New configuration of submerged electro-bioreactor (SMEBR) for nutrient removal in water recovery

Zohreh Arian

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By: Zohreh Arian

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Dr. S. Li___________________________________ Chair

Dr. S. Ibeid________________________________ Examiner

Dr. L. Tirca________________________________ Examiner

Dr. Z. M. Kabir______________________________Examiner External (to program)

Dr. M. Elektorowicz__________________________ Supervisor

Approved by Dr. F. Haghighat

Graduate Program Director

____

Dean of Faculty of Engineering and Computer Science

Date ____

Abstract

New configuration of submerged electro-bioreactor (SMEBR) for nutrient removal in water recovery

Zohreh Arian

Submerged Membrane Electro-Bioreactor, or SMEBR, is a compact hybrid unit that biological process, membrane filtration and electrokinetic phenomena are carried out for wastewater treatment. In this novel submerged membrane electro-bioreactor, interactions of three fundamental processes -- biodegradation, membrane filtration and electrokinetics -- control the removal of nutrients. This thesis focuses mainly on nutrients (total nitrogen and phosphorus) removal by new design of the electro-bioreactor with multiple electrical units.

Two new configurations of electro-bioreactor were tested upon different technical parameters and operating conditions. In the SMEBR system, relationship between different levels of current density and electrode configuration played an important role for nutrient removal. Based on the results obtained in this study, under the best operating conditions the average removal of ammonia, nitrates, phosphorous and COD reached 98%, almost 100%, 93%, and 97% respectively. Nitrogen was removed by transformation of ammonia nitrogen into the nitrogen gas through nitrification and denitrification process. The SMEBR system could accomplish almost complete nitrification of ammonium and denitrification of nitrate. Phosphorous was removed by the formation of aluminum phosphate. COD was removed through oxidation of biomass and microbial activities in the electro-bioreactor.

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Dedication

This thesis is dedicated to my mother; she has always been for me through ups and downs of my life. It is also dedicated to my father, who taught me that even the largest task can be accomplished if it is done one step at a time. I would like to thank them immensely for their caring and their support. I would also like to dedicate this study to my sister and brother.

Glossary

Abbreviations and symbols	Definitions
Anammox	Anaerobic Ammonium Oxidation
AS	Activated sludge
BOD	Biological Oxygen Demand
C	Carbon
CD	Current Density
COD	Chemical Oxygen Demand
CFV	Cross Flow Velocity
DC	Direct Current
DO	Dissolved Oxygen
EC	Electrical Conductivity
EKDIM	ElectroKinetic Dewatering, Inactivation of pathogens and Metal removal
EPS	Extracellular Polymeric Substances
F/M	Food/Mass ratio
HRT	Hydraulic Retention Time
MBR	Membrane Bio-reactor
MCL	Maximum Concentration Level
MF	Microfiltration

MFI MLSS MLVSS	Membrane Fouling Index Mixed Liquor Suspended Solids (mg/l) Mixed Liquor Volatile Suspended Solids (mg/l)
N	Nitrogen
P PSD	Phosphorus Particle Size Distribution
SMEBR SMP SRT SS SVI	Submerged Membrane Electro-Bioreactor Soluble Microbial Products, Soluble EPS Sludge Retention Time Suspended Solid Sludge Volume Index
TMP	Trans-Membrane Pressure
WWTP	Wastewater Treatment Plants

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Chapter 1: Introduction and research objectives

1.1 Introduction

According to the Canadian Council of Ministers of the Environment (CCME, 2006), three trillion liters of wastewater are discharged annually into the surface water in Canada. Municipal wastewater effluents have several adverse impacts on human health, animal life and ecosystems because of its composition and the total volume discharged. To reduce these impacts it is necessary to remove excess amount of nutrients from water and wastewater (CCME, 2006). Otherwise, the effluents may introduce significant amount of nutrients (nitrogen and phosphorous), pathogens (viruses and bacteria) and decaying organic materials into the water body.

Nutrients are vital for algae, other aquatic plants and biological organisms. Excessive amount of algae, planktons or aquatic plants provoke "eutrophication" of lakes and rivers. Ammonium and nitrate should be removed to avoid producing toxic algal bloom and eutrophication. Nitrogen in a form of urea and proteinaceous matter can be found also in fresh wastewater. As time passes, organic compounds are decomposed by bacteria to ammonia. Fertilizers are known as a source of nitrates, nitrites and phosphate that can be entered into waters. All municipal sewage contains phosphorous. Several form of phosphorus can be found in aqueous solutions, e.g.: orthophosphate, polyphosphate and organic phosphate. Usually phosphorous is found as mono-hydrogen phosphorus (HPO_4^{2-}) in sewage (Davis and Cornwell, 2008). The implementation of effective wastewater treatment technologies presents a major challenge all over the world in terms of having high quality effluent Different biological and physicochemical processes have been used for removing nutrients from sewage: BNR; filtration; stripping, chemical coagulation, etc. One of most common treatment of COD is activated sludge (AS) treatment (Metcalf and Eddy 2003) which is used for both municipal and industrial wastewater. This process was first developed in Great Britain in 1914, and improved over time (Liu and Lipták 2000).

. The main goals for biological treatment of wastewater are: a) Transform species into the acceptable end products, b) Make biological flocs or biofilm by capturing the suspended particles and non-settleable colloidal solids, c) Remove nutrients (nitrogen and phosphorus), and d)

Stabilize the organic compounds.. The following factors make wastewater a complex system for microbial activities: 1) Wastewater has a great variety of microorganisms with a great number of substrates 2) Microbial processes occur under changing aerobic and anaerobic conditions. 3) The microbial activates proceed in different phases such as suspended solids, biofilms, and sludge. 4) The microbial processes in the wastewater are interaction across boundaries [electron donors (NH_3, Fe^{+2}) and electron acceptors (SO_4, NO_3, CO_2 and O_2)] (Akamatsu *et al*. 2010).

For designing a WWTP, other factors (besides producing a high quality effluent) are important: tendency to reduce the consumed energy; costs (which are minimized by decreasing the requirements of aeration in conventional SMER systems); footprint (saving the land needed for construction); and emission of waste materials from different parts of the operational units within WWTP. In some places where advanced treatment is needed, cost plays an important role because wastewater treatment plant requires a higher number of operation units. However, many municipalities cannot afford this very often. Then, it is necessary to develop new approaches to boost the performance of wastewater treatment plants.

Activated sludge process is known for biological treatment of municipal and industrial wastewaters. The basic activated sludge system consists of three parts: 1) one aeration tank in which microorganisms are kept for microbial wastewater treatment; 2) a liquid-solid separation tank; 3) a recycle system that returns activated sludge to the reactor (aerated tank). Importantly, the activated sludge system produces flocculent settleable solids which are removed based on gravity and sedimentation. Several factors affect the activated sludge process, including: wastewater characteristics (such as: alkalinity, volatile suspended solids, carbonaceous, nitrogenous and phosphorus compounds); the food to microorganism ratio (F/M); the value of biological oxygen demand (BOD); the value of chemical oxygen demand (COD); the level of oxygen; the organic loading rate; the returns rate; the wasted activated sludge (WAS); the sludge returning time (SRT); and the hydronic retention time (HRT). According to Metcalf and Eddy (2003), the mass balance analysis of activated sludge process is based on factors including: mixed-liquor suspended solids; secondary influent flow rate; return sludge flow rate; return activated-sludge suspended solids; waste activated-sludge slow rate; and effluent flow rate and effluent suspended solids.

In the activated sludge process, suspending growth of organisms removes biological oxygen demand (BOD) and total suspended solid (TSS). The mixture of raw wastewater and suspended biomass in the reactor is usually called mixed liquor suspended solids (MLSS) (Metcalf and Eddy 2003). During activated sludge processes, flocs are formed; these flocs have bacteria which held together by extracellular polymeric substances (EPS). Large amounts of EPS can be found in mixed liquor of sludge (cell lysis of bacteria and hydrolysis products) derived from bio-treatment of wastewater.

However, conventional secondary (biological) treatment process cannot remove all nutrients in one reactor. In tertiary treatment, nutrients can be removed with carefully designed chemical reactions, but this is a very costly process. Then, advanced treatment technology like biological nutrient removal (BNR) can reduce the amount of nutrients by modifying the suspended growth treatment system, and nitrification/denitrification processes. Phosphorus can be removed by phosphate accumulating organisms (PAOs) in anaerobic and aerobic reaction (EPA, September 2008). In a BNR process, nitrogen can be removed in an anoxic/aerobic condition.

In the last two decades, Membrane Bioreactor Technology (MBR) was implemented for water and wastewater treatment (Wei *et al.* 2012). In the aerobic activated sludge reactor, organic materials are oxidized by the microbial biomass for removing carbon (Ibeid 2011). In recent years, chemical coagulation has been applied for wastewater treatment. This uses materials such as alum and iron salts, or adsorptive materials such as powdered activated carbon and zeolite are used for sewage treatment industry. The addition of chemical coagulants like alum or ferric chloride to the wastewater may cause several effects: they can increase the volume of sludge in the reactor (Aguilar *et al*. 2002), reduction of soluble microbial products (SMP) as a result of alum addition to wastewater (Holbrook *et al*. 2004), and also increase the mean particle size of the mixed liquor, therefore reduce the membrane fouling problem (Song *et al.* 2008).

One of the modern techniques for wastewater treatment is electrocoagulation process (EC), which replaces conventional chemical coagulation (Elektorowicz *et al*. (2009a); and Elektorowicz *et al*. (2011)). Colloidal particles, metals, and soluble inorganic compound are the most common compounds removed by electrocoagulation. The electrocoagulation unit interacts directly with biological processes and membrane filtration (Salamati 2010). Electrical technologies such as electrofiltration, electrophoresis (charging the particles to move to the opposite electrode and aggregation of particles), and electrocoagulation, help reduce membrane fouling and minimize use of chemicals. Electrocoagulation is a useful substitute alternative for conventional chemical coagulation process which used Fe and Al as coagulants. In a new system, Fe and Al are applied as sacrificial anode material to generate coagulants in the reactor (Bani-Melhem and Elektorowicz (2010, 2011); Hasan (2012); and Liu *et al.* (2012)).

In the northern environment of Canada, the electro-bioreactor system is one of the applicable technologies for management of water (*i.e.*: treatment and supply of water) (Elektorowicz *et al.* 2013). Working in remote northern applications in Canada, Wei *et a*l. (2012) show electrical enhancements of the MBR system that improve total nutrient removal and reduce the problem of membrane fouling. They introduced a new technology for their goals: an electrically enhanced membrane bioreactor (EMBR). In conventional treatment plants, removing nutrients (nitrogen and phosphorus) requires several biological reactors working at different conditions. These are costly and require substantial energy consumption and labor (Elektorowicz *et al.* 2011). Alternatively, new submerged membrane electro-bioreactor (SMBER) technology operation can reduce costs by eliminating some units, like clarifier and sludge thickening (Salamati 2010). The SMEBR is a combination of electrokinetic principles and membrane in one bio-reactor. This modern technology was introduced for reducing wastewater's organic and mineral micro-contamination, nutrients, and microorganisms (Elektorowicz *et al.* 2009). This technology reduces membrane fouling, which is considered as a main problem for conventional membrane bioreactor (MBR) (Bani-Melhem and Elektorowicz 2011). This technology can also achieve high quality effluent.

The submerged electro-bioreactor (SMEBR) also resolves the problem of membrane fouling, according to the current researchers (Ibeid *et al.* (2012); Hasan *et al.* (2012); Bani-Melhem and Elektorowicz (2010, 2011); Salamati (2010); Hirzallah and Wei *et al.* (2012)). Previous researchers already studied electrocoagulation, biological transformation of organics and ammonia (Bani-Melhem and Elektorowicz 2011), and characteristics of flocs that influence the fouling problem (Ibeid *et al.* 2013; Hasan 2012). However, their work was done using a single membrane/electrical unit. Although it was proven that the SMEBR system solves many actual problems defined at WWTP, current facilities have not yet adapted the system at full scale. To do so, various configurations of the system should be tasted.

1.2 Wastewater regulation and limitation

Municipal sewage usually has adequate levels of nutrients (organically bound) to support biological treatment for the removal of unwanted materials. The level of nitrogen and phosphorus are important when wastewater is treated by biological processes. Municipal untreated wastewaters contain about 20 to 70 mg/l N as nitrogen and 4 to 12 mg/l P as phosphorous (Metcalf and Eddy 2003).

Based on EPA (2013), the maximum concentration level (MCL) in wastewater for nitrate is 10 mg-N/L as nitrogen; MCL for nitrite is 1 mg-N/L as nitrogen in drinking water. In municipal wastewater, the total concentration of organic and ammonia as nitrogen is between 25 to 45 mg/l, based on a flow rate of 450 L/capita.d (EPA, 2013). Typically, limits are based on toxicity to aquatic life. Provincial regulations vary across Canada. Some provinces, like Ontario, Quebec, and British Columbia, base their limits on the *Environmental Quality Act.* This act enacted in 1972, focused mainly on water and air pollution, hazardous waste disposal, and contaminated soil.

The water policy in Quebec aims to protect water quality and aquatic life. Concentration of phosphorous in effluents produced by different wastewater treatment process should be between 0.1 to 1 mg TP/L. This range depends on the: wastewater treatment methodology, the location of lakes or rivers (some lakes must be protected), the volume of treated effluents discharged into the rivers or lakes (Développement durable, environment et lutte contre les changements climatiques, 2014).

1.3 Motivation

In order to build and set up a large scale of submerged membrane electro-bioreactor (SMEBR) system for wastewater treatment plants, multiple electrical units and membranes should be applied. Therefore, the SMEBR should undergo the scale up process in several steps, while volume of the reactor is change, number of membrane modules and electrical units increase. Thus, there is a necessity to investigate the SMEBR’s performance under different design configurations with multiple units.

1.4 Research objective

The main objective of this thesis is related to investigate reduction of the nutrients (nitrogen and phosphorus) from wastewater by using new designs of membrane electro-bioreactor (SMEBR) with multiple electrical units for water recovery. The new configurations should be tested in the presence of different technical parameters and operating conditions (HRT; SRT; dissolved oxygen concentration; current density; reactor dimensions; and etc.) in order to generate information for scale up the system.

1.5 Thesis layout

This thesis consists of seven chapters. The topic of each chapter is as follows:

Chapter One covers the statement of problems and objectives, and introduces the submerged membrane electro-bioreactor (SMEBR).

Chapter Two provides background and a review of the relevant research works.

Chapter Three describes an experimental set up performed in the laboratory.

Chapter Four demonstrates the results obtained from different experimental tasks in the laboratory.

Chapter Five shows conclusions of this study within the context of the research, and discusses the potential of nutrient removal by redesign submerged membrane electro-bioreactor. It also contains contributions and recommendations for future research.

Chapter Six lists the references used in this study.

Chapter Seven consists of appendices.

Chapter 2: Literature reviews

As it was mentioned in Introduction and Objectives, this study focuses on the application of submerged membrane electro-bioreactor (working based on activated sludge processes, membrane filtration and electrokinetic phenomena) to nutrients and COD removal. The main nutrients in wastewater are nitrogen and phosphorous. There are different methods for removing nutrients from sewages. Therefore, this section is about the general information and overview of conventional methods for removing nutrients (N and P). Typical values of untreated domestic wastewater are shown in Table 2.1.

Table 2.1: Typical values of domestic wastewater (Metcalf and Eddy 2003)

Contaminants	Concentration (mg/l)		
	Low strength	Medium strength	High strength
Nitrogen as N	20	40	70
Organic	8	15	25
Free ammonia	12	25	45
Phosphorus as P	4	7	12
Organic	1	2	4
Inorganic	3	5	10

2.1 Methods for phosphorus removal

Current methods for phosphorus removal include: chemical phosphorous removal (using trivalent metal ions, and lime addition); physicochemical adsorption; biological assimilation; and enhanced biological phosphorus removal (EBPR).

2.1.1 Chemical phosphorus removal

Phosphorus can be removed by precipitation processes. Orthophosphates are removed by trivalent metal cations, usually with ferric ions (Fe^{3+}) in the form of ferric chloride, or with aluminum ions (Al^{3+}) in the form of aluminium sulfate (alum). The precipitation reactions depend on the variability of phosphorus species which convert to PO_4^{3-} .

Through this process, alkalinity (hydroxide ion) is consumed; therefore, for this reaction, sufficient alkalinity should be present in the solute. Several reactions occur for chemical phosphorus removal (EPA, September 2008) (Eqs. 2.1 to 2.6):

$$H_nPO_4^{(3-n)} + n\,OH^- \rightarrow PO_4^{3-} + n\,H_2O \qquad (2.1)$$

Phosphorus removal using alum:

$$Al_2\,(SO_4)^3 \rightarrow 2\,Al^{3+} + 3\,(SO_4^{2-}) \qquad (2.2)$$

$$Al^{3+} + PO_4^{3-} \rightarrow AlPO_4\,(s) \qquad (2.3)$$

Phosphorus removal using ferric chloride:

$$FeCl_3 \rightarrow Fe^{3+} + 3\,Cl^- \qquad (2.4)$$

$$Fe^{3+} + PO_4^{3-} \rightarrow FePO_4\,(s) \qquad (2.5)$$

Ferrous ions converted to ferric ions:

$$2Fe^{2+} + \tfrac{1}{2}\,O_2 + 2H^+ \rightarrow 2Fe^{3+} + H_2O \qquad (2.6)$$

Chemical addition requires high costs of treatment and increases the presence of Al and Fe in bio-solids.

2.1.2 Biological phosphorus removal

Biological phosphorus removal carried out by two reactions (Pattarkine and Randall 1999):

1- Anaerobic reaction: phosphorus can be released by phosphate accumulating organisms (PAOs):

PAOs + stored polyphosphate + Mg^{+2} + K^+ +VFS → PAOs + stored biopolymers + Mg^{+2} + K^+ + CO_2 + H_2O + PO_4^{-3} (Released) (2.7)

2- Aerobic reaction: this reaction is done by PAOs:

PAOs + stored biopolymers + Mg $^{+2}$ + K^+ + PO_4^{-3} + O_2 (or NO_3)→ PAOs + stored polyphosphate + Mg $^{+2}$ + K^+ + CO_2 + H_2O + glycogen (2.8)

PAOs play an important role for biological phosphorus removal, which is carried out first in anaerobic condition and then by aerobic conditions. Sufficient amount of volatile fatty acids (VFAs) are needed for biologically removing phosphorus. In an anaerobic case, microorganisms break the high energy bonds of polyphosphate. Organic matter is consumed as VFAs or other biodegradable organic matter. The result of this reaction is phosphate. In an aerobic condition, microorganisms use phosphate in making the internal polyphosphate molecules. Thus, the concentration of total phosphate in the solute is decreased. Beyond this reaction, microbes also can remove phosphate as part of their BOD removal. In the conventional activated sludge system, just a small amount of phosphorus is removed (1.5 to 2 percent on a dry weight basis). But by using the PAOs in anaerobic zone followed by aerobic zone, phosphorus removal is 2.5 to 4 percent higher than it is in conventional methods for activated sludge processes (EPA, September 2008).

2.1.3 Phosphorus removal technology

2.1.3.1 Enhanced biological phosphorus removal technology (EBPR)

There have been many technologies for phosphorus removal. Activated sludge system is designed for biological phosphorus removal; the process is known as an enhanced biological phosphorus removal (EBPR). Yet, more studies still are needed for the ecology of EBPR and microbial population dynamics of EBPR (Sathasivan 2009; Seviour *et al.* 2003). EBPR is a technology which has a potential to get low levels of phosphorus (even less than 0.1 mg/l), it is more cost effective and also it has less sludge. Mino *et al.* (1998) made a detailed review of this process.

Fundamentally, anaerobic-aerobic conditions provided for bacteria are the main requirement for the EBPR process (Seviour et al. 2003). Large amount of phosphorus was found in extracellular polymeric substances (EPS). They can act as a reservoir for phosphorus in the anaerobic/ aerobic conditions. Zhang *et al*. (2013) found that in an aerobic phase, 5 to 9% of sludge's phosphorus was remained in the EPS. Also, an important amount of long chain of intracellular polyphosphate was reduced in an anaerobic phase (Zhang *et al*. 2013).

In EBPR process, phosphate accumulating organisms (PAOs) store polyphosphate as energy in intracellular granules. In the presence of fermentation compounds and under anaerobic conditions, PAOs can release other forms of phosphate (i.e. orthophosphate). PAOs can use energy for accumulation of simple organic compounds and store them as a polyhydroxy alkanoates (PHAs) (Strom 2006).

The phosphate can be removed during EBPR process from the waste activated sludge. This methodology has been applied in several systems such as: bardenpho process (this process is also used for removing nitrogen), anaerobic/oxic or poredox (A/O), and A/A/O or A_2O (this process is also used for removing nitrogen), sequence batch reactors (SBRs), and the PhoStrip process (a combination of EBPR with phosphate stripping and chemical removal) (Strom 2006).

In the A/O process, anaerobic zone is followed by aerobic zone. RAS and influent enters into the anaerobic zone. The operational problem of A/O system is related to the inhabitation of anaerobic growth of PAOs due to the recycle of nitrate in the aerobic zone. To improve this problem, anaerobic zone is split into an anoxic zone for nitrate denitrificatin process, and an anaerobic zone for phosphorus removal.

2.1.3.2 Physical-chemical technologies

- Membrane filtration technologies

MBRs used activated sludge biological processes however secondary clarifier is substitute by membrane module. By this process, complete solids-liquid separation is carried out. Phosphate is remained in the reactor as polyphosphate. To get low effluent phosphorus, sometimes, chemical compounds are needed to add to the wastewater, they should be added before the membrane. A MBR was used in the Utah's WWTP, the annual effluent concentration of phosphorus was 0.07 mg/l. Another example was related to the Lone Tree Creek, Colorado, the annual concentration of phosphorus's effluent was 0.027 mg/l (EPA, September 2008).

- Chemical addition

Aluminium sulfate (alum) or iron salts (ferric chloride) are usually added in to the secondary clarifier or tertiary filters. The drawbacks of adding chemical is related to the production of excess amount of sludge, cost of chemical compounds, and O and M cost of process.

- EBPR with filtration technology

Enhanced biological phosphorus removal with filtration technologies is used for phosphorus removal in wastewater treatment plants. In this technology, chemical compounds are added to the system, and/or filtration process is used (EPA, September 2008). Drury *et al.* (2005) reported that, after chemical polishing of EBPR in WWTP of Nevada, the final concentration of phosphorus was lower than 0.1 mg/l. For this process, 10 mg/l alum was added into the system for prevention of filter clogging. With chemical polishing and filtration in Durham, the final concentration of phosphorus after removal was lower than 0.07 mg/l (EPA, September 2008). However, both biological and chemical removal of phosphorous requires building additional facilities and/or supplying additives.

2.2 Nitrogen removal technology

Total nitrogen includes nitrate, nitrite, ammonia and organic nitrogen. Physical-chemical technologies for nitrogen removal include stripping, ion exchange and oxidation. The biological methods for removing nitrogen are relying on nitrification, denitrification and anammox processes. For biological treatment of wastewater, type and number of organisms are important. Nitrification and denitrification are two important reactions in reactors that deal with ammonia and nitrate removal.

2.2.1 Biological nitrogen removal (BNR)

Nitrogen can be removed biologically in an anoxic/aerobic process through nitrification and denitrification.

2.2.1.1 Biological nitrification

Biological nitrification is a two-step process in which ammonia (NH_4 - N) is oxidized to nitrite (NO^-_2 -N) and then nitrite is oxidized to nitrate (NO_3^--N). Nitrification has a major influence on the quality of water due to the: a) fish toxicity due to the presence of ammonia, and b) eutrophication (Metcalf and Eddy 2003). Nitrification requires the presence of oxygen, and an alkaline environment to neutralize the resulting acids (electron donor and electron acceptor).

Nitrification process description

It should be noted that the two steps of this biological process are carried out by two different groups of autotrophic bacteria: *Nitrosomonas* and *Nitrobacter.* They can oxidize ammonia to nitrite and then to nitrate. Dytczak *et al.* (2008) studied activated sludge operational regime and nitrification rates. They staged sequence batch reactor under two conditions: alternating anoxic/aerobic conditions. They found that rapid nitrifiers bacteria like *Nitrosomonas* and *Nitrobacter* had a population around 79.5% of nitrifying bacteria in the alternating reactor. By comparison, slower nitrifiers like *Nitrosospira* and *Nitrospira* had occupied only 78.2% in the aerobic reactor. Nitrifiers in the aerobic reactor were negatively affected by ammonium or nitrate, while nitrifiers in alternating anoxic reactor were commensurable with the amount of ammonium or nitrate. The alternating anoxic reactor was more practical because the oxidation, growth, and decay rates of *Nitrosomonas* and *Nitrobacter* were faster than *Nitrosospira* and *Nitrospira* (Dytczak *et al.* 2008). Ammonia exists in natural waters due to the direct discharge of the pollutants or the decomposition of organic matter in various forms. Ammonia is oxidized to nitrite by *nitroso-bacteria* under aerobic condition, as the following reaction (Metcalf and Eddy 2003).

$$2NH_4 + 3O_2 \rightarrow 4H^+ + 2H_2O + 2NO_2^- \qquad (2.9)$$

The nitrite is then oxidized by *nitro-bacteria* to form nitrate, as the following reaction:

$$2NO_2^- + O_2 \rightarrow 2NO_3^- \qquad (2.10)$$

Total oxidation reaction:

$$NH_4^+ + 2O_2 \rightarrow 2H^+ + H_2O + NO_3^- \qquad (2.11)$$

Based on the above reaction (Eq. 2.11), the oxygen required for oxidation is 4.57 g O_2/g N for ammonia, 3.43 g O_2/g NO for nitrite, and 1.14 g O_2/g NO_2 for nitrate. In a biological nitrification process, important factors include: concentration of nitrogen, COD, alkalinity, temperature, and the presence of toxic compounds in wastewater. For nitrifier bacteria growth, CO_2 and phosphorus are required, as other trace elements: Ca = 0.50 mg/l, Cu = 0.01 mg/l, Mg = 0.03mg/l, Mo = 0.001 mg/l, Ni = 0.10 mg/l, Zn = 1.0 mg/l (Metcalf and Eddy 2003). Gao *et al.* (2004) observed that by using submerged membrane bioreactor (SMBR) system, the removal rate for ammonium was 99% at a influent NH_4^+- N from 180 mg/l to 1300 mg/l (the nitrifying bacteria were dominant into the reactor), hydraulic retention time (HRT) of 24 h, and MLSS varied from 3000-5000mg/l. They also showed that the number of ammonia oxidizer and nitrite oxidizers increased during their experiments.

2.2.1.2 Denitrification process

During denitrification, nitrate is converted to nitrite, then to nitric oxide, to nitrous oxide, and to nitrogen gas. For biological nitrogen removal, this denitrification process is very important. For nitrate reduction, an electron donor is needed, which can be supplied from carbon in the influent of wastewater (BOD), by internal respiration, or through an external source of carbon.

Denitrification process description

In biological nitrogen removal, two factors are important, which are assimilating and dissimilating nitrate reduction (Fig. 2.1).

Biological denitrification is summarized in Eq. 2.12 (Metcalf and Eddy 2003). Briefly: aerobic oxidation only needs organic compounds and a mere few days as a solid retention time (SRT). To achieve better nutrient removal, bacteria should grow in sufficient numbers. Nitrifying bacteria need 10 to 20 days for nitrification (Metcalf and Eddy 2003).

$$NO3- \rightarrow NO2- \rightarrow NO \rightarrow N2O \rightarrow N2 \qquad (2.12)$$

Barr et al. (1996) studied about the effects of HRT, SRT and treatment on the performance of activated sludge reactors. They run the reactor at 35°C in steady state condition for a period of two months with HRT of 10–12 hours and SRT of 12–15 days. During this time, the average level of removal for BOD, COD and toxicity compounds was 87.9%, 32.4%, and 97.7%, respectively (Barr et al. 1996).

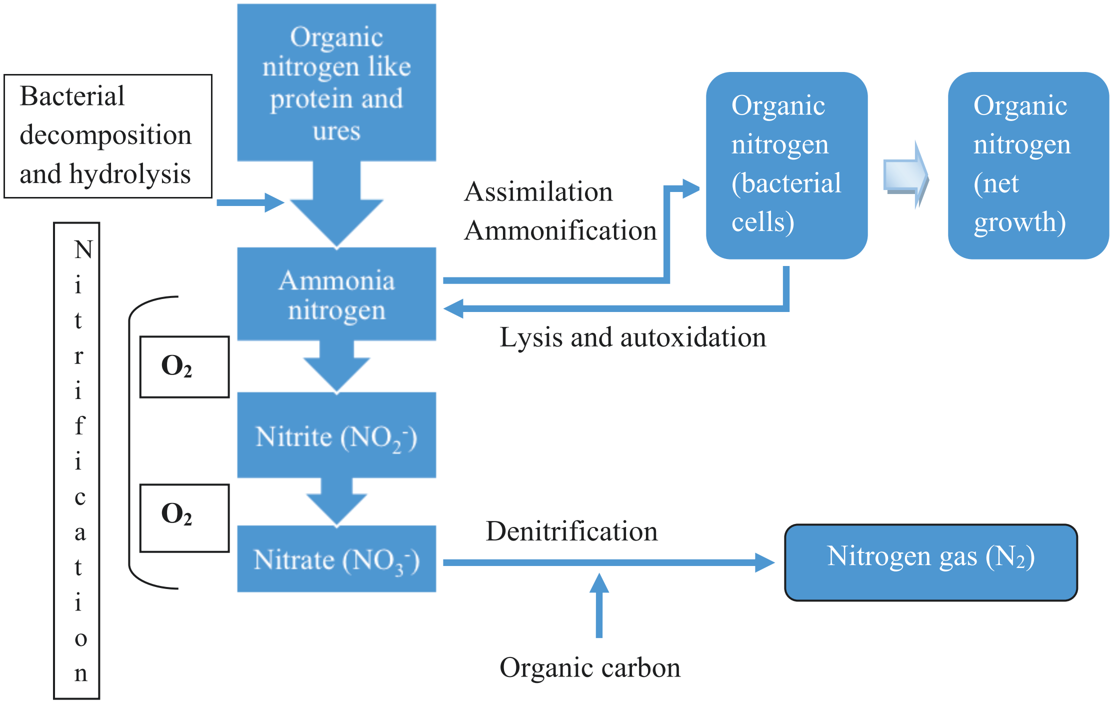

Fig. 2.1: Schematic illustration of the various forms of nitrogen in sewage and their reactions in the activated sludge process (after Metcalf and Eddy 2003)

Temperature plays an important role in the nitrification process of the activated sludge reactor. Kos *et al*. (1998) showed that during the winter, SRT values must be 7 to 10 days instead of 13 to 18 days for conventional nitrification process. Therefore, the aeration tank must be smaller than in a conventional nitrification. Sears *et al.* (2003) found a relationship between SRT and nitrification process in aerobic activated sludge. In their experiments, the nitrification was carried out in a minimum SRT of 12 days.

2.2.2 Anaerobic ammonium oxidation (Anammox)

For nitrogen removal in conventional wastewater treatment technology, lots of energy is needed to create the aerobic condition for nitrifier bacteria (nitrification). Organic carbon is used for removing nitrate by denitrifier bacteria (denitrification).

Another biological process for nitrogen removal is anammox (anaerobic ammonium oxidation) by using anaerobic ammonium oxidizing bacteria, which need less energy but grow slowly (Kartal *et al.* 2010). Ammonium can be removed via Anammox process in a condition of high concentration of ammonia. *Anammox* bacteria cannot use oxygen for ammonia oxidation;

instead, they use nitrite as an electron acceptor. The optimum temperature for growing anammox bacteria is 30 to 40° C. This process also has been seen at 20°C (Metcalf and Eddy 2003). Dosta, 2008 indicated that anammox bacteria can work successfully at 18°C, while at 15°C activity of bacteria is reduced. The bacteria which are activated in anammox process are different than the autrotrophic nitrifying bacteria and they cannot grow in the pure culture. Anammox bacteria can oxidize ammonia into the nitrogen gas 6 to 10 times faster than *Nitrosomonas Europaea (N. Europaea)* bacteria under anaerobic conditions (Metcalf and Eddy, 2003). The disadvantage of anammox process is related to the slow start-up period for anammox bacteria (10 to 12 days at 35° C) (Kartal *et al.* 2010). Kartal *et al.* (2010) explored processes to speed up the anammox process; to achieve this goal they used all organic matter to produce more energy.

In the anommox process, ammonium and nitrite are converted to the nitrogen gas thanks to the inorganic carbon source (CO_2). This bacteria use CO_2 as their carbon source, therefore they do not need organic carbon for their growth (Kartal *et al.* 2010). The following reactions show the anammox (Eq. 2. 13) and nitrification processes (Eq. 2. 14).

$$NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O \qquad (2.13)$$

$$NH_4^+ + 0.5\ O_2 \rightarrow NO_2^- + 2H^+ + H_2O \qquad (2.14)$$

Together:

$$2NH_4^+ + 0.5\ O_2 \rightarrow N_2 + 2H^+ + 3H_2O \qquad (2.15)$$

In the conventional activated sludge process, organic matter is converted to carbon dioxide by microorganisms which grow in the flocs. This process needs lots of electrical energy for providing oxygen. Also in the final step of denitrification process, more organic matter is needed for converting nitrate into nitrogen gas.

In marine environments, the anammox process causes at least 50 percent of the nitrogen turnover to other products. The common nitrogen removal process is illustrated by the following figure (Fig. 2.2).

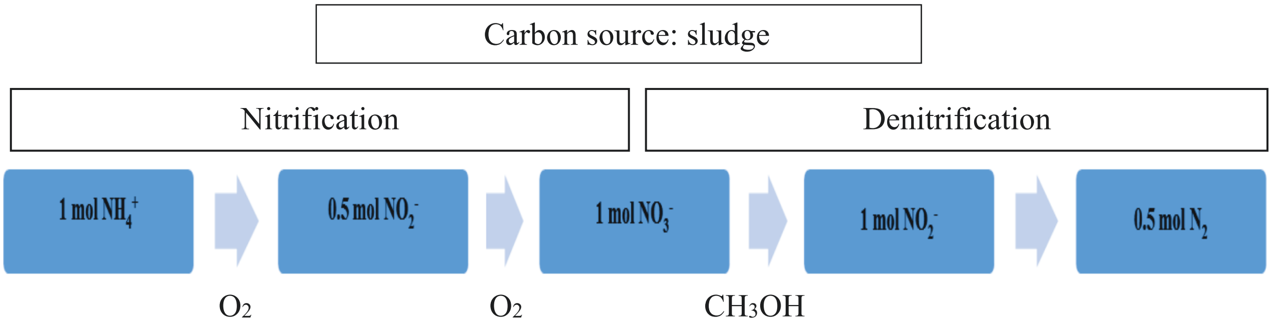

Fig. 2.2: Conventional nitrogen removal cycle

When ammonium (NH_4^+) is released into the environment, it might be oxidized by nitrifier and archaea bacteria to nitrite (NO_2^-) and nitrate (NO_3^-) respectively. In anaerobic condition (denitrification), nitrate and nitrite may be converted again to ammonium or nitrogen gas (N_2). Nitrite can have a reaction with ammonium to produce nitrogen gas in the anammox reaction (Fig. 2.3).

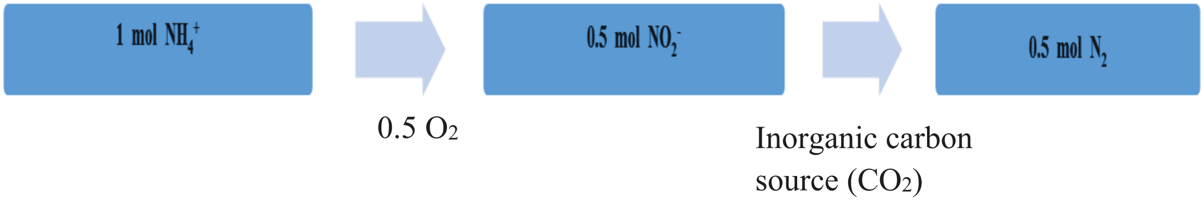

Fig. 2.3: Anammox process

In an anaerobic treatment process, most of the organic compounds are converted to the biogas. Therefore, the organic compounds may not be available for the denitrification process. In this situation, we have large amounts of ammonium in the system plus the possibility of a nitrogen pollution problem. The anommox process can solve this problem, because the anammox process does not need organic compounds for removing nitrogen (Kartal et al. 2010). It seems that Anammox bacteria are suitable to be present in electro-bioreactor system due to the possibility of alternative aerobic/ anaerobic conditions in a reactor.

2.3 Processes of membrane filtration

2.3.1 Type of processes

Study of membrane characteristics is based on both physical and chemical factors. Physical factors include porosity, pore size and membrane configuration. Chemical factors include hydrophobicity, divalent ions (Ca^{2+}, Mg^{2+}), ionic strength and pH (Hai and Yamamoto (2011); Hasan (2012)). Filtration is the mechanical or physical system which separates solids from liquids or gases by a thin layer or barrier. Fluids can pass through the filter because of the difference between pressures. Liquids or gases move from the high pressure side to the low pressure side of the filter. Based on particle size, the membrane filtration is classified into four cases: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). Microfiltration is usually applied to eliminate particles 0.1 to 10 μm in size. Ultrafiltration can separate large particles like protein, bacteria and viruses that range in size from 0.01 to 0.1 μm. Nanofiltration is applied to remove small particles and herbicides that range in size from 0.001 to 0.01 μm. Reverse osmosis can separate the smallest molecules or particles, which are solute in wastewater even in a size of 0.0001μm (Hasan 2012).

2.3.2 Membrane modules

Membranes are usually made up from inorganic materials (ceramic) or organic materials (polyethylene, polypropylene and polyvinylidene fluoride). Of these, inorganic membranes work properly for water and wastewater treatment in terms of thermal stability and mechanism, but they are more expensive than organic ones. On the other hand, organic membranes are more appropriate for water and sewage treatment due to their flexibility and high surface area. They can be used as a plate and frame membranes, hollow fiber membranes, tubular membranes and so on (Hasan 2012).

Plate-and-frame membranes include two membrane material flat sheets, usually with a thin layer of organic polymer stretched across it. The vacuum creates a motivation force, which is placed between the spaces of two membrane sheets. This membrane filters compounds from outside to the inside. For this kind of membrane, several plates are submerged in the mixed liquor (Hasan 2012).

Hollow fibers are like several long strands, and are usually made up from organic materials. This fiber has a specific structure which serves as an air delivery system and permeates the transportation system. These functions cause the lifetime of the membrane to increase; there is also the possibility of cake layer decrease. The driving force is based on the vacuum. This kind of membrane filters compounds from the outside to the inside (Hasan 2012).

Tubular membranes are made from inorganic compounds like ceramic. The membrane is placed on the surface area of tubes. Under the membrane area there are several porous for supporting the structure of tubular membranes. For tubular membrane the, motive force is not based on vacuum. Materials can be separated under high velocity and pressure. Thanks to this force the water can pass through the membrane while the other particles are rejected. This kind of membrane filters particles either inside to outside or in a converse way (Hasan 2012).

Spiral-wound is used for the reverse osmosis (RO) system. A small laboratory scale spiral-wound membrane includes a single membrane covered with a collection tube. This membrane is placed inside a tubular pressure vessel. The surface area of this module ranges from 0.2 to 1.0 m^2. This type of membrane is widely used for removing calcium and other divalent ions from hard water (Baker 2012).

FiberPlate ™ membrane was recently used by Anaergia Inc.; it is a *hybrid-membrane* technology which is used in membrane bioreactor facilities in Victor Valley Wastewater Reclamation Authority (VVWRA). In other words, FiberPlate ™ is a new generations of membrane technology. It is a combination of flat and hollow fiber technology. In the MBR, the FiberPlate ™ will provide the ultrafiltration barrier. This ultrafiltration barrier has several advantages in MBR such as: low transmembrane pressure, high packing density, and high backwashing capacity, easy operation condition of flat sheet membranes, and reduced infrastructure and operational costs. The FiberPlate ™ membrane is designed by Fibracast Inc., who is a member of the Anaergia group (Anaergia Inc. 2014).

2.3.3 Operating condition of membrane bioreactor

For designing and operating of the membrane bioreactor, important elements include: configuration of reactor, cross flow velocity (CFV), aeration, HRT, SRT, TMP, unsteady state operation, and organic loading. Liu *et al.* (2000) reported that CFV and aeration can reduce membrane fouling. They found that at CFV values lower than 0.3 m/s, TMP sharply increased. Therefore suspended solids were deposited on the surface of the membrane. Aeration could affect the biological and physical characteristics (shape and size) of the sludge. Hai and Yamamoto, (2011) found that SRT, and thereby the food per microorganisms ratio (F/M ratio), had a great influence on membrane fouling by controlling the biomass characteristics. Since the mid-1990s, further improvements in the membrane bioreactor have been made, such as controlling fouling by using two phase bubbly flow. Before 1990s, MBRs were operated at very long SRTs (100 days) with a high MLSS concentration (up to 30 g/l), while after the mid-1990s, MBRs were operated with SRTs of 10-20 days and MLSS of 10-15 g/l. Due to the controlling of the fouling the membrane cleaning process has been simplified (Hai and Yamamoto 2011). Increase the SRT might causes increase the MLSS and biodegradation of compounds, while producing less sludge. But in this circumstance viscosity of suspended solids might be increased (Hasan 2012).

Chang and Lee (1998) found that when SRT was decreased the tendency of membrane fouling was increased. Unsteady state operation brings a variation in flow input, HRT, and organic loading rates (Hai and Yamamoto 2011). If the HRT increased, the organic loading and MLSS concentration increased. Thus, the possibility of membrane fouling increased; consequently transmembrane pressure (TMP) value increased (Hasan 2012).

2.3.4 Process description in bioreactor

In biological reactor, microorganisms can be degraded by activated sludge processes, consequently membrane filtration process is used for direct separation of solid/liquid compounds. Membrane ultrafiltration is widely used for wastewater treatment with pore size ranging from 0.04 to 0.4 µm. Until now, different configuration of membrane system is designed. The most common ones are internally (submerged) and side-stream units. For submerged membrane, mostly hollow fiber or plate and frame membranes are used. In the submerged membrane system, the separation unit (membrane) is located inside the vessel where particles moves towards the membrane under a vacuum pressure (Hasan 2012).

The advantages of the submerged membrane include: small footprint, better control of oxygen demand, and 28% reduction of the cost of operation and liquid pumping (Gender *et al.* 2000 and Hasan 2012). Therefore, the operation cost and energy usage are reduced .The energy consumption rates for the internal membrane are around 0.2 - 0.4 kWh/m^3) (Chua *et al.* 2002). The main disadvantages of MBRs are related to the fouling problem and high aeration cost (Hasan 2012).

In contrast, in the side-stream membrane, the membrane is installed outside the unit. In this configuration particles are passed through the membrane to the outside and sludge is returned again to the system by the high flow rate through tubular membrane. Because of this, more power is required for side-stream membrane. Also the energy cost increased from 2 - 10 kWh/m^3 based on internal diameters of the tube used (Côté *et al*. 1997; Hasan, 2012). Chua *et al*. (2002) found that in side-stream membranes, 60 % to 80% of total costs are spent for pumping and operation. But in spite of these disadvantages, side-stream membranes have some advantages: almost complete removal of COD and nutrient from a single unit; small footprint; high loading rate capacity; low sludge producing and no sludge bulking.

2.3.5 Membrane fouling

One of the restrictive factors when using MBRs in wastewater treatment is that membrane fouling occurs (Meng *et al.* 2007). Three fouling phenomena are summarized below: 1) Cake formation, which is related to the filtration force and biomass characteristics. 2) Blockage of strands of fiber, which works like deep bed filter, blockage the filter depends on biomass characteristics and configuration of filter, and 3) Biofilm formation (Hai and Yamamoto 2011). Carbohydrates and proteins are very small in size; therefore they deposit more rapidly than microbial flocs and colloids on the surface of membrane. Membrane fouling is affected by the retention, deposition or accumulation of suspended solids or colloidal compounds into the surface area of the membranes or membrane's pores (Hernandez *et al*. 2005). In membrane fouling, permeate flux decreases, then trans-membrane pressure (TMP) increases; therefore, the final effluents do not have good quality (Guo *et al.* 2008; Hasan *et al.* 2012). According to the previous study (Arabi *et al.* 2008), MLSS influenced filterability, but did not directly affect the membrane fouling. In MBR, the following elements influence membrane fouling: membrane material; membrane configuration; F/M ratio (Food to Microorganisms ratio); Supernatant Chemical Oxygen Demand (COD_{cr}); Extracellular Polymeric Substance (EPS) (Hernandez *et al.* 2005); size of particles and viscosity; sludge characteristics; and also operating conditions (indirect impact). Akamatsu *et al.* (2010) developed novel fouling suppression system (electro-ultrafiltration) in membrane bioreactors system by electric field. They found that the surface of the activated sludge was charged negatively. In other words, it might be possible to regulate the motion of activated sludge by using an external electric repulsive force. By this action, sludge moves away from the membrane when the permeate flux is reduced due to the membrane fouling. Moreover, Akamatsu *et al.* (2010) found that membrane fouling significantly improved by switching the electric field of 6 V cm $^{-1,}$ ON/OFF every 90 seconds. This electric field was expected to reduce cake layers around the membrane and charged macromolecules like extracellular polymeric substances (EPS). Applying an excessive amount of air in tank also plays an important role for avoiding membrane fouling not only by providing oxygen for the sludge producing bacteria, but also by shaking the submerged membrane mechanically to remove any sludge stuck to the membrane surface (Akamatsu *et al*. 2010). Cake layer may be removed by using air scouring and backwashing or using chemical cleaner, like combination of hydrochloric acid, water and bleach (Hai and Yamamoto 2011; Ibeid 2011).

In the full scale operation, fouling will be controlled by applying the following methods: 1) pre-treating (by adding acid due to the formation of calcium carbonates), 2) decreasing the permeate flux, 3) increasing the aeration, 4) using backwashing technology, 5) changing the MLSS characteristics, 6) using chemical and physical cleaner for membranes, 7) chemical modification, and 8) operating optimization (Hasan 2012; Wei *et al*. 2012 ; Guo *et al*. 2008). Backwashing is one of the physical methods for cleaning the membrane of the deposited foulants and cake layer. In this process, filtration is stopped for a specific time, and then a part of the permeate water is pumped in the reverse direction. A combination of air sparging and backwashing can provide 4 times better cleaning than the conventional air sparging for hollow-fiber modules (even in the low air velocity of 0.08 m/s), and it also can reduce the backwashing time (Guigui *et al.* 2003).

Moreover, when membrane and organic compounds or colloids in solute have the same charge, they repulse each other because of the electrostatic force. Therefore, the possibility of fouling problem is reduced. Most of the colloids and organic compounds in solute have negative charges. If solutes have low ionic strength (proteins), the membranes will adsorb less colloids and organic compounds. In this circumstance, protein molecules are repelled by the membrane.

2.3.6 Sludge characteristics

Several parameters effects on sludge characteristics, including: Extracellular Polymeric Substance (EPS); soluble microbial products (SMP; feed characteristics; biomass characteristics (MLSS concentration, viscosity, temperature, dissolved oxygen); flocs characteristics (flocs size and dewaterability/surface charge), and settelability of biomass. EPS is an insoluble compound which is polymerized by microorganisms, while SMP is a soluble and large molecule which is produced by cell metabolism (Hai and Yamamoto 2011). The common aspect between EPS and SMP is that both produce colloidal particles which settle down at the surface of membrane. EPS are comprised of carbohydrate, protein, humic substances, small amount of uranic acid and DNA. Heavy metals (including: Pb, Ni, Cd, and etc.) and organic materials can be removed by EPS in the neutral pH because they have large groups of molecules that possess both adsorptive and adhesive characteristics (Chang and Lee 1998). EPS can create a barrier against permeate flow in the membrane process and interact with the hydrophobic phenomenon among microorganisms' cells (Hasan 2012).

Liu and Fang (2003) reported that, if EPS level decreased, possibility of flocs formation decreased. Then, dissolved oxygen plays an important role in electro- bioreactor technology. It not only can provide oxygen for microorganism's activities but also it can reduce the possibility of clogging on the membrane surface. DO concentration has an influence on changes in the biofilm structure, flocs size division, and SMP level. Increasing the concentration of MLSS mostly has negative effects on the MBR hydraulic conditions. The importance of MLSS viscosity is related to the modification of the bubble size and reducing the movement of submerged membrane hollow fiber. Therefore, MLSS viscosity can increase the risk of clogging by decreasing the movement of particles in the hollow fiber in the submerged form (Wicaksana *et al.* 2006; Hai and Yamamoto 2011). Also, increasing the viscosity in the suspension caused DO reduction. Fouling becomes worst at low DO (Germain and Stephenson 2005; Hai and Yamamoto 2011).

Particle size is distributed based on the average number or average size of particles in the sludge suspension. Colloids and biological flocs cause fouling as they are released from EPS compounds into the reactor. Besides, if particle size decreased in the sludge suspension, it would cause much more disposition on membrane and therefore lower permeate flux (Hasan 2012) and it would also produce more irregular flocs shape and higher hydrophobicity (Hai and Yamamoto, 2011). Hydrophobicity of the membrane can be improved by the adsorption of organic compounds and colloids (Hasan 2012). Optimally, dissolved oxygen would reduce the chance of cake layer formation, with producing the large particle sizes, and greater porosity (Hai and Yamamoto 2011; Ibeid 2011).

Elektorowicz and Oleszkiewicz (2009) used electrokinetic technology for sludge dewatering, inactivation of pathogen and metal removal (EKDIM). After a while, sludge is dewatered until 99% and metals compounds are trapped (from 80 to 100%). This system also claimed simultaneous disinfection of sludge (Elektorowicz and Oleszkiewicz 2009).

2.4 Electrokinetics/ Electrocoagulation (EC) vs. Chemical Coagulation

2.4.1 Chemical coagulation

There are many negatively charged fine colloids (0.01 to 1μm) in wastewater. Coagulation process destabilizes this colloidal suspension. Coagulation reactions have numerous side reactions with other compounds in wastewater. These reactions depend on the characteristics of sewage. Chemical destabilization is caused by chemical coagulation of compounds which are added into the wastewater to form larger particles (flocs) through perikinetic flocculation (aggregation of fine particles in the size of 0.01 to 1 μm). Typical coagulants and flocculants include natural and synthetic organic polymers, metal salts such as aluminium sulphate or alum ($Al_2 (SO_4)_3$), ferric sulphate ($Fe_2 (SO_4)_3$), and ferro sulphate ($FeSO_4$).

Ferric chloride and polymers (anionic, cationic) can be added into the system, while alum and ferric chloride are commonly used for wastewater treatment (Metcalf and Eddy 2003). Chang (1998) reported that colloids between 0.1 μm to 2 μm were converted to the large particles by adding coagulants like alum. Aguilar *et al.* (2002), studied nutrient removal and sludge production in the coagulation-flocculation process. They used alum, ferric sulphate and polyaluminium chloride as coagulants. However, they used several inorganic products as coagulant aids like activated silica and powder activated carbon. Based on their research, approximately 100% of orthophosphate was removed and total phosphate was removed between 98.93% - 99.90%, while, the ammonia and nitrogen removal was very low (between 73.9 – 88.77 %). Aguilar and their team also showed that 41.6% of the sludge volume was reduced by using coagulants (Aguilar *et al.* 2002).

Song *et al. (*2008) studied the effect of coagulant addition on membrane fouling and nutrient removal in the submerged membrane bioreactor. They introduced two coagulants alum and ferric chloride (inorganic coagulants) into the aerobic tank of a pre-anoxic nutrient removal unit. Song and their team found that alum in a concentration of 200-500 (mg/l) could remove nutrients more effectively than ferric chloride can. Through their experiments, 98% of phosphorus was removed (Song *et al.* 2008).

2.4.1.1 Electrical double layer

Two main forces which are acting between colloids are the Van der Waals attraction forces and the electrical repulsion forces. Later forces are usually large enough to keep colloids apart. However, if the particles are given enough energy to overcome this repelling force, then the Van der Waals forces will dominate and bring the particles together. Therefore, if sufficient energy is supplied by heating or mixing, the colloids may begin to agglomerate. Van der Waals forces cannot be increased; if the electrical forces are reduced then the energy barrier can be decreased or totally removed. If colloidal particles are close enough together, Van der Waals forces will cause them to agglomerate, but the effect will be opposed by the electrostatic repulsion of the zeta potential. Most colloids are electrically charged, either positively or negatively. These charged colloids then attract ions in the solution of opposite charge to form a surrounding layer of counter-ions (opposite charge). This layer of counter-ions surrounding the colloids is called the ‘diffuse layer’. The surface charge of the colloid and the diffuse layer together form the ‘electrical double layer’, A large proportion of the counter-ions are situated at close distance to the colloid, called the ‘Stern layer’. The separating boundary between these two layers is referred to as the ‘Stern plane’ (Metcalf and Eddy 1972).

As colloidal particles approach one another, no repulsive force acts between them, and the Van der Waals forces are allowed to bring colloids together. To reduce the electrical forces, zeta potential must be lowered. This can be achieved by adding a higher concentration of ions with a higher charge. These additional ions can replace to the existing counter-ions and they can also reduce the thickness of the diffuse layer. This in turn reduces the Stern or zeta potential and the repelling force, causing stabilisation.

2.4.2 Description of EC

Introducing the electrocoagulation (EC) process in activated sludge unit is one approach to improve the performance of the wastewater treatment system. There are several physicochemical technologies for wastewater treatment like filtration, ion-exchange, chemical precipitation, etc. The common aspect for all of them is that they require additional chemical compounds. In electrochemical technology like electrocoagulation (Holt 2005) and electrofiltation (Li *et al.* 2009), it is not necessary to add chemical compounds. Between these two technologies, electrocoagulation is grabbing the attention due to its lesser capital cost and better performance in comparison with electrochemical technology. In electrocoagulation process, less sludge is produced, and colloidal particles can be removed by binding with the opposite charged ions or by adsorption to the metallic hydroxide. Thanks to the EC technology, metals, soluble inorganic compounds and colloidal particles would be removed (Mollah *et al,* 2001). Electrocoagulation starts with electrolysis reactions around the anode (made of iron or aluminum) area to produce trivalent cations (hydroxide flocs) as coagulants once the DC field is activated.

Electrocoagulation has many advantages compared to the chemical coagulation processes. For example: liquid chemical compounds are not added in reactor; alkalinity is not consumed; the operation is easy to perform; the EC process does not need coagulation agents and thus produces less sludge; the operation cost is reduced because no coagulant is added; EC with six iron plates electrods, has been proven to be a cost effective technology (0.85-1.11 kWh/m^3) (Irdemez *et al*. 2006), generation of by-products are reduced in the effluent as well as wasted solids (Bani-Melhem and Elektorowicz 2010). Microbial populations and colloids are usually charged negative, therefore in the presence of the electrical field (by continuous or intermittent electrical field), their behavior can be changed. They diverted away from the surface of the membrane, therefore. Reducing membrane fouling and filtration flux (Wei *et al.* 2012). The electrical field might also have impact on the activated sludge components. The sludge produced by electrocoagulation consists of large flocs with less bounds of water. As a consequence, the volume of produced sludge becomes modest (Mollah *et al.* 2001).

2.4.3 Mechanisms of EC

Electrocoagulation process depends on the current density, which is based on the current [A] and surface area of each electrode [m^2]. The range of current density might be between 10 to150 A/m^2 (Ibeid, 2011). Lower current density is required when separation units (such as sedimentation tank, sand/coal filtration and membrane filtration) work with electrocoagulation processes. In contrast, higher current density usually is required for separation of coagulants particles, especially in a flotation process. In the electrocoagulation process, no chemical salts are added into the system, thus the value of salt and ion in solids and the supernatant are decreased.

The mechanisms of electrocoagulation can be summarized as follows (Molla 2008; Li *et al.* 2009):

- Electrophoresis: once the current is available in reactor, particles can move toward the oppositely charged electrodes to make large particles (flocs)
- Electro-osmosis: movement of water in reactor due to the electrical gradient forces and presence of more cations than anions in solute.
- Electromigration: when the current is available, ions and polar molecules are transported to the opposite direction electrode.
- Releasing the materials of cathode (hydroxyl ions) and anode (metallic cations) over time and reaction of these compounds with other elements which already existed in the solute.
- The metallic ions which are realized from anode have a reaction with OH^- .
- Long chain of metallic hydroxides compounds can grab unwanted elements such as colloids.
- Oxidation of unwanted organic compounds.
- Electrofiltation: removing the unwanted compounds by sticking them to bubbles which are generated during the EC process.

2.4.3.1 Reactions occurred around electrodes

When current is applied, some of the cationic monomeric forms such as aluminum ions (Al^{3+}) and ($Al\ (OH)_2^+$) can release from anode into the reactor (anode material depends on the wastewater characteristics) this condition causes flocculation of colloidal particles (Bouamra 2012). By oxidation of water, hydrogen ions and oxygen gas are produced around the anode zone. Hydroxides (OH^-) and hydrogen gas are produced around the cathode zone, due to the water reduction process (Eq. 2.16).

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \qquad (2.16)$$

In the suitable pH condition, $Al\ (OH)_3$ (Eq.2.17) afterward, $Al_n(OH)_{3n}$ will produce (Eq. 2.19) (Hasan 2012).

$Al_{(s)} \rightarrow Al^{3+} + 3e^-$ (2.17)

$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+$ (2.18)

$n\,Al(OH)_3 \rightarrow Al_n(OH)_{3n}$ (2.19)

Several reactions are carried out in solute based on level of pH (Eq.s 2.20 to 2.23) (Hasan 2012).

$Al^{3+} + H_2O \rightarrow Al(OH)^{2+} + H^+$ (2.20)

$Al(OH)^{2+} + H_2O \rightarrow Al(OH)_2^+ + H^+$ (2.21)

$Al(OH)_2^+ + H_2O \rightarrow Al(OH)_3 + H^+$ (2.22)

$Al(OH)_3 + H_2O \rightarrow Al(OH)_4^- + H^+$ (2. 23)

Solubility of aluminum compounds depends highly on pH values. In the pH around 6.3, the minimum solubility of aluminum (0.03 mg/l) occurs. AL^{3+} and $Al(OH)^+_2$ at low pH are generated while, as soon as pH increased these compound converted to $Al(OH)_3$.

Holt (2002) pointed out that in pH between 5 to 9, predominant compounds are polymeric complexes species of aluminium and aluminum hydroxide $Al(OH)_3$ (Holt 2002). Flocculation of colloids is a phenomenon in which Al^{+3} are released into the reactor through reduction in the value of zeta potential. In this case, the Vander Waal's forces are bigger than repulsive forces between the negative charged colloids. In a reactor, aluminum ions have a reaction with free hydroxide ions to mostly make monomeric form like: $Al(OH)^{+2}$, $Al(OH)_2^{+1}$, and $Al(OH)^-_4$. Then these compounds can be converted into the polymeric form such as: $Al_8(OH)^{+1}_{20}$, $Al_{13}(OH)^{+5}_{31}$. These processes can be continued until long chain of $Al(OH)_{(s)}$ are finally produced (Eq. 2.20) (Ibeid *et al.* 2012).

2.4.4 Factors affecting in EC process

There are many factors that affect the performance of electrocoagulation process. The current density is one of the important factors for this issue. It is derived from surface area of anode (m^2) and current (A).Current density represents the number of aluminum or iron ions that are realized in solute from the electrodes. If the current density increases, the number of ions increases too. The current efficiency (CE) also reduces when high current density is applied in the system. Increasing the current density requires wasting energy in heating up the wastewater. It can also reduce the removal efficiency. Electrodes which are made up of aluminum have current efficiencies around 130%, while, for iron it can be 100%. This means that aluminum sheets oxidize over time due to the oxidation of chlorine anions. In order to have optimum performance, the current density should be from 10 to 150 A/m^2; this range is suitable for a long period of time, excepting the periodical cleaning of the electrode's surface (Chen 2004).

Chemical characteristics of the solute, conductivity, temperature, the pH value (should be in a range of 6 to 8) and also formation of sludge plays an important role in electro coagulation process (Ilhan *et al*. 2008). For increasing the conductivity, sodium chloride (NaCl) is sometimes added into the water or wastewater. The benefit of adding NaCl is an increase in the ionic contribution in moving the electric charge. This shows which chloride ions have more ability to reduce the adverse effect of the other ions like bicarbonate (HCO_3^-) and sulfate ion (SO_4^{2-}). Sodium chloride also reduces the energy consumption because it increases conductivity (Chen 2004). The level of pH has an influence on the EC processes from two perspectives: current efficiency (CE) and solubility. Aluminum electrodes have higher current efficiency (CE) in both acidic and alkaline environment than in neutral conditions. In alkaline condition more OH^- can release from cathode, while CO_2 is oversaturated in acidic condition. The impact of temperature on EC process has not been wildly assessed. But Chen (2004) found that in temperatures higher than 60^o C, current efficiency might increase, and thus power consumption decreases. On the other hand, the aluminum oxide layer which is formed on the anode surface is destroyed in high current efficiency. At high temperatures $Al(OH)_3$ shrinks and makes denser flocs, which have more affinity to settle down on the electrode's surface. Higher temperature also results in higher conductivity and thus less energy consumption. Other important parameters for the electrocoagulation process are power supply and exposure time. The power might be used as a continuously mode (without any OFF/ON timer), the negative point for this mode is oxidation of anode and passivation of cathode. Therefore the current should be in a mode of OFF, and then ON after some minutes. Low exposure times would decrease the efficiency of EC tasks, while high exposer time might increase the amount of sludge production. Therefore this element is very essential for the performance of EC. Previously, many researchers studied the direct current exposure time (Ibeid *et al.* (2012); Hasan *et al.* (2012); Wei *et al.* (2012), Bani-Melhem and Elektorowicz (2010, 2011); Salamati (2010), and). The types of materials which are used in EC process as electrodes are very important because they release the ions in solute and hence generate the coagulants. Other factors like particle size and electrode spacing have an effect in EC process.

2.5 Operational cost

In the large scale, operational cost is considered by energy demand, treatment of sludge and disposal, and chemical cleaner for membranes (Hai and Yamamoto 2011). Over time, as the application of MBR has increased, the installation cost has decreased. By reducing the footprint, the water treatment cost is reduced (Hai and Yamamoto 2011). Mannina and Cosenza (2013) investigated different strategies for energy saving like influences of aeration, time for backwashing and the frequency of membrane cleaning. They found that by using low air flow rate at the first time of operation, operational cost was reduced by 20%. Until now, several studies about MBR and SMEBR have been performed, but studies about saving energy are still needed. Energy demand was calculated based on aeration, pumping, and mixing energy. Over more than two decades, the accomplishments of Membrane Bioreactor (MBRs) have increased. However, in comparison with the conventional activated sludge process, MBRs need more energy for operation and aeration for scouring the membranes (34%) and for biological activities (42%) (Wallis-Lage and Levesque 2009). These percentages are shown in Table 2.2.

Table: 2.2: Energy used in MBR (Haribljan 2007)

Processes	**Energy used**
Bio-process aeration	42%
Membrane aeration	34%
RAS pumping	10%
Anoxic mixing	9%
Permeate pumping	4%

Meunier *et al*. (2006) pointed out that the cost of energy requirement for electrocoagulation process is up to five times lower than for processes using chemical precipitation for removing chemical and metallic compounds. Liu *et al.* (2012) noted that electric energy consumption can be reduced by using a more effective electric field, better configuration for electrodes, and using a more matched membrane, or maintaining an appropriate electrostatic repulsive force against EPS or foulants on the surface of membranes. They also found that, for having less fouling problems via electric field, electro-coagulation can be used. In their research, the energy consumption in the electric field was between 2×10^{-3} kWh/m^3 and 8×10^{-3} kWh/m^{3,} this energy consumption depends on the level of voltage (0.4 V and 0.2 V respectively). It was designed based on the side stream membrane (Liu *et al*. 2012).

2.5.1 Treatment of sludge and disposal

Sludge dewatering is one of the most expensive processes in wastewater treatment plants. If bound water in the sludge reduced, the volume of sludge can be reduced as well, consequently reducing the cost of treatment facilities. Liu *et al.* (2012) pointed out that, thanks to electro-coagulation, size of sludge was increased and zeta potential was reduced. Therefore, the cost of sludge treatment and disposal might be reduced.

2.6 Membrane electro-bioreactors

The electro-bioreactor is a modern technology for simultaneous reducing organic and mineral contaminants, and nutrients (nitrogen and phosphorus), in wastewater. In general, electro-bioreactor’s performance depends on wastewater characteristics, biological parameters, and operating conditions. Submerged Membrane Electro-Bioreactor, or SMEBR, is a compact hybrid unit for biological processes, membrane processes (membrane filtration) and electrokinetic processes. Before 2009, no one applied these 3 fundamental processes in one unit for wastewater treatment. Elektorowicz’s research team studied about the combination of these 3 processes (biodegradation, membrane filtration and electrokinetics) for controlling and removing nutrients (nitrogen and phosphorus) (Ibeid (2012); Bani-Melhem and Elektorowicz (2011); Hasan (2011); Salamati (2010); Elektorowicz *et al.* (2009)). One of the interesting aspects of electro-bioreactor technology is observed when an arctic area generates a large amount of sewage. Elektorowicz *et al.* (2013) studied sustainable water management within a mining area in the arctic region. SMEBR system can adapt to the flow rate and quality of influent, which change over times. When mining operations in one area end, the electro-bioreactor system can be disassembled and transported into another place. Besides this wastewater treatment technology, another EKDIM technology can be applied for wasted sludge management (biosolids). It can almost completely remove water, pathogens and metal as well as odors (Elektorowicz *et al.* 2013).

2.6.1 Important factor for SMEBR

Previous study has shown that SMEBR was designed in a cylindrical polyethylene container with one submerged membrane placed inside the bioreactor (Ibeid, 2011). For aeration, Hasan (2012) and Ibeid (2011) used 4 and 2 fine bubble air diffusers, respectively, at the bottom of SMEBR. Electrodes were connected to a power supply which worked in a direct current mode. An electrical timer was applied to achieve the appropriate current density and exposure time (time ON and time OFF). During the time ON, the constant current was activated for a few minutes and then stopped as a time OFF for another few minutes. Balancing between Times OFF and time ON should generate sufficient amount of Al^{+3} for removing phosphorus and transforming N into gas. Ibeid (2012) tested current densities in several stages. Hassan (2012) found that in current densities around 10, 15 and 27 A/m^2 ,sufficient amount of aluminium ions were produced, in comparison with current densities of 5 A/m^2. Ibeid (2011) showed that with 5 min ON: 20 min OFF, the electrical mode or exposure time had better results for controlling effluent at minimum cost. Moreover, the values of current between electrodes had an effect on electrokeinetic process like electro-osmosis and electromigration. Thus, the current density had an effect on sludge properties, membrane fouling and quality of effluent. Previous works (Ibeid 2011, Hasan 2011) showed that the novel SMEBR system can reduce the fouling problem by applying direct current (DC) in a medium current density (15 to 25 A/m^2) into the bioreactor. He compared the application of SMEBR and conventional MBR system in terms of reducing the fouling problem (up 8 times less than conventional method based on the concentration of soluble microbial products (SMP) and volatile suspended solids (VSS)) and removal efficiencies of nutrient from effluent (95% of carbon, 99% of phosphate and 97% of nitrogen).

2.6.2 Mechanisms of nutrient and carbon removal in SMEBR

When direct current in SMEBR is activated, various forms of electrochemical reactions are carried out because aluminum is selected as an anode and iron is selected as a cathode. The main goal for this reactor is removing the nutrients from wastewater. There are three main elements for removing phosphorous and nitrate in new submerged electro-bioreactor technology:

1) Current density. It is an electric current (A) per unit area of cross section (m^2) (CD = I/A). If the current density increases, the level of Al^{+3} and electrons around the anode increase. Moreover, the concentration of hydrogen gas around the cathode increases, too. Electrochemical reactions which are carried out in the anode and cathode are shown below [Eqs. 2.24 to 2.27]:

- At the anode:

$Al \rightarrow Al^{+3} + 3e^-$ (2.24)

$2H_2O \rightarrow O_{2\,(gas)} + 4H^+_{(aq)} + 4e^-$ (2.25)

- At the cathode:

$3H_2O + 3e^- \rightarrow 3/2\ H_{2\,(g)} + 3OH^-$ (2.26)

$\frac{1}{2} O_2 + 2e^- + H_2O \rightarrow 2OH^-$ (2.27)

2) Timer for showing the electrical exposer time (ON/ OFF). Microorganisms are very sensitive when they are exposed to the continuous current density. Therefore, enough OFF time should be given. During this time, microorganisms can recover from electrical shock and renew their activities. This mode also affects the production of aluminum ions, hydrogen gas, and electrons in reactors.

3) The amounts of dissolved oxygen are very important. DO is responsible for transformation of nitrogen gas and phosphate in reactor. This parameter should not be too high nor too low, in order to provide a level for the transformation of nitrogen in the reactor. The DO level in SMEBR is usually between 0 to 1.5 mg/l, depending on the operating conditions. The main nutrients which are removed in SMEBR are: nitrate, ammonium, phosphate and carbon.

2.6.2.1 Phosphorous removal

Earlier researchers have reported that a submerged membrane bioreactor (MBR) can remove phosphorous but it has difficulty removing nitrogen (Elektorowicz *et al.* 2011 and Gao *et al.* 2004). The SMEBR system increases the possibility of removing the phosphorous by electrocoagulation and deposition on electrodes (Hasan *et al.* 2012). Phosphorus can be removed in the SMEBR system when it creates complexes with the Al^{+3}. In the bioreactor, metal ions (Al^{+3}) would have a reaction with the phosphate to form insoluble aluminum phosphate ($AlPO_4$) or form calcium hydroxyapatite in the presence of calcium (Eqs. 2.28 and 2.29, respectively), which would also be presented as follows (Hasan *et al.* , 2012):

$$Al^{+3} + H_nPO_4^{3-n} \leftrightarrow AlPO_4 + nH^+ \qquad (2.28)$$

$$10\ Ca^{2+} + 6PO_4^{3-} + 2OH^- \leftrightarrow Ca_{10}(PO_4)_6(OH)_2 \qquad (2.29)$$

Hasan *et al.* (2011) tested a novel submerged membrane electro-bioreactor at pilot scale in l'Assomption (Quebec). They showed that the SMEBR system removed over 99% of phosphorous while exposed to daily fluctuation of sewage quality and temperature. The influent concentrations of phosphorous varied between 2 to 10 mg PO_4^{-3} -P/l.

2.6.2.2 Nitrogen removal

The nitrification process is affected by two groups of microorganisms: ammonia-oxidizing bacteria and nitrate-oxidizing bacteria. Both of them are characterized as aerobic autotrophs bacteria (Dytczak *et al*. 2008). The population of nitrifiers is affected by enzyme kinetics, growth rates, and decay rates. Nitrifirers like *Nitrosomonas* and *Nitrobacter* grow quickly. In contrast, *Nitrosospira* and *Nitrospira* grow slowly but they can live longer (Dytczak *et al.* 2008). Kartal *et al.* (2010) reported that nitrogen can be removed by using anoxic ammonium-oxidizer (anammox) bacteria. This process needs less energy but anoxic ammonium-oxidizing bacteria grow slowly. Conventional wastewater treatment systems need more energy for nitrogen removal to provide aerobic conditions for nitrification processes, and also need organic carbon for denitrification process. The anammox process can transform 50% of nitrogen in marine environments. Briefly: this process is a shortcut in the nitrogen cycle. Anammox bacteria are suitable to be present in electro-bioreactor because of the possibility that they will create alternate aerobic and anaerobic conditions in such a reactor. For this reaction, partial nitrification of ammonium needs to obtain the appropriate ratio of ammonium/ nitrate equal to 1. Nitrogen can be removed in six steps in SMEBR, which are explained as follows (Elektorowicz *et al.* 2011):

Step 1: In the submerged electro-bioreactor unit, when a direct current is ON, electrons are discharged from the anode area into the reactor (Eqs. 2.24 and 2.25). Most of the dissolved oxygen has a reaction with electrons to produce hydroxide (Eq. 2.27). Based on Eqs. 2.26 and 2.27, DO concentration is reduced as time passes with the electrical exposer showing the time is ON (Ibeid 2011).

Step 2: For removing the nitrogen gas, the level of DO is very essential. As long as the current is off, the concentration of DO will increase. The solute still has enough oxygen to act as an electron acceptor for the biological activates (nitrification and denitrification processes). One of the important ways of removing nitrogen from wastewater is nitrification in an aerobic condition through two steps of biological activity; ammonium can oxidize to nitrate (Eqs. 2.30 and 2.31). Optimum aerobic concentration in a submerged electro-bioreactor (SMEBR) appears during the times OFF; this time is appropriate for nitrification process, because in this time the oxygen level is recovered. During the times ON, electrons are produced and the DO level is reduced to create anoxic environment for the denitrification process. Aerobic and anoxic environments are alternated by switching the electrical exposer timer (ON/OFF) and current density (Elektorowicz *et al.* 2011).

$$NH_4 + 3/2\ O_2 \rightarrow NO_2^- + H_2O + 2H^+ \qquad (2.30)$$

$$NO_2^- + H_2O \rightarrow NO_3^- + 2H^+ + 2e^- \qquad (2.31)$$

Step 3: Once the dissolved oxygen in the solute decreases, oxygen cannot act as an electron acceptor, and nitrate therefore gets this responsibility in the system (anoxic condition). In this case, heterotrophic denitrifiers play an important role to reduce nitrogen concentration. Nitrification is followed by denitrification (Eq. 2.32). This process is carried out in an anoxic condition wherein nitrogen can be converted to nitrogen gas (Ibeid 2011).

$$NO_3^- \rightarrow NO_2^- \rightarrow NO + N_2O \rightarrow N_2 \qquad (2.32)$$

Microorganisms for denitrification process use hydrogen as an electron donor. Previous studies show that ammonium can be removed in anaerobic conditions; also nitrate can be removed in the alternative aerated membrane bioreactor, more easily (Elektorowicz *et al.* 2011).

These aerobic /anoxic conditions can be achieved also in a Sequence Batch Reactor (SBR). Previously research showed that, for effective nitrate removal in a sequence batch reactor, anoxic conditions (for denitrification) must operate before aerobic conditions (for nitrification) (Elektorowicz *et al.* 2011; Ibeid 2011; and Dytczak *et al.* 2008).

Step 4: In the anammox process, ammonium and nitrate can convert to the nitrogen gas. Anammox bacteria use an inorganic carbon source (CO_2) for growing. The combination of anammox and nitrification process is shown in Eq. 2.34. Anammox bacteria start to nitrify the ammonium which already exists in the solute (Ibeid 2011).

$$NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O \tag{2.33}$$

$$2NH_4^+ + 3/2\ O_2 \rightarrow N_2 + 2H^+ + 3H_2O \tag{2.34}$$

Step 5: The system needs aerobic conditions for nitrification processes. Besides, it needs anoxic conditions for supporting the anoxic heterotrophic denitrifers and the anammox. During the time-ON, concentration of dissolved oxygen changed as direct current available into the system. During the time-ON, enough electrons released into the reactor to introduce electron acceptors like NO_3^- and nitrite at the anammox conditions. During the time-OFF no more electrons are released into the system. Exposure time-OFF is necessary for the system to recover its oxygen concentration to a desired level which can support the aerobic conditions (Ibeid 2012).

Step 6: For their activities, some bacteria use the hydrogen gas as an electron donor which is produced at the cathode zone (Eq. 2.26). In this reaction, nitrate works as an electron acceptor. The final product is nitrogen gas (Eq. 2.35).

$$2NO_3^- + 5H_2 + 2H^+ \rightarrow N_2 + 6H_2O \tag{2.35}$$

Pilot tests of SMEBR in l'Assomption (Quebec) by Hasan *et al.* (2011) showed 99% of ammonia was removed when the influent concentrations of ammonia were between 30 to 70 mg NH_3-N/l. Wei *et al.* (2012) also demonstrated in lab scale that by applying electro-bioreactor (EMBR) more than 99% of ammonium-N is removed.

2.6.2.3 Carbon removal

Since additional oxidation of organic carbon can be observed, biodegradation processes are not the only way for removing the carbon (Bani-Melhem and Elektorowicz 2011). Furthermore, in the reactor, aluminum ions can be converted to polymeric species such as Al_8 $(OH)^{+1}_{20}$, Al_{13} $(OH)^{+5}_{31}$.. Eventually they are transformed to the long chains of $Al\ (OH)_S$. These cationic compounds can adsorb the colloids and other organic compounds that are charged negatively. Hasan *et al.* (2011)'s pilot tests of SMEBR performance in l'Assomption showed that the system can remove 92% of COD when the influent concentration of COD fluctuated between 160 to 700 mg/l.

For better performance of microbial activities, current density should be as low as possible, and also the time exposer mode (time OFF) should be as long as possible. More than 95 % of dissolved COD can be removed by membrane electro-bioreactor technology (Wei *et al.* 2012).

Chapter 3: Experimental setup

3.1 SMBER system

The Submerged Membrane Electro-Bioreactor, or SMEBR, is a compact hybrid reactor where biological processes (biodegradation), membrane processes (membrane filtration) and electrokinetic phenomena are carried out. In this study, the SMEBR was designed in a rectangular tank. Depending on the configuration of the bioreactor, there were 3 or 2 pairs of electrodes; 60% perforated aluminum flat sheets were applied as anodes, while stainless steel mesh was applied as cathodes. The distance between electrodes was 5 cm. The level of the current density was chosen based on previous investigations (Ibeid 2012). Two major designs were investigated in this research. The electrical exposure modes were tested several times during the investigation of the first and the second design. The optimum mode for the first design of reactor was 5 min ON: 20 min OFF and in the second design was 5 min ON: 15 min OFF. In order to achieve the objectives described in Chapter 1, the methodology consist of 3 phases as shown in Fig. 3.1. These phases are as follows: Phase 1: Preparation and set up (2 stages) of installations; Phase 2: Investigations of the first design of electro-bioreactor (EBR) (2 stages) followed by analysis of effluent (discussed in Chapter 4 in subdivisions 4. 1 and 4.2) and Phase 3: Investigations of the second design of electro-bioreactor (2 stages) followed by analysis of effluent (discussed in Chapter 4 in subdivisions 4. 1 and 4.2).

3.2 Preparation and set up of installations

3.2.1 Phase 1 (Stage 1): preliminary determination of the SMEBR

The main objective of the laboratory scale study was to assess the operating factors of submerged membrane electro-bioreactor by analyzing the effects of current density, the reactor's dimension, HRT, SRT, aeration intensity, membrane performance, and sludge properties. In this experiment, synthetic wastewater was applied. For this study, activated sludge was taken from the municipal wastewater treatment plant in Saint Hyacinthe (Quebec). The synthetic wastewater was selected based on similarity to real wastewater concentration of organic compounds, phosphorus, nitrate and soluble salts.

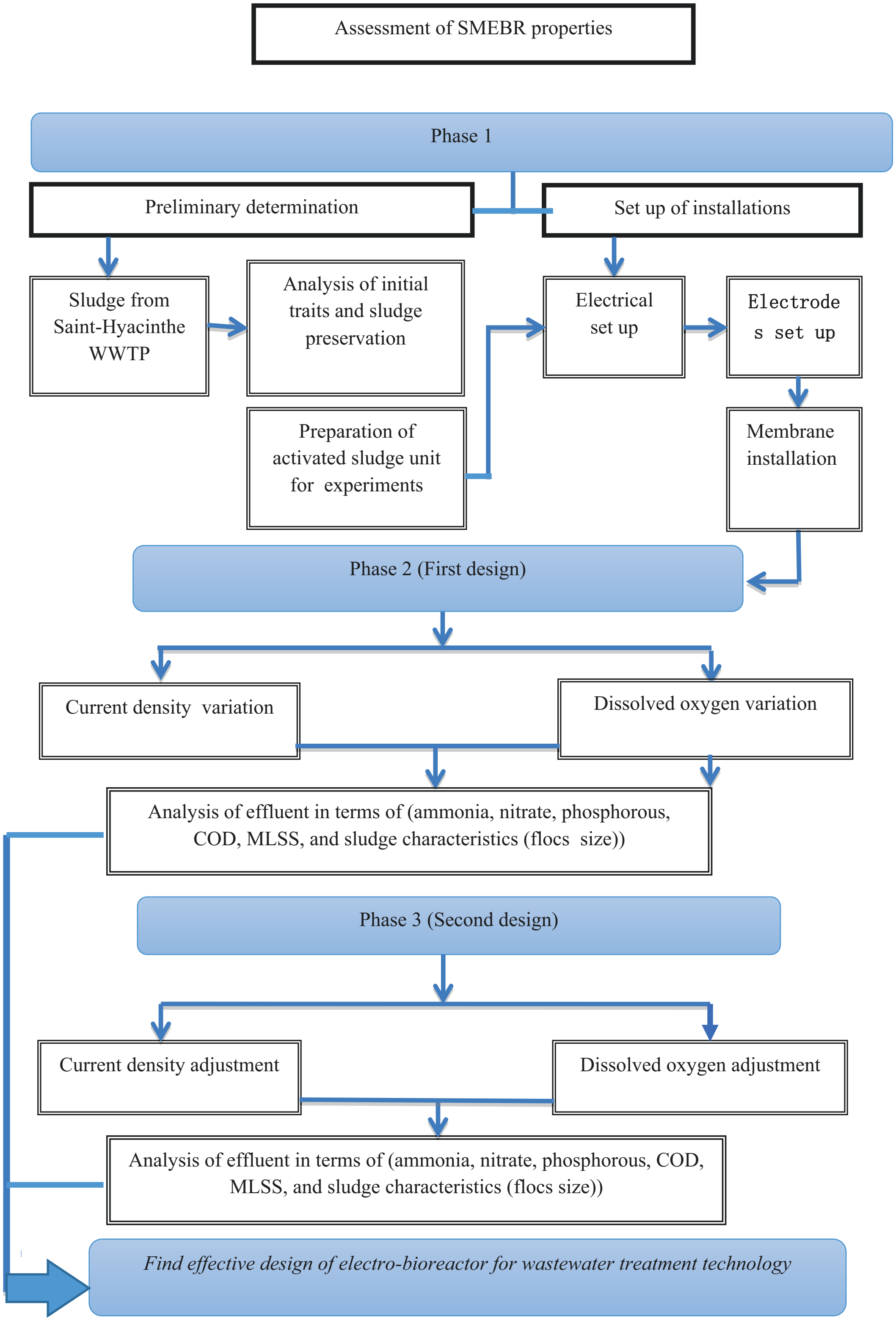

Fig. 3.1: Work plan

The gradients of synthetic wastewater were: glucose, peptone, yeast extract, ammonium sulfate $(NH_4)_2SO4$, potassium phosphate KH_2PO_4, magnesium sulfate $MgSO_4 \cdot 7H_2O$, manganese sulfate monohydrate $MnSO_4 \cdot H_2O$, iron sulfate $Fe_2O_{12}\ S_3 \cdot 5H_2O$, calcium chloride $CaCl_2 \cdot 2H_2O$, potassium chloride KCl, and sodium bicarbonate $NaHCO_3$ (Table 3.1).

Table 3.1: Composition of synthetic wastewater

Component	Concentration (mg/l)	
	Stage 1	Stage 2
Glucose*	665	864.5
Peptone*	85	72.25
Yeast extract*	100	85
Ammonium sulfate	100	100
Potassium phosphate*	37	22.8
Magnesium sulfate	40	40
Manganese sulfate monohydrate	4.5	4.5
Iron sulfate	0.4	0.4
Calcium chloride	4	4
Potassium chloride	25	25
Sodium bicarbonate	25	25

*: Concentration of glucose, peptone, yeast extract and potassium phosphate changed in the synthetic wastewater over time

The synthetic wastewater was prepared by adding all the above compounds into 1L of pure water, and mixing them with a shaker for at least 15 minutes to ensure that all compounds dissolved in the water. Then, the contents of the 1L were increased to 100 L by adding extra pure water and mixing them manually again for at least 5 minutes before beginning the experiments.

These contents were pumped into the bioreactor tank by means of peristaltic pumps (MasterFlex pump, from Cole-Parmer, USA). In addition, effluents (treated water) were pumped out by MasterFlex suction pumps, from Cole-Parmer, USA. In the SMEBR, compressed air was injected into the bioreactor through 14 fine air stone which were located at the bottom of a rectangular cubic container. For adjustment of air and pressure in the system, an air flow meter and pressure regulator were used.

Transmembrane pressure (TMP) was measured by a digital gauge pressure. Ultrafiltration ZeeWeed-1 (GE, Canada) membrane modules were submerged vertically at the left side (in the first design) and at the middle of the bioreactor (in the second design). This membrane module has 0.04µm pore size and 0.047m^2 surface areas. The laboratory design of SMEBR is illustrated in Figs. 3.2 and 3.3.

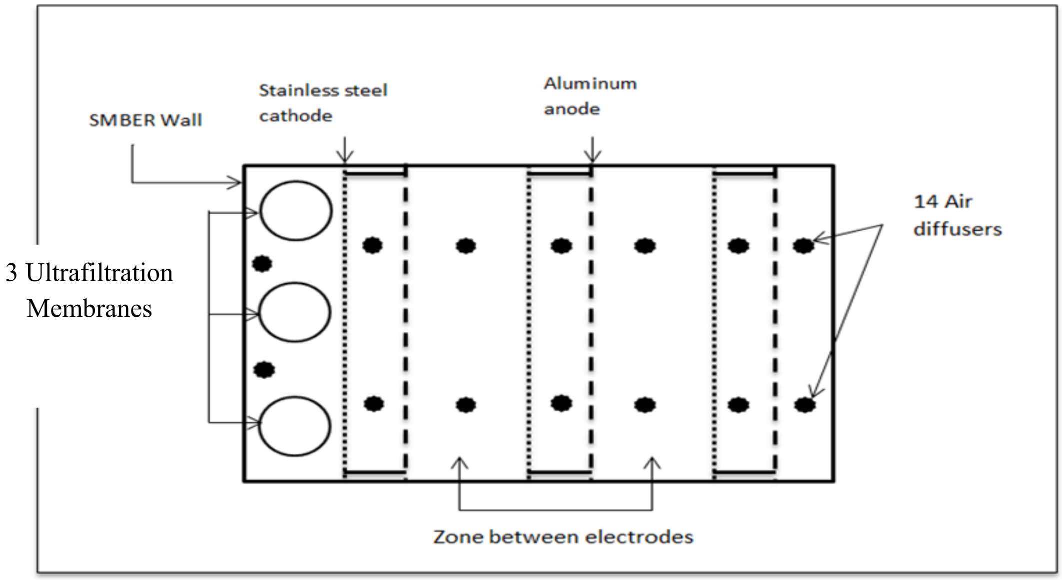

Fig. 3.2: Plan view of the first design of SMEBR

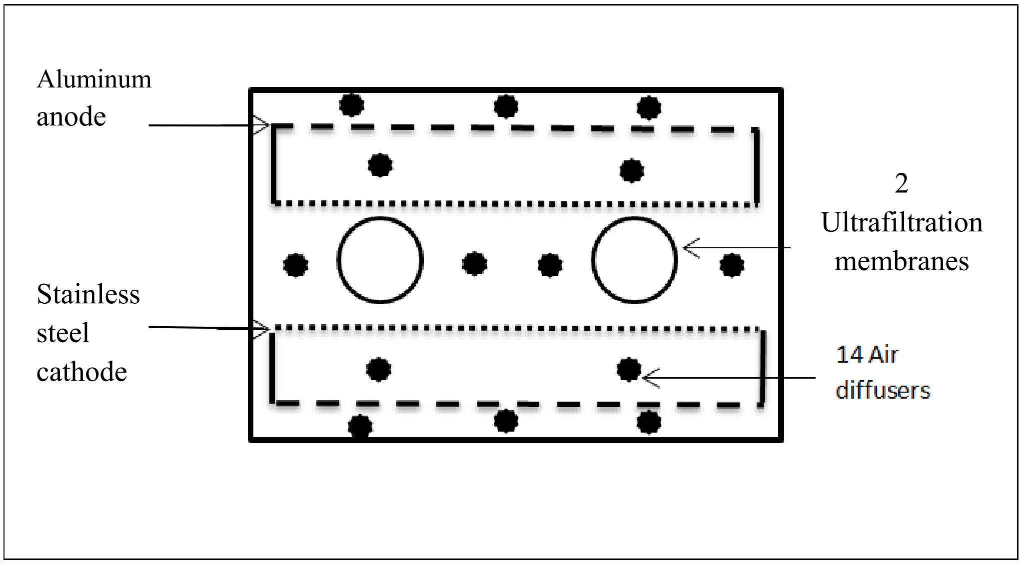

Fig. 3.3: Plan view of the second design of SMEBR

Primary design of laboratory electro-bioreactor was done by previous researchers of Dr. Elektorowicz's team: Ibeid *et al.* (2012); Hasan *et al.* (2012); Bani-Melhem and Elektorowicz (2010, 2011).The membrane was cleaned physically and chemically when it is clogged. In the small pore size of membranes, the cake layer feature a higher resistance compared to the large pore size of membranes. However, cake layer which is produced in small pore membranes is removed more easily than cake layer which is produced in large pore size membranes due to the internal pore clogging (Hai and Yamamoto 2011). Time of cleaning was indicated when transmembrane pressure increased up to 50 - 70 kPa (Hasan 2012). Furthermore, during the experiment, the aluminum perforated sheet (as an anode) was changed 16 times because of the corrosion.

3.2.2 Phase 1 (stage 2)

In Phase 1, the electro-bioreactor was designed by a perforated (60%) aluminum as an anode, and an iron sheet (made from stainless steel mesh) as a cathode. Both electrodes were attached to a wooden frame in order to keep them in a vertical position at a constant distance of 5 cm.

In the electro-bioreactor, air was supplied through air stones which were located at the bottom of bioreactor to maintain adequate DO (dissolved oxygen) conditions. In total, 14 cubic air stones were installed for mixing, evacuating gases generated by microorganism, improving biodegradation, controlling dissolved oxygen (DO) level, and decreasing membrane fouling. Laboratory scale design of electro-bioreactor was implemented in a 22.5 L (Shape 1, Fig. 3.2) and 15 L (Shape 2, Fig. 3.3) vessels; both were designed with a rectangular container shape. The characteristics of reactor and electrodes are shown in Table 3.2.

Table 3.2: Characteristics of reactor and electrodes

Reactor	Material: Plastic Shape : Rectangular container Dimension (cm^3): 54 × 35 × 13 and 37 × 20 × 20 Effective Volume(L): 22.5 and 15 Reactor mode: Complete-mixed Aeration system : Cubic air diffuser
Electrodes	Material: Aluminum as anode and stainless steel as cathode Shape: Rectangular Number of electrical units (a): 3 or 2 Effective anode surface area (m^2)(b) : 0.0216 or 0.0342 Electrodes arrangement: Parallel Distance between electrodes (cm): 5

(a) Number of electrical units is depending on first and second design. 3 units used for first configuration and 2 units used for second configuration of electro-bioreactor.

(b)For the first design: 0.12 m × 0.3 m × 0.60 = 0.0216 m^2 and for the second design: 0.19 m × 0.3 m × 0.60 = 0.0342 m^2.

This phase was monitored at different current density (7.71, 12.34, 15.43, 28.93, 18.51 and 21.92 A/m2) and dissolved oxygen values (3 to 8 mg/l). All electrodes and the DC power supplier (TES 6230) were connected to the distribution panel. The switch-timer was connected to the power supplier (See Appendix A, Fig. 1). In order to obtain reliable results, operating parameters like current density and exposure mode should be adjusted with other parameters like organic loading, HRT, SRT and MLSS. The influent concentration of ammonia, nitrate, phosphorus and COD are shown in Table 3.3. The samples were taken from synthetic wastewater to perform analysis of the influents.

Table 3.3: General operating conditions in SMEBR

Design #	Duration (weeks)	Timer (min.)	Average influent NO_3^- (mg/l)	Average influent NH_4-N (mg/l)	Average influent PO_4^{-3} (mg/l)	Average influent COD (mg/l)
1	1 to 21	5′-ON/ 20′-OFF	0.13 ± 0.25	17.6 ± 1.6	20.25 ± 2.25	931 ± 65
2	22 to 27	5′-ON/ 15′-OFF	*Various 0.228 ± 0.010	*Various 16.8 ± 0.6	*Various 18.7 ± 3.4	945 ± 53.5

*: Changing the concentration of *glucose*, *peptone*, *yeast extract* and *potassium phosphate* in the synthetic wastewater.

During this experiment the electro-bioreactor was run for about 6 months. The operating conditions of the electro-bioreactor are shown in Table 3.4. In stages 1 to 5, the electro-bioreactor was operated at 5′-ON/20′-OFF and in stage 6, the electro-bioreactor was operated at 5′-ON/15′-OFF. HRT and SRT were not changed during the experiment; they were adjusted to 12 h and 15 days respectively.

Table 3.4: Operating condition based on current density

Stage #	Duration (weeks)	Total current (A)	Current density (A/m^2)	Average MLSS (mg/l)
1	1 to 7	0.5	7.71	4551
2	8 to 10	0.8	12.34	4310
3	11 to mid of 12	1	15.43	5480
4	Mid of 12 to mid of 18	2.5	28.94	6920
5	Mid of 18 to 21	1.2	18.52	6480
6	22 to 27	1.5	21.93	6950

3.3 Phase 2 (fist design of submerged membrane electro-bioreactor)

For this phase, a series of experiments was carried out for 21 weeks. In these experiments, the impact of electrokinetics was tested in two ways: regarding variation of current density and regarding variation of dissolved oxygen (Fig. 3. 4). Several direct currents imposed into the system. Variations in mode (ON/OFF) change the exposure of sludge to the electrical field. The system was assessed based on analyses of ammonia, nitrate, phosphorous, COD, MLSS, and flocs characteristics in terms of size.

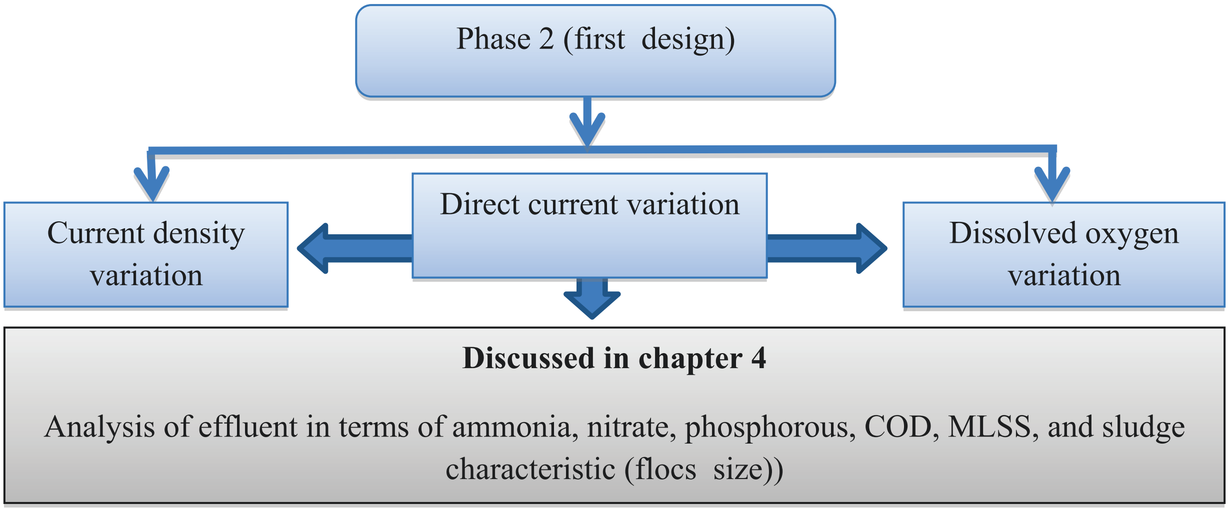

Fig. 3.4: Flow chart of the methodology approached in phase 2

3.3.1 First design of reactor

In order to reduce the nutrients (nitrogen and phosphorus) in a new design of membrane electro-bioreactor with multiple electrical units, several stages and experiments were carried out. In this research, the idea for the system and operation conditions for a new submerged membrane electro-bioreactor (SMEBR) comes from Hasan (2012), Ibeid (2012), Oleszkiewicz *et al.* (2012), and Bani-Melhem and Elektorowicz (2010, 2011). However, a new design of SMEBR is necessary to expand the application of the SMEBR. The first design of the SMEBR (Fig. 3.6) has specific parameters which are shown in Table 3.5. In the initial first stage, the system ran for 48 hours to ensure sufficient microorganism presence in the system. This stage ran without current, and after this time, effluent and sludge were sampled for further assessment. As seen in Fig. 3.5, the electro-bioreactor was designed with three membranes and three pairs of electrodes (aluminum anode and stainless steel cathode). Synthetic wastewater was pumped from the feed tank to the bioreactor tank via one suction pump (MasterFlex pump, Cole-Parmer, USA).

Table 3.5: Specific parameters for the first design of electro-bioreactor

Parameters	1st design	Parameters	1st design
Number of air diffusers	14	SRT	15 days
Sludge wastage	1.5 L/d	HRT	12 h
Effective length of anode (submerged in sludge)	12 cm	Current density (A/m^2)	7.71, 13.88, 15.43, 28.93, 18.51, 21.92
Width of tank (cm)	35 cm	Exposer mode	5’ ON: 20 ’OFF
Area of anode (cm)	20 × 30	Area of cathode(cm)	20 × 30
Distance between electrical zone	5 cm	Number of electrical units and membranes	3
Effective area	60%	Temperature	19 - 21°C
(a) Effective area (As) of each anode	0.0216 m^2	Volume of reactor	22.5 L
Flow wastage	1.5 (L/d)	Flow rate (L/d)	45

(a) For the first design, the effective area of each anode was 0.0216 m^2

In each tests, wastewater underwent biological and electrokinetic processes in the tank. Then, liquid was separate from the sludge due to membrane filtration. The effluent was pumped by three suction pumps (MasterFlex pump, Cole-Parmer, USA).

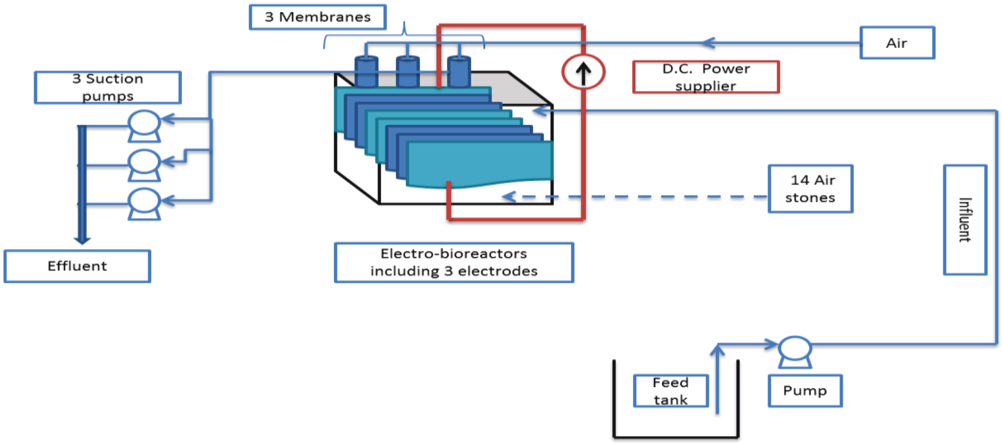

Fig. 3.5: Sample design of new SMEBR in laboratory (First design)

3.3.2 Investigation of the impact of current density in the first design electro-bioreactor

The laboratory design of a new electro bioreactor system was based on the HRT of 12 hours, SRT of 15 days, and different current densities (7.71, 12.34, 15.43, 28.93, 18.51 A/m^2). These values were selected according to the previous studies ((Hasan (2012), Ibeid (2011) and Salamati (2010)). During the experiment, current should change to make sure that the SMEBR operates at an optimal way under adequate current density, and an electrical exposure mode that does not inhibit the biological process. This experiment was controlled at the exposure time of 5 min ON: 20 min OFF. In the SMEBR, the current passing through three pairs of electrodes was commensurate to the concentration of soluble ions, and to the suspended solid's concentration (Ibeid *et al.* 2012). In the electro-bioreactor unit, Al^{3+} is released from aluminum anode sheet and subsequently, several electrochemical reactions occurred inside the sludge liquor such as: electrocoagulation, electro-migration, electrophoresis, and electro-osmosis. Therefore, current density can change sludge properties, membrane fouling and quality of effluent, because the value of the current can affect electrokinetic processes (Ibeid *et al.* 2012). Current density is calculated as follows:

CD = I/A (Eq. 3.1)

Where: I = Current showing on power supply for each electrode (A)

A = Effective electrode surface area for each electrode (m^2)

Different levels of current densities were applied into the first design of electro-bioreactor (within 21 weeks). Therefore, based on Eq. 3.1, the current density can be calculated (from 7.71 to 18.52 A/m^2).

3.3.3 Investigation of the impact of dissolved oxygen (DO) on SMEBR

Compressed air was injected into the reactor through 14 fine bubble air diffusers that supplied oxygen not only for the sludge but also for mechanical vibration of the submerged membrane; this removed sludge which was stacked onto the surface of membrane. Dissolved oxygen was monitored by using a multi parameters digital DO meter (hQ30d, HACH, USA). In the first design of the electro-bioreactor five ranges of dissolved oxygen were operated at the system : 7- 8 mg/l; 3-4 mg/l; 5-6 mg/l; 4-5 mg/l; and 3-4 mg/l. Table 3.6 represent different level of dissolved oxygen within 21 weeks of experiment.

Table 3.6: Variation of DO in the first design of electro- bioreactor

Time (weeks)		DO (mg/l)
a	Week 1 to 7	7 to 8
b	Week 8 to 10	3 to 4
c	Week 11 to mid12	5 to 6
e	Week mid12 to mid18	4 to 5
f	Week mid18 to 21	3 to 4

3.4 Phase 3 (second design of SMEBR)

In order to achieve optimal performance, a second configuration of the reactor was designed. In phase 3, as in phase 2, current density and dissolved oxygen were investigated. Duration of the laboratory experiments for this phase was almost 6 weeks (from week 22 to 27).

The effluent and sludge were sampled for analysis of ammonium, nitrate, phosphate, COD, MLSS, and flocs characteristics in terms of size (Fig. 3.6).

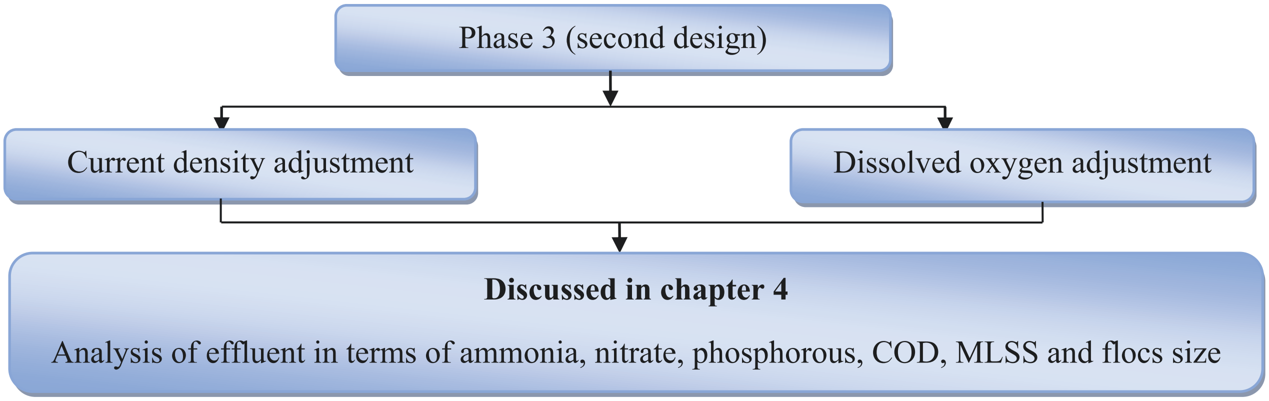

Fig. 3.6: Flow chart of the methodological approach in phase 3

Simple design of new SMEBR system is illustrated in Fig. 3.7 and is based on the information shown in Table 3.7. Like the first design of electro-bioreactor, the effluent can be extracted by MasterFlex pumps (Cole-Parmer, USA).

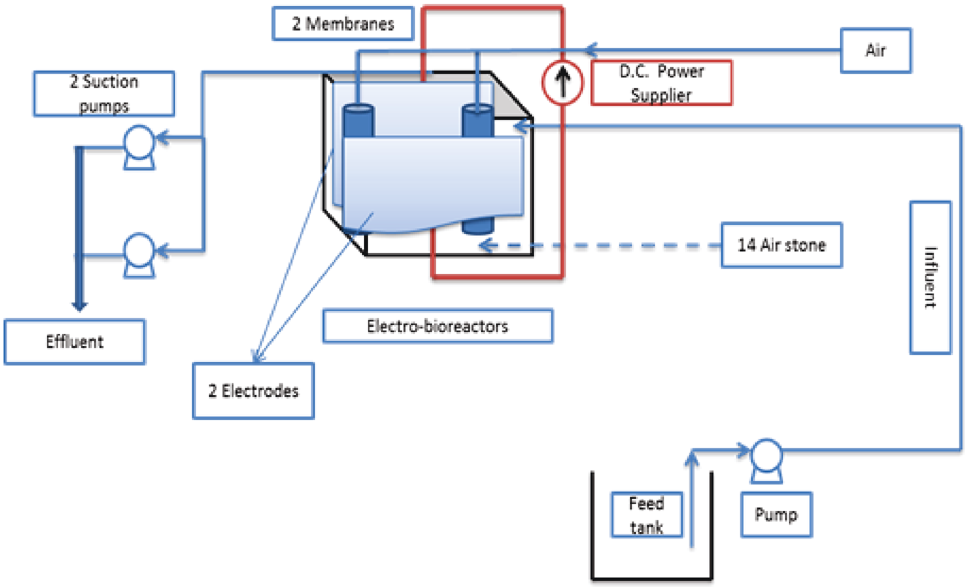

Fig. 3.7: Lab-scale of new SMEBR in laboratory (Second design)

Table 3.7: Specific parameters for the second design of SMEBR

Parameters	2^{nd} design (Fig. 3.7)
Number of air diffusers	14
Number of electrical units and membranes	2
Effective length of anode (cm) (submerged in sludge)	19
Width of tank (cm)	20
Area of anode (cm)	20 × 30
Area of cathode (cm)	20 × 30
Distance between electrical zone (cm)	10
SRT (days)	15
HRT (h)	12
Effective area	60%
Current (I) (A)	1.5
Exposure mode	5 min ON:15 min OFF
Temperature	From19 to 21°C
Volume of reactor (L)	15
Effective area (As) of each anode (m^2)	0.03742
Flow wastage (L/d)	1
Flow rate (L/d)	30

3.4.1 Investigation of the impact of current density in novel SMEBR

The second design of bioreactor had a current density of 21.92 A/m^2. Like the previous electro-bioreactor, perforated aluminum sheets were used as an anode, and stainless steel mesh was applied for cathodes.

3.4.2 Investigation of impact of dissolved oxygen (DO) in SMEBR

In the second design of the bioreactor, dissolved oxygen was between 3 to 3.91 mg/l from week 22 to week 27. In this design of electro-bioreactor, like the previous one, an air diffuser was installed at the bottom. A pressure regulator and air flow meter were also used to adjust the air pressure and flow rate, respectively.

3.5 Sample assessment and materials

The activated sludge used in this research was taken from the wastewater treatment plants in Saint-Hyacinthe (Quebec). The initial characteristics of this activated sludge are shown in Table 3.8.

Table 3.8: Initial values of activated sludge from Saint-Hyacinthe

Parameters	Values
pH	6.64
Dissolved Oxygen (mg/l)	8.32
Electrical Conductivity (µS/cm)	443
Temperature (^{0}C)	19
MLVSS (mg/l)	1580
MLSS (mg/l)	2482
Sludge Volume Index (SVI) (ml/g)	411
Particle Size Distribution (PSD) (mean size µm)	38.67

For this study, several assessments were carried out. Some assessments were done before the experiments, and some were done during the experiments. Temperature, level of pH, dissolved oxygen (DO), electrical conductivity (EC) were assessed before the experiments. The values of nitrate, ammonium, phosphate, COD, flocs particle size in electro-bioreactor, MLSS, MLVSS, and SVI were measured during the experiments.

3.5.1 Basic analysis of activated sludge

Level of pH, DO and EC were measured every day by single input multi-parameter digital meter (hQ30d, USA). A direct power supply was used for monitoring the current. All electrodes and also switch-timers were connected to the DC power supply (TES 6230) by distribution panel. Current density was measured based on level of current (A) which passed through anode surface area (m^2) [Eq. 3.1]. Pressure regulator and air flow meter were used to adjust the air pressure and flow rate respectively. Changes of sludge characteristics were assessed during the experiments. Standard Methods (APHA 1998) were used for the examination of initial values of activated sludge like: MLSS, MLVSS, TSS and VSS. For MLVSS measurement, the temperature of oven increases to 500^{o}C, and at this temperature, organic compounds can be expected to burn. The organic compounds will be converted to water and carbon dioxide as a gas form. In an activated sludge plant, nearly 80% of mixed-liquor may be organic (Barnes and Wilson, 1978). The analytical methods are shown in Table 3.9.

Table 3.9: Analyses of sludge properties

Sludge properties	Analytical method
pH	Analyzed by using HQ30d, multi- parameters meter (Hach, USA)
EC (µS/cm)	Analyzed by using HQ30d, multi- parameters meter (Hach, USA)
DO (mg/l)	Analyzed by using HQ30d, multi- parameters meter (Hach, USA)
Flocs mean size distribution	Analyzed by using LA-950V2 (Horiba, USA)
Total suspended solids (TSS)	Sludge filtered + drying at 105^{0}C (water and wastewater treatment, method 2540 D of standard method)
Volatile suspended solids (VSS)	After putting in desiccator, burning at 550^{o}C (water and wastewater treatment, method 2540 E of standard method)

SVI has been used for assessment of settling characteristics of sludge. In practice, SVI is the volume in milliliters occupied by one gram of the sludge in a mixed-liquor sample after 30 min of settling in the graduated cylinder. It is calculated as milligrams per liter (mg/l) (Metcalf and Eddy 1991). SVI of 150-200 mg/l is considered poor values, but in values of 50 mg/l and below, sludge volume index have a good condition for settling (Barnes and Wilson 1978). Analyzing the sludge should be carried out immediately in terms of assessment of flocs size and characteristics of sludge. To measure the flocs mean size distribution, samples of sludge were taken every two weeks from the electro-bioreactor to control and monitor the variations of the flocs size within the system. Particle size distribution was assessed by using Laser Scattering Particle size-distribution analyzer LA-950V2 (Horiba, USA). For this experiment, 10 ml of the sludge was taken from the electro-bioreactor. Then, before injecting few drops into a sampling bath, the sample was shacked for optimum analyzing of flocs size.

3.5.2 Sampling

During this experiment, many samples were taken from influent, from effluent, and from the sludge. To measure the value of phosphate, samples were taken twice per week from effluent. To measure the value of ammonium and nitrate, samples were taken four times per week from effluent. To measure the value of COD, samples were taken two times per week from effluent. The value of nutrient and COD were measured by TNT plus vials, DR 2800 and DRB 200 spectrophotometer from Hatch (USA Company), as shown in Table 3.10.

Table 3.10: Laboratory tests for assessments the concentrations of nutrients and COD

Elements	Laboratory kits	Conditions
Nitrate (NO_3^-)	TNT 835, LR for influent and effluent	Stored at +15 or +25 ºC. It can cover the nitrate in a range of 0.23 to 13.50 mg/l as NO_3-N or 1.00 to 60.00 mg/l as NO_3
Ammonia (NH_4^+)	TNT 832, HR for influent and effluent	Stored in refrigerator. It can cover the ammonia in a range of 2 to 47 mg/l as NH_3-N
Phosphorous (P)	TNT 844,HR for assessment of the influent	Stored at +15 or +25 ºC. It can cover the phosphorus in a range of 0.5 to 5.0 mg/l as PO_4-P or 1.5 to 15.0 mg/l as PO_4
	TNT 843, LR for assessment of the effluent	Stored at +15 or +25 ºC. It can cover phosphorus in a range of 0.05 to 1.5 mg/l as PO_4-P or 0.15 to 4.50 mg/l as PO_4
COD	TNT 822, HR for assessment of the influent	Stored at +15 or +25 ºC. It can cover the COD in a range of 20 to 1500 mg/l.
	TNT 821, LR for assessment of the effluent	Stored at +15 or +25 ºC. It can cover COD in a range of 3 to 150 mg/l

Chapter 4: Results and discussion

This experimentation was carried out in the laboratory for almost 7 months to obtain the optimum results in terms of removing nutrients and COD under various operational conditions. The influent concentrations of nitrate, ammonia, phosphorous and COD in both designs were mentioned in Chapter 3.2.2, Table 3.3. It was demonstrated that a superior quality of effluent can be achieved in one hybrid reactor without recirculation and without the addition of coagulants. Such effluent can be used successfully for water recovery. The first design of electro- bioreactor run for 21 weeks while second design of electro-bioreactor run for 6 weeks. It was proven that the SMEBR has an ability to remove nutrients to the desired level, while the conventional membrane bioreactor system (MBR) cannot remove nutrients to the acceptable level (Hasan 2012; Ibeid 2011; Elektorowicz *et al.* 2011). From a different perspective, Hasan (2012) found that in an optimum condition, MBR at pilot scale could remove 80.4% of COD, 97% of ammonia, and 59% of phosphorous, from raw wastewater. He found that by using SMEBR at the steady state condition, the removal of COD, ammonia, and phosphorous were 92%, 99% and 99% respectively. Also, Ibeid (2011) at lab scale found that 97% of COD and only 25% of phosphorus could be removed from synthetic wastewater by MBR system. He found that by using SMEBR, at the steady state condition, the removal of COD, nitrogen, and phosphorous were more than 95%, 97% and 99% respectively.

4.1 COD and nutrient removal in the SMEBR

4.1.1 COD removal

Carbon can be removed through biological activities (biodegradation) and flocculation (physicochemical process) in activated sludge. As it was mentioned in Chapter 2.4.3.1, aluminum ions can be converted to the polymeric compounds (cationic hydroxide (*i.e.*: aluminum hydroxide). As time passes by, eventually, they transform to the long chain of Al complexes. These compounds can adsorb colloids and other organic compounds. In the first and second design of the electro-bioreactor, COD was removed in an optimum rate (>97%), it might be derived from the highly performance of microbial activities. Microbial flocs were able to recover from the electrical shock in the electrical field under the specific operating condition (*i.e.*: current density was 21.92 A/m^2 under the exposer time of 5′-ON/15′-OFF).

In the first design of the electro-bioreactor, the maximum and minimum levels of COD removal were 99.5% and 97% respectively (Fig. 4.1).

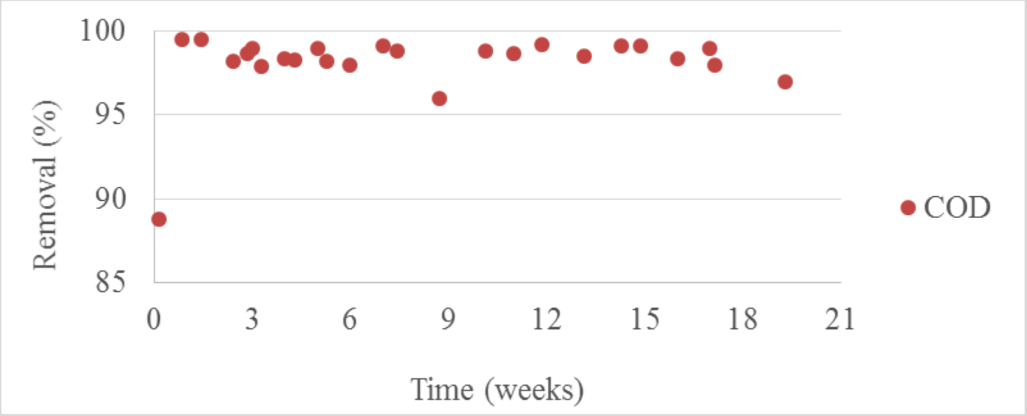

Fig. 4.1: COD removal percentage in the first design of the electro- bioreactor

The maximum level was achieved in week 2, at influent and effluent concentration of 749 mg/l and 3.41 mg/l respectively. The minimum level was achieved in week 20, at the influent and effluent concentration of 918 mg/l and 22.4 mg/l respectively (Fig. 4.2). Both concentrations are below the acceptable limit for effluent in Quebec.

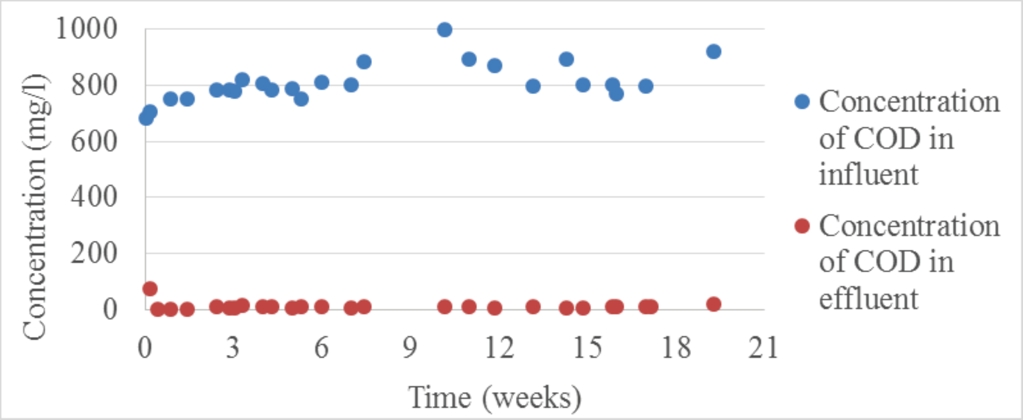

Fig. 4.2: COD concentration in the first design of the electro-bioreactor

In the second design of the electro-bioreactor system, the maximum and minimum levels of COD removal were 97.8% and 96.7% respectively (Fig. 4.3).

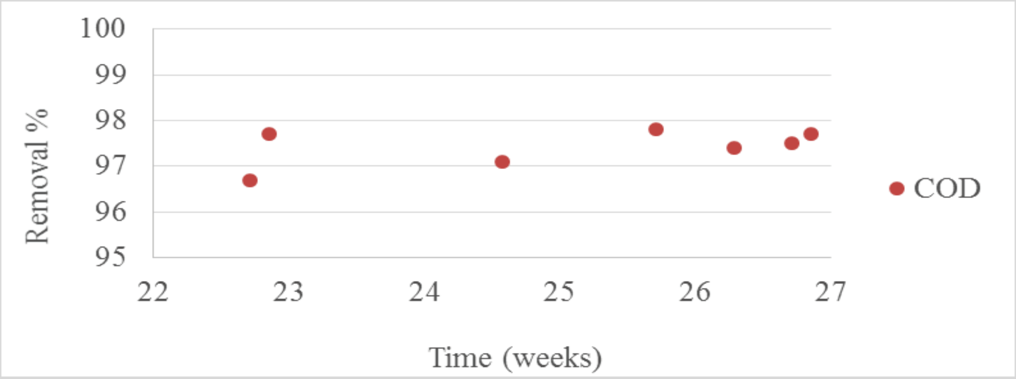

Fig. 4.3: COD removal percentage in second design of the electro- bioreactor

The SMEBR had high removal efficiency because of the ability of the system to coagulate the colloidal organic compounds or even those which had high molecular weight. The maximum level was achieved in week 26, at the influent and effluent concentration of 892 mg/l and 19.2 mg/l, respectively. The minimum level was achieved in week 23, at the influent and effluent concentration of 992 mg/l and 25 mg/l, respectively (Fig. 4.4).

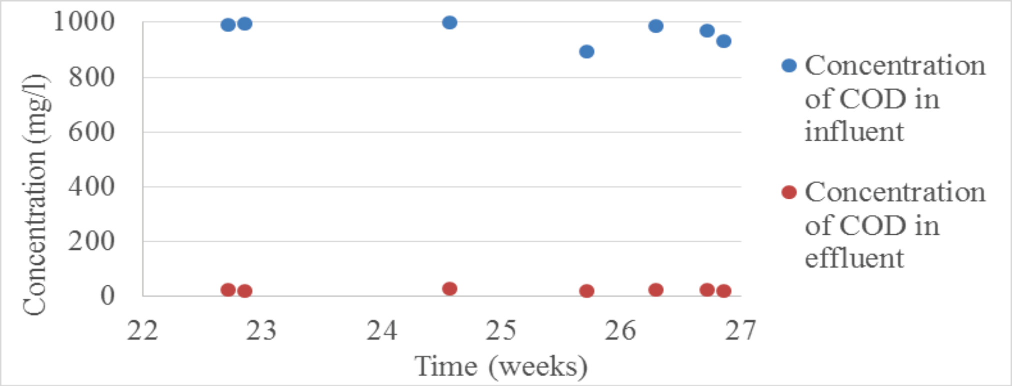

Fig. 4.4: COD concentration in the second design of the electro- bioreactor

It could be concluded from the Figs. 4.1 and 4.3, that the COD removal for both designs was not affected by the changing of current density.

4.1.2 Phosphorous removal

Phosphorus can be removed through the formation of $AlPO_4$ or phosphorus complexes with Al $(OH)_S$. In a reactor, aluminum ions have a reaction with free hydroxide ions (OH^-) to mostly make monomeric forms like Al $(OH)^{+2}$, Al $(OH)_2^{+1}$, and Al $(OH)^-_4$. These compounds can then be converted into polymeric forms such as: $Al_8(OH)^{+1}_{20}$, Al_{13} $(OH)^{+5}_{31.}$ These processes can be continued until a long chain of Al $(OH)_{(s)}$ is finally produced. Accordingly, phosphorus became a part of the suspended solids in the reactor that could be recovered after using membrane modules (liquid separated from solid). The biological removal of phosphorus and electro-deposition on electrode is also proven (Hasan 2012). However, the common membrane bioreactor system (MBR) cannot remove phosphorus to an acceptable level (Hasan 2012; Ibeid 2011 and Elektorowicz *et al.* 2011). In the first design of the electro-bioreactor, the maximum and minimum levels of phosphorus removal were 99.8 and 85 % respectively (Fig. 4.5).

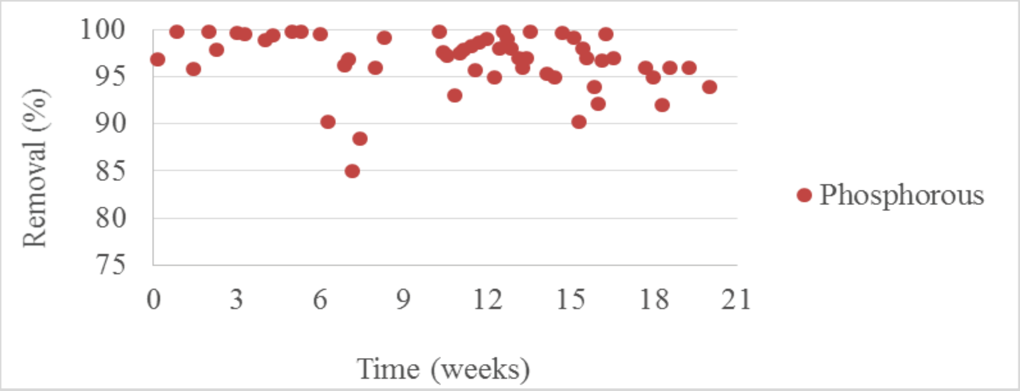

Fig. 4.5: Phosphorous removal percentage in the first design of the electro-bioreactor

The maximum level was achieved in weeks 1, 2, 5, 6, 11, 13 and 14. The minimum level (which is still high in comparison to a conventional MBR, based on Hasan (2012), MBR could remove only 59% of phosphorus) was observed in week 8, at the influent and effluent concentration of 16.3 mg/l and 2.44 mg/l, respectively (Fig. 4.6). In the week 8, the rate of phosphorous removal was reduced, it might be related to the corrosion of electrode, and therefore less aluminum was produced.

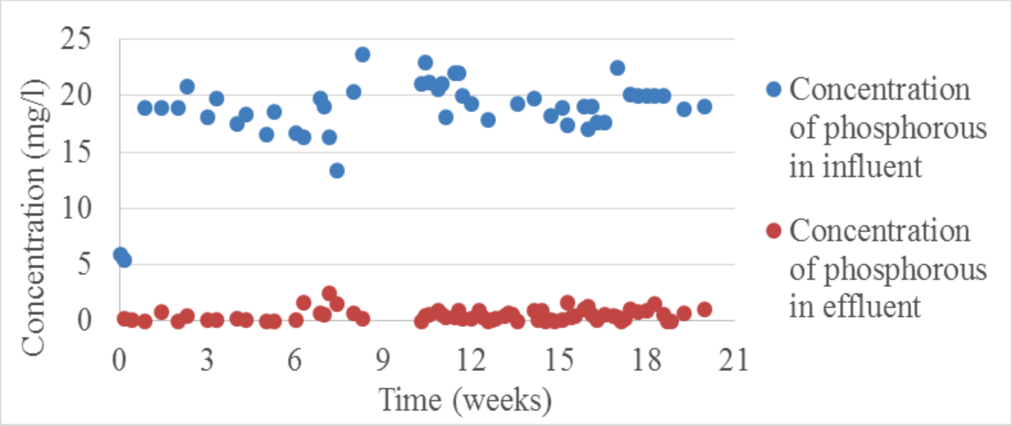

Fig. 4.6: Phosphorus concentration in the first design of the electro- bioreactor

In the second design of the SMEBR, under the operating condition of current density of 21.93 A/m^2, HRT of 12 h and exposure time of 5′-ON/15′-OFF, the removal percentage of phosphorus becomes higher than the first design of SMEBR. Dissolution of electrodes and generation of aluminum ions in the electro- bioreactor provided sufficient conditions to form complexes with phosphorus. Results showed that the maximum level (99.8%) of phosphorus removal was achieved in week 27 (Fig. 4.7).

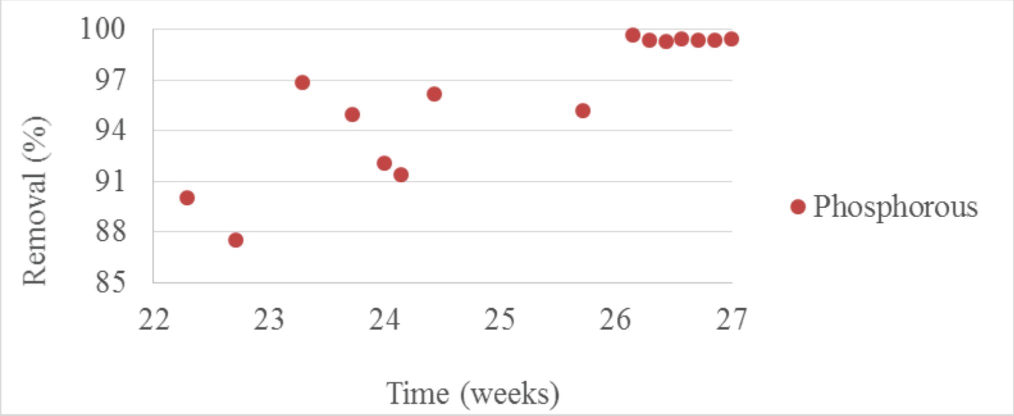

Fig. 4.7: Phosphorous removal percentage in the second design of the electro- bioreactor

99.8 % of phosphorous can be removed at the influent and effluent concentration of 15.3 mg/l and 0.04 mg/l, respectively (Fig 4.8).

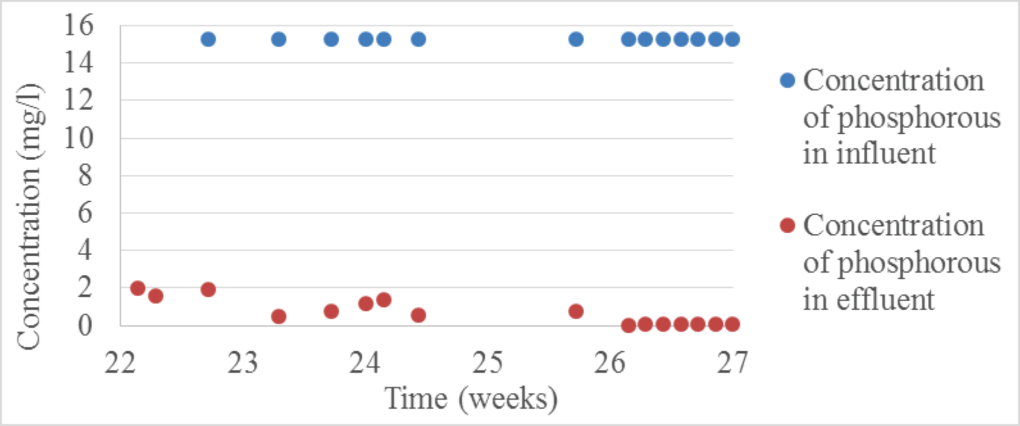

Fig. 4.8: Phosphorous concentration in the second design of the electro- bioreactor

It could be concluded that both designs of SMEBR could remove phosphorus at optimum levels through the formation of phosphate hydroxide complex. Moreover, under the exposure of direct current field, phosphorus could be deposited on the surface of electrodes as also confirmed by Hasan (2011) or form complexes with the Al species in the sludge liquor generated by the electrocoagulation process.

4.1.3 Ammonia removal

First design of SMEBR showed nearly complete nitrification after week 12. After this time, the ammonia concentration was high; this means that the bioreactor still needed some modification to achieve the optimum level before the new design of SMEBR (week 22 to 27). The operating conditions after the week 12 can be summarized as follows: dissolved oxygen concentration was less than 5 mg/l (as time passed by the DO concentration reduced to 3 mg/l), current densities were 28.93, 18.5185 and 21. 92 A/m^2 and the exposure mode was 5′-ON/20′-OFF. As shown in Fig. 4.9, the maximum and minimum levels of ammonia removal were 97.1 and 61.5 %, respectively.

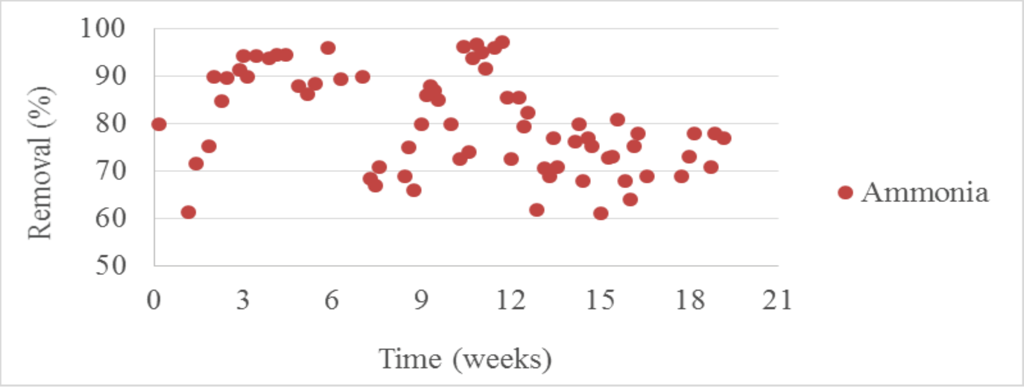

Fig. 4.9: Ammonia removal percentage in first design of the electro-bioreactor

The maximum level was achieved in week 12, at the influent and effluent level of 18.2 and 0.524 mg/l, respectively. The minimum level was achieved in the week 2, at the influent and effluent level of 17.31 and 6.62 mg/l, respectively (Fig. 4.10).

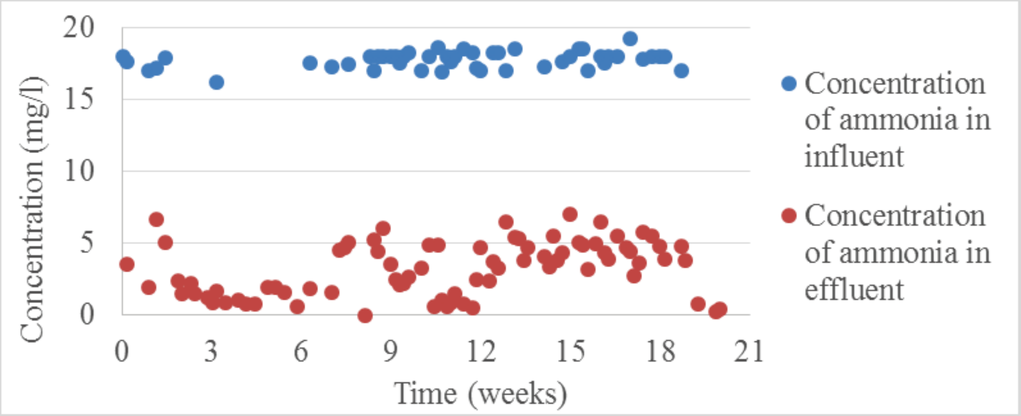

Fig. 4.10: Ammonia concentration in the first design of the electro-bioreactor

As shown in Fig. 4.11, in the second design of the SMEBR (week 22 to 27), the maximum and minimum levels of ammonia removal was 99.9% and 96.2%, respectively.

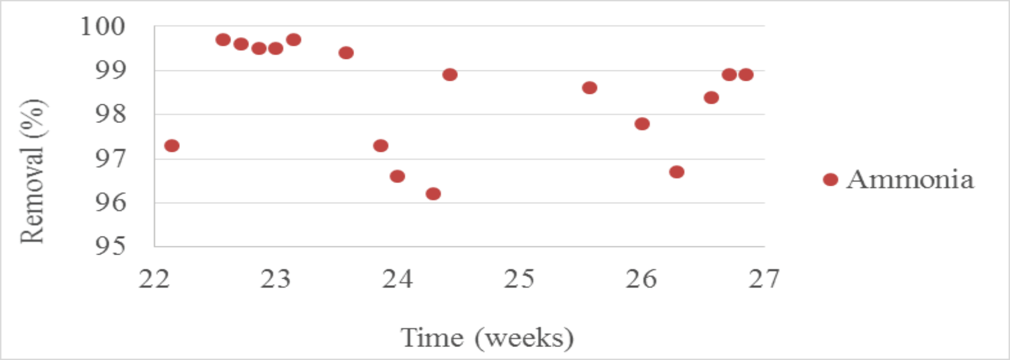

Fig. 4.11: Ammonia removal percentage in second design of the electro- bioreactor

The maximum level was achieved in week 22, at the influent and effluent concentration of 16.80 and 0.014 mg/l, respectively. The minimum level was achieved in week 25, at the influent and effluent concentration of 17.40 and 0.65 mg/l, respectively (Fig. 4.12).

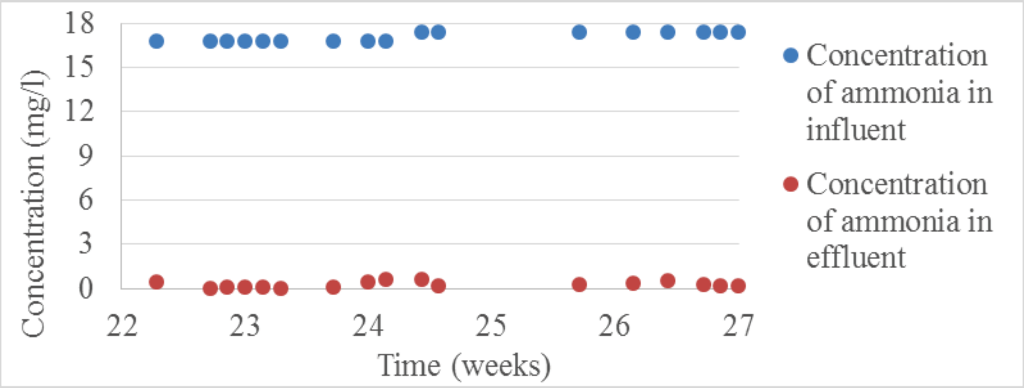

Fig. 4.12: Ammonia concentration in the second design of the electro- bioreactor

The results of the first design of the SMEBR showed that the loading of ammonium (NH_4^+) into the electro-bioreactor was higher than their potentials for nitrification processes under the specific operating conditions (DO = 8-3 mg/l and current density < 19 A/m^2). More than 99% of ammonia and almost 100 % of nitrate were removed when the DO concentration was around 3 mg/l, current density was 21.93 A/m^2 and exposure time was 5′-ON/15′-OFF. In anoxic conditions, the concentration of oxygen gas decreased while the level of nitrate or other oxidized nitrogen compounds increased. Thus, nitrate concentration plays an important role in anoxic transformation. In the denitrification process, nitrite is temporarily accumulated until the concentration of nitrate decreases. The highest reduction of nitrate took place when the electro-bioreactor provided good conditions for denitrifiers by having minimal dissolved oxygen concentrations. Results showed that the second design of SMEBR system allows almost complete nitrification of ammonium and denitrification of nitrate in one reactor.

4.1.4 Nitrate removal

Nitrification and denitrification conditions in the electro-bioreactor play an important role for nitrogen removal. Conditions of SMEBR in terms of DO could help nitrifiers and denitrifiers bacteria to transform ammonium into nitrogen gas. The complete removal of nitrogen required the fluctuation of oxygen between the anoxic/anaerobic and aerobic conditions. The complete transformation of nitrogen into the nitrogen gas in one reactor relies highly on the level of oxygen at anoxic/anammox and the aerobic condition. Exposure mode, current density and voltage gradient should be adjusted continuously with other operating parameters like HRT and MLSS. Working at low MLSS required high dissolved oxygen levels, since the diffusivity of electrons in the electro-bioreactor is high. In this case, it might be easier to interrupt the aim role of the oxygen as an electron acceptor. In contrast, at high MLSS, low dissolved oxygen levels are required because the movement of the electrons is hindered by the low diffusivity of the electro-bioreactor. As shown in Fig. 4.13, the maximum and minimum levels of nitrate removal for the first design are 17 and 69 % respectively.

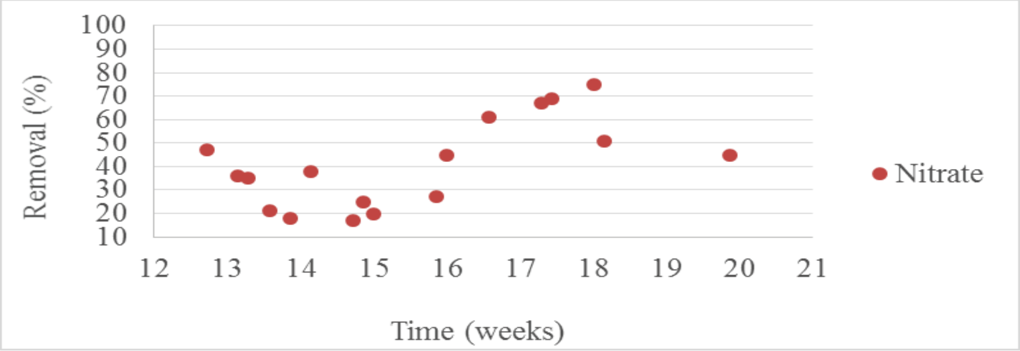

Fig. 4.13: Nitrate removal percentage in first design of the electro- bioreactor

As shown in Fig. 4.14, the minimum level of nitrate was achieved in week 15, at the influent and effluent concentration of 0.207 and 0.177 mg/l, respectively. The maximum level was achieved in week18, at the influent and effluent concentration of 0.266 and 0.082 mg/l, respectively.

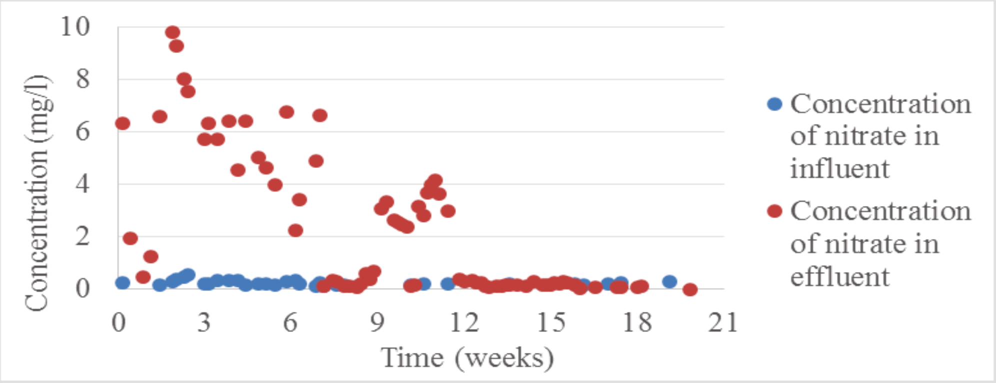

Fig. 4.14: Nitrate concentration in the first design of the electro-bioreactor

In the second design of SMEBR, as shown in Fig. 4.15, the maximum and minimum levels of nitrate removal are 100% and 95.6%, respectively.

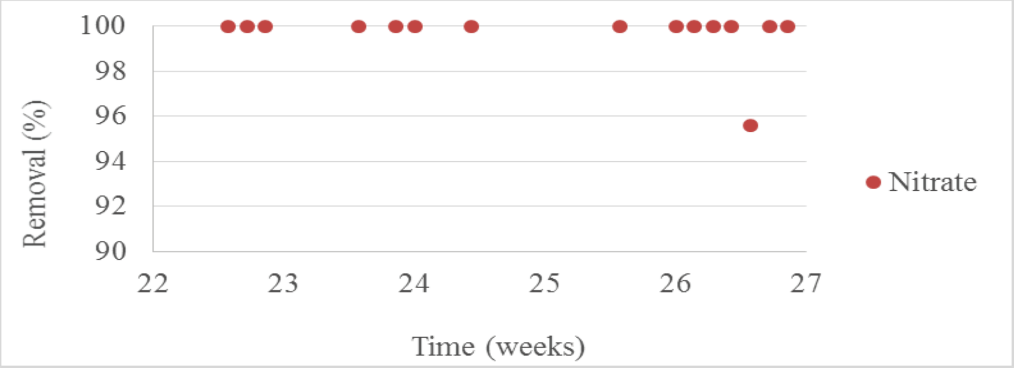

Fig. 4.15: Nitrate removal percentage in second design of the electro-bioreactor

As shown in Fig. 4.16, the maximum level was achieved in week 27, at the influent and effluent concentration of 0.228 and 0 mg/l, respectively. The minimum level was also achieved in week 27, at the influent and effluent concentration of 0.228 and 0.010 mg/l respectively.

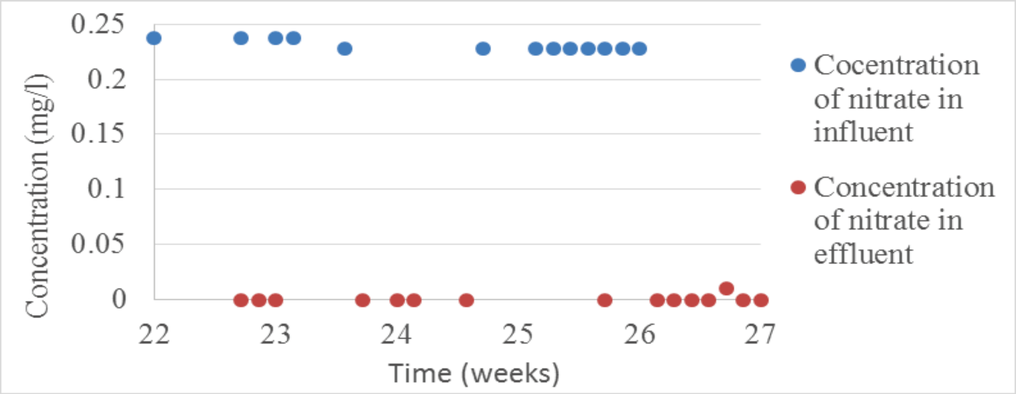

Fig. 4.16: Nitrate concentration in the second design of the electro-bioreactor

The complete removal of nitrate required fluctuation of level of dissolved oxygen between the anoxic/aerobic and aerobic conditions.

4.2 Overall results for first and second design of the electro-bioreactor

Overall, results achieved from the first design of SMEBR system (during the five months of investigations between weeks 1 to 21 are as follows (Fig. 4.17): the average percentage removal of ammonia was 79%; the average percentage removal of nitrate was 43%; the average percentage removal of phosphorus was 92%; the average percentage removal of COD was 98%.

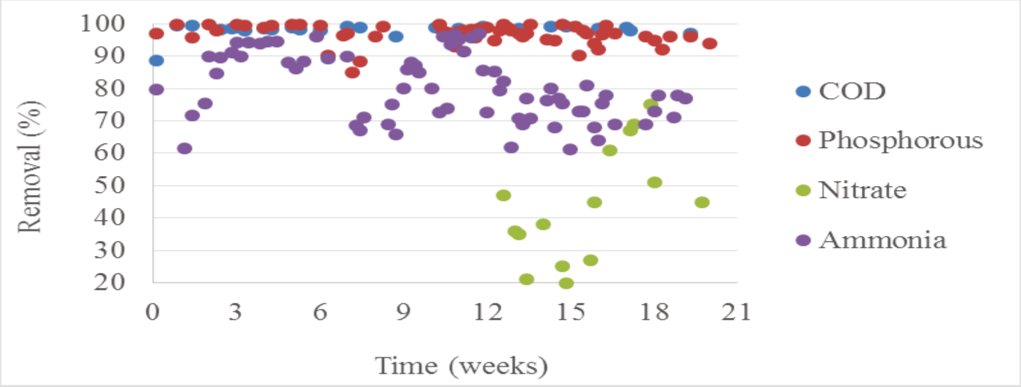

Fig. 4.17: Removal of nutrients and COD in the first design of electro-bioreactor

However, during the last 2 months of investigation of SMEBR (weeks 22 to 27) the following results were achieved (Fig. 4.18): the average percentage removal of ammonia was 98%; the average percentage removal of nitrate was almost 100%; the average percentage removal of phosphorus was 93%; and the average percentage removal of COD was 97%.

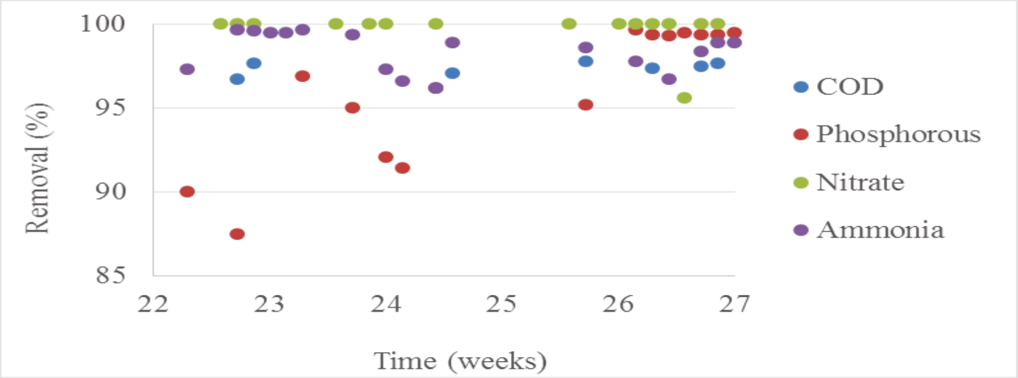

Fig. 4.18: Removal of nutrients and COD in the second design of electro-bioreactor

Fig. 4.19 represents the combined results of nutrient and COD removal in both the first and second designs of the electro-bioreactor. This new configuration of SMEBR demonstrates the high efficiency of nutrients removal as well as highly efficient COD removal. One of the important factors for removing ammonia and nitrate is concentration of dissolved oxygen.

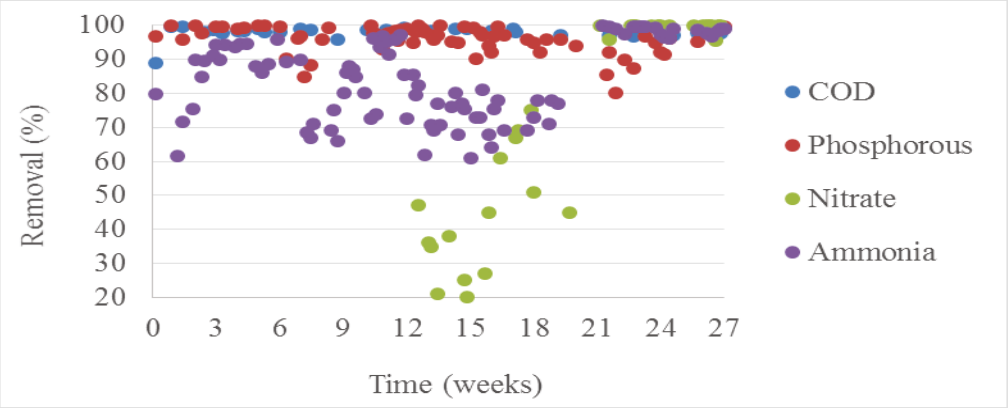

Fig. 4.19: Nutrients and COD removal in first and second designs

As shown in Fig. 4.20, the DO level around 3 mg/l provides a better condition for denitrification process; up to 90% of ammonia and nitrate was removed.

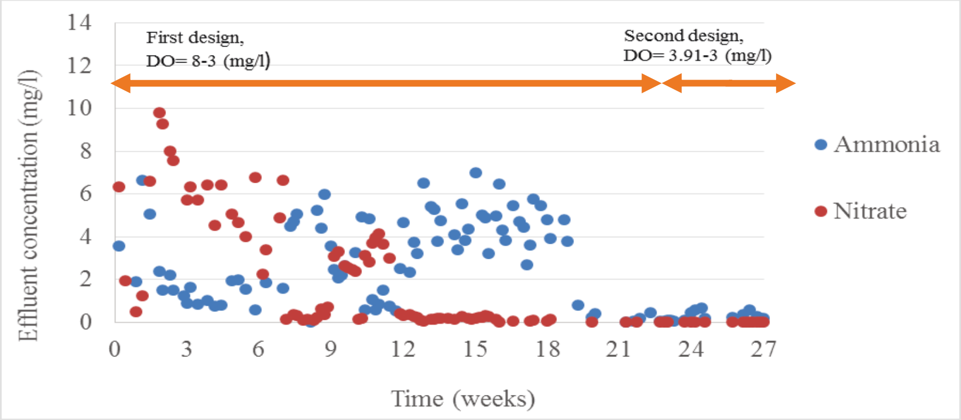

Fig. 4.20 Effluent concentration of nitrate and ammonia in the first and second design

The comparison of nitrate and ammonia removal percentage is illustrated in Fig. 4.21. Nitrogen was removed by transformation of ammonia into nitrogen gas through nitrification and denitrification processes. Removal rate increased because of the anammox process, presence of denitrifier bacteria, and operating conditions (DO level and electrical exposer time). The SMEBR system showed an ability to almost complete nitrification of ammonium and denitrification of nitrate.

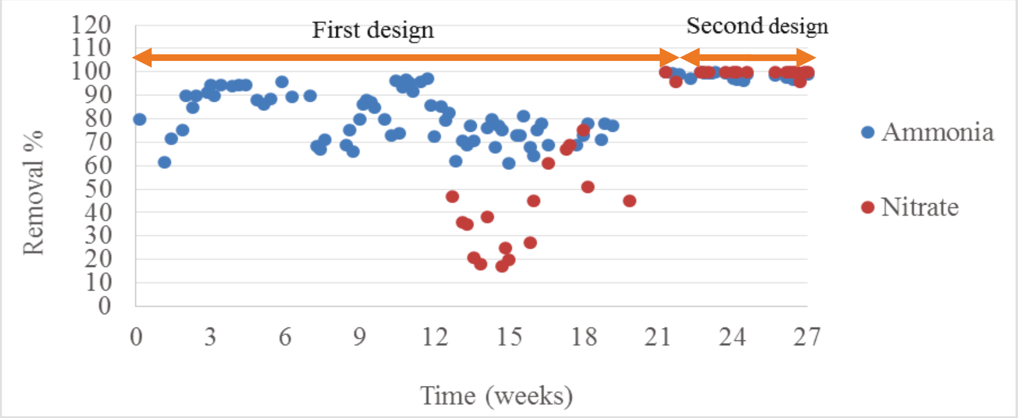

Fig. 4.21: Percentage removal of ammonia and nitrate in the first and second design

Based on Figs. 4.17 to 4.21, the following statements are concluded:

- Under the specific operating condition (i.e.: current density of 21.93 A/m2; HRT of 12 h; SRT of 15 days; electrical exposer time OFF/ON (5'-ON/15'-OFF)), sufficient amount of aluminum ions are produced for removing phosphorus, and enough electrons are produced for transforming nitrogen into the gas form.

- The results were strongly related to the dissolved oxygen (DO) concentration. A high fluctuation of DO concentration in the system caused several errors and also it has an influence on nutrient's removal levels. The effluent's value of DO, P, and N measured 3 to 5 times in a day. In this study, the high variation of DO appears in the week 10, and 19. For example, in the week 19, the variation of DO concentration was between 3.81 to 4.71 mg/l. The effluent concentration of phosphate was between 0.578 to 2.37 mg/l, and the standard deviation of phosphate was 0.89. The effluent concentration of ammonium was between 3.99 to 4.32 mg/l, and the standard deviation of ammonium was 0.13. The effluent concentration of nitrate was between 0.027 to 0.216 mg/l, and the standard deviation of nitrate was 0.07. Best results of the lab tests were achieved in week 26 and 27, when the concentration of DO was stabilized (3.15 mg/l). During this period, the effluent concentration of ammonium was between 0.18 to 0.57 mg/l (standard deviation of 0.19). The effluent mean concentration of nitrate was zero mg/l. The effluent mean concentration of phosphate was 0.040 to 0.073 mg/l (standard deviation of 0.015).

- Electro-bioreactors run with three (first design) and two (second design) pair of electrodes connected in parallel. Energy costs at lab scale (according to calculations in Appendix B, 7.2) for treatment wastewater, the energy per each cubic meters has been estimated for 0.6 kWh this is based on the second design of lab scale experiment. Considering, the price for 1 kWh is cost 3 cents and 5 cents for industrial and domestic usage, respectively. Then, the energy cost per day has been estimated for 0.03$/ m3. d. However, pilot tests with one operation unit, showed total energy around 1 kWh/m^3 (Hasan 2011).

4.2.1 Relationship between total current and nutrient removal percentage

Changes of nutrients and COD concentration, as affected by different levels of total current, are shown in Figs. 4.22 to 4.27. Fig. 4.22 represents a relationship between current density and removal percentage in the first stage. A current of 0.5 A was applied to the system from week 1 to week 7. Because of the corrosion of the anode in week 6, the current became less than 0.5 A. Fig. 4.23 represents a relationship between current density and removal percentage in the second stage. A current of 0.8 A was applied to the system from week 8 to week 10. The current turned off for about 3 days, because the effluent concentration of ammonia became high.

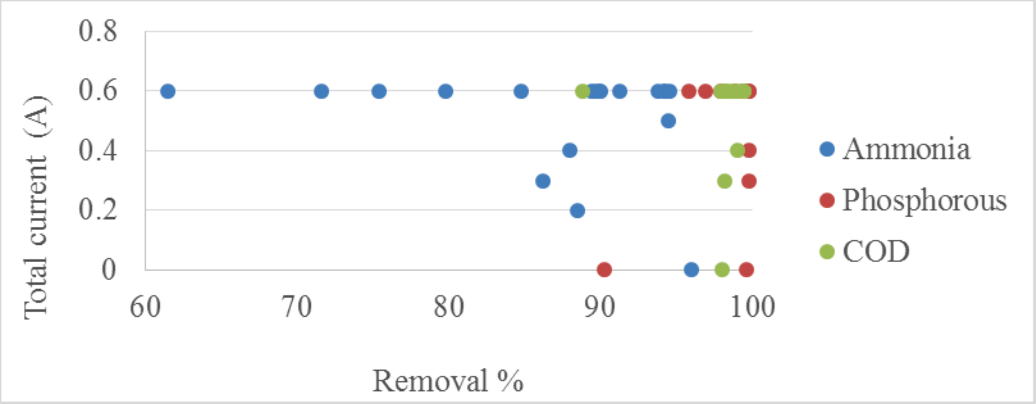

Fig. 4.22: Removal percentage in the first stage (week 1 to 7, total current of 0.5 (A), current density of 7.71 (A/m^2))

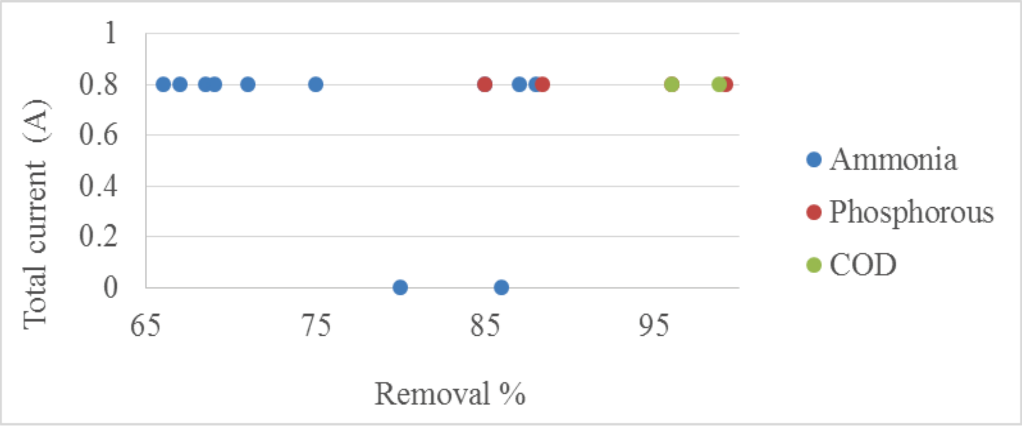

Fig. 4.23: Removal percentage in the second stage (week 8 to 10, total current of 0.8 (A), current density of 12.34 (A/m^2))

Fig. 4.24 represents a relationship between CD and removal percentage in the third stage, at the current of 1 A, from week 11 to mid-week 12. Fig. 4.25 shows the stage of the system in a current of 2.5 A, from mid-week 12 to mid-week 18.

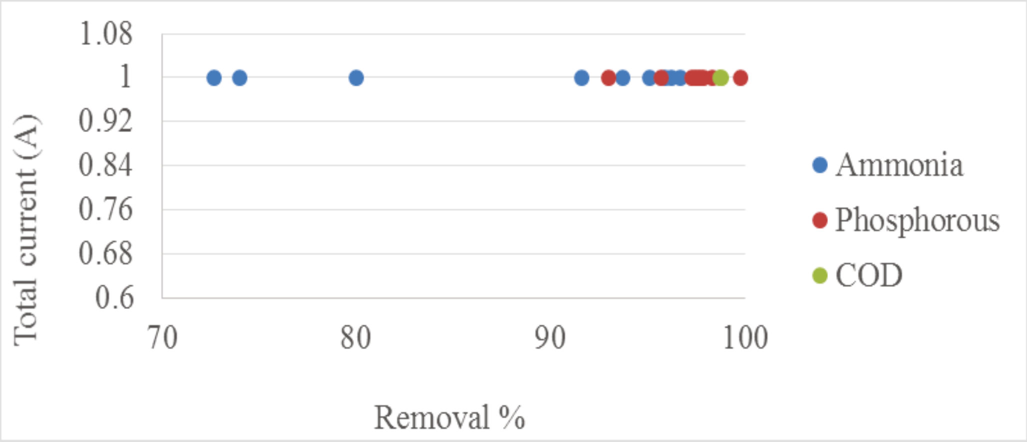

Fig. 4.24: Removal percentage in the third stage (week 11 to mid of the week 12, total current of 1 (A), current density of 15.43 (A/m^2))

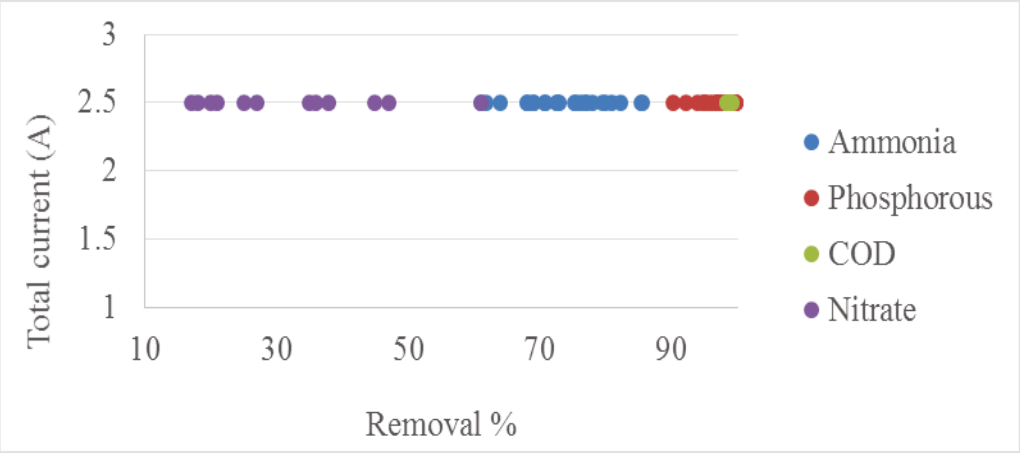

Fig. 4.25: Removal percentage in the fourth stage (week 12 to mid of the week 18, total current of 2.5 (A), current density of 28.92 (A/m^2))

Fig. 4.26 represents a relationship between current density and removal percentage in the fifth stage, at the current of 1.2 A, from mid-week 18 to week 21.

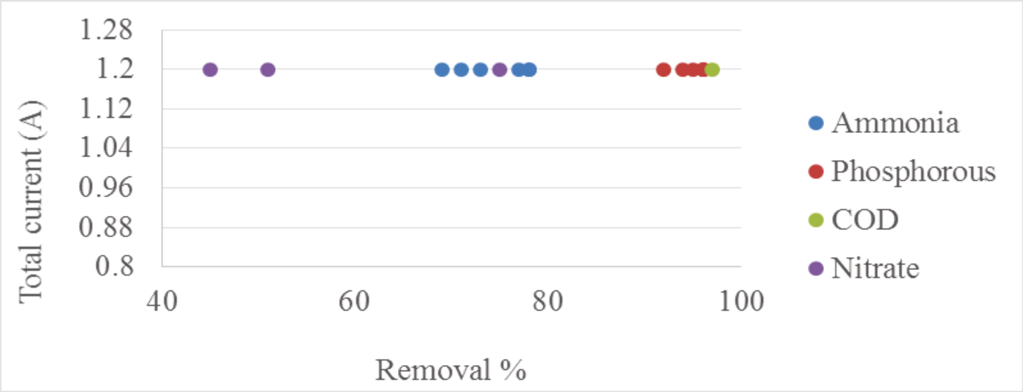

Fig. 4.26: Removal percentage (total current of 1.2 (A), CD of 18.51 (A/m^2))

As seen in Fig. 4.27 (stage number 6, weeks 22 to 27), a better result was achieved in terms of removing nutrient and COD. This figure shows that, under the operating conditions of a total current of 1.5(A), exposure time of 5′-ON/15′-OFF, and the current density of 21. 92 A/m^2, enough Al^{+3} ions were produced for removing phosphorus. The figure also shows that the electro-bioreactor has the ability for almost complete nitrification of ammonium and denitrification of nitrate.

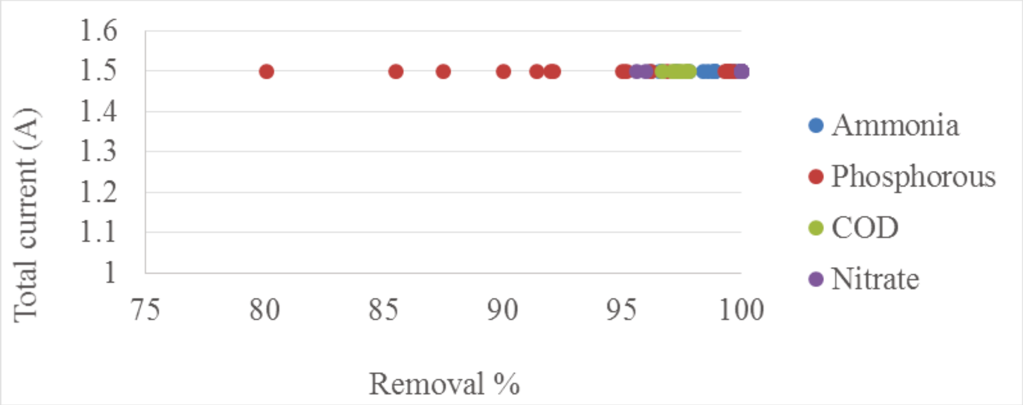

Fig. 4.27: Removal percentage (total current of 1.5 (A), CD of 21.92 (A/m^2))

4.3 Scale up the SMEBR

4.3.1 Impact of current density over time on SMEBR

In this study, the impact of current density in the first and second designs of the SMEBR is considered by understanding the relationship between the total current and the nutrient removal percentage. The value of the current which passes through electrodes in the SMEBR is commensurate to the concentration of soluble ions, the concentration of suspended solids and the voltage which is applied to the system. These three factors determine the final value of the current. Therefore, the current which was used in this experiment was a base principle for effluent quality. For removing nutrients and COD, the current was changed 6 times (Fig. 4.28). Stages 1 to 5 are related to the first design of the electro-bioreactor; and stage number 6 is for the second design of the SMEBR. The first and last total currents (TC) were 0.5 and 1.5 A, respectively.

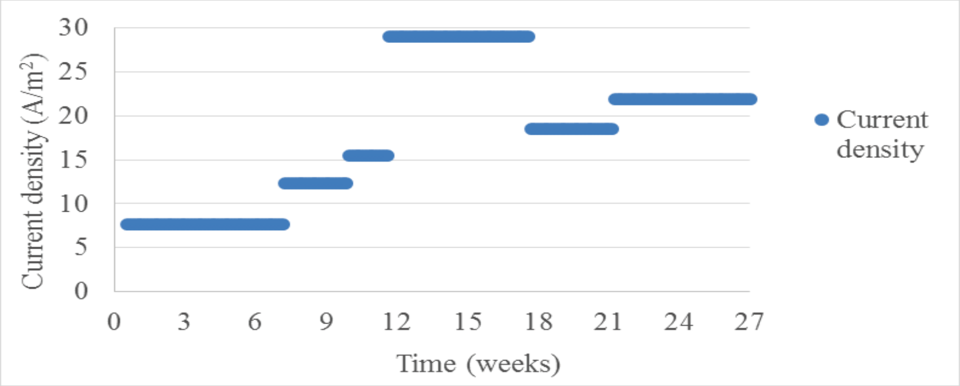

Fig. 4.28: Different levels of current density used in the SMEBR system (stage 1 to 6)

4.3.2 Impact of effluent concentration over ratio of total current per volume in SMEBR

Scale up the SMEBR can be based on the ratio of total current per volume (TC/V). For example, in the first design of electro-bioreactor, in the presence of 0.11 mg/l of dissolved oxygen, nitrate concentration was 0.03 mg/l with the ratio of total current per volume of 0.11 (current per volume or Ohms/l) (Fig. 4. 29).

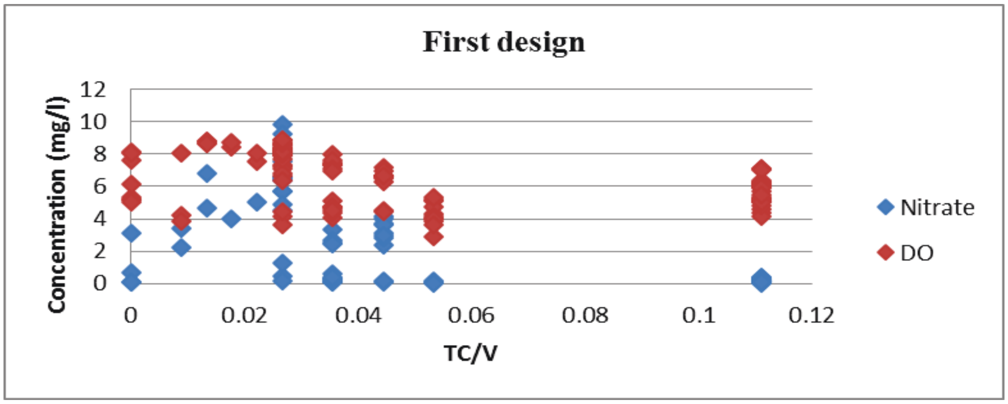

Fig. 4.29: Scale up parameters (nitrate): best in total current per volume equal to 0.11 (ohms/l)

The second design confirmed such ratio, since in the present of 0.03 mg/l of nitrate concentration, when dissolved oxygen was 0.1 mg/l and the ratio of total current per volume was 0.1 (ohms/L) (Fig. 4. 30). Scale up the SMEBR based on the second design with a ratio of total current per volume equal to 0.1 (ohms/L) confirms adequacy of this approach (another examples are provided in Chapter 7, Appendix C).

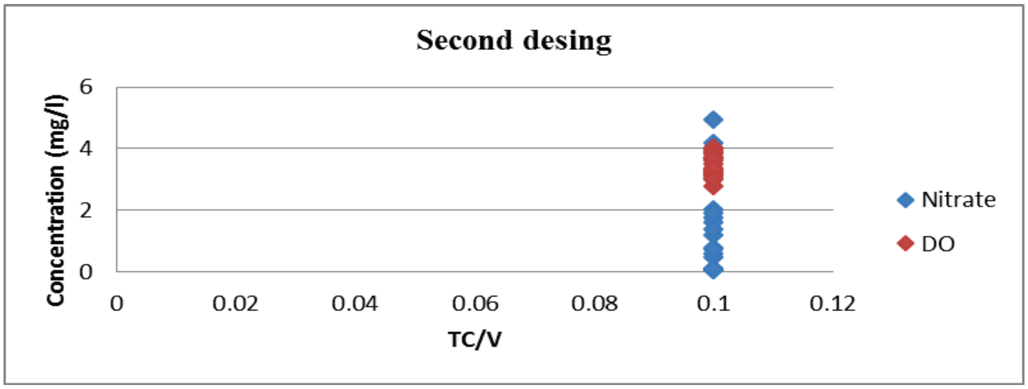

Fig.4. 30: Scale up parameters (nitrate): best in total current per volume equal to 0.11 (ohms/l)

4.3.3 Impact of ratio of total surface area of anode per volume over time in SMEBR

The volume of the electro-bioreactor tank for the first design of the SMEBR is 22.5 liters with 3 parallel electrodes, while the electro-bioreactor tank for the second design of SMEBR has a 15 liter volume with 2 parallel electrodes. The submerged anode surface area for the first design is 216 cm^2 or 0.0216 m^2. This number should be multiplied by 3 because 3 electrical unit were submerged into the tank. For this part, current density ranged from 7.71 to 18.51 A/m$^{2.}$ The submerged anode surface area for the second design was 342 cm^2 or 0.0342 m^2; this number should be multiplied by 2 because 2 electrical unit were submerged into the tank. For this part, current density was 21.92 A/m^2. The ratio of total anode surface area per volume for the first and second design of electro-bioreactor was the same: between 3.95 ± 0. 61(m^2/m^3).

4.4 Sludge characteristics

4.4.1 Relationship between current density and flocs size

Converting the small flocs into the large ones (flocculation) is mainly accomplished by reducing the repulsive forces between the flocs; this is achieved by reducing the zeta potential. Gradually, as time passed, the size of flocs decreased because of the removal of bound water by electroomosis (Ibeid 2012). Accordingly, the current density should be strong enough to cause flocculation. It should be noted that the maximum PSD showed from week 8 to 9 due to the increasing of current density and the possibility of anode corrosion in the bioreactor. Once the flocs were affected by interrupted adequate current, bound water was removed through electroosmosis and consequently, floc size reduced. Fig. 4.31 (“a” and “b”) show that microbial flocs mean particle size distribution (PSD) reduced over time.

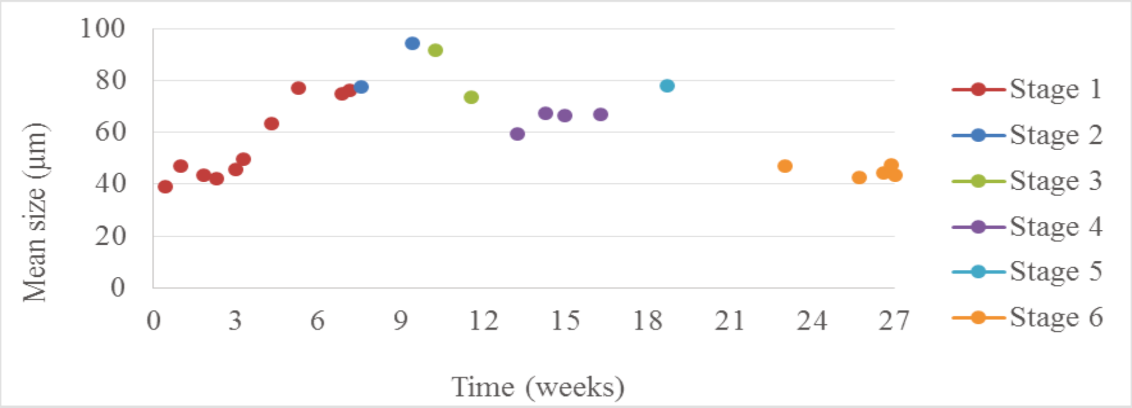

Fig. 4.31a: Variation of flocs size in all stages

In the electro-bioreactor, several factors have direct or indirect effects on microbial growth. It was assumed that microbial growth can be affected in function of fluctuation of current density. A higher electrical field means the higher aluminum ions enter into the system. However, there are different forms of aluminum in the electro-bioreactor. Some of them are deposited at the cathode. Feeding the electro-bioreactor by the synthetic wastewater can affect the level of soluble salts and ions over time.

Wei *et al.* (2011) proved that in a current density above 20A/m^2, microbial activities and COD removal decrease. The MLSS concentration is also related to aluminum production from the anode, the presence in sludge and the deposit on the cathode.

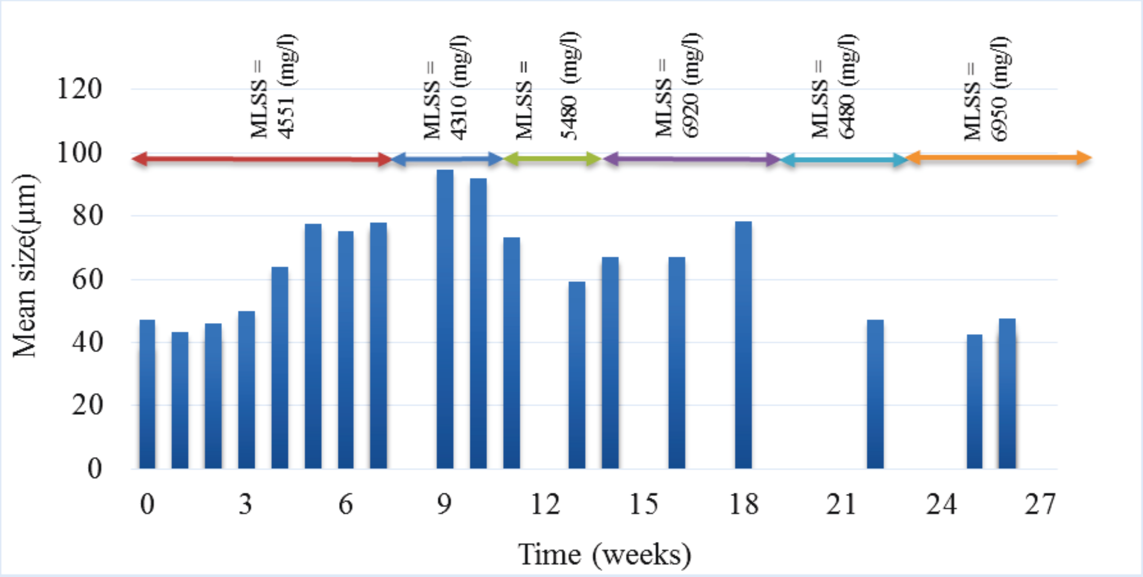

Fig. 4.31b: Variation of flocs size in the first and second design of the electro-bioreactor over average MLSS

The size of flocs in the electro-bioreactor was changed over the operating time following the level of the current density. Low current density of 7.7A/m^2 to 18.5 A/m^2 in the first design of the electro-bioreactor showed a slight increase of flocs size at the average MLSS of 4551 mg/l (Fig. 4.30). In week 10, because of the heavy corrosion of aluminum anode, the size of flocs slightly increased. Conversely, at the current density of 21.93 A/m^2 in the second design of the electro-bioreactor (Fig. 4.31), the mean particle size distribution (PSD) of flocs size decreased at the average MLSS of 6950 mg/l. The variation of current in the first design of the electro-bioreactor is shown in Fig. 4.32:

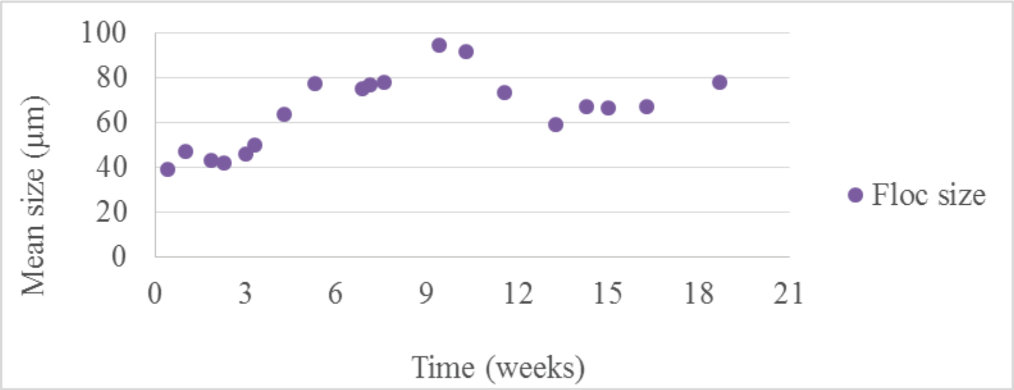

Fig. 4. 32: Variation of flocs size in the first design of the electro-bioreactor at current density range of 7.71 A/m^2 to 18.52 A/m^2

- The first stage: current density of 7.71 A/m^2 (week 1 to week 7). In this stage, the size of the floc increased because the electrical density increased. The reactions around the anode intensified because aluminum ions were dissolved into the system.

- The second stage: current density of 12.34 A/m^2 (week 8 to week 10). In this stage, the size of the floc slight increased because aluminum ions produced more. Flocculation of aluminum ions accelerated.

- The third stage: current density of 15.43 A/m^2 (week 11 to mid-week 12). In this stage, the size of the floc slightly decreased. Around week 10, due to the electroomosis phenomena, bound water (presented between flocs) was removed, therefore, flocs became smaller.

- The fourth stage: current density of 28.93 A/m^2 (mid-week 12 to mid- week 18). In this stage, the size of the floc slight increased because the system involved to a high dance colloidal system and electrokinetic phenomena occurred. More aluminum ions presented into the electro-bioreactor system.

- The fifth stage: current density of 18.51 A/m^2 (mid-week 18 to week 21). In this stage, the size of the floc slightly decreased, due to the electroomosis phenomena. The water which is presented between flocs was removed. Therefore, flocs became smaller.

The above current levels were applied to the system with the exposer time of 5 min ON and 20 min OFF, from week 1 to week 21. In that period, the minimum mean PSD was 35 µm and the maximum mean PSD was 95 µm.

In the second design of electro-bioreactor, the current density was 21.92 A/m^2 (from week 22 to week 27) with the exposer time of 5 min ON and 15 min OFF. In that period, the minimum mean PSD was 40 µm and the maximum mean PSD was 50 µm (Fig. 4.33).

- The sixth stage: current density of 21.92 A/m^2 (week 22 to week 27).). In this stage, the size of the floc slightly decreased, due to the electroomosis phenomena. After 27 weeks, the system became strong enough to withstand to the electrical shocks. The water which is presented between flocs was removed. Therefore, flocs became smaller (Fig. 4.33).

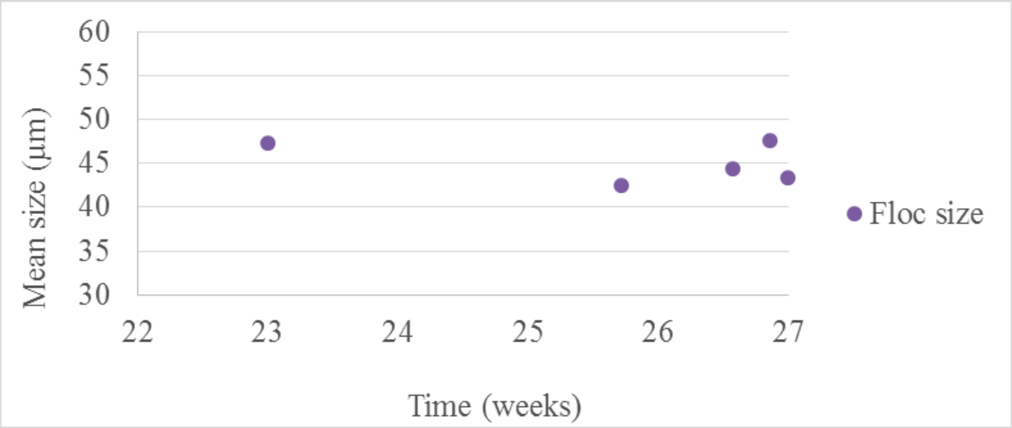

Fig. 4.33: Variation of flocs size in the second design of the electro–bioreactor at current density of 21.92 A/m^2

Chapter 5: Conclusions, contributions and future works

5.1 Conclusions

- The SMEBR (submerged membrane electro-bioreactor, patented by Elektorowicz *et al*. 2011) showed removal of nitrogen, phosphorus and carbon in one vessel for water recovery, due to the combination of biodegradation, membrane filtration and electrokinetic phenomena.
- In this study, SMEBR under the following operating condition had an ability to remove nutrients and COD: current density of 21.93 A/m^2; HRT of 12 h; SRT of 15 days; electrical exposer time OFF/ON (5'-ON/15'-OFF) These conditions were good enough to produce sufficient amount of aluminum ions for removing phosphorus and producing enough electrons for transforming nitrogen into the gas form.
- First design of electro-bioreactor was designed with 3 membranes and 3 electrodes in one rectangular bioreactor tank. According to results for first design, the average removal percentages of ammonia, nitrate, phosphorous and COD were: 79 %, 43%, 92% and 98% respectively.
- Second design of electro-bioreactor was designed with 2 membranes and 2 electrodes in one rectangular tank. The results proves SMEBR had an ability to optimally remove of nutrients (N and P) and COD from wastewater. The following results showed the average removal percentages of ammonia, nitrate, phosphorus and COD. They were 98%, almost 100%, 93% and 97% respectively.
- Nitrogen was removed by transforming ammonia into nitrogen gas through nitrification and denitrification processes. Removal rate might increase because of the anammox process. The SMEBR system showed an ability to almost complete nitrification of ammonium and denitrification of nitrate.
- Phosphorus was removed by the formation of aluminum phosphate since the electrical parameters were adjusted to produce sufficient aluminum ions from the anode.
- Carbon was removed through biodegradation and flocculation in the electro-bioreactor.
- For scaling up the SMEBR the ratio of total current per volume (TC/V) is suggest to be between 0.1 to 0.11 in the steady state conditions.

- The optimum level of dissolved oxygen (DO) for removing nitrogen found to be around 3 mg/l.
- The energy estimated based on the lab scale electro-bioreactor was 0.6 kWh/m^3.
- Effluent of SMEBR can be considered as a water recovery (depends on usage). It can be used for agriculture, some industries, or even for drinking water with a different membrane.

5.2 Contributions

- Designing of new electro-bioreactor systems with multiple electrical units and multiple membrane modules.
- Demonstrating that the SMEBR system can be scaled up, since new configuration with multiple electrical units and membrane modules confirmed high removal of COD and nutrients.
- Demonstrating for the first time that a multi membrane system can completely remove of C, P, and N in one hybrid reactor.

5.3 Future works

- Testing different sources of raw wastewater by using electro-bioreactor technology.
- Investigating better mixing processes in all zones of the electro-bioreactor.
- Studying more about the relationship between the influent concentrations of carbon and nitrogen, with the objective of achieving a complete denitrification process.
- Studying more about the biological nitrogen removal by anammox process.

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Chapter 7: Appendices

7.1 Appendix A: Experimental pictures

Fig. 7.1: Experimental set-ups of SMEBR system

Fig.7.2: Ultrafiltration ZeeWeed-1 (GE, Canada)

7.2 Appendix B: Power and cost estimation method for bench scale SMEBR system

For calculation the SMEBR's power (P) cost several factors are considered (i.e.: resistivity (ρ), electrical conductivity (σ), resistance (R), length between the electrodes (L), cross section of material (A), electric potential (V), and electric current (I)).

Resistivity is calculated based on the following formula: $\sigma = 1/\rho$

Resistance is derived from resistivity:

$R=\rho L/A$

Where: R= Resistance (ohms)

L= Length between the electrodes (cm)

A= Cross section of material (cm^2)

σ = Conductivity (Siemens/cm)

ρ= Resistivity (ohm.cm)

Voltage can be calculated from resistance formula:

$V=IR$

Where: V= Electrical potential (voltage)

I= Electrical current (amperes)

Finally, power as watts calculates from the following formula:

$P=VI$

7.3 Appendix C: Figures for scale up the system

The other examples for scale up the SMEBR based on the ratio of total current per volume (TC/V) are mentioned in Figs. 7.3 to 7.4.

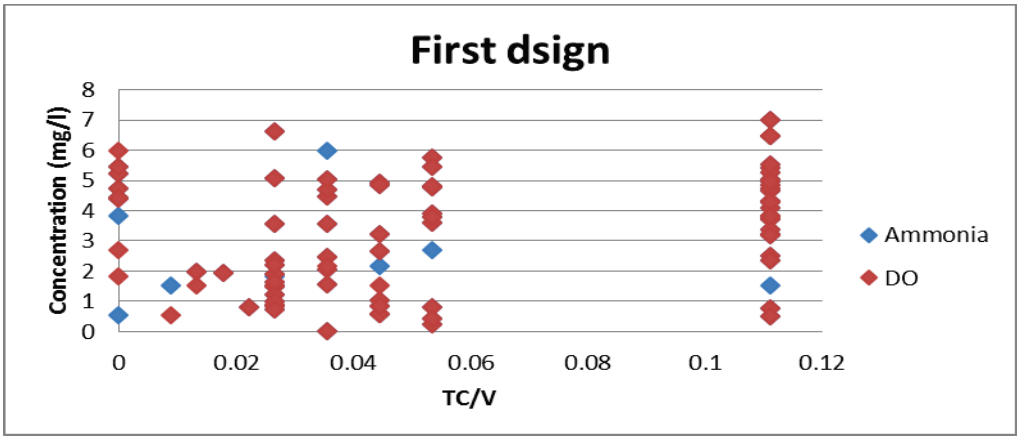

Fig. 7. 3: Scale up parameters (ammonia): best in total current per volume equal to 0.11 (ohms/l)

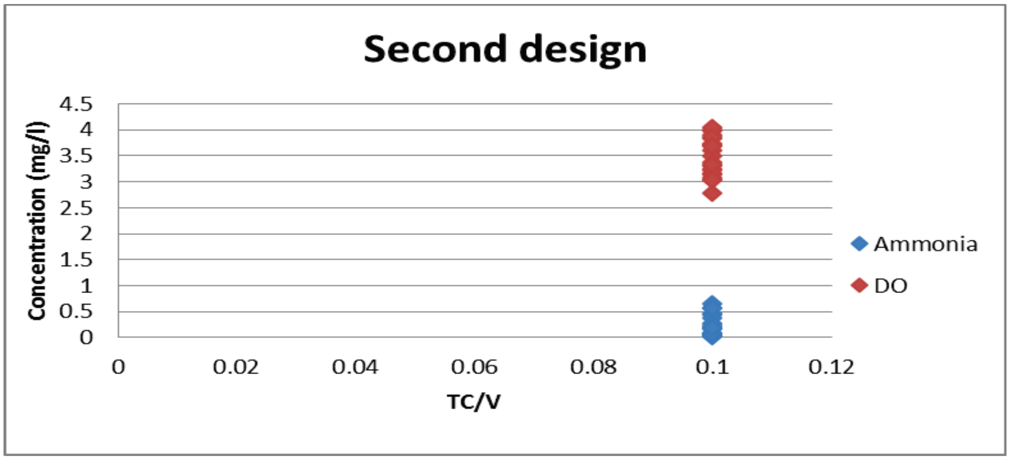

Fig.7. 4: Scale up parameters (ammonia): best in total current per volume equal to 0.1 (ohms/l)