 5 minutes between each reading. The trends of DO variation were presented by the records of reading, also the maximum, minimum and average DO was defined in each day from the records.

3.4.6 ORP

ORP is another indicator of the nitrification/denitrification process and also indicates the level of DO. During the experiment, ORP measurement was conducted through an electrode test (IntelliCAL[™] MTC101 Standard Gel-Filled ORP electrode, HACH) with an HQD meter. Considering that the new electrode had been delivered in the middle of stage two and because of the malfunctioning of the old electrode applied in the first stage, valid measurement results can only be achieved for stage two and three. During the measurement, At least three reading were conducted to guarantee the accuracy of the results.

3.4.7 TSS

Similar to other studies on activated sludge (Zhang et al., 2014), a drying process was implemented in this study through an electrical heating desiccator and a furnace to measure the amount of suspended solids. Filter paper (regular and glass fiber filters for different purposes)

was utilized to separate sludge from water. As the standard procedure, the sludge was left in the desiccator at 105 °C for approximately 12 h to measure the dry weight of the sludge. To calculate the percentage of organic compounds in the sludge, a glass fiber filter was utilized instead of regular filter paper. After 12 h of heating at 105 °C, the sample was heated in a furnace at 550 °C for 1 h to remove the volatile compounds, which are mostly organic. The residue contained inorganic compounds only.

3.4.8 Nitrogen

3.4.8.1 Measurement of ammonia nitrogen

Two ammonia nitrogen measurement methods were applied in this study. In stage one and two, analysis was conducted with TNT Hach method (TNT 832 for 2–47 mg NH₃⁻-N/L). In stages two and three, the assessment of ammonia concentration was performed with an ammonia electrode (IntelliCALTM Ammonia ISE Electrode, Hach) and an HQD meter for the measurement. In each measurement, at least three reading were conducted to guarantee the accuracy of the result.

3.4.8.2 Measurement of nitrate nitrogen

Nitrate nitrogen is also a major form of nitrogen compound in the environment and biological wastewater treatment. It is a product of the nitrification process when ammonia is oxidized. In stage one and two, analysis was performed with TNT Hach methods (TNT 835 for

 $0.23-13.50 \text{ mg NO}_3$ -N/L). In stages two and three, the concentration of nitrate was evaluated with an electrode (IntelliCALTM Nitrate Ion Selective Electrode, HACK) and an HQD meter for the measurement. In each measurement, at least three reading were conducted to guarantee the accuracy of the result.

3.4.8.3 Measurement of total nitrogen

Total nitrogen (TN) was measured to provide a direct observation of the total amount of nitrogen compounds in the sample, including organic and inorganic nitrogen compounds. The test result reveals the overall nitrogen removal by comparing TN with inflow nitrogen concentration, which is the standard operational condition in this experiment.

TN analysis was conducted before operational condition adjustment, such as the application of the aeration input and new composition of the synthetic wastewater. Considering the cost and the stability of the results, daily measurement was not necessary. Thus, analyses were conducted every 3 to 5 days. The analyses were performed with TNT Hach method (TNT 826 for 1–16 mg TN/L). Dilution was necessary at times because of the limitation of the test. To maintain the high accuracy, each sample was measured at least three times, and 10 minutes between each reading. The average value from the reading was recorded as TN.

3.4.8.4 Measurement of TKN

TKN measurements were conducted to specify the amount of and relationship between organic and inorganic nitrogen compounds in total nitrogen under specific operational conditions. In experiment stage three, five TKN tests were conducted to determine the percentage removal of organic nitrogen in both reactors under a low carbon source input. Analysis was performed with TNT Hach method (TNT 880, s-TKN for 0–16 mg TKN/L). Dilution of the sample was necessary at times. Same as other measurements, the average from multiple reading was conducted to guarantee the accuracy.

3.4.9 Phosphorus

In this study, weekly analyses were conducted to measure phosphorus removal. Considering that the source of phosphorus in the synthetic wastewater was soluble compounds, the measurement focused on soluble reactive phosphorus removal (orthophosphates) only. The analyses were performed with TNT Hach method (TNT 844, Phosphorus Reactive, 1.4–15 mg PO4³⁻/L). Sometimes, sample dilution was necessary when the phosphorus concentration beyond the range of measurement. The average from multiple reading of the sample was recorded as the concentration of orthophosphorus.

3.5 Analyses

3.5.1 Current density

Control of the current input is necessary in the SMEBR system. As indicated in a previous experiment, the changes in current input are directly related to the treatment results (Hasan et

al., 2014). In the current experiment, the level of current supply was controlled by directly adjusting the voltage.

As expected, an electrical field is generated between the anode and cathode according to previous experiments. The electrical field generated by the DC current input transfers the small particles to large flocs, provides a large area for sorption, provides metals from the reaction on anode to react with phosphorus, oxidizes organic compounds and makes them bioavailable, facilitates the removal of sludge stuck to the membrane, reduces fouling, controls the settling of sludge particles on the anode and cathode, changes the sludge properties and controls the sludge pH, controls the form of the flocs and the viscosity of the sludge, and changes the zeta potential in the reactor (Hasan et al., 2014).

The calculation of anode information was based on observation and the information provided by the anode material, and also the direct measurement. Given that the shape of the hand-made anode module is not a perfect circle, the assumption of its diameter was established by calculating the average of the longest and shortest diameter of the anode section area.

A minimum of 5cm distance was maintained between the anode and cathode to provide sufficient space for the electrokinetic process. With different current supply values, current density was directly changed (Table 7).

Current (A)	Current density A/cm ²	Current density A/m ²
0.5	0.000530517	5.31
0.6	0.00063662	6.37
0.7	0.000742724	7.43
0.8	0.000848827	8.49
0.85	0.000901879	9.02
0.9	0.00095493	9.55
0.95	0.001007982	10.08
1	0.001061034	10.61
1.1	0.001167137	11.67
1.2	0.001273241	12.73
1.3	0.001379344	13.79
1.4	0.001485447	14.85
1.5	0.001591551	15.92

Table 7: Current density in each level of current input

The current density depends on both the design of the reactor, particularly the anode, and the current input from the power supply. In this experiment, the optimal current input was 1 A, which provides an optimal current density of 10.61 A/m^2 . However, the adjustment of voltage was limited during the experiment. Extremely high voltage and current density would kill the microorganisms in the sludge and affect the treatment result (Wei et al., 2011). Meanwhile, the

reactor conditions were changed at times, such as the volume of the sludge (because of fouling or accidents). Hence, when the experiment was executed, a range of 0.5A to 1.5 A of current input was maintained.

3.5.2 DO

Most of the results were compared with the level of DO, which was one of the variable operational conditions that affect the treatment results. During the experiment, MBR was provided sufficient DO (5 mg/L to 8 mg/L) to allow the sludge to maintain the optimal reaction condition. Meanwhile, the adjustment for aeration support was conducted in SMEBR to balance the concentration between ammonia nitrogen and nitrate nitrogen. The average DO level in each experimental stage was defined by the different carbon source input. Given that more carbon sources means more food for microorganisms and more DO is required by the microorganisms to digest the food. A balanced DO level, which is when total nitrogen removal reaches the optimal point under the certain level of carbon input, was found by calculating the average of multiple tests and the optimal treatment results. Then, the optimal results from SBEBR were compared with the results from MBR to assess the degree of treatment improvement.

Instead of the slow trend of change caused directly by the adjustment of aeration supply, a small DO switch at high speed and low amount was also observed during the measurement under different current (ON/OFF) phases. Between the on and off phases, the DO in SMEBR was affected by the electrokinetic process. A trend was observed: when the current is on, DO

decreases and denitrification begins under a relatively low DO condition; when the current is off, the DO concentration is recovered and nitrification begins instead of denitrification. The time of current on and off directly controls the time of each reaction and the DO concentration. To analyze the impact, the DO measurement requires multiple readings in each phase. The result shows not only DO switching by the current but also the daily trend of DO change by the aeration supply.

3.5.3.1 Relationship with ORP

ORP can be utilized to determine the level of DO under different circumstances, such as when DO is too low to be measured, or to double check the DO measurements. Theoretically, when the current is on, DO decreases and the reactor is in the heterotrophic phase (denitrification); the carbon source is the organic compounds, such as the glucose applied in this experiment, and the ORP should be less than 50 mV and could be as low as -150 mV. When the current is off, DO concentration increases, and the reactor is in the autotrophic phase (nitrification); this condition means that the carbon source is the inorganic compounds, and ORP at this moment should be more than 100 mV (Ibeid, 2011). However, during the experiment, the accuracy of the ORP measurement was not always satisfactory because of the limitations and inaccuracy of the equipment and unexpected reaction conditions. Although the result was not very accurate, an increasing/decreasing trend of ORP was still observed, which is necessary for the treatment system's manipulation as the operational control condition of nitrogen removal.

3.5.3 TSS

As indicated by most studies on activated sludge, TSS needs to be measured to determine the concentration and type of suspended sludge. By preceding the experiment, a series of TSS measurements could present the changes in sludge quality, which in turn could be utilized to denote the reaction condition in the reactor.

During the present study, the initial concentration of suspended solids in each stage differed, because the new sludge applied at the beginning of the second and third stages was collected as "take-out" sludge from MBR in the previous stage of the experiment. However, the same concentration was prepared in both the comparative reactor and SMEBR at the beginning of each stage; at the least, a comparison can be made between the two reactors in each stage. One of the main purposes of conducting a TSS test, especially at 550 °C furnace heating, is to prove that the SMEBR system can generate additional inorganic sludge and provide better settling capability than the sludge from the regular MBR process.

3.5.4 COD

The organic carbon input is the control factor for the C/N ratio. The COD/N ratio was calculated to double check the results. In each stage, the amount of applied carbon was reduced by 30%, which is in the COD range of 500 mg/L to 100 mg/L. The amount of organic carbon input in each stage was calculated before starting the experiment; this calculated amount was used in the entire stage. Thus, COD was expected to remain similar during each stage. Weekly

COD analyses were performed for checking purposes. Meanwhile, when the state of the sludge was changed, a COD test was conducted for analysis purposes.

To achieve the aim of this study, the relationship between nutrient removal and carbon source input, which could also be explained as the relationship between total nitrogen and COD, requires an analysis that combines COD and the remaining nitrogen compounds; hence, nitrogen removal under different COD levels is presented as the proof of the reliability of the measured treatment results.

3.5.5 Nitrogen

The nitrogen present in the wastewater and effluent was analyzed as ammonia, nitrate, TN, and TKN. The samples from both reactors were then separated for use in four different tests. For the daily measurements, the changes in ammonia nitrogen and nitrate nitrogen concentration presented two comparative curves. These curves show the level of nitrification and denitrification when combined with the analysis of DO and allow for the prediction of total nitrogen.

The measured results of comparative MBR and SMEBR were compared to directly reveal the improvement in nitrogen removal. Moreover, the results were compared with the records of other operational conditions, such as DO, pH, and EC. The most influential factor was highlighted, and the controlling range to directly determine the optimal rate of nitrogen removal was recommended as the conclusion of the study.

3.5.6 Phosphorus

Previous studies (Bani Melhem et al., 2011) have proven that SEMBR has a high capacity for phosphorus removal; unlike those of nitrogen removal, the result of phosphorus removal can be stabilized for a long period of time under similar operational conditions. Thus, daily analysis was not required.

Similar to the analysis of nitrogen removal, studies on phosphorus removal were also conducted by comparing the results of MBR and combining them with the analysis of other operational conditions. However, unlike nitrogen removal, phosphorus removal is based on different mechanisms. Thus, the factors that affect nitrogen removal might not affect phosphorus removal. The analysis only presented whether phosphorus removal can reach optimal when the best nitrogen removal was acquired in SMEBR.

Chapter IV

Results and discussions

4.1 Overview

The results from the three stages were successfully recorded after six months of experiments. Each measurement was individually discussed, which included the results of current density, electrical conductivity, pH, DO, ORP, COD, nutrient removal, TSS, and membrane pressure. Meanwhile, the nutrient removal, which was the target of the experiment, was also discussed with other factors, especially carbon input and aeration supply. These two variable conditions were considered in the experiment. In conclusion, a suggested range of DO under the certain C/N ratio and current density was provided with the optimal results of nutrient removal. The result of phosphorus removal was also presented within these conditions.

4.2 Current density

As the main operational condition and major mechanism of SMEBR system, the manipulation of the current supply was simpler than the control of C/N ratio, which was only needed to adjust the current of power supply. However, maintaining stabilized current density remains the key to a successful experiment. Hence, the daily check was conducted and recorded.

Based on the previous experiment (Ibeid, 2011), the researcher maintained the current in 1 A from the power supply for the control of current input. The current was kept at 1 A most of the time in the first two stages. However, the voltage was adjusted as high as 25 V at the end of stage two to keep the level of current input. In addition, the microorganisms had been affected by the strong voltage, which reduced the nutrient removal efficiency. Thus, the voltage level had been kept as low as 15 to 20 V in stage three, instead of maintaining current support level. In this stage, the current input dropped from 0.5 to 0.85 A.

To facilitate easier calculation and to provide a general range of current density in each stage, the calculation used the average current input of each stage. The results are shown in Table 8.

Stage	Average current input A	Submerged area m ²	Current density A/m ²
1	0.94	0.09	10.45
2	1.03	0.09	11.44
3	0.68	0.09	7.58

Table 8: Average current input and current density in each stage

This study assumed that the expected current around 1 A means that the theoretical current density should in between 10 and 11 A/m^2 . Based on the calculation, the current density could be kept similar with the theoretical value in stage one and two. However, the current density had become lower in stage three, but the treatment result was still acceptable. If the current input can be adjusted higher, a better result might be observed despite the increase in voltage.

4.3 Electrical conductivity

Electrical conductivity reduced compared with the use of MBR by applying the SMEBR system. This reduction was caused by the electrokinetic process, which involves the provision of electrical field. In this method, the particles with charges combined as large flocs. Those flocs would settle at the bottom as waste sludge and will no longer affect the suspended sludge. The results of the experiment are shown in Table 9.

	Influent	MBR	SMEBR
	(µS/cm)	eff. (µS/cm)	eff. (µS/cm)
		Stage I	
Average	637	551	473
Maximum	688	642	690
Minimum	509	477	395
		Stage II	
Average	610	537	432
Maximum	665	575	518
Minimum	565	503	373
		Stage III	
Average	609	552	483
Maximum	645	585	570
Minimum	490	501	410

Table 9: Electrical conductivity in influent and effluent from both reactors, stage I, II, III

Based on the record of the measured electrical conductivity, the value was stabilized at all stages of the experiment at around 600 μ sS/cm in the synthetic wastewater, 550 μ S/cm in the MBR, and 450 μ S/cm in SMEBR. The variation between the minimum and maximum measurements denoted the changes of the reactor's condition, especially when the synthetic wastewater was exhausted and new synthetic wastewater was prepared.

The electrical conductivity had been reduced by approximately 10% by applying the MBR system, as shown in Table 10. This reduction was caused by the activated sludge and microfiltration process. However, the SMEBR system had doubled this result by conducting the electrical field. More than 20% of electrical conductivity reduction had been maintained. The difference between MBR and SMEBR had denoted the improvement under the electrokinetic processes.

	MBR	SMEBR
Stage I	13.57%	25.72%
Stage II	11.96%	29.18%
Stage III	9.31%	20.63%

Table 10: Reduction of electrical conductivity in percentage, in stage I, II and III

Electrical conductivity was mostly obtained from the chemical compounds that dissolved in the wastewater. In this case, the SMEBR still had higher removal of electrical conductive compounds than the MBR system, which had presented better electrical conductivity reduction. In addition, the possibility of reducing electrical conductivity under the higher input has been presented, such as wastewater with high concentration of heavy metal compounds.

4.4 pH

The pH measurement was another indicator of the steady operational conditions in the reactor. The result was stabilized, as shown in Table 11.

	pH in MBR eff.	pH in SMEBR eff.
Stage II		
Average	5.01	7.19
Maximum	6.47	8.85
Minimum	4.08	6.18
Stage III		
Average	5.91	7.35
Maximum	6.31	7.72
Minimum	5.24	6.88

Table 11: pH in both SMEBR's and MBR's eff., in stage I, II and III

The record in Table 11 and Figure 5 shows that the pH value in MBR controlled at around 4 to 6. The value increased from 6 to 8 in SMEBR. This difference was related to the reaction

in both reactors. The increase of pH in the SMEBR compared with that in MBR was caused by the production of hydroxide ion through the electrochemical reactions which is:

$$3H_2O + 3e^- \rightarrow 3/2 H_2(g) + 3OH^-$$

$$1/2 \text{ O}_2 + 2e^- + H_2O \rightarrow 2OH^-$$



Figure 5: pH vs DO in the sludge of SMEBR and MBR

From the observation (Figure 5) in MBR, the average pH decreased with C/N reduction. Nevertheless, under the same C/N reduction condition, the average pH in SMEBR was stabilized in both stages. It had presented that instead of the common factors like carbon input or DO, the electrokinetic processes were the major factor of pH variation in SMEBR. This result had proven that SMEBR was capable of neutralizing acidic sludge.

4.5 Dissolved oxygen

The control of dissolved oxygen is one of the most important factors of wastewater treatment because the aeration level is directly related to the nitrification/denitrification process. Hence, adequate aeration had constantly been provided in both reactors during the experiment. In SMEBR, aeration supply was carefully controlled to balance the amount of DO required for nitrification and denitrification processes, which was necessary to remove ammonia nitrogen and nitrate nitrogen. Furthermore, anaerobic ammonium oxidation (anammox) bacteria (Ibeid, 2011) that leads to the improvement of treatment results was also expected under the low dissolved oxygen. Thus, the required aeration in SMEBR was much lower than MBR.

Finding the dissolved oxygen values adequate to particular C/N ratio for nitrogen removal allows the aeration in SMEBR to be adjusted when the treatment results were below expectations. The adjustments were minimal and the response of microorganisms was slow. Hence, the observable impact from the adjustment is seen only in the subsequent days. Two trends of dissolved oxygen changes were seen in this study. One change was caused by current switching, which was changing fast and with small amount. The other change was caused by the adjustment, which was slow and smooth, but the changed amount was relatively large.



Figure 6: Average dissolved oxygen fluctuation in each stage in SMEBR

Figure 6 shows that the average dissolved oxygen in each stage was mainly maintained in three levels. In stage I, the average DO was mostly around 2 to 4 mg/L. In stage two, the level dropped to 0 to 1 mg/L. In stage three, the concentration was kept at approximately zero most of the time. However, the actual DO value between two different days when measured DO was 0 mg/L were mostly varied between deeply zero and barely zero DO condition (Section 4.8.1.6), which was caused by sludge settling under the weak or no aeration supply.

In each stage, the DO curve fluctuations were mainly caused by adjustment of the aeration supplies during the experiment. These adjustments were conducted based on the observation of the experiment, which was related to the direct observation of the reactor conditions and the records of the treatment results, especially the relationship between different DO and nitrogen removal. Finding balanced ammonia nitrogen and nitrate nitrogen removal under the certain DO level is necessary to obtain an optimal TN removal, which was the major objective of this study.

The concentration of dissolved oxygen in SMEBR slightly changed with the current input switching. The dissolved oxygen would start to decrease continuously after a few seconds when the current input was ON. The changed amount was small in this situation. When the current input was OFF, the dissolved oxygen increased after a few seconds. These changes were small and would recover when the phase switched. A typical example is shown in Figure 7, where the changes of ORP value enabled the changes of dissolved oxygen to be observed clearly. The changes of dissolved oxygen in SMEBR were constantly following these trends with the exception of a few unexpected interruptions in the process.

The dissolved oxygen can be recovered when the current switch. Because of the adjustment and environmental changes, the overall average dissolved oxygen could hardly remain at a constant level. A trend of dissolved oxygen curve that slowly increased or decreased was observed in this study. A decreasing trend of dissolved oxygen can be observed from the curve in the typical example provided in Figure 7.



Figure 7: An example of DO and ORP daily fluctuation in SMEBR

Theoretically, the average dissolved oxygen should be maintained at a certain level as stabilized condition when operating a treatment system. The control of current input and aeration is not extremely accurate in this study. The size of the container was too small to distribute the aeration equally. The unexpected changes can hardly be avoided. The accumulation of these impacts led to the slow change of dissolved oxygen. The adjustments of aeration input with experimental purposes also changed the stability of dissolved oxygen concentration in the sludge. These changes could be avoided in real operation when the dissolved oxygen input can be stabilized. The dissolved oxygen level can be maintained much easier within the bigger container, better aeration system, and more accurate current control.

4.6 Oxidation reduction potential (ORP)

The ORP measurement had been conducted in this experiment to assess the conditions of sludge. The theoretical value had been assumed based on previous studies (Ibeid, 2011). In this study, the ORP value could remain in the ON and OFF phase at a certain level, as shown in the typical example presented in Figure 7.

A clear distinction between the current ON and OFF phases on the ORP curve has been observed. The ORP in each phase was mostly stabilized in the same level. Adjustment was made once the measured ORP demonstrated a big difference with the expected value. The possible factors of changing ORP could be the changes of DO and current input. The measurement of ORP also permitted the accuracy of dissolved oxygen measurements in the reactor to be checked, because the average dissolved oxygen concentration is predictable under certain ORP. The electrode was left in the reactor with the same location and depth, which was close to DO electrode, because the location of measurement of ORP electrode was important.

The change of ORP was related to the changes of current input in the SMEBR under a similar aeration support, because the current will affect the micro-organisms and will change the dissolved oxygen uptake. ORP changed with current in several instances, and dissolved oxygen input adjustment had been recorded during the experiment. Figure 8 shows that a comparison between stages was conducted.



Figure 8: Average ORP vs average DO under different current input, Stage II & III

As result, different levels of dissolved oxygen in SMEBR facilitated ORP changes in a certain range. The average dissolved oxygen in stages two and three was as low as 0 to 1 mg/L (Figure 8). The dissolved oxygen in stage two was slightly higher, which resulted in slightly higher overall ORP than that in stage three. The maximum ORP could reach more than 200 mV in stage two when current input increased to 1.4 A, and DO was 0.23 mg/L. The minimum ORP in stage two was approximately 100 mV under the current input of more than 1 A, with DO approximately equaled to zero. The minimum ORP in stage three could reach as low as -300 mV at current input between 0.5 and 0.8 A and DO was zero. The highest ORP occurred when current input was almost 1 A, and DO reached 1.39 mg/L. The ORP variation could be caused by the different levels of dissolved oxygen; however, the current could also be a potential cause.
The dissolved oxygen was in a barely zero level once the dissolved oxygen was measured as zero in stage three and when the ORP value was in positive or close to zero. Therefore, the dissolved oxygen at the measuring position was zero. The dissolved oxygen might not be zero in some positions, such as the area close to the air diffuser. Dissolved oxygen was in deeply zero level when the ORP value was negative and as low as around -300 mV. Dissolved oxygen cannot be tested anywhere in the sludge. This phenomenon during the experiment caused the ORP measurements to be constantly conducted to find the condition of dissolved oxygen in SMEBR, especially in stage three. The discussion of barely zero and deeply zero dissolved oxygen, as well as the effects to the treatment result, would continue at the discussion of nutrient removal (Section 4.8.1.6).

4.7 COD removal

4.7.1 COD measurement

The COD measurement in this study was mostly the indicator of the amount of carbon source input. The dissolving and removal rates in the reactors were also important in studying the treatments. Three levels of COD had been defined in the entire experiment. The treatment result is shown in Table 12.

Value	COD MBR eff.	COD SMEBR eff.	COD input (mg/L					
	(mg/L COD)	(mg/L COD)	COD)					
	Stage I, C/N=3:1							
Average	19.4	22.4	426.5					
Maximum	38.9	46.0	538.0					
Minimum	4.2	2.6	356.0					
	Stage II, C/N=2:1							
Average	13.4	8.3	248.6					
Maximum	21.1	16.2	379.0					
Minimum	8.2	1.6	99.4					
Stage III, C/N=1:1								
Average	24.3	26.9	121.2					
Maximum	31.9	41.2	186.0					
Minimum	17.7	19.2	94.9					

Table 12: COD measurements in influent and effluents of both reactor, stage I, II and III.

Table 12 shows that COD removal using SMEBR can reach the same level as MBR. The variation between minimum and maximum COD input was mainly caused by the incomplete dissolution of carbon sources in synthetic wastewater and the losses from preparation process of new synthetic wastewater. The use of water sample for measurements might also be a factor. This position of taking sample constantly took place at approximately 10 cm below the surface.

However, the concentration of dissolved carbon will change at the same position with the consumption of synthetic wastewater. For instance, the measured concentrations of synthetic wastewater between the day when new synthetic wastewater was prepared to refill the container, and the day when synthetic wastewater was consumed consistently, had differences at around 100 mg/L COD. This difference may be caused by the self-quality change in synthetic wastewater and by biological reactions and water evaporation, as well as by an incomplete dissolution of glucose when preparing new wastewater. However the SMEBR system continuously positively responded to nutrient removal in spite of COD fluctuation, which is the case of real world conditions.



Figure 9: COD input (black) and removal by MBR (red) and SMEBR (blue) in each stage

Figure 9 shows the three levels of COD in the influent and effluents from both reactors. COD in SMEBR and MBR could consistently have extremely low concentration in all stages. Also, the record in table 12 shows that a lower remaining COD was obtained from effluent of both reactor in stage two. This low remaining COD in SMEBR effluent that less than 10 mg/L might denote the best range of COD input for COD removal, which was around 250 mg/L. The abrupt increase of COD from the effluent of both reactors in stage three could also prove this assumption.

4.7.2 COD removal

According to the previous experiment (Hasan et al., 2014), the SMEBR system has a high COD removal rate. This statement has been proven in this study. Both reactors had high COD removal rate in this experiment. The results of COD removal are shown in Figure 10.



Figure 10: COD removal by MBR and SMEBR, in stage I, II and III

COD removal in MBR and SMEBR could reach 90% to almost 100% in stages one and two. In stage three, the removal for both reactors was around 80%. The decrease of carbon source input influenced removal rate because COD is the limiting factor of biological process. With a higher carbon source input, the sludge could maintain higher growth of microorganism, which would consume more carbon sources. When carbon source input decreases without sufficient food supply, the slow generation and faster death rate will decrease the amount of microorganisms in the sludge. This situation would lead to the decrease of COD removal. The result shows that the insufficient carbon input has affected the COD removal in experiment stage three. Based on the comparison with the result from the previous stages, this condition was still acceptable.

4.8 Nutrient removal

4.8.1 Nitrogen removal

Nitrogen removal was investigated by conducting four different tests, namely, ammonia, nitrate, TN, and TKN tests. Daily measurements included ammonia and nitrate nitrogen, which are the two major forms of nitrogen compounds in wastewater. TN was measured in an interval of three to seven days depending on the condition of the experiment. Several TKN tests were conducted by the end of the study to determine the amount of organic nitrogen compounds in the effluent of the reactors. TKN test can identify the amount of organic nitrogen by deducting the measured ammonia nitrogen. Knowing the percentage of organic nitrogen can help provide

insight into the relationship between the input of organic/inorganic nitrogen and efficiency of nitrogen removal.

In each stage of the study, the average nitrogen removal was varied because the organic carbon input was different owing to the diverse growth and death rates of microorganisms. Ibeid (2011) proposed that nitrogen removal highly depends on the concentration of carbon in the sludge, and the system might be very sensitive on the variation of carbon source under a low C/N ratio. In his study, Ibeid (2011) analyzed the best nitrogen removal efficiency with submerged membrane electro bioreactor (SMEBR) at different C/N ratios, as well as looked into the major factors that could primarily control the removal under a particular C/N ratio.

4.8.1.1 Ammonia nitrogen

One of the major goals of wastewater treatment is to remove ammonia nitrogen, given that these waste liquids affect the environment when they oxidize in nature. The nitrification/denitrification process treats most ammonia nitrogen into nitrate nitrogen and then converts this into nitrogen gas. In this study, the researcher expected to accomplish the same normal process with only one reactor. The major nitrogen compounds in the synthetic wastewater were in the form of ammonia nitrogen because the source of input nitrogen was ammonium sulfate. The differences in the observed treatment results reflected how strong the reaction was, and how the operational conditions of the reactor changed.



Figure 11: Daily measured ammonia nitrogen in SMEBR eff. in three stages



Figure 12: Daily measured ammonia nitrogen in MBR eff. in three stages

In stage one, the average level of ammonia nitrogen, particularly in the membrane bioreactors (MBR), was lower than those in stages two and three. Moreover, the concentration value in stage two was lower than or similar to that in stage three (Figures 11 and 12). These findings can be attributed to the reduction of organic carbon input. In particular, the difference between stages two and three was less than the difference in stage one because of the low organic carbon input.

Figures 11 and 12 show the fluctuations of the concentration on a daily basis. These changes indicate the operational condition changes, some of which were due to accidents such as sludge volume changes or synthetic wastewater exhaustion. Nevertheless, most of the changes in the reaction condition were caused by the attempted adjustments on the condition with experimental purposes, such as the adjustment of current input and aeration supply. The majority of the adjustments were conducted in SMEBR; thus, more changes were observed in the SMEBR-related curve.

Figures 13 to 15 present a comparative presentation of ammonia nitrogen removal in both reactors at each stage.



Figure 13: Ammonia nitrogen removal in stage I



Figure 14: Ammonia nitrogen removal in Stage II



Figure 15: Ammonia nitrogen removal in Stage II

The percentage of ammonia nitrogen removal in Figures 13 to 15 was calculated with the formula specified in Appendix A2. In the calculations, expect for stage three, the inflow concentration was the average value from the measurements. This study assessed the inflow concentration in stage three on a daily basis by conducting electrode tests. Accordingly, the ammonia nitrogen removal was directly determined by using the exactly measured inflow concentration.

The results in stage three were better compared with the results in stages two and one. In particular, the removal rate in stage three attained as high as 80%, and the SMEBR consistently maintained a higher removal rate than MBR. In stage one, the DO was frequently changing. Similar results were observed in the first few days of stage two. The removal rate in stages one and two was probably reduced by the unsteady aeration supply. When the operational conditions, such as sludge condition and synthetic wastewater input were stabilized (at the last 10 days of stage one or after Day 16 of stage two), the result improved, and all operational conditions attained a high removal rate ranging from 60% to 80%. In the paper, the effect of DO is also conferred in the discussion about TN removal under the certain C/N ratio and current density.

4.8.1.2 Nitrate nitrogen

Nitrate nitrogen is another major form of nitrogen compound in wastewater, which is the final product of ammonia nitrification process. In this study, the source of nitrogen in synthetic wastewater was ammonia sulfate. Therefore, the amount of measured nitrate nitrogen in the effluent was mostly derived from the nitrification process in the reactor.

Nitrate nitrogen removal is also remarkably important because of its conversion to nitrogen gas. In the regular treatment of nitrogen removal, anoxic digester is added to begin denitrification after the aerobic digestion process. However, in SMEBR, both nitrification and denitrification are generated in the same reactor. Thus, identifying the balance between nitrification and denitrification is necessary for the success of a treatment. Figures 16 and 17 display the measured results of nitrate nitrogen in both SMEBR and MBR.



Figure 16: Daily measured nitrate nitrogen in MBR in all three stages



Figure 17: Daily measured nitrate nitrogen in SMEBR in all three stages

Similar to ammonia nitrogen, concentration curves indicated fluctuations of nitrate nitrogen from both reactors, denoting the changes of operational conditions. The concentration value of nitrate nitrogen in SMEBR at stage one was the lowest, whereas the third stage had the highest value. This phenomenon presented the relationship between nitrification and denitrification processes in each stage. When DO decreased, the reduced nitrification process

directly led to the increase of remaining ammonia nitrogen in effluent. Both less nitrate nitrogen generation from reduced nitrification and more nitrate nitrogen consumption from the stronger denitrification process decreased nitrate nitrogen at the same time, whereas the other conditions remained constant. By contrast, under the higher DO, the increased nitrate nitrogen exhibited a stronger nitrification process, and the amount of ammonia nitrogen was decreased. Unfortunately, , when one form of nitrogen compounds reached optimal removal, the TN removal didn't reach optimal, which means too strong or too weak nitrification and denitrification processes were not welcomed.. The studies on the combination between the concentration of ammonia nitrogen and nitrate nitrogen could help determine the optimal treatment condition. Instead of providing operational conditions for optimal removal of only ammonia or nitrate nitrogen, maintaining a balanced removal between ammonia nitrogen and nitrate nitrogen was the most effective means to control TN removal.

Figures 18 to 20 illustrate the comparison of remaining nitrate nitrogen in the effluent between MBR and SMEBR in each stage.



Figure 18: Remaining nitrate nitrogen in both reactors' effluent, stage I



Figure 19: Remaining nitrate nitrogen in both reactors' effluent, stage II



Figure 20: Remaining nitrate nitrogen in both reactors' effluent, stage III

Based on the above analyses, the overall nitrate nitrogen generation in MBR was consistently higher than that in SMEBR. Therefore, the sludge under a higher DO level in the MBR system had stronger nitrification process, and most of the nitrate nitrogen generated from nitrification in MBR flowed out with effluent without undergoing the denitrification process. SMEBR had higher ammonia nitrogen removal and lower remaining nitrate nitrogen. Therefore, the deducted amount of nitrogen was the removed extra nitrogen. This instance is presented in the discussion of TN removal.

As shown in Figures 18 to 20, the nitrate nitrogen in SMEBR at stage one generally remained at a very low level. A stronger nitrification process was expected considering that the first stage in SMEBR had higher DO input. However, the findings revealed less final products of the nitrification process, particularly in the first half of the stage. Meanwhile, the transfer of nitrogen from NH₃ -N to NO₃ -N was not effectively processed given the lower efficiency of nitrogen removal by SMEBR and the presence of relatively higher ammonia nitrogen in the effluent. The reason for this occurrence could be the ineffective control of operational condition and sludge quality. At the beginning of stage one, the SMEBR system was frequently adjusted because the researcher was looking for the best carbon sources and was trying to reduce fouling problem. Thus, the reaction condition was not optimal, which affected the sludge quality. In this event, the electrical system was shut down between Days 13 and 20 of stage one to recover the sludge quality in SMEBR. Nevertheless, the result was not good because the reactor was same as in MBR. Thus, on Day 25, 2 L of activated sludge was added into both reactors to increase the amount of microorganisms. By providing the same aeration and nutrient source input, nitrate nitrogen was increased in the subsequent half of the stage. Based on the observations, the nitrogen compounds experienced a strong nitrification/denitrification process, and low nitrate nitrogen concentration was illustrated in the second half of the test. After the sludge conditions were stabilized, SMEBR exhibited a stronger denitrification under this particular level of carbon source and DO input.

The situation improved and became predictable in stages two and three, in which the DO constantly remained between 0 and 1 mg/L. Subsequently, the stronger denitrification process reduced the amount of nitrate nitrogen than with the conventional MBR. However, because of the lower carbon source input, less food supplies reduced the activity of microorganisms, leaving relatively more nitrate nitrogen in the effluent.

4.8.1.3 Ammonia nitrogen versus nitrate nitrogen in SMEBR

The concentration changes in ammonia nitrogen and nitrate nitrogen directly indicate whether the sludge is under nitrification or denitrification process. SMEBR is only a system, in which both processes occur in the same reactor. Similar to a process that occurs in a common system, an increase of nitrate nitrogen and decrease of ammonia nitrogen can directly show the level of nitrification in the reactor under oxidation conditions. Moreover, a reduction of nitrate nitrogen with the decrease of ammonia nitrogen can show the level of denitrification in anoxic conditions. In this study, the DO concentrations were monitored in relation to nitrogen removal. The efficiency of denitrification was related to the carbon concentration in wastewater. Hence, this research assessed the ability of nitrogen removal by SMEBR in various C/N ratios.

4.8.1.3.1 Stage I



Figure 21: Ammonia nitrogen vs nitrate nitrogen under average DO, Stage I, SMEBR

In stage one, the level of remaining ammonia nitrogen (NH₃⁻-N) in effluent was largely maintained between 15 and 25 mg/L NH₃⁻-N; 50% of ammonia nitrogen was removed compared with the average influent concentration (45 mg/L NH₃⁻-N). Meanwhile, nitrate nitrogen was kept between 0 and 5 mg/L NO₃⁻-N, which was generated by nitrification process. Two clear trends of generating nitrate nitrogen were observed between Days 32 and 57 when the reactor attained a steady state condition. Simultaneously, the amount of ammonia nitrogen was decreased with the relatively high DO input. Under the conditions provided in this period, the nitrification process was strongest. The results between Days 45 and 50 also revealed a decrease of both nitrate and ammonia, implying that the reactor experienced a good nitrification/denitrification process. This particular observation once again verified the best range of DO concentrations.

The observation of the DO curve presented in Figure 21 demonstrates that the DO was maintained in a very low level between 2 and 4 mg/L. However, some levels of the DO at 4 or 5 mg/L affected the reactor. Overall, within a relatively higher organic carbon input and adequate DO (i.e., C/N = 3:1, DO = 2 to 4 mg/L), the concentration of both ammonia nitrogen and nitrate nitrogen in this stage could maintain a constant level when the system was more tolerable at sudden environmental changes of the reactor.



4.8.1.3.2 Stage II

Figure 22: Ammonia nitrogen vs nitrate nitrogen under average DO, SMEBR, Stage II

In the first 30 days of stage two, ammonia nitrogen (NH_3^--N) decreased, whereas nitrate nitrogen (NO_3^--N) increased. This trend indicates that the denitrification process did not sufficiently convert nitrate to gas nitrogen. A relationship between the concentration of DO and level of nitrification process was observed on Day 21. During this interval, ammonia nitrogen rapidly decreased, whereas nitrate nitrogen increased because of the sudden increase of the DO.

Another good example of the trend was observed between Days 28 and 31. During this period, from the measurements of effluent quality, the concentration of nitrate nitrogen reached highest of this stage, and the Ammonia nitrogen concentration reached minimum, when the DO rapidly increased on Day 28. In this case, the nitrification reaction was severe, and removing TN would not be good without converting nitrate nitrogen into nitrogen gases. The result indicated in Figure 22 suggests that the improved nitrogen removal was less than 20%, proving the assumption of this study. Accordingly, the DO should be reduced to balance the nitrification/denitrification process: with less carbon input, less DO would be consumed by biological processes. Compared with stage one, maintaining a lower and stabilized oxygen input in stage two with less organic carbon input (C/N = 2:1, DO = 0 to 1 mg/L) becomes more important, and the system is more sensitive on the environmental changes of the reactor.

4.8.1.3.3 Stage III



Figure 23: Ammonia nitrogen vs nitrate nitrogen under average DO, SMEBR, Stage III

The curve of effluent concentrations of ammonia and nitrate nitrogen in stage three were smooth and more stable than those in stage two. In this event, the DO was generally maintained close to zero. The overall trend of ammonia and nitrate nitrogen was decreasing at the same time, thus indicating that the system underwent a good nitrification/denitrification process.

However, the data presented in Figure 23 express that, some changes still occurred, although DO constantly remained zero. These changes were due to the operational conditions as deeply and barely zero DO.

On Day 22, nitrate nitrogen under the zero DO condition increased, without any adjustment, when ammonia nitrogen was still decreasing. This result indicates that the sludge was under stronger nitrification, which had a better capacity to remove nitrate nitrogen than the denitrification process. The sludge between Days 22 and 26 (Days with increasing nitrate nitrogen and decreasing ammonia nitrogen) was supposed to be under the deeply zero DO condition.

On Day 26, the concentration of nitrate nitrogen was 13 mg/L NO₃^{--N}, which was numerically similar to the concentration of ammonia nitrogen at 13.7 mg/L NH₃^{--N}. This finding reveals that the reactor underwent an extremely strong nitrification, which affected the TN removal. After Day 26, the DO was increased to 0.14 mg/L without any adjustment in previous lab work. This particular circumstance implies that, between those days, the DO in the sludge exhibited an increasing trend and induced changes in the concentration of the compounds. After Day 27, the DO was adjusted. Subsequently, ammonia nitrogen was increased evidently, whereas nitrate nitrogen was decreased. Such event led to an ineffective nitrogen removal.

At the end of this stage, the aeration input was increased to identify the maximum allowable DO. At this point, the high concentration of remaining nitrate nitrogen was observed in the effluent for several days, and the influence of the increased DO was much higher than that on Day 27. The reactor was under the strong nitrification completely, and without denitrification, nitrate nitrogen was not released as nitrogen gases, but flowed out with the treated effluent.

When the sludge in SMEBR was under low organic carbon input (i.e., C/N = 1:1), the system was more sensitive to variation of the aeration supply (DO close to 0 mg/L). The observation result of all three stages reveals that, under the low organic carbon source input condition, the sensitivity of sludge on the DO increased with the decreasing carbon input. A good treatment result may be obtained by carefully controlling the DO in the sludge with low C/N ratio than running the reactor with wastewater containing relatively higher C/N ratio.

4.8.1.3.4 Sensitivity of DO variation in the sludge of SMEBR

A simple calculation was conducted to present the sensitivity of DO in number, which is:

Sensitivity of dissolved oxygen variation

= (present concentration – previous concentration) (present dissolved oxygen – previous dissolved oxygen)

By taking results from daily measurements, the sensitivity of DO in each stage are shown in Table 13, and these results provide a better understanding of the increasing sensitivity of the DO to C/N deduction.

Stage I, C/N = 3:1							
	NH3 ⁻ -N changing rate/DO changing	NO3 ⁻ -N changing rate/DO changing					
	rate	rate					
Max	16.39	11.73					
Min	0.00	0.00					
Ave	2.39	1.19					
Stage II, C/N = 2:1							
Max	780.00	413.00					
Min	0.00	0.00					
Ave	42.77	25.23					
Stage III, C/N = 1:1							
Max	2969.30	4720.00					
Min	0.09	0.07					
Ave	290.74	259.61					

Table 13: Results of sensitivity on DO variation in SMEBR's sludge, stage I, II and III

The results presented in Table 13 indicate that the sensitivity number was increased with C/N reduction, in which the larger number denotes the faster changes of nitrogen (ammonia or nitrate) with smaller variation of DO. The results were dramatically increased from maximum 10 to 4000 when the C/N ratio was suddenly reduced from 3:1 to 1:1. This finding directly proves the relationship between organic carbon input and sludge sensitivity.

4.8.1.4 TN

The TN in the effluent was measured as a means to monitor the removal of all forms of nitrogen removal. In this study, a group of two samples from both reactors' effluent was

measured in each measurement, and the difference between the two results was considered the improvement of nitrogen removal.

The measurement results illustrate that the TN concentration in influent varied from 43 mg-N/L to 46 mg-N/L. However, the theoretical input was assumed 40 mg TN/L as shown by the previous calculation (Section 3.2.1). The higher concentration of TN was probably due to the residuals from previous synthetic wastewater or the incomplete dissolution of nutrient sources. A difference emerged between the theoretical and measured values. In the calculations, 44.5 mg/L TN was used as the TN input to prevent inaccuracy from the measurement and induce an easier computation. Thus, the TN removal was calculated by simply deducting the measured effluent TN from the average TN in the influent; the results are displayed in Figures 24 to 26.



Figure 24: Removed TN in SMEBR & MBR, stage I



Figure 25: Removed TN in in SMEBR & MBR, stage II



Figure 26: Removed TN in in SMEBR & MBR, stage III

The results of the calculation demonstrate that the TN removal with SMEBR was always better than that with the conventional MBR comparative system. In the experiment, the change of C/N ratio or organic carbon input influenced the TN removal by SMEBR and was evidently reduced. Nevertheless, the MBR system was more affected by the carbon source reduction, and its TN removal rapidly decreased with the devaluation of C/N ratio. Meanwhile, the differences between the TN removal in SMEBR between stage I and II or stage II and III were around 5 mg-N/L (average 20, 15, and 10 mg/L TN removal in stage I, II, and III, respectively), depicting that a similar removal efficiency was achieved. Thus, this finding strongly verifies that SMEBR can significantly tolerate low organic carbon supplies and can provide a better treatment result under such condition.

Table 14 specifies that the maximum TN removal in each stage was presented with the measured average DO; yet, the maximum removal in stage two was the highest. This particular observation contradicts the expected condition, that is, the highest removal will occur with a higher level of carbon input. The highest TN removal in stage one was slightly lower than that in stage two; yet, the average TN removal in the first stage was approximately 5 mg/L higher than that in the second stage (Figures 24 to 25).

Stage	Maximum TN	Maximum TN	DO with max TN	
	removal	removal	removal	
	(mg/L N)	(%)	(mg/L DO)	
I	21.5	48.31%	3.01	
II	24.3	54.61%	0.11	
III	13.4	30.11%	0.03	

Table 14: Maximum TN removal by SMEBR in each stage.

In the third stage of the conventional MBR, some measurements of the effluent remaining TN were equal to or more than the average TN input. Except the possibility of having residues from previous synthetic wastewater, the above finding may denote that the MBR reactor under this condition has no nitrogen removal. The DO input was constantly high in MBR reactor, but the low organic carbon input (as food supply) insufficiently supported the activities of the microorganisms. In this event, the system failed to remove the nitrogen from wastewater.

4.8.1.5 Improved rate of TN removal by SMEBR

A direct comparison between SMEBR and MBR can demonstrate how the former system can improve the TN removal. Using the formula specified in Appendix A3, the TN removal was calculated in percentage; the result is presented in Figure 27.



Figure 27: The improvement of TN removal vs average DO in all stages

In particular, Figure 27 shows that the TN removal in stage one improved with the increase of DO until it attained 3 mg-N/L; still, the improvement began to decrease. Consequently, under the condition that C/N ratio is around 3:1, the suggested DO concentration should be kept between 2 and 4 mg/L, and the optimal concentration must be around 3 mg/L DO. Compared with the conventional MBR system under the same influent quality and sufficient DO (average

between 5 and 8 mg/L DO), the SMEBR with less DO as suggested can maintain the improvement of TN removal between 25% and 30%.

In stage two (C/N = 2:1), when the DO was closed to zero, the SMEBR attained the best removal improvement, but was decreased when the DO was more or less than the range of 0 and 1 mg/L. When DO was higher than 1 mg/L, the improvement of TN removal rapidly dropped to approximately 8%. Meanwhile, when DO was equal to zero, the improvement once again dropped to a low level, due to no effective nitrification process that transferring ammonia nitrogen to nitrate nitrogen. The best result of TN removal occurred when the DO was closed to zero, which was in the average of 0.2 mg/L. Most of the high TN improvements were determined under the DO lower than 0.5 mg/L. Thus, when C/N ratio is around 2:1, the range of DO should be kept between 0.1 and 0.5 mg/L, but not zero, to maintain the improvement of TN removal more than 20% with the conventional MBR process.

The experimental method performed in stages one and two was also conducted in stage three (C/N = 1:1). The results in the third stage, however, were slightly different from those in other two stages. Thus, the analysis was different and related to the discussion of deeply and barely zero DO condition, which was a special problem in this study.

4.8.1.6 Deeply and barely zero DO condition

The trend line of TN removal improvement by SMEBR (compared with the conventional MBR) in stage three was initially increasing, but became evenly decreasing when the DO conditions were more than 1.5 mg/L (Figure 27). This initial decreasing trend was induced by

the fact that the value of DO was mostly equal to zero. Some changes still occurred when the sludge was in deeply or barely zero DO condition although the DOs were equal to zero. This situation explains why some of the measured points for low improvement affected the trend of TN removal. The trend line, which began from the observation of points of improved percentage of TN removal in stage three (Figure 27), did not reveal improvement in the TN removal. When DO was equal to zero, some results showed a significantly high improvement rate of more than 20%, whereas other results indicated lower improvement that was less than 10%. This phenomenon was caused by the unequal distribution of aeration in relatively small reactor. The DO was zero at the measuring position, which was far from the aeration unit at the bottom sludge. By contrary, the DO was not exactly zero at the aerated position, which was particularly close to the aeration points and at the sludge surface exposed to the air. This phenomenon was described by considering the sludge, which had DO equal to zero that was generally located anywhere, as deeply zero DO. Meanwhile, the sludge located only in some position with DO equal to zero, including the measuring position, was defined as barely zero DO. This aeration problem hardly occurs in a large reactor, in which air can be equally distributed. However, in a small reactor, maintaining the DO as low as possible may induce aeration to become extremely weak and cannot be equally distributed in the entire reactor. In this study, a small lab scale reactor was used so that problem had occurred.

Figure 28 displays the trend lines without the consideration of the effect from the unbalanced air distribution under the zero DO. In particular, this figure expresses that the new trend line of stage three (none zero trend line) acquired the best improvement at the beginning, but subsequently decreased when DO increased. When the DO reached 0.36 mg/L, the

improvement of TN removal reduced to 24%, which was much less than the optimal result (37.10% improvement), but still acceptable. The best results in stage three were obtained when the DO was as low as zero, and the suggested DO range was between barely zero and 0.5 mg/L DO.



Figure 28: The improvement of TN removal vs DO in all stages,

(Including the trend line of none zero points in stage III)

The means of how DO changes influence the TN removal can be determined by defining the differences of effects with respect to deeply and barely zero DO by measuring the oxidation reduction potential (ORP) in the sludge.

4.8.1.6.1 Proven by ORP measurement

Previous discussions on ORP claim that the ORP measurement can help define the level of DO when such level was remarkably low to be accurately measured in the sludge. High and positive ORP indicates the barely zero DO, whereas the low or negative ORP presents the deeply zero DO condition.

Day	SMEBR	MBR	Improvement of		ORP in SMEI	BR mV
	TN mg/L	TN mg/L	TN removal	Max	Min	Average
28	33	43.2	23.61%	126.3	88.2	107.25
29	37.1	41.6	10.82%	76.3	-85.8	-4.75
31	32	39.25	18.47%	-270	-321	-295.5
37	32.35	40.4	19.93%	-273	-304.3	-288.65
39	31.65	34.95	9.44%	134.8	104.7	119.75
45	34.15	40.45	15.57%	-156.1	-212	-184.05

Table 15: Improvement of TN removal and ORP under average DO equals to zero, Stage III

Table 15 specifies that all improvements of TN removal occurred under DO conditions that were equal to zero. In particular, the table evidently suggests that in Days 28 and 39 (Stage three), the sludge was under a barely zero DO condition, in which the ORP level was maintained as high as 100 mV. The measurement from Day 37, however, revealed that the sludge was under a deeply zero DO condition because it had ORP level as low as –300 mV. The result of

this study reveals that the best TN removal improvement occurred on Day 28, which was the time when the sludge was under a barely zero DO condition, as proven by ORP. The measurement obtained on Day 39 was also good because the sludge was under a barely zero DO condition, as verified by the ORP measurement. Nevertheless, the TN removal rate by MBR was also high on this day; hence, the TN removal improvement was not high.

4.8.1.6.2 Conclusion

Under the low C/N ratio at 1:1, aeration must be carefully controlled and maintained close to zero, but not exactly at zero level. Table 16 shows that the highest TN removal improvement occurred on Day 28, in which the sludge was under a barely zero DO condition. Likewise, TN removal was remarkably good on Day 39, but since the comparative MBR on that day also provided good results, the improvement was not shown. Under deeply zero DO conditions, the SMEBR could also maintain the improvement of TN removal between 10% and 18%. This finding is acceptable and can still be improved.

When the DO was not zero, the highest TN removal improvement occurred on Day 24, in which the average DO concentration was close to zero (average 0.03 mg/L) (Tab. 16). Meanwhile, the highest TN removal improvement ensued on Day 4 when the value of DO was close to 0.6 mg/L. These two findings were both higher than the improvement when DO was zero, implying that the best DO range for TN removal by using SMEBR under C/N = 1 is between 0 and 0.5 mg/L and close to zero as much as possible, but not in deeply zero DO.

Day		SMEBR		SMEBR	MBR	Improvement of
	DO max	DO min	DO ave	TN mg/L	TN mg/L	TN
4	0.79	0.49	0.57	31.6	44.9	29.62%
8	5.76	5.27	5.59	40.9	42.6	3.99%
11	2.27	1.98	2.12	35.4	42.5	16.71%
14	0.4	0.11	0.22	39.8	48.5	17.94%
17	0.05	0.02	0.03	37.8	43.4	12.90%
21	0.08	0.02	0.04	35.1	40.1	12.47%
24	0.07	0.02	0.03	31.1	50.2	38.05%
42	1.81	1.35	1.57	31.1	38.95	20.15%
44	0.61	0.53	0.56	38.45	46.2	16.77%

Table 16: TN removal improvement under DO higher than zero, stage III

Another observed effect on the TN removal was the possible low mixing (settling) of the sludge. Deeply zero DO pertains to weak aeration in the reactor. The aeration in the experimental utilized reactor is the only method of maintaining the sludge in suspension. Thus, most sludge might settle within insufficient aeration, thereby affecting the treatment results. This problem would not arise in a large reactor because other stirring methods can be used. In a small reactor, however, stirring can hardly be implemented at lab scale. Thus, future research under deeply zero DO condition must be conducted at pilot scale facilities.

4.8.1.7 Total Kjehldahl Nitrogen (TKN)

In this study, few TKN tests were conducted at the end of stage three to better define the forms of nitrogen compounds in the effluent (Tab. 17).

		SMEBR eff.			MBR eff.			Inflow	
Date	TN	NO3 ⁻ -N+NO2 ⁻ -	TKN	TN	NO3 ⁻ -N+NO2 ⁻ -	TKN	TN	NO3 ⁻ -N+NO2 ⁻ -	TKN
	mg/L	N mg/L	mg/L	mg/L	N mg/L	mg/L	mg/L	N mg/L	mg/L
31	32	5.65	26.35	39.25	15.25	23.95			
37	32.35	9.75	22.6	40.4	15.35	25.05	43.95	1.565	42.35
39	31.65	12.9	18.75	34.95	13.85	21.1			
42	31.1	13.45	17.65	38.95	15.45	23.5			
44	38.45	14.55	23.9	46.2	15	31.2	46.4	1.905	44.5

Table 17: Remaining forms of nitrogen in influent and effluent, stage III

Results show that the TKN concentration in MBR was generally higher than that in SMEBR. However, on Day 31, the TN was low in SMEBR, but the TKN was relatively high because the higher ammonia concentration under deeply zero DO condition. Such circumstance resulted in slightly higher concentration of TKN in SMEBR compared with MBR. Nevertheless, all concentrations were below the acceptable limit in Quebec. In definition, TKN consists of ammonia and organic nitrogen; by calculation, the results of organic nitrogen are shown in Table 18.

	Organic nitrogen in	Organic nitrogen in	Organic nitrogen in
Stage III	effluent mg/L	effluent mg/L	effluent mg/L
	MBR	SMEBR	Inflow
31	1.3	1.3	-
37	4.2	6.3	9.4
39	-1.8 (0)	2.4	-
42	3.1	6.7	-
44	11.5	9.0	16.4

Table 18: Organic nitrogen in influent and effluents, stage III

By deducting ammonia nitrogen, the organic nitrogen in MBR was predominantly lower than that in SMEBR, except for the measurement on Day 44. This occurrence was probably due to the high TKN in the influent and MBR on that day, which led to the high overall nitrogen concentration. The results of the other four groups signify that the MBR contained higher TKN, but lower organic nitrogen. Accordingly, the nitrogen in MBR was mostly in the form of ammonia nitrogen, and the treatment was not effective. Meanwhile, less TKN and more organic nitrogen not only presented a better ammonia nitrogen removal by SMEBR, but also confirmed that the effluent from SMEBR would be more eco-friendly when it contains more organic nitrogen instead of ammonia.

4.8.1.8 Improvement of nitrogen removal

The relationship between ammonia nitrogen removal, nitrate nitrogen removal, and total nitrogen removal is extremely important in the improvement of nitrogen removal in SMEBR. Controlling the amount of nitrogen compounds should strike balance between nitrification and denitrification processes (either too high or too low ammonia and nitrate nitrogen removal is not allowed), which depend on the control of aeration and carbon supply. Total nitrogen removal focuses on the amount of nitrogen source input that can finally be transformed and released as nitrogen gas. This amount will be the combination of nitrification and denitrification processes.

The discussion of each stage's result has provided the level of improvement of ammonia nitrogen removal, nitrate nitrogen removal and total nitrogen removal by comparing the SMEBR and MBR results. By taking observation of all the results, it would be helpful to find the balance of ammonia nitrogen removal and nitrate nitrogen removal that can provide optimal total nitrogen removal.

4.8.1.8.1 Stage I

Stage one requires more oxygen to maintain good removal rate (Figure 29) with a relatively high organic carbon supply (C/N = 3:1). The suggested range of dissolved oxygen in this experiment has been defined as 2 to 4 mg/L. Ammonia nitrogen and nitrate nitrogen removal by SMEBR could improve 20% to 40% compare to MBR when dissolved oxygen was around 3 mg/L. For instance, when dissolved oxygen was between 1.99 and 2.93 mg/L, improved

ammonia nitrogen removal reached 25.93% in day 57 (a typical example of good TN removal). Improved nitrate nitrogen removal has reached 43.82%. Total nitrogen removal improved by 25.5%, which was one of the best results in this stage.



Figure 29: Improvement of NH₃⁻-N, NO₃⁻-N and TN removal with average DO, SMEBR, stage I

Trend shows that total nitrogen removal would be more effective when the improved percentage of ammonia nitrogen removal (red curve) and nitrate nitrogen removal (blue curve) are becoming close. This trend is shown in Figure 29. With sufficient dissolved oxygen, the nitrification would transfer more ammonia nitrogen to nitrate nitrogen. Hence, ammonia nitrogen removal could be better improved. However, nitrate nitrogen cannot be transferred to nitrogen gases without a good condition for denitrification. Therefore, nitrate nitrogen removal would be low and treatment result would be limited. More nitrate nitrogen could be transferred
to gases under extremely low dissolved oxygen. Ammonia nitrogen could not be transformed to nitrate form without nitrification. In this case, ammonia nitrogen removal would be limited and nitrate nitrogen would be reduced further, which will produce poor results. The optimal condition is when the improvement levels of ammonia nitrogen removal and nitrate nitrogen removal are close in percentage. This situation is present in strong nitrification and denitrification. Ammonia nitrogen removal will not be extremely high because of controlled dissolved oxygen. Therefore, the denitrification process transfers more nitrate nitrogen to gases.

Another example that supports this assumption was the final day of the stage when the dissolved oxygen had been reduced to an almost zero level to observe the effects. Consequently, the improvement of ammonia nitrogen removal reached 76.52%. Therefore, the concentration of NH₃⁻-N in the effluent of SMEBR was higher than that in MBR. However, nitrate nitrogen reached 98.31% improvement. This result is typical in the absence of nitrification. Most of the nitrogen were in the form of ammonia nitrogen. The difference between the percentage of ammonia nitrogen removal improvement and nitrate nitrogen removal improvement was almost 200%. In this case, the total nitrogen removal using SMEBR reactor improved by only 0.34%, which means that nitrogen removal in SMBER was the same as in MBR. SMEBR could maintain the same result without aeration considering the sufficient dissolved oxygen supply in MBR, which was also an improvement.

In conclusion, if the total improvement of nitrogen removal can be maintained to as high as 30% compared with the regular MBR treatment, then the ammonia nitrogen removal will most likely improve in the range of around 20% by using SMEBR under the condition that the C/N ratio is equal to 3:1 and current density is around 10 A/m². Moreover, the amount of dissolved oxygen is between 2 and 4 mg/L. The generated nitrate nitrogen removal improvement will be in the range of 40% to 80%.



4.8.1.8.2 Stage II

Figure 30: Improvement of NH₃⁻-N, NO₃⁻-N and TN removal with average DO, SMEBR, stage II

Carbon input was reduced by 35% in stage two (C/N = 2:1). This reduction rapidly decrease the required oxygen input to approximately 0 to 0.5 mg/L. Figure 30 shows that several measurements of dissolved oxygen before day 30 were more than 1 mg/L. These measurements were caused by the process of seeking for the appropriate dissolved oxygen input for the best nutrient removal. The big difference between the improvement of ammonia nitrogen and nitrate nitrogen removal in this period, which were caused by excessive DO in SMEBR, led the improvement of TN removal to as low as between 0% to 20%. The dissolved oxygen from day

30 to day 50 was mostly maintained at between 0 and 0.5 mg/L. The improvements of ammonia nitrogen, nitrate nitrogen, and total nitrogen removal stabilized.

The best TN removal improvement (37.1%) happened on day 32 (Figure 30). Ammonia nitrogen removal improved by 45.8%, and the generated nitrate nitrogen removal was improved by 46%, which was close to that of ammonia nitrogen removal's improvement. These results prove that when the percentage of ammonia nitrogen and generated nitrate nitrogen removal improvement were close, both decreased at a similar rate compared with that in the conventional MBR. The balance between nitrification and denitrification was maintained under this condition, and so total nitrogen removal was better improved.

The dissolved oxygen on day 32 was between 0.14 and 0.23 mg/L. This DO level allowed for the assumption that the sludge was mostly in anoxic conditions. Theoretically, denitrification reaction will be stronger than nitrification under the low DO. Increased ammonia nitrogen and less remaining nitrate nitrogen (presented in Figure 30 as decreasing ammonia nitrogen removal improvement and increasing nitrate nitrogen removal improvement in percentage) proved this assumption. However, the total nitrogen removal could also be improved by approximately 37% of the treatment results from comparative MBR under this condition. Hence, this DO level was the best level to the balancing reaction under low carbon input at C/N = 2:1.

The dissolved oxygen had been adjusted higher at the end of the experiment to observe the changes of treatment result after 50 days of successfully running the reactor. This adjustment was applied to prove that the variation of TN removal was not affected by the unstable sludge condition, but only by the increased dissolved oxygen. When the dissolved oxygen was around

0.5 mg/L, the total nitrogen removal improvement dropped to less than 20%. These results were the same in the first 30 days. The reduction of the improvement of ammonia nitrogen removal and increase of nitrate nitrogen removal improvement could also be observed from the curves on Figure 30. Therefore, the nitrification process had become stronger. This result proved that the best dissolved oxygen input under C/N ratio equal to 2:1 should be kept between 0 and 0.5 mg/L. Under the current density around 11 A/m², the SMEBR within this DO input can effectively remove ammonia nitrogen and nitrate nitrogen. Total nitrogen removal improved to more than 30%. Ammonia nitrogen removal and nitrate nitrogen removal can be improved from 30% to 40%.

4.8.1.8.3 Stage III

In stage three (C/N = 1:1), the control of dissolved oxygen became harder than in previous two stages because the appropriate aeration was approximately zero. However, aeration input should exist for mixing purposes. Hence, minimizing dissolved oxygen without shutting down the aeration was the only way to achieve the required dissolved oxygen. Figure 31 shows that the improvement curve of ammonia nitrogen removal and nitrate nitrogen removal were continually changing. However, dissolved oxygen mostly remained zero. This result shows that the sludge would be more sensitive to environmental changes, especially dissolved oxygen input change, under the low carbon supply.



Figure 31: Improvement of NH3--N, NO3-N and TN removal with average DO, SMEBR, stage III

The best TN removal improvement happened on day 24, which showed 38.05% improvement (Figure 31). Improved ammonia nitrogen removal was around 40.77% under dissolved oxygen of around 0.03 mg/L, and improved nitrate nitrogen removal was 31.82%. Figure 31 shows that these two improvement curves were becoming closer on day 24. This occurrence proved the previous assumption, which claims that TN removal could be optimally improved by SMEBR when the improvement of both ammonia nitrogen and nitrate nitrogen removal was getting close. Under the low carbon input (C/N = 1:1), the microorganisms were exceptionally sensitive to dissolved oxygen. The minimum aeration supply (DO close to 0) was enough to support the nitrification process in the small reactor at experimental scale.

Other results of TN measurement could not reach more than 30% of improvements because of dissolved oxygen control. Figure 31 indicates the direct observation of the percentage improvement curves, shows that the percentage of improvement of ammonia nitrogen removal and nitrate nitrogen removal had a big difference. However, the dissolved oxygen from the measurement was mostly zero. This difference is related to the discussion of barely zero and deeply zero dissolved oxygen (Section 4.8.1.6). As expected from a bigger reactor, the higher improvement of the total nitrogen removal could be achieved when better aeration system is applied.

Similar to the procedure in stage two, the aeration supply was increased at the end of this stage to define the maximum DO under C/N =1:1. Unlike the measurement for the first 10 days, which exhibited high DO without the impacts from the unstable sludge condition at the beginning, the result occurred only because of the changes of aeration supply. The dissolved oxygen had increased from day 41 to 42. As expected from the result shown on the curves in Figure 31, the improvement of ammonia nitrogen removal has suddenly increased. In this case, the improvement of nitrate nitrogen removal rapidly dropped to the bottom (day 41, where DO = 3.01 mg/L). The dissolved oxygen slowly decreased in the next few days, and the two curves were also getting close. Under this condition, TN removal by SMEBR could still maintain more than 15% improvements compared with the comparative MBR system. This result proved that the SMEBR under C/N = 1 was sensitive to excessive aeration supply. However, recovery was fast when aeration returned to the suggested level.

According to the records of stage three, the control of SMEBR condition in this phase was not optimal. Numerous problems occurred, such as sludge settling and unequal distribution of the aeration. The average TN removal improvement oscillated between 15 % and 20 %. These percentages were not better than the results of the previous stages, but proved that the SMEBR was still better than the conventional MBR system. Therefore, the results not only proved the nutrient removal capability of SMEBR, but also showed the high tolerance of SMEBR of worse condition, and its high recovery speed after experiencing negative impacts.

By using SMEBR reactor, the total nitrogen removal could be averagely improved at around 15% to 20%, under the conditions that the C/N ratio is equal to 1:1 and current density is around 7.58 A/m², and also the dissolved oxygen is barely zero (where ORP is around 100 mV). Total nitrogen removal could be increased to 30% to 40% when the condition can be better controlled. The best ammonia nitrogen removal improvement can be assumed to be around 30% to 40%, and the best nitrate nitrogen removal improvement will be similar in percentage under these conditions.

4.8.1.9 Mechanisms of nitrogen removal improvement

The comparative study of MBR and SMEBR shows that nitrogen removal could constantly improve independently to different C/N. However, the level of improvement is highly dependent on the reactor conditions, including dissolved oxygen input, electrical current input, organic carbon input and other conditions, such as pH and electrical conductivity. Controlling these conditions, especially DO, C/N and current density, allows the nitrification/denitrification process in SMEBR to successfully convert more ammonia nitrogen to nitrogen gas.

According to previous studies (Ibeid, 2011), the dissolved oxygen should be consistently kept at a relatively low level because the SMEBR is capable to switch reactor conditions between these adequate for nitrification and these adequate for denitrification. It is assumed that the majority of nitrogen removal took place through nitrification/denitrification process. In some point, the created in reactor conditions might be also adequate for anammox organisms growth as it has been discussed in the previous studies. In spite that the conditions for anammox bacteria growth were limited, it was possible that these bacteria also contributed in removal of nitrogen following the reaction (Ibeid, 2011).

$$NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O$$

Moreover, the SMEBR system generates the electrooxidation reactions. Thus, it is speculated that ammonia and nitrites might also have been exposed to electrooxidation leading to additional nitrogen removal. Theoretically, the nitrogen removal improvement in SMEBR could be from both anammox and the electrooxidation processes, which can give explanations to fluctuations of ammonia removal in Figures 13, 14 and 15 comparing to the remaining nitrate concentration (Figures 18, 19 and 20).

4.8.2 Phosphorus removal

Phosphorus, another important nutrient compound aside from nitrogen, must be removed during most of the wastewater treatment process. In conventional MBR systems, phosphorus removal is not effective and extra treatment units or additional processes are required to remove the remaining phosphorus. However, in the SMEBR, the problem can be solved using only a single reactor. Compared with other treatment methods, such as conventional MBR, SMEBR can provide better results in terms of total phosphorus removal. This study thus aims to prove the high efficiency of phosphorus removal under condition that provides optimal nitrogen removal by using the SMEBR under the low C/N ratio, and then compare its performance with that of conventional MBR.

Good phosphorous removal is expected in the current study, in accordance with previous findings (Wei et al., 2009; Ibeid, 2011). The results (Figure 32) proved this assumption because the concentration of the orthophosphorus in SMEBR effluent remained as low as zero most of the time. By contrast, the concentration of remaining phosphorus in the effluent from the MBR was mostly the same as that of the influents. Thus, the MBR treatment was unable to effectively remove phosphorus.



Figure 32: Remaining orthophosphorus in MBR effluent (black) and SMEBR effluent (red) in all stages

Figure 32 shows the obvious drop in phosphorus concentration in the effluent from the SMEBR reactor, which could be observed at the beginning of each stage. These results showed that the preparation time of the SMEBR reactor for phosphorus removal could only be a few 103

days. The effective phosphorus removal had to rely on the appropriate control of the operational conditions, including electrokinetic process. Nevertheless, some sudden increases, especially those that occurred during the second stage, also proved that the system can be very sensitive to changes in treatment conditions and demonstrated fast recovery speed when these conditions were back to normal. Hence, during the operation, such conditions as temperature, pH, current density, and so on, should be carefully controlled.

4.8.2.1 Stage I

At the beginning of the study, there were many conditions needed to be adjusted. In this stage, before day 31, the conditions such as sludge concentration, dissolved oxygen input, organic carbon input and phosphorus source input had been changed many times. In spite of such unstable operational conditions, the phosphorus removal reached a good a good level: almost all phosphorus had been removed.

The results on Figure 33 demonstrate the high efficiency phosphorus removal capability of the SMEBR, even under unstable conditions. This can be attributed to the electrokinetic process given that other conditions were changed frequently.

After Day 7, dissolved oxygen was recorded, after which a comparison of the results from the phosphorus measurement in Day 31 and the level of dissolved oxygen was conducted. Meanwhile, the dissolved oxygen in MBR was maintained between 5 mg/L to 8 mg/L (not shown in Figure 33).



Figure 33: Orthophosphorus concentration in both reactors' effluent, stage I

The formula shown in Appendix (A4, 1) was used to calculate phosphorus removal in each reactor, whereas the formula shown in Appendix (A4, 2) was used to calculate the improvement of the phosphorus removal rate through the application of SMEBR. During calculation, phosphorus input was assumed to have an average of 20 mg PO_4^{3-}/L . The calculated results with measured concentrations are shown in Table 19.

DO mg/L	Orthophosphorus	Orthophosphoru	Improved	Removal by
SMEBR	in SMEBR	s in MBR	removal %	SMEBR
	(mg/L PO4 ³⁻)	(mg/L PO4 ³⁻)		%
2.53	0.651	18.1	96.40%	96.75%
3.01	1.22	14.4	91.53%	93.90%
2.36	0.052	21.3	99.76%	99.74%
3.00	0.164	14.8	98.89%	99.18%
0.21	0.564	14.7	96.16%	97.18%
0.00	2.78	13.3	79.10%	86.10%

Table 19: Orthophosphorus removal while C/N = 3:1, stage I

The results show excellent phosphorus removal that way below acceptable limits in Quebec confirmed previous studies (Hasan, 2011; Ibeid, 2011). Subsequently, no daily measurements of phosphorus were performed, assuming the same trend of phosphorus removal under dissolved oxygen from 2 mg/L to 3 mg/L (i.e., the best range defined for nitrogen removal). Thus, the successful removal of phosphorus was observed in the first stage under the condition C/N=3:1. Compared with conventional MBR, when dissolved oxygen ranged between 2 mg/L and 3 mg/L, under the current input of 1 A (current density average: 10.45 A/m²), 90% to 99% phosphorus removal improvement was reached. This finding suggested that, under optimal operational conditions for nitrogen removal in SMEBR, phosphorus removal can also reach optimal level. In fact, phosphorus removal did not seem to be related to DO concentration, but

was more improved by ideal electrical conditions, including current density and current ON/OFF phase switching. As long as the electrical condition was stabilized, the effect from dissolved oxygen was negligible.

4.8.2.2 Stage II

After adjustments in the first stage, the sludge conditions, such as the current input and sludge concentration, were stabilized. Thus, the reactor conditions in the second stage were much easier to control, and the results were mostly as good as expected. Figure 34 shows the relation between phosphorous removal and average dissolved oxygen concentration in both the MBR and the SMEBR reactor's effluent.



Figure 34: Orthophosphorus in both reactors' effluent, Stage II

In comparison, conventional MBR was unable to achieve complete phosphorus removal (Figure 34). Specifically, high remaining phosphorus was observed in the effluent of MBR under both the high and low dissolved oxygen conditions. Often, phosphorus removal for this reactor is as low as none. By contrast, the proposed SMEBR maintained high phosphorus removal efficiency under any DO concentration.

Dissolved oxygen	Orthophosphorus	Orthophospho	Improved	Removal by
in SMEBR	in SMEBR	rus in MBR	removal	SEMBR
mg/L	(mg/L PO ₄ ³⁻)	(mg/L PO ₄ ³⁻)	%	%
3.44	2.21	20.1	89.00%	88.95%
0.53	1.16	0.434	-167.28%	94.20%
0.02	0.945	1.9	50.26%	95.28%
4.83	1.98	7.05	71.91%	90.10%
1.56	1.58	7.66	79.37%	92.10%
0.09	7.6	10.8	29.63%	62.00%
0.17	0.004	10.2	99.96%	99.98%
0.17	0.01	13.5	99.93%	99.95%
1.06	0.011	12.6	99.91%	99.95%
0.24	0.008	16.3	99.95%	99.96%
0.57	0.765	17	95.50%	96.18%

Table 20: Orthophosphorus removal while C/N = 2:1, stage II

Under the C/N ratio of 2:1, with the best dissolved oxygen range for nitrogen removal of between 0 mg/L to 0.5 mg/L, and a current input around 1 A (current density average: 11.44 A/m²), the phosphorus removal in the SMEBR also reached the best removal rate; it showed more than 99% improvement compared with the regular MBR system (Table 20). Thus, the suggested operational condition for nitrogen removal under this level of C/N was also good for phosphorus removal.

4.8.2.3 Stage III

In the third stage, phosphorus removal in SMEBR was even more effective compared with the two previous stages. In these cases, the concentration of phosphorus in the SMEBR effluent was close to zero most of the time, except when the initial measurement was taken, during which the electrical system was not applied into the SMEBR and the system was running as a conventional MBR. Under the same conditions, the phosphorus concentration in MBR effluent remained close to the input phosphorus (20 mg/L PO4³⁻ on average). This result proved the ineffectiveness of phosphorus removal using conventional MBR. By contrast, under the same situation, most phosphorus was removed in the SMEBR through an electrokinetic process.



Figure 35: Orthophosphorus in both reactors' effluent, Stage III

Meanwhile, phosphorous removal was close to 100% for dissolved oxygen between 0 mg/L to 6 mg/L (Figure 35). When C/N=1:1, the suggested dissolved oxygen for nitrogen removal was between 0 mg/L and 0.5 mg/L. Thus, phosphorus removal was also optimal under this DO level. Moreover, the current input of this stage was reduced from 0.6 A to 0.8 A, which led to less current density. However, under this condition, the result remained at a relatively good removal rate of 99% (Table 21). The high tolerance of inappropriate operational condition by conducting SMEBR had been proved.

Dissolved oxygen mg/L	Orthopho	osphorus,	Improved	Removal by
SMEBR	mg/L PO4 ³⁻		removal	SMEBR
	SMEBR	Control	%	%
6.62	12.8	15.4	16.88%	36.00%
5.59	0.015	17.8	99.92%	99.93%
0.22	0.047	19.9	99.76%	99.77%
0.04	0.022	16.5	99.87%	99.89%
0.00	0.033	17.7	99.81%	99.84%
0.00	0.031	17.5	99.82%	99.85%
1.57	0.015	17.8	99.92%	99.93%
0.00	0.05	19.38	99.74%	99.75%

Table 21: Orthophosphorus removal while C/N = 1:1, Stage III

Thus, with C/N ratio =1:1, when the sludge was in an adequate range of dissolved oxygen (between 0 to 0.5 mg/L) and current input between 0.6 A and 0.8 A (current density as average 7.58 A/m²), the SMEBR removed more nitrogen compounds than regular MBR, demonstrating an improved phosphorus compound removal rate of over 99%. Although the suggested current input from DC power supply was 1 A, a slight change on the current did not greatly affect the results.

4.8.2.4 Conclusion of phosphorus removal

Under low organic carbon input, the SMEBR can remove more than 99% of reactive phosphorus. Furthermore, phosphorus removal in SMEBR is more dependent on the electrokinetic process, and a different sludge quality or dissolved oxygen does not affect the results contrary to regular MBR.

Moreover, based on the observation of treatment results, unlike nitrogen removal, phosphorus removal using SMEBR can be maintained even under some unstable operational conditions, especially sudden changes in levels of dissolved oxygen. The tolerability of unsteady state condition is beyond the regular treatment. Moreover, under such conditions as low carbon source or dissolved oxygen input, SMEBR still provided reliable results. Thus, when designing treatment systems using the SMEBR under the low carbon source input, tertiary treatment for phosphorus removal is no longer required.

4.8.3 Overall nutrient removal performance of the SMEBR

This study showed that under a low C/N ratio (between 1:1 and 3:1), the SMEBR could improve approximately 30% to 40 % of nitrogen removal and more than 99% of phosphorous removal compared with the conventional MBR process. For nitrogen removal, the results are highly dependent on controlling dissolved oxygen: too high or too low dissolved oxygen reduces treatment efficiency. With a lower carbon source input, the range of optimal dissolved oxygen support is also lower. The system would be more sensitive to DO changes. Thus, the nitrification/denitrification process should be placed under a more critical control.

For phosphorus removal, as a result of the applied electrical field, the effect from dissolved oxygen and activated sludge are insignificant, and the major factor of effective phosphorus removal is electrokinetic process. In conclusion, under an adequate control of dissolved oxygen, current input and sludge suspension, the overall nutrient removal performance of the SMEBR is much higher than that obtained using the MBR system.

C/N=3:1	MBR	SMEBR	Improvement
Current density (CD) (SMEBR): 10.45 A/m ²			
Suggest range of DO (mg/L)	5 to 8	2 to 4	
Ammonia N removal % (NH3 ⁻ -N)	40 % to 50%	60%	Improvement up to 20%
Remaining nitrate N mg/L (NO3 ⁻ -N)	10 to 12	0 to 5	Reduction40% to 80%
Total nitrogen removal mg/L (TN)	5.3 to 14.8	13.5 to 21.5	Improvement 31%
Phosphorus removal mg/L (PO4 ³⁻)	6.9 to 21.3	0.052 to 2.78	Improvement 90% to 99%
C/N=2:1, CD (SMEBR): 11.44 A/m ²	MBR	SMEBR	Improvement
Suggest range of DO (mg/L)	5 to 8	0 to 0.5	
Ammonia N removal % (NH3 ⁻ -N)	40%	60% to 80%	Improvement 20% to 40%
Remaining nitrate N mg/L (NO ₃ ⁻ -N)	10 to 15	5 to 10	Reduction 30% to 40%
Total nitrogen removal mg/L (TN)	6.6 to 17	12.1 to 24.3	Improvement 37%
Phosphorus removal mg/L (PO4 ³⁻)	0.434 to 20.1	0.008 to 7.6	Improvement > 99%
C/N=1:1, CD (SMEBR): 7.58 A/m ²	MBR	SMEBR	Improvement
Suggest range of DO (mg/L)	5 to 8	close to 0	
Ammonia N removal % (NH3 ⁻ -N)	40%	40% to 80%	Improvement up to 40%
Remaining nitrate N mg/L (NO3 ⁻ -N)	12 to 17	6 to 12	Reduction 30% to 40%
Total nitrogen removal mg/L (TN)	1.1 to 9.55	3.6 to 13.4	Improvement 38%
Phosphorus removal mg/L (PO4 ³⁻)	16.5 to 19.9	0.0015 to 0.05	Improvement > 99%

Table 22: Overall nutrient removal

Operational conditions	C/N = 3:1	C/N = 2:1	C/N = 1:1	
Current density (A/m2)	10.45	11.44	7.58	
Dissolved oxygen (mg/L)	2 to 4 0 to 1 close to 0			
рН	around 7			
HRT (hour)	12 to 14			
SRT (day)	15 to 20			
Electrical conductivity (µS/cm)	430 to 480			
TSS (mg/L)	Initial: 3000- 4000, increase with operation time			

Table 23: Suggested operational condition for effective nutrient removal, SMEBR

4.9 Total suspended solid (TSS)

Several TSS tests had been conducted to balance the initial concentration of sludge in both reactors. The measured TSS could be the same as the mixed liquid suspended solid (MLSS) (Zhang et al., 2014).

Changes in the quality and concentration of suspended solid were observed during the experiment. The same amount (700 mL/d) of sludge from each reactor had been taken daily in order to maintain the same sludge residence time of 20 days. The results showed that the generation of sludge from biological and electrokinetic processes in SMEBR was consistently higher than in conventional MBR. The sludge from SMEBR contained more inorganic compounds, which were mostly from the dissolution at the anode. This phenomenon could be directly observed from different sludge colors (Appendix A6, Figure A1).

The observation of settling tests showed that the sludge from SMEBR was more easily settled than the MBR sludge. These results were caused by the high sludge concentration and quality of sludge particles in SMEBR sludge (Table 24). A good settling ability provided the sludge a good dewatering rate. Hence, less time and energy would be required by applying SMEBR in the sludge dewatering process.

A trend of increasing suspended solid concentration in SMEBR could be observed in each stage, as shown in Table 24. Simultaneously, the sludge concentration in MBR decreased because of the daily sludge take out. This decrease is caused by low organic carbon input. The generation of organic sludge could also be low in SMEBR under the same condition. Hence, the increased of sludge amount is mostly due toelectrokinetic process. The aluminum material would react with the source of nutrients in the sludge under electrokinetic process. Normally, the material could only hold for a certain period, which depended on its size and quality. The similar anode unit was conducted in stages one and two had been used for 5 months.

Stage I				
Day	SS MBR (g/L)	SS SMEBR (g/L)		
0	1.782	1.996		
14	2.816	6.084		
48	1.14	4.66		
	Stage II	t		
Day	SS MBR (g/L)	SS SMEBR (g/L)		
13	2.302	0.534		
40	1.028	2.436		
47	0.654	2.742		
	Stage II	I		
Day	SS MBR (g/L)	SS SMEBR (g/L)		
0	0.854	1.006		
14	0.186	0.804		
28	0.044	1.288		
31	0.142	0.858		
37	0.046	1.234		
39	0.366	2.694		
42	0.272	2.36		
44	0.292	1.796		

Table 24: Suspended solids concentration in in both reactor's sludge, Stage I, II, III

Three TSS organic percentage tests had been conducted at the end of experiment stage three. The residue from SMEBR sludge was higher than in MBR after incubation. Table 25 provides the measurement results of organic sludge in the total sludge of both reactors. In the activated sludge from MBR, the organic sludge were as high as 85% of the total amount of sludge. The SMEBR sludge only contained 37% of the organic sludge. The original sludge in each reactor contained the same quality and volume of sludge. The synthetic wastewater input also contained no sludge input. Hence, the extra inorganic sludge would be obtained mostly from the SMEBR system (corrosion of anode material) during the operation.

Day	MBR	SMEBR
39	0.83	0.38
42	0.86	0.37
44	0.86	0.36

Table 25: MLVSS/MLSS ratio in both reactors

The organic sludge can also be considered as mixed liquid volatile suspended solid (MLVSS), and at the same time, the total sludge can be considered as MLSS. In this case, the average MLVSS/MLSS ratio was around 0.85 in MBR demonstrating typical ratio for activated sludge while average 0.37 ratio was observed in SMEBR (Tab. 25). This result might not be very accurate because of limitations in laboratory equipment's accuracy. However, this result

showed the difference between regular activated sludge and SMEBR sludge. In addition, this result could be used as the basis for the range of future studies.

4.10 Membrane fouling reduction

Membrane fouling reduction was not the major target of this study. However, some indications still proved that the SMEBR system could effectively reduce the impact from fouling. The pressure gauge was connected to the membrane module to measure transmembrane pressure, which indirectly indicated the fouling rate as transmembrane pressure (TMP). Two gauges were connected between the membrane module and effluent pump. Unfortunately, the malfunction caused one of the gauges to fail to present the value accurately after half of stage one. Therefore, the comparison could be made at only the first half of stage one. The record indicated that the gauge readings and observations showed an increase of transmembrane pressure in the membrane module of both reactors at the beginning of experiment (day 11). Later, the increased TMP for MBR was by average 5 times higher, and increased TMP for SMEBR was less than 2 times higher comparing to the initial conditions.



Figure 36: Gauge pressure in both reactors, stage I, 0 to 33 days

The results in Figure 36 show the frequent changes of TMP in the gauge of MBR caused by membrane fouling. Meanwhile, the TMP pressure has also changed significantly in SMEBR in the first 11 days under the same cleaning process. Subsequently, the pressure change curve has become smooth and steady. This difference was mostly attributed to sludge quality changes in SMEBR, which were caused by the electrokinetic process.

. The gauge pressure in MBR has stabilized as high TMP at the end of the measurements (Figure 36) without backwash process. Under the same condition, the TMP in SMEBR has dropped slowly due to lower fouling. The results have proven that using SMEBR system can extend the lifecycle and processing time of a membrane without cleaning. In the study, conventional MBR conducted daily membrane cleaning. The period between two cleanings in SMEBR could be doubled. However, the longer period (more than two days) was not tested with small reactors and spilling prevention consideration. This long period studies was left to future experiments.

4.11 Reproducibility of results

To minimize the impact from inaccurate measurements, the evaluation of reproducibility had been conducted. Since the removal of COD and orthophosphorus has reached more than 95% in most of measurements, their reproducibility was clear and no more evaluation was required. The evaluation focused mainly on nitrogen removal, including ammonia nitrogen and nitrate nitrogen.

To double check the accuracy of measurements, on Feb 8th 2014, stage three, three readings had been conducted in both samples of influent, SMEBR effluent and MBR effluent. The time between two readings was approximately 10 minutes, the result has presented in table A5.1 (Appendix) and Figure 37.



Figure 37: Comparison of ammonia nitrogen remaining between influent, SMEBR and MBR effluent.

To calculate standard deviation, multiple measurements of each chemical had been conducted. In the measurements with electrode tests, the daily measurements' results were taken from the average of multiple reading: At least three reading were conducted in the same sample, and result was calculated as:

$$Average = \frac{X1 + X2 + \cdots Xn}{n}$$

Where n was the number of measurements, and X was the measured result.

This method had applied in measurements of ammonia nitrogen, nitrate nitrogen, electrical conductivity and pH.

For DO and ORP measurement, since the electrode was directly inserted into the sludge, and results were recorded in every 2 to 5 minutes, the deviation can be minimized by taking average as well. However, for measurements that conducting TNT Hach tests, because of the limited number of measuring kits, multiple measurements was not available. To minimize the impact from inaccurate measurement, in each measurement, the sample would be measured in three times, and approximately 10 minutes between each reading. The average value from three readings would be the final result of measurement, which was calculated by conducting similar formula as the electrode measurements.

This method had applied in measurements of COD, orthophosphorus, TN, TKN, and ammonia/nitrate nitrogen in stage one.

To calculate the standard deviation, conducting formula as (Ibeid, 2011):

$$\sigma = \sqrt{\frac{\sum (x - \gamma)^2}{n}}$$

Where x is the measured value, ne is the number of measurement,

 γ is the mean of the value, same formula as average calculation are used for calculation of mean.

From the experiment, the measurements by conducting TNT Hack tests had only small difference in 0.001 between each day's results. Thus, the deviation was always less than 1; by conducting electrode measurements, the difference between measurements, was slightly higher than TNT Hack test, which was in range of 0.1 to 1. In that case, the standard deviation would be extended, but still mostly less than 1.

In measurements of DO and ORP, each reading would have relatively higher difference (1 to 10 mV difference in ORP, 0.1 to 0.5 mg/L difference in DO), This difference was caused by

electrokinetic process, and by taking average, the inaccurate reading would no longer affect the results.

As a result, the difference in each measurements was between 1 to 1.7 mg/L. So the standard deviation was calculated as shown in table 26:

Reading	NH ₃ ⁻ -N in SMEBR	NH3 ⁻ -N in MBR	NH ₃ ⁻ -N in
	effluent mg/L	effluent mg/L	influent mg/L
Standard deviation	0.42	0.80	0.70

Table 26: Result of standard deviation in ammonia nitrogen measurements.

As expected, the standard deviation between electrode measurements were all less than 1.

To know the difference between measurements by TNT HACH method and electrode method, the comparison was always made by conducting both measurement in the same sample. Table A5.2 (Appendix) has presented the measured results in three days. From the results, the difference between TNT HACH measurement and electrode measurement has presented. By calculation, the difference has shown as standard deviation in Table A5.3 (Appendix).

The standard deviation between TNT HACH measurements and electrode measurements has reached as high as 2.75. From the mechanisms of the two types of measurement, these results presented the relatively less accuracy in electrode measurements. However, comparing with previous studies (Ibeid, 2011), this difference was acceptable.

Similar measuring methods had been applied on the measurements of nitrate nitrogen. From the measured result, the measurements done by the same method with different readings were close and with difference of less than 1 mg/L, which was similar to previous discussion about measurement of ammonia nitrogen done by electrode tests. Thus, the calculation of standard deviation to the individual measuring method was meaningless. However, the difference between TNT HACH test and electrode test was also presented in the measurements. Three comparative measurements had been done on stage two, when new electrode had been delivered to the lab. The results have been presented in table A5.4 (Appendix). Based on the results from measurement, the mean value and standard deviation has been calculated and presented in table A5.5 (Appendix).

In conclusion, both TNT HACH and electrode measurement could provide stabilized results in each day's measurement. However, between two types of measurements, the measured result was not same. The highest standard deviation between two measurements had reached 2.75. Although this result was acceptable as a successful experiment output, to minimize the impact from inaccurate reading, multiple measurements had been conducted for all the time.

Chapter V

Conclusion, contribution and future work

5.1 Conclusion

This study confirmed that SMEBR could provide effective nitrogen removal also under the lower C/N ratio from 3:1 to 1:1, which presented various real characteristics of many municipal wastewater. Generally, biological nitrogen removal decreases with decrease of C/N ratio. In spite of such principle, the results showed that SMEBR had better (by 40%) nitrogen removal than MBR due to SMEBR specific redox conditions.

Under the C/N ratio between 3:1 and 1:1, the SMEBR successfully removed TN (up to 50%), and also more than 99% of orthophosphorus under the current density between 7.5 and 10.5 A/m². Moreover, similar to conventional MBR, SMEBR could maintain almost complete COD removal, in spite that the required aeration was as low as 0 mg/L in DO, which was much less than that in the conventional MBR.

The results of this study indicate that, under a low organic carbon input, the regular MBR system cannot process enough nitrogen and phosphorous compounds. However, by using SMEBR, the nitrogen was removed with an average improvement of 35% compared with MBR, and a maximum 54.61% removal was acquired. In addition, the phosphorus removal attained more than 99% because of the electrokinetic processes.

Compared with the conventional MBR system, SMEBR reactor requires lower aeration support because it demands the balance between nitrification and denitrification. In this study,

the average DO in SMEBR and MBR ranged from 0 mg/L to 3 mg/L and 5 mg/L to 8 mg/L, respectively.

Subsequently, the energy required for aeration was conserved. The SMEBR system was supported by the electrical current input that consumes energy; yet, considering the high energy requirement for the aeration in large reactors, the SMEBR might also reduce the energy consumption.

Higher volume of generated sludge was observed in SMEBR compared with MBR. This occurrence was due to the electrokinetic dissolution of the aluminum anode. Although the total sludge amount was increased due to inorganic compounds, the sludge undergone self-thickening process and could be easier settled and dewatered improving the management and decreasing the costs of sludge treatment.

This study did not focus on fouling reduction. Nevertheless, direct observations of membrane modules, and changes of sludge properties, as well as frequency of cleaning process showed the lower fouling in SMEBR comparing to MBR.

5.2 Contribution

This study provided the first assessment of operation conditions for low C/N ratio (3:1, 2:1,

1:1) for simultaneous removal of nitrogen, phosphorous and carbon by electro-bioreactor.

Then, it proved that the SMEBR was a technology capable to replace conventional MBR under low C/N conditions.

5.3 Future work

The results of this study should be applied and further confirmed at full scale wastewater treatment facilities under the real wastewater inflow. Normally, there are many differences between small and large reactors, particularly with respect to control DO supply, current value, and sludge quality. This study showed that controlling current density and DO must be considered in scale up process and must be enhanced in the future.

Future work should include a new solution for better mixing process at the pilot scale, which is other than aeration supporting sludge suspension. It may provide greater results with respect to nitrogen removal under the low aeration condition. Thus, using, other mixing mechanisms, better DO distribution will be provided when treating wastewater with low C/N ratio at low DO.

The effective treatment depends on an adequate control of DO in the SMEBR. Accordingly, a new system of diffusers should be applied at pilot and full scale facilities for uniform distribution of DO.

Chapter VI

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Chapter VII

APPENDIX

A1: Preparation and operational conditions

Stage	Glucose	Glucose	Ammonia	Ammonia	Potassium	Potassium
	input	concentration	Sulfate	Sulfate	phosphate	phosphate
	g/160L	g/L	input	concentration	input	concentration
			g/160L	g/L	g/160L	g/L
1	64	0.4	30	0.1875	6.16	0.0385
2	41.6	0.26	30	0.1875	6.16	0.0385
3	19.2	0.12	30	0.1875	6.16	0.0385

A1.1 Chemical composition of synthetic wastewater

A1.2 Calculation of the average water volume

To calculate the average water volume, conducting the interpolation formula as:

$$\frac{X1}{Y1} = \frac{X2}{Y2}$$

X1 is the average water volume.

Y1 is the measured concentration, in this study, it was 45 mg/L of TN

X2 is the theoretical volume, in this study, it was 160 L of water.

Y2 is the theoretical concentration, in this study, it was 39.9 mg/L.

A2 Calculation of percentage of ammonia nitrogen removal

To calculate the percentage of ammonia nitrogen removal, conducing the formula as:

 $\frac{Inflow \ concentration - Measured \ concentration}{Inflow \ concentration} \times 100\%$

Where inflow concentration is the ammonia nitrogen concentration in the synthetic

wastewater, mg/L NH₃⁻-N;

Measured concentration is the ammonia nitrogen concentration in thec9ollected effluent sample, mg/L NH₃⁻-N;

The result will be in percentage.

A3 Calculation of improved TN removal by SMEBR

The calculation conducted formula as:

$$TN \ removal \ improved = \frac{MBR - SMEBR}{MBR}\%$$

Where MBR denotes the TN removal by conventional MBR system (mg/L N)

SMEBR is the TN removal by SMEBR system (mg/L N)

TN removal improved will be presented in percentage.

A4 Calculation of phosphorus removal and improvement

 For the calculation of phosphorus removal (for example: in SMEBR), conducting the formula as:

Removal by SEMBR
$$\% = (1 - \frac{\text{Removal by SMEBR mg/L}}{\text{Phosphorus input mg/L}})\%$$

Phosphorus input is the influent phosphorus concentration.

The calculated result is in percentage

2. To calculate how much phosphorus removal has been improved by SMEBR, conducting the formula as:

$$Improved \ removal \ \% = \frac{\text{Removal by MBR mg/L} - \text{Removal by SMEBR mg/L}}{\text{Removal by MBR mg/L}} \ \%$$

Removal by MBR is the orthophosphorus removal by conducing conventional MBR;

Removal by SMEBR is the orthophosphorus removal by conducting SMEBR

The calculated result is in percentage.

A5 Calculation of Standard deviation

	NH3 ⁻ -N in	NH3 ⁻ -N in	NH ₃ ⁻ -N in
Reading	SMEBR effluent	MBR effluent	influent
	mg/L	mg/L	mg/L
1	10	20.4	31
2	10.3	18.7	29.3
3	11	20.4	30
Average (mean)	10.43	19.83	30.10
Difference between max & min	1	1.7	1.7

Table A5.1: Ammonia nitrogen remaining on Feb. 8th 2014, stage III

	NH3 ⁻ -N,	NH₃⁻-N,	NH3 ⁻ -N,	NH3 ⁻ -N,	NH3 ⁻ -N,	NH3 ⁻ -N
Reading	SMEBR	SMEBR	MBR	MBR	inflow,	inflow,
	TNT,	electrode,	TNT,	electrode,	TNT,	electrode,
	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
Jan 17th	15.5	16.5	24.9	24.5	-	-
2014						
Jan 23th	13.7	9.13	19.8	21.1	34.5	33.6
2014						
	-	9.56	-	15.7	-	24.4
	average:	9.35	average:	18.4	average:	29
Feb 4th	14.9	12	21.2	15.9	32	27.4
2014						

Table A5.2: Results of ammonia nitrogen measurements

Table A5.3: Results of mean and standard deviation calculation, ammonia-N

SMEBR						
	NH3 ⁻ -N,	NH3 ⁻ -N SMEBR	Average	Standard		
Reading	SMEBR TNT,	electrode, mg/L	(mean),	deviation,		
	mg/L		SMEBR,	SMEBR		
			mg/L			
Jan 17th 2014	15.5	16.5	16	0.5		

Jan 23th 2014	13.7	9.35	11.52	2.1775				
Feb 4th 2014	14.9	12	13.45	1.45				
MBR								
	NH3 ⁻ -N, MBR	NH3 ⁻ -N, MBR	Average	Standard				
Reading	TNT, mg/L	electrode, mg/L	(mean), MBR,	deviation, MBR				
			mg/L					
Jan 17th 2014	24.9	24.5	24.7	0.2				
Jan 23th 2014	19.8	18.4	19.1	0.7				
Feb 4th 2014	21.2	15.9	18.55	2.65				
	Influent							
	NH3 ⁻ -N,	NH3 ⁻ -N inflow,	Average	Standard				
Reading	inflow, TNT,	electrode, mg/L	(mean),	deviation, inflow				
	mg/L		inflow, mg/L					
Jan 23th 2014	34.5	29	31.75	2.75				
Feb 4th 2014	32	27.4	29.7	2.3				

Table A5.4: Nitrate nitrogen removal's measurement

Reading	NO ₃ ⁻ -N,	NO ₃ ⁻ -N, SMEBR	NO ₃ ⁻ -N, MBR	NO ₃ ⁻ -N, MBR
	SMEBR TNT,	electrode, mg/L	TNT, mg/L	electrode, mg/L
	mg/L			
Oct. 22th 2013	3.35	4.24	10	8.86

Oct. 25th 2013	4.53	4.91	9.06	10.3
Oct. 26th 2013	6.09	6.31	11.7	11

Table A5.5: Results of mean and standard deviation calculation, nitrate-N

SMEBR								
Reading	NO ₃ ⁻ -N,	NO ₃ -N SMEBR	Average	Standard				
	SMEBR TNT,	electrode, mg/L	(mean)	deviation				
	mg/L			between two				
				measurements				
Oct. 22th 2013	3.35	4.24	3.795	0.445				
Oct. 25th 2013	4.53	4.91	4.72	0.19				
Oct. 26th 2013	6.09	6.31	6.2	0.11				
	MBR							
Reading	NO ₃ ⁻ -N, MBR	NO ₃ ⁻ -N, MBR	Average	Standard				
	TNT, mg/L	electrode, mg/L	(mean)	deviation				
				between two				
				measurements				
Oct. 22th 2013	10	8.86	9.43	0.57				
Oct. 25th 2013	9.06	10.3	9.68	0.62				
Oct. 26th 2013	11.7	11	11.35	0.35				

A6 Pictures from experiment



Settling after 15 mins

Settling after 30 mins

Settling after 45 mins

Settling after one day

Figure A1: Example of the settling test, Dec. 18th 2014, SMEBR



Figure A2 Experimental system setup