Innovative sludge disinfection approach to generate Class A biosolids for land applications

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A thesis

In

the Department

of

Building, Civil and Environmental Engineering

Presented in Partial Fulfillment of the Requirements

For the Degree of Master of Applied Science (Civil Engineering) at

Concordia University,

Montreal, Quebec, Canada

June 2018

CONCORDIA UNIVERSITY

School of Graduate Studies

This is to certify that the thesis prepared

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Entitled: Innovative sludge disinfection approach to generate Class A biosolids for land applications

And summitted in partial fulfillment of the requirements for the degree of
Master of Applied Science (Civil Engineering)

Complies with the regulations of the University and meets the accepted standards with respect to originality and quality.

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Abstract

Innovative sludge disinfection approaches to generate Class A biosolids for land applications

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Beneficial use of biosolids (sludge), generated by municipal wastewater treatment plant, requires adequate disinfection before its land application. Traditional methods for sludge disinfection are either time consuming or cost demanded. To overcome the drawbacks, novel technologies which uses electrical field phenomena, were proposed to achieve Class A biosolids. Electro-Fenton disinfection and electrokinetics combined with biocide treatment were investigated in this thesis. The lab scale results demonstrated better effectiveness of Electro-Fenton disinfection than Fenton oxidation. Class A quality of biosolids (with 5.8 log reduction) has been achieved within 30 min when Electro-Fenton system was applied in presence of H_2O_2 (30%). It was found that technological parameters such as pH, current, hydrogen peroxide concentration, total solids content, ratios of Fe^{2+}/H_2O_2 influenced effectiveness of sludge disinfection. The results showed that electrokinetic phenomena combined with biocide achieved faster disinfection efficiency reaching log 7.2 fecal coliform reduction within 30 to 50 min while the internal temperature rose to 40°C only. Then, an optimization of current, as well as biocide dosage were conducted in this study. Scale up of the system has also demonstrated an effective sludge disinfection. The study showed that both systems can be applied to WAS (or potentially to other types of sludge) to convert it to Class A biosolids. The systems are particularly beneficial for sludge thickened with iron containing coagulant. The novel technologies produce fertilizing materials which are safe for environment and public health when landfarming.

Acknowledgments

I would like to express my deepest gratitude and respect to my supervisor Professor Maria Elektorowicz for her continuously support and instructions through my research and study in Concordia University. I very appreciate her patience and encouragement during my entire work.

I very appreciate the supported by Dr. Maria Elektorowicz's Natural Sciences and Engineering Research Council of Canada (NSERC) Discovery Grant 156161 as well as graduate students' financial support (FPRST) from the Faculty of Engineering and Computer Science at Concordia University.

I am also grateful to receive the helps from our laboratory technician Mrs. Hong Guan as well as my laboratory colleagues Mrs. Wen Ma, Mrs. Tiantian Chen, Mr. Arash Fellah Jahromi, Mr. Esmaeel Kariminezhad, and all of the others laboratory colleagues.

I also would like thank the Canadian Society for Civil Engineering (CSCE) for recognition of my work by discerning the Honorable Mention in 2018 Best Paper Competition during the CSCE Environmental Specialty Conference in Fredericton, NB, Canada.

At last, I would like to express my thanks to my family and friends for their continuously encouragement and support.

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Abbreviations

AAS	Atomic absorption spectrophotometer
AOP	Advanced oxidation process
AN	Ammonia salts
BDD	Boron-doped diamond
BS	Biocide/Bioxy^{TM}
CCME	Canadian Council of Ministers for the Environment
DC	Direct current
EPA	Environmental Protection Agency
EPS	Extra polymeric substance
EDL	Electric double layer
PEF	Pulsed electric field
ROS	Reactive oxygen species
RCS	Reactive chlorite species
WAS	Waste activated sludge
TS	Total solids
TAN	Total ammonia nitrogen

Chapter 1 Introduction

1.1 Background Statement

Management of the municipal sludge produced during the wastewater treatment can represent up to 60% of the total operating costs (Andreoli et al., 2007). In the US, more than 7.7 million dry tons of municipal sludge is generated annually, of which 55% is disposed of via land application (Lu et al., 2012). However, sludge has also been proven to have potential applications as an energy source and a fertilizer if it is adequately treated.

According to both Sopper (1993) and Henry et al. (1993), municipal sludge can be used to promote plant establishment and growth. In addition, it can improve soil quality and structure (Haering et al., 2000). Because municipal sludge contains nitrogen and phosphorus, it can be used as a soil amendment to provide plants with nutrients. Other nutrients such as potassium, sulphur, calcium, and magnesium, essential components for plant growth, are present in sludge as well. In Canadian wastewater treatment facilities, the nitrogen concentration in sludge typically ranges from 17,100–59,000 mg/kg of total solids (TS) on a dry weight basis, while the phosphorus concentration is 4,120–32,500 mg/kg dry TS (CCME, 2012). With such large amounts of nutrients present in municipal sludge, it is desirable to find a beneficial use for it.

Before sludge can be put to a useful purpose, however, it must be properly treated to prevent public health problems. Obstacles to the beneficial use of municipal sludge are primarily related to the possible presence of heavy metals and pathogens. Heavy metals that can be found in sludge normally include cadmium, lead, chromium, arsenic, mercury, nickel, copper, and zinc. Without proper treatment to remove heavy metals, soil would be contaminated by sludge application, eventually causing health problems for consumers of contaminated drinking water.

Pathogens such as bacteria, viruses, protozoans, spores, and worms (helminths) may also exist in sludge. This can lead to infections of individuals exposed to insufficiently treated sludge. For this reason, sludge must be disinfected before it is applied to protect human health. The US Environmental Protection Agency (USEPA, 1993), which regulates sludge properties, classifies it into Class A and Class B biosolids. Depending on the purpose of a land application, different classes of biosolids should be used. Class A biosolids can be directly applied to agricultural lands as fertilizer, whereas the application of Class B biosolids is limited by certain conditions.

Usually, municipal sludge management includes dewatering, pathogenic disinfection and nutrient recycling, if possible. Among these processes, the reduction of pathogens might be the most important with respect to public health and environmental protection. Several treatment methods, including lime treatment, aerobic and anaerobic digestion, and composting, have been designated as standard treatment methods by EPA regulations (EPA, 2003).

In spite of different treatment methods, new approach to sludge treatment is required for more sustainable use of sludge.

Chapter 2 Literature review

2.1 Introduction

Biosolids, or sludge, are a by-product of municipal wastewater treatment that contain significant amounts of organic matter, nutrients and trace elements. They represent a potential resource for agriculture usage due to high contents of ammonium and phosphate. However, pathogens, heavy metals, antibiotics or other contaminants may also be present without adequate treatment. In Canada, more than 660,000 dry metric tons of sludge are generated per year (CMWC, 2015), in addition to 6.5 million tons in the US. Therefore, with such large amount of potential usage of organic resource and the concern of the potential health problems, proper treatment must be applied to decontaminate biosolids before land application.

2.2 Problem Statement

2.2.1 Water in Sludge

There are four types of water in the sludge matrix: free, absorbed, capillary, and cellular water. It is vital to remove water from the sludge matrix because a large amount of water will cause problems such as reduced anaerobic digestion efficiency and increased capital cost of sludge transportation. Free water can easily be removed simply by using gravity settling but absorbed water and capillary water can only be removed by applying extra mechanical force or using a coagulant to eliminate the electrical double layer so as to release the captured water. By removing absorbed water and capillary water, it can increase the solid percentage by up to 30%. For cellular water, only thermal evaporation can remove the water (Sanin et al. 2011).

2.2.2 Heavy Metal Contamination

heavy metals may contaminate plants if they are present in sludge applied to the land. Trace heavy metals absorbed by the root system will eventually transfer to animals via the food chain, which can lead to accumulation inside the human body (Jamali et al., 2007). Due to the toxicity of heavy metals, this accumulation can be a health hazard. From a regulatory standpoint (Tsadilas et al. 2011), heavy metal contamination primarily concerns cadmium, copper, zinc, nickel, mercury, chromium, lead, arsenic and selenium. To satisfy the safety criteria for land application, the concentrations of heavy metals in sludge must be lower than the legally designated maximum levels. Depending on the area, (EPA 40CFR503, 1993) and (CCME, 2012) are usually the regulations that limit heavy metal concentrations for sludge disposal within the US and Canada, respectively.

2.2.3 Trace Organics Pollutants

As growing up consumption of organic matter increases, problems associated with the disposal of organics are becoming more serious; these materials are eventually discharged into municipal wastewater treatment plants. Although most organic matter can be decomposed by bacteria through aeration tank treatment or anaerobic digestion, a certain number of high-resistance organics are difficult to treat. Common organic pollutants include cyanide, phenol, methyl chloride, 1,1,1,-trichloroethane, toluene, and ethyl benzene. In recent years, more and more organic compounds have come to be considered as pollutants (Tsadilas et al. 2011). For example, the accumulation of PCBs in birds increases the fragility of the shells of their eggs (Custer et al., 2014).

2.2.4 Pathogenic Microbes

Pathogens from sludge may contaminate groundwater by infiltration during rainfall (Straub et al., 1993). In this way, diseases can be introduced to water supplies, causing risks to human and animal health. Direct contact with pathogens may also occur when people handle crops or other plants that were fertilized with the sludge. Sludge may harbor diseases or parasites without a proper treatment to eliminate pathogenic microbes.

Five major groups of pathogenic organisms are present in sludge: helminths, protozoans, fungi, viruses and bacteria. The density of pathogenic organisms depends on the source of the sludge and the treatment process. Table 2.1 shows the concentrations of pathogenic organisms in different types of sludge.

Table 2.1 General characteristics of sludge regarding pathogens (Andreoli et al. 2007)

Pathogen	Type of sludge	Density of pathogens
Helminth eggs	Primary sludge	10^3 - 10^4/kg TS
	Digested sludge	$10^2 – 10^3$/kg TS
	Partially-dewatered sludge	$10^1 – 10^3$/kg TS
	Partially-dewatered sludge from aerobic treatment	$10^2 – 10^3$/kg TS
	Anaerobic sludge	$6.3*10^3 – 1.5*10^4$/kg TS
Protozoan cysts	Primary sludge	$7.7*10^4 – 3*10^6$/kg TS
	Digested sludge	$3*10^4 – 4.1*10^6$/kg TS
	Dewatered sludge	$7*10^1 – 10^2$/kg TS
Bacteria	Sludge	$10^1 – 8.8*10^6$/kg TS
	Extended aeration sludge	10^8/kg TS
Viruses	Primary sludge	$3.8*10^3 – 1.2*10^5$/L
	Digested sludge	$10^1 – 10^3$ /L
	Biological sludge	$10^1 – 8.8*10^6$ /kg TS

2.3 Conventional Methods for Sludge Treatment

Decontamination of sludge for land application should begin with sludge stabilization/sanitation. A few methods are currently in worldwide use: pasteurization, aerobic/anaerobic digestion, chemical treatment, composting, and thermal drying (Table 2.2).

Table 2.2 Summary of sanitary technologies and requirements (EPA, 1993)

Treatment Method	Operational Parameters
Pasteurization	Temperature ⩾70 ℃ for 30min to 1h
Aerobic Thermophilic Digestion	First phase with HDT⩾5d; temperature 50℃ for 23h; 55℃ ⩾10h; 60℃⩾4h
Anaerobic Thermophilic Digestion	Temperature 50-55℃⩾48-72h; 70℃⩾1h; or 1h > 70℃ and 55℃ > 2h or 50℃ > 4h
Lime Treatment	pH > 12 and T > 55℃ for > 2h; pH > 12 for 3 months
Composting	Temperature to 55℃ > 10d; Temperature to 60℃ > 2d
Thermal Drying	Heat to 80℃ and drying to > 90℃; Heat to 80-90℃ for 30min

2.3.1 Sludge Pasteurization

In pasteurization, sludge is heated to a minimum temperature at 60 ℃ for 30 minutes. Other combinations of temperature and time can also be used, such as 70℃ for 25 minutes, 75℃ for 20 minutes, or 80℃ for 10 minutes. However, a minimum disinfection time of 10 minutes is required regardless of temperature.

2.3.2 Aerobic/Anaerobic Digestion

Of the types of treatment processes listed above, sludge digestion is the most common method for sludge stabilization. This is because it is considered environmentally friendly. The mechanism for both aerobic and anaerobic digestion is to apply certain conditions to the sludge and use microorganisms to consume organic matter, converting it into either carbon dioxide in aerobic digestion or methane in anaerobic digestion, respectively. The advantages of aerobic digestion are greater ease of operation compared to anaerobic digestion and lower retention time during the process, resulting in lower container volume requirements for the sludge, which could significantly decrease construction costs. However, the drawback of aerobic digestion is its high operating costs relative to anaerobic digestion, since aerobic digestion requires providing the aerobic bacteria with a continuous supply of air/oxygen. Additionally, the primary by-product of aerobic digestion is carbon dioxide, which is considered a main greenhouse gas driving global warming.

Anaerobic digestion is a very common approach for sludge stabilization. This process operates under oxygen-free conditions so that anaerobic bacteria can produce methane. There are multiple stages of anaerobic digestion: hydrolysis, acidogenesis, acetogenesis and methanogenesis. First, bacteria break down complex organic compounds and generate simple organic chain materials such as cellulose, protein and lipids. Then, another type of microorganism converts products from this first stage to acetic acid, propionic acid, volatile fatty acids, hydrogen and carbon dioxide. In the last stage, one group of methanogenic bacteria transform carbon dioxide and hydrogen into methane, while another group transforms acetates into methane and bicarbonates. Because this process produces methane, anaerobic digestion is commonly utilized for serving large metropolitan areas. A sufficient source of organic material can easily generate enough methane to supply the wastewater treatment plant with electricity (Cakir and Stenstrom, 2005). This is an ideal method for stabilizing sludge without further contaminating the environment. However, operation issues always arise because the overall process is a very complex stepwise reaction, in which any changes in operation will cause poor results. This is especially true because of the high sensitivity of methanogens to the presence of heavy metals, ammonia, and volatile fatty acids, which will inhibit their activity and cause poor generation of digested sludge (Chen et al., 2008). Furthermore, environmental changes such as pH and temperature can also result in poor sludge digestion.

Aerobic thermophilic stabilization of sludge

As noted above, aerobic treatment requires a sufficient oxygen supply to allow microbes to grow and consume organic matter. These exothermal microbial and metabolic processes result in the production of heat. To meet sludge quality criteria regarding safe pathogen density for disposal, temperature and retention time must be considered. According to regulation, it is necessary to maintain a temperature of 50°C for at least 23 h, 55°C for 10 h, or 60°C for 4 h (EPA, 1993).

A two-phase process includes both aerobic thermophilic and anaerobic digestion. Before the anaerobic treatment process, if the pasteurization or aerobic treatment satisfied the temperature and retention time conditions, the anaerobically treated material must be maintained at 30°C without feeding additional sludge into the batch reactor.

2.3.3 Composting and Thermal Drying

The composting method for sludge stabilization has been used for more than a thousand years and is the oldest method for managing human waste. The mechanism for this method is similar to that of aerobic digestion. Aerobic bacteria break down complex organic matter for their own metabolism, and organic matter is converted into carbon dioxide and water. Because all of the complex long-chain organic compounds are converted into simple short-chain compounds, it can be applied to land as fertilizer. Composting can generally be used in rural areas with small-scale wastewater treatment plants but is not suitable for large wastewater treatment plants because of the space required for composting. Also, if the wastewater treatment plant is close to the community, the public may be exposed to disease vectors (Epstein et al., 1983). Therefore, although composting is an economical treatment method, it cannot satisfy the requirements of mega cities generating large amounts of sludge.

Thermal methods such drying and incineration are efficient methods for sludge stabilization regarding the time consuming. Thermal drying can sanitize sludge rather fast by applying heat to evaporate the water inside the sludge and stabilizing it. Incineration is even faster sludge treatment, however, high operational costs and the transformation of heavy metals into fly ash are still limiting factors for using this method (Fytili and Zabaniotou, 2008).

2.3.4 Treatment with Lime (CaO)

By adding a large amount of CaO to sludge, the exothermic reaction of calcium oxide with water heats the lime-sludge mixture to a temperature between 55 and 70°C. At the same time, the pH increases to at least 12.6. Under such conditions, the retention time is at least two hours.

2.4 Others Treatment for Beneficial Use of Sludge

2.4.1 Pre-treatment of Anaerobic Digestion

Due to anaerobic digestion is a stepwise process, the production of methane in the methanogenesis process is usually limited by the hydrolysis efficiency. Because insufficient of biodegradable substrate in methanogenesis stage can lower the methane production efficiency. Therefore, most of the recent research are focusing on increase the available biodegradable subtract for methanogenesis stage. Methods are generally including physical, chemical, biological as well as some others different combinations of extra conditions for promoting methane production.

Physical pre-treatment process includes such as sonication, lysis-centrifuge, liquid shear, collision, high-pressure homogenizer, microwave, and thermal treatment in order to produce more available organic carbon for anaerobic digestion. Among all of this pre-treatment process, thermal treatment is one of the most studied pre-treatment methods. The mechanism is thermal treatment can disintegrate the cell membrane, subsequently it increase the solubilization of the organic compounds (Ferrer et al., 2008), which provide more biodegradable materials for methanogenesis bacteria. Cambi^{TM} thermal hydrolysis is one of the most worldwide applied process for pre-treatment of anaerobic digestion. It is by using high temperature steam and pressure pass through the sludge matrix, so that it could disintegrate cell walls and release the organic compounds. By using such application, retention time of anaerobic digestion can be shortened, and it could increase the organic loading in anaerobic digester as well as reduce the sludge production after anaerobic digestion treatment.

Chemical pre-treatment includes injection of strong acid, alkalinity and oxidant for destroying the long chain organic compound to which made it more available for microorganism. However, chemical pre-treatment usually requires huge amount of reagent which resulting in a high capital cost. Therefore, comparing physical pre-treatment, chemical pre-treatment is not commonly used in worldwide.

Biological pre-treatment has basically the same mechanism as the previous two methods, which is increase the hydrolysis of the complex substrate by applying specific microbe growth before the anaerobic digestion process. Though, since biological pre-treatment causing decrease of certain amount of organic compounds, proper operational time should be concerned regarding the possibility of reducing of methane production.

2.4.2 Nutrient Recovery

Nutrient recovery technology recently focuses on phosphorus recovery. One of the application was patented by university of British Columbia spinoff company (Ostara). They developed a method for precipitating struvite and recover it as fertilizer for agricultural plantation. This application already has 15 commercial installation in worldwide and it’s being more acceptable for more and more wastewater treatment facilities.

2.5 Quality Control of Microbes within the Sludge Matrix

The US EPA has regulated sludge since 1993; they define different classes of sludge quality for land application. There are two types of sludge, or biosolids. Class A biosolids are considered fully treated sludge in which microorganisms have been sufficiently reduced. Therefore, Class A sludge can be directly used for agricultural purposes and exposed to humans. Class B biosolids are of lower quality than Class A; the applications for Class B biosolids are limited by regulation, and direct human exposure is prohibited. For this reason, proper measures should be taken after land application of Class B sludge, such as fencing to prevent direct contact with the public. Additionally, disposal time should be considered with regard to forecasted rainfall so as to prevent pathogens from being transported by surface overland flow and contaminating groundwater by infiltration or surface water by direct runoff. Table 2.3 below illustrates the standard for classifying sludge quality.

Table 2.3 Criteria for sludge disinfection equivalency (EPA, 1993)

	Class B	Class A
Process Efficiency Parameters:	≥2 log reduction of fecal coliform bacteria	(1) ≥3 log reduction of total *enteric* virus, and (2) ≥3 log reduction of viable helminth (*Ascaris*) ova, and (3) ≥3 log reduction of fecal coliform bacteria
	And/or	And/or
	≥2 log reduction of E. coli bacteria	≥3 log reduction of fecal coliform bacteria
	And/or	And/or
	≥2 log reduction of *Enterococcus spp.* bacteria	≥3 log reduction of *Enterococcus spp.* bacteria
		And/or
		≥3 log reduction of *Salmonella spp.* bacteria
Process Compliance Parameters (EPA 40CFR503)	≤2,000,000MPN or cfu/g total solid (TS) of fecal coliform in the treated sludge	Microorganism densities in the treated sludge of: (1) ≤1cfu/4 g TS of total enteric virus, and (2) ≤3 viable helminth (*Ascaris*) ova/4 g TS, and (3) ≤1,000 MPN fecal coliform/g TS or 3MPN *salmonella* spp./4g TS

In Canada, regulations regarding biosolids vary by province. Some of the provinces refer to the US EPA regulations, whereas others have their own legislation. In Quebec, biosolids land application is classified using the C-P-O classification. C stands for chemical contaminant content, P for pathogen content, and O for odor value. There are 3 categories for pathogen contents in biosolids: P1, P2, and P3. The equivalent of the US Class A quality standard for biosolids in Quebec is the P1 category, and P1 quality biosolids should meet the requirements of the US EPA standard according to legislation (Environment Quebec, 2004).

2.6 Disinfection

2.6.1 Introduction to Disinfection in the Water/Wastewater Industry

Among the available processes for water/wastewater industry disinfection, chlorine disinfection is the most common method used by the water/wastewater treatment industry due to its cost-effectiveness and ease of operation. However, recently, many studies have demonstrated that chlorine disinfection generates numerous by-products that have negative impacts on human health (Bougeard et al., 2010). Therefore, more and more water treatment entities are seeking alternative means of disinfection.

UV and ozone are the two commercial methods for either water or wastewater disinfection. The feasibility of these methods has already been proven, with years of operation experience. However, both options are associated with high costs and operation problems. For that reason, researchers are still investigating other methods for disinfection.

Advanced oxidation processes (AOPs) are gaining attention for a few reasons. First, AOPs have much higher disinfection efficiency compared to ozone or chlorination treatment of bacteria (Diao et al., 2004). Second, while wastewater treatment targets pathogenic microbes, concern is increasing about other substances such as pharmaceuticals with regards to their resistance to degradation and their toxicity in natural environments (Yuan et al., 2013). AOP treatment generates oxidant species that oxidize pollutants, including pathogenic microbes as well as pharmaceuticals and some macro organic pollutants like phenol. Most AOPs are aimed at generating oxidant species called hydroxyl radicals (^{-}OH) to remove pollutants. The application of (^{-}OH) for pollution treatment has gained interest in recent years. It can be produced through different methods, such as anodic oxidation, Fenton reaction, Fenton-like reaction, and photo-Fenton (Ortega-Gómez et al., 2012; Polo-López et al., 2012; Selvakumar et al., 2009).

[2.1] $\mathrm{Fe} \rightarrow \mathrm{Fe}^{2+} + 2\mathrm{e}^{-}$

[2.2] $4\mathrm{Fe} + 10\mathrm{H_2O} \; + \; \mathrm{O_2(g)} \rightarrow 4\mathrm{Fe(OH)_3(s)} + 4\mathrm{H_2(g)}$

2.6.2 Introduction to Electro-disinfection

Novel remediation technologies for emerging contaminants have also arisen in recent years. Electrochemical treatment is being considered as an alternative method for treating contaminants such as organic matter, pharmaceuticals and pathogenic microbes. Specifically, a literature review regarding pathogen disinfection will be the focus here.

There are four different types of configuration for electro-disinfection, each of which has its own mechanism for microbe disinfection.

(1) *Electrocoagulation disinfection*

By sacrificing the anode in an electrical circuit system, an iron or aluminium electrode will rapidly dissolve into electrolyte (Eq. [2.1]). Iron will dissolve as ferrous iron. In the presence of dissolved oxygen, ferrous iron will be oxidized to ferric iron (Brillas and Martínez-Huitle, 2015). Ferric iron will then hydrolyze (Eq. [2.2]) and exist in different forms of $Fe(OH)_n$ depending on the pH. A high surface area structure has the function of absorbing and capturing containment such as virus bacteria as well as arsenic. Delaire et al. (2015) used TEM images to demonstrate that the formation of ferric hydroxyl flocs physically removes *E. coli* from groundwater as precipitate. This mechanism is similar to using an aluminum anode and forming $Al(OH)_3$ to capture bacteria. Ghernaout et al. (2008) compared the results of using different electrode materials as anodes, finding that an aluminum electrode yields better results for bacteria removal. Moreover, Delaire et al. (2016) compared bacteria removal efficiency between coagulation using $FeCl_3$ and electrocoagulation using an iron electrode; they note that physical removal is not the only mechanism of bacteria removal, as oxidant agent generation also affects bacterial disinfection. Oxidant agents specifically refer to the generation of ^-OH, Fe(IV) from a Fenton-type reaction in the presence of oxygen. It has been demonstrated that all of these methods have extremely high disinfection efficiency (Huang et al., 2016; Ortega-Gómez et al., 2012; Schmalz et al., 2009).

(2) Anodic Oxidation Disinfection

In summary, methods for applying anodic oxidation are implementing high oxidation potential electrode to generate oxidant for the microorganism elimination. One of the method is applying the dimensional stable anode such as Pt, Ti, IrO_2, or RuO_2 plate. Another method is by using so-called non-active electrode, such as PbO_2, SnO_2 or boron-doped diamond (BDD), as the anode for the electrolysis cell. Depending on the activity of the electrode, non-active electrodes such as BDD easily form reactive oxygen species (ROS), which include hydroxyl radicals (^-OH), hydrogen peroxide (H_2O_2), and ozone (O_3) (Huang et al., 2016). However, electrodes such as Pt, Ti, IrO_2, or RuO_2 are more likely to produce reactive chlorite species (RCS) such as HClO or ClO^-. Since (^-OH) molecules are bound to the surface of the active electrode, they act as a mid reactant to oxidize chlorine into RCS (Chen et al. 2004). Therefore, depending on electrode component, oxidant generation would be varied, and it would affect the operating parameters.

Table 2.4 Summary of anodic disinfection processes

Type of electrode	Electrolyte	Target Microorganisms	Remarkable finding	Reference
Titanium with thin layer of $IrO_2/Sb_2O_3/SnO_2$	NaCl	MS2	With assisting of excess Chlorite presence, Chlorine dioxides is the main disinfectant	(Fang et al., 2006)
TiO_2/BiO_x	Cl^-/NH_4^+	MS2, *E.coli*, rACl5, *Enterococus*	Under presence of NH_4^+, free chlorite will be converted to Chloramine so that decrease disinfection efficiency	(Huang et al., 2016)
Pt (Platinum-tipped copper)	Cl^-	MS2, *E.coli*, PRDI, *salmonella*	Disinfection efficience affected by initial microorganism population density	(Drees et al., 2003)
BDD	Cl^-	*E.coli*	Under presence of free chlorite, disinfection oxidant from hydroxyl radical generation can be neglectable	(Schmalz et al., 2009)
BDD	Cl^-	*E.coli*	Hydroxyl radical (^-OH) has much higher disinfection efficiency than free chlorite, lowering temperature increase production of Hydroxyl radical (^-OH)	(Jeong et al., 2006)
BDD, Ti/IrO_2, Ti/RuO_2, Ti/Pt, IrO_2, Pt	Free Chlorine	*E.coli*	BDD electrode has the highest disinfection efficiency since it generates the most Hydroxyl radical (^-OH) compare other electrode	(Jeong et al., 2009)
Ti	Cl^-	Total coliform	In terms of NaCl presenting in saline water, free Chlorite and Hydroxyl radical (^-OH) co-functioning on microorganism disinfection	(Li et al., 2002)
BDD	Cl^-/NO_3	*E.coli*	Treatment on wastewater disinfection efficiency depends on concentration of total nitrogen	(Cano et al., 2012)

Table 2.4 summarizes the methods for anodic oxidation disinfection and their efficiency. It is obvious that the BDD electrode method has garnered the most attention because it has the highest thermodynamic potential for the formation of hydroxyl radicals (^{-}OH) (E=2.38V, SHE) (Kapalka, 2008) compared to the other electrodes. Jeong et al. (2006, 2009) demonstrated that of the anodic oxidation disinfection processes, the BDD electrode has the best results because it generates the highest amount of hydroxyl radicals (^{-}OH). Furthermore, they also proved that compared to free chlorite species, (^{-}OH) has better disinfection results.

However, some researchers have considered that with the presence of chloride in the electrolyte, the hydroxyl radical (^{-}OH) is not the only type of reactive oxidant species. The hydroxyl radical only acts as a mid reactant to oxidize chlorite into HClO and ClO^{-} (free chlorite), and they interact with the microorganism. This has been demonstrated by Schmalz et al. (2009). They explain that the chlorite ion acts as a hydroxyl radical (^{-}OH) scavenger. That is why their results show that as temperature increases, disinfection efficiency also increases. This was also indirectly proven by Jeong et al. (2006), who reported that at higher temperatures, fewer hydroxyl radicals (^{-}OH) are present. Therefore, for a non-active anode electrode, to improve disinfection efficiency, it is necessary to have chlorite ions in the treating matric.

(3) Pulsed Electric Field (PEF) Technology

Commercially, another technology is being used for disinfection, which operates by applying high voltage pulsed electric fields to the treatment matrix. Under the influence of a pulsed electric field, the microbial wall structure changes. Charge builds up on the surface of the microbial membrane, causing an electrical potential difference. When the electrical potential difference exceeds 1V, the structure of the lipid bilayer changes and pores form on the surface of the microbe, permitting ions and others macromolecules to cross freely (Rubinsky, 2007). Irreversible cells damage will cause cell death and inactivate the microbe. This technology is primarily applied in the field of food processing and has been studied for decades, but most work has focused on disinfection of fluids such as fruit juice and milk (Mohamed and Eissa, 2012). It is unheard of to use such technology on waste-activated sludge.

(4) Heat and Electrokinetic Technology

The configuration for heat generation within electrical field is similar to that for a pulsed electric field, which is simply the application of an electric field to electrolytes. The difference is that heat generated from electrical field applies a lower voltage compared to PEF technology, and it mainly uses heat for microbe disinfection. The mechanism is much simpler and easy to operate, which is why it has been used for the last few decades in the food industry for disinfection of vegetables, meats, soup, milk, and fruit juice. There has been very little study of the use of electrical heat for pathogenic disinfection for the municipal wastewater industry. Some previous work has already demonstrated that direct current applied to sludge controls *E.coli* via electrokinetic stress (Esmaeily et al., 2006), desiccation (Huang et al., 2008), or ohmic heat stress (Daneshmand et al., 2012). Furthermore, Yin et al. (2018) demonstrated that the corrosion rate of the electrode varies using different electrolytes. According to their energy consumption evaluation, the most important process for ohmic heating disinfection of sludge is heat preservation. At the same time, pH is also an important factor affecting microbe disinfection efficiency, as a lower pH at a higher temperature will result in greater disinfection (Daneshmand et al., 2012). Recent study for improvement on such technology was conducted by Safaei et al. (2012, 2013), who used electrokineitc combined with biocide and ammonia salt enhancer for spores disinfection in WAS, and she successfully achieved 3 log reduction on *C.perfringens* reduction.

2.6.3 Fenton/Photo-Fenton for Disinfection

The Fenton reaction was discovered over 100 years ago, but its application for oxidizing organic pollutants only began in the 1960s (Neyens and Baeyens, 2003). The mechanism for the Fenton reaction, following Eq. [2.3], is the combination of H_2O_2 with ferrous iron to generate hydroxyl radicals for oxidizing organic pollutants (Eq. [2.6]), which are then further oxidized and fully converted to water and CO_2. Therefore, the amount of hydroxyl radical generation is extremely important for the effectiveness of organic compound removal. A series of reactions are involved in the generation of hydroxyl radicals in a Fenton reaction.

[2.3] $Fe^{2+} + H_2O_2 + H^+ \rightarrow Fe^{3+} + H_2O + {}^{\bullet}OH$

$k_1 \approx 70 M^{-1} s^{-1}$ (Rigg et al., 1954)

[2.4] $Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2 \bullet + H^+$

[2.5] $Fe^{3+} + e^- \rightarrow Fe^{2+}$

[2.6] $RH + OH\bullet \rightarrow H_2O + R\bullet$ (Walling, 1975)

$k_6 = 10^7 - 10^{10} M^{-1} s^{-1}$

[2.7] $R\bullet + H_2O_2 \rightarrow ROH + OH\bullet$

[2.8] $R\bullet + O_2 \rightarrow ROO\bullet$

[2.9] $OH\bullet + Fe^{2+} \rightarrow OH^- + Fe^{3+}$

$k_7 = 3.2 \times 10^8 M^{-1} s^{-1}$ (Buxton et al., 1988)

k----Reaction rate constant

Because hydroxyl radicals are a strong oxidant, they would also oxidize the ferrous iron (Eq. [2.9]). Furthermore, ferric iron can react with hydroxyl radicals (OH•) (Eq. [2.4]), which is referred to as a Fenton-like reaction. Hydroxyl radicals (OH•) can also react with H_2O_2 by Eq. [2.10]; therefore, hydrogen peroxide can act as an OH• scavenger as well as an initiator.

[2.10] $OH\bullet + H_2O_2 \rightarrow H_2O + HO_2\bullet$

$k_{10} = 10^7 M^{-1} s^{-1}$

In the presence of organic matter, hydroxyl radicals (OH•) will first react with organic matter because $k_6 > k_{10}$. Consequently, an excess of organic matter in the matrix will consume all of the hydroxyl radicals, and H_2O_2 and ferrous iron will only act as reactants. The application of hydroxyl radicals (OH•) to bacterial disinfection has been studied by Cho et al. (2004). They demonstrated that the Photo-Fenton reaction is an improvement on the Fenton reaction. In the presence of UV irradiation, ferric iron is photo-catalyzed to ferrous iron (Eq. [2.11]). Therefore, if sufficient H_2O_2 is present in the matrix, hydroxyl radicals (OH•) can be continuously generated without adding more ferrous iron. The applications of Photo-Fenton and Fenton reactions to bacterial disinfection have previously been studied by Ortega-Gómez et al. (2012) and Selvakumar et al. (2009), respectively.

[2.11] $Fe^{3+} + H_2O + hv \rightarrow Fe^{2+} + H^+ + HO^{\cdot}$

2.7 Conductive Heat Transfer

There are three modes for heat transfer: conduction, convection and radiation. Conduction occurs when heat is exchanged between two media with a temperature difference.

[2.12] $q_{conductive} = -kA\frac{\Delta T}{\Delta x}$

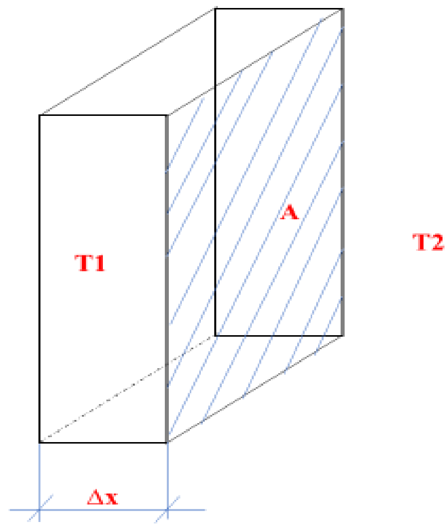


Figure 2.1 Schematic representation of the heat transfer

The rate of heat transfer rate by conduction is defined in Eq. [2.12] (Incropera et al., 2007). The effectiveness of heat transfer is determined by thermal conductivity k, and it varies in different materials. Good conductors tend to have high conductivity, such as iron materials. Poor conductors have low conductivity and could be used as insulators. For any materials, thermal conductivity is determined by the cross-section area (A) over which heat is conducted and the temperature difference (T) between the two surface areas.

2.8 Electrolytic Conductivity

Electrolytic conductivity is a reliable measurement of the ionic content in a solution. Electrolytic conductivity is proportional to the distance between electrodes, as shown in Eq. [2.13-2.15], where l represents the distance between two electrodes and A represents the cross-section area of the electrode. Additionally, electrolytic conductivity is highly affected by temperature, where higher temperatures tend to result in higher conductivity.

[2.13] $R = \rho\frac{l}{A}$

$$[2.14]\ \sigma = \frac{1}{\rho}$$

$$[2.15]\ \sigma = \frac{l}{RA}$$

2.9 Motivation and Objectives

Process for sanitizing/stabilizing municipal sludge in a conventional method is both energy and time consuming. Conventional process such as aerobic treatment needs to have at least 5 days retention time for thermal stabilization purpose, which might cause very high constructive cost in terms of reactor size and installations. Furthermore, since aerobic treatment requires a sufficient oxygen demand for microorganism, it also needs high amount energy for operating as well. Therefore, anaerobic thermal digestion is mostly being considered as another option for sludge sanitization, because it could also generate methane at the same time so that to compensate the operational energy consumption. However, since anaerobic digestion process is very complicated, its practical implementation might be affected by a lot of environmental conditions, therefore, production of methane varied significantly following the variation of influent characteristics. Besides, anaerobic digestion process requires a high capital cost, which is not suitable for many small and medium size wastewater treatment plants. Anaerobic digestion are mostly mesophilic, which requires to operate at around 35°C. Smith et al., (2005) pointed out that under such temperature, pathogens satisfy 2 log reductions of the total population. Therefore, to, satisfy regulations, pathogens should be exposed to further treatment. In such cases wastewater treatment plant usually introduces lime in the sludge matrix for stabilization purpose. An important amount of lime is required to obtain adequate high pH to create conditions in which free ammonia affect pathogens membranes. Lime treatment usually will increase the volume of the overall sludge, increase the transportation fees. Therefore, an innovative process for a rapid sludge stabilization should be developed to reach the land application standard and simultaneously permit on a full recovery of nutrients for soil amendment. Scheme on Figure 2.2 shows possible implementation of the proposed novel sludge management system.

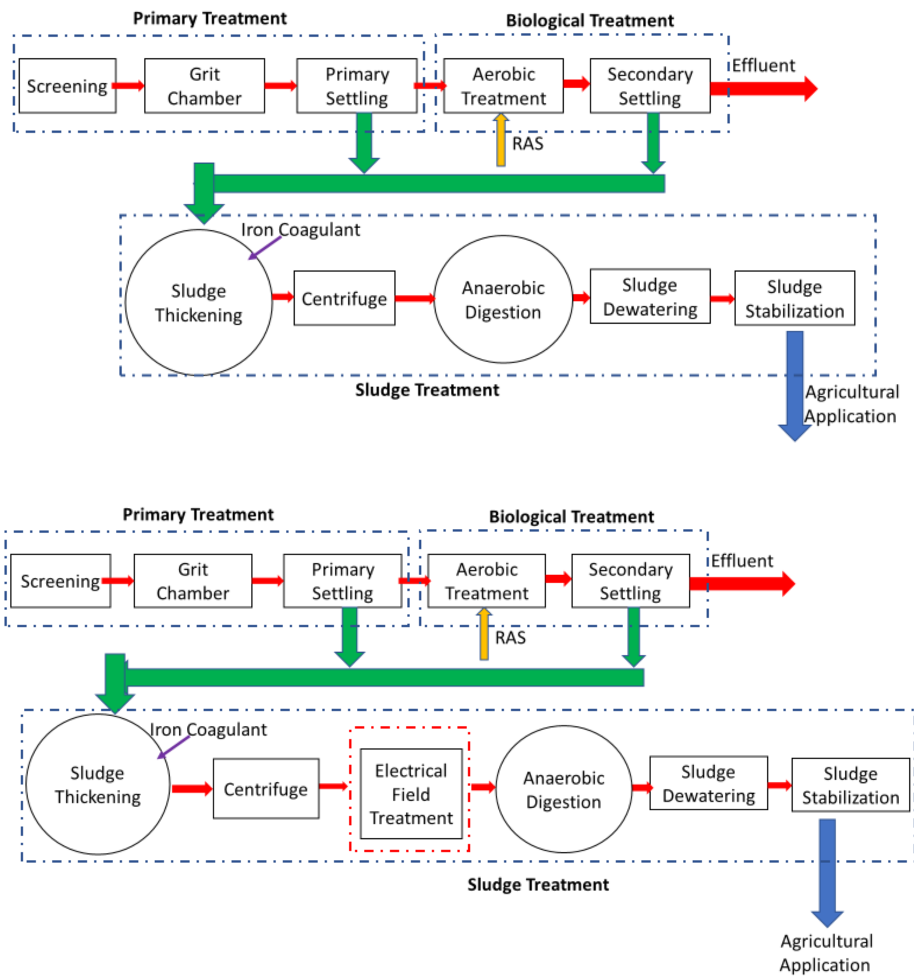


Figure 2.2 Schematic representation of the developed innovative process

A new method could deal with such conditions, which need only a short retention time and fast sanitization of sludge. Hydroxyl radicals ($^{\bullet}OH$) show their high oxidization potential, which can rapidly react with organic matrix as an effective disinfectant (Selvakumar, 2009). The previous study demonstrated that ($^{\bullet}OH$) can achieve 2 log *E. coli* reduction with the concentration of 0.8×10^{-5} mg/L.min (Cho et al., 2004). Generation of hydroxyl radicals can be done through Fenton reaction, Eq. [2.3], where ferrous iron, acting as a catalyst, reacts with hydrogen peroxide to generate ferric iron and hydroxyl radicals, while introduced hydrogen peroxide reacts with ferric iron to regenerate ferrous irons. Then, the reaction with ($^{\bullet}OH$) leads to pathogens' disinfection. Fenton reaction has been used in different situations for disinfection, e.g. water/wastewater. Recent studies on water and wastewater disinfection by using Fenton reaction focus more on Photo-Fenton reaction by applying extra UV light to accelerate the production of ferrous iron to generate more hydroxyl radicals (Ortega-Gómez et al., 2012; Polo-López et al., 2012). Since UV light cannot penetrate through the medium, photo-Fenton is not efficient for treatment of highly turbid water with a high organic load. In this case, Electro–Fenton oxidation process is considered as a substitute technology for the Fenton-based application. Similar to the Photo-Fenton reaction, ferrous iron reacts with hydrogen peroxide to produce hydroxyl radicals. However, reduction of ferric iron is conducted by electro-reduction. Equation [2.5] shows that ferric iron is reduced at the cathode in the presence of continuously applied hydrogen peroxide and produces hydroxyl radicals ($^{\bullet}OH$) (Brillas et al., 2009).

One of the advantage of Electro-Fenton and Photo-Fenton processes compared to traditional Fenton reaction is the feasibility of using smaller amount of ferrous iron at the beginning as a catalyst, which can be regenerated by the system. For example, Electro-Fenton process were applied for treatment of both leachate and industrial wastewater having obvious turbidity and high concentration of COD (Zhang et al. 2006; Li et al. 2010; Jahromi 2016). However, there is always a concern to find technological conditions which inactivate pathogens during a short retention time at low costs. Therefore, the aim of this study was to find such an Electro-Fenton oxidation method which might transform waste activated sludge (WAS) into Class A biosolids during a short exposure period in order to advance the sludge management methods

In spite of this, an electrical field was applied to a colloidal system, which here, it has been considered as an electrokinetic technology. Some previous works have already demonstrated that direct current (DC) applied to sludge generates electrokinetic stressors (Esmaeily et al. 2006), desiccation stressors (Huang et al. 2012), or heat stressors for disinfection of fecal bacteria (Daneshmand et al. 2012) as well as multi-stressors for disinfection of spores (Safaei et at. 2013). Therefore, potential usage for such technology applied on WAS were studied in this study to achieve Class A biosolids.

Therefore, the main objective of this work is to develop an innovated process for a rapid sludge stabilization based on an electrical field application. Two methodological approaches were defined.

The first approach was to determine optimal conditions for the application of Electro-Fenton process to disinfect thickened sludge before its land application. The second methodological approach permitted to investigate the electrokinetic phenomena with respect to disinfection where biocide (Bioxy^{TM}) was directly applied to sludge subjected to electrical field.

Specific objectives for this thesis

Approach one:

- Evaluation of Electro-Fenton effect on the fecal coliform disinfection efficiency, which is understand by shortening time of the process, while the costs of energy and chemicals are minimized
- Investigating of an impact of the H_2O_2 concentration, initial pH value, different H_2O_2 feeding modes and different ratios of Fe^{2+}/H_2O_2 regarding Fenton disinfection.
- Defining the impacts of a ratio of Fe^{2+}/H_2O_2, TS%, pH, and current on Electro-Fenton disinfection.

Approach two:

- Assessing feasibility and efficiency of the fecal coliform disinfection using an in-site H_2O_2, PAA generator (Bioxy^{TM}) combined with the electrokinetic process
- Demonstrating the effect of temperature on the fecal coliform disinfection efficiency in the system
- Investigating the fecal coliform disinfection efficiency in at a larger scale reactor
- Evaluating the energy consumption of the reactor

Chapter 3 Materials and Methodology

3.1 Materials

3.1.1 Municipal Sludge

Municipal sludge samples were collected from a wastewater treatment plant in Saint-Hyacinthe, Quebec. The collection point was located after the activated sludge centrifuge process. The process before the centrifuge is the thickening/settling tank, in which $Fe_2(SO_4)_3$ was used for coagulation. The characteristics of the sampled sludge are summarized below in Table 3.1. The sludge was stored at 4°C until use, with an aeration period of at least 3 days implemented before usage to create aerobic conditions and simulate on-site operating conditions.

Table 3.1 Measured characteristics of sludge sample

Parameters	Values
pH	6.39
Total ammonia nitrogen (TAN) mg/L	51
Fecal Coliform (cfu/100ml)	4.00×10^7
Total Coliform (cfu/100mL)	4.17×10^7
Cl^- (mg/L)	14.5
Conductivity (mS/cm)	1.2
TS (%)	5.5
Fe^{2+} (g/kg TS)	16.9
Total Fe (g/kg TS)	37.5

3.1.2 Materials Analysis and Preparation

The pH value was monitored using an accumet^{TM} AB2000 gel-filled AgCl combination electrode (Fisher Scientific, Canada). Conductivity electrode was used for conductivity measurement. TAN were measured via the colorimetric method using a Hach kits (TNT 832) and following the instructions in the manual (Boyd and Daniels, 1988), while a spectrophotometer (DR2800, Hach) was used for ion concentration determination. The concentration of Cl^- was analyzed with an ISE chloride probe (Fisher Scientific). TS measurement was according standard method 2540A (APHA/AWWA/WEF, 2012). Temperature and current were recorded by a data logger system (Keysight 34970A). According to the current value, current density has been calculated base on the current value divided by the electrode surface area. For ferrous iron measurement, certain sample preparation steps were implemented. Subsequently, dilution of thickened sludge was sometimes required to make the sample homogenous. The diluted sample was then transferred to a centrifuge tube, and a centrifuge tube mixer was used to homogenize the sample. Then, the tubes were placed in a centrifuge (Thermo Fisher Scientific), which was operated at 8000 rpm for 4 minutes. The supernatant was collected, and a standard 0.45 um syringe (APHA, 2012) filter was used to remove the suspended particles in the matrix. Next, 0.5 ml of filtrate was transferred to a 15 ml centrifuge tube and chelated with 1 mM of 2,2-bipyridine (Cagle and Frederick Smith, 1947). After chelation, the mixed liquid reacted as defined in Eq. [3.1] to obtain red color solution in ferrous iron was present in the matrix. Then, 3 ml of the mixed liquid was transferred to a cuvette and analyzed with a UV-VIS spectrophotometer (Thermo Fisher Scientific Evolution 201). For the spectrophotometer operation, the wavelength was set to 522 nm, the absorbance peak for bipy-ferrous chelated chemicals showed in Figure 3.1.

[3.1] $3bipy + Fe^{2+} \Longleftrightarrow Fe(bipy)_3^{2+}$ (Red)

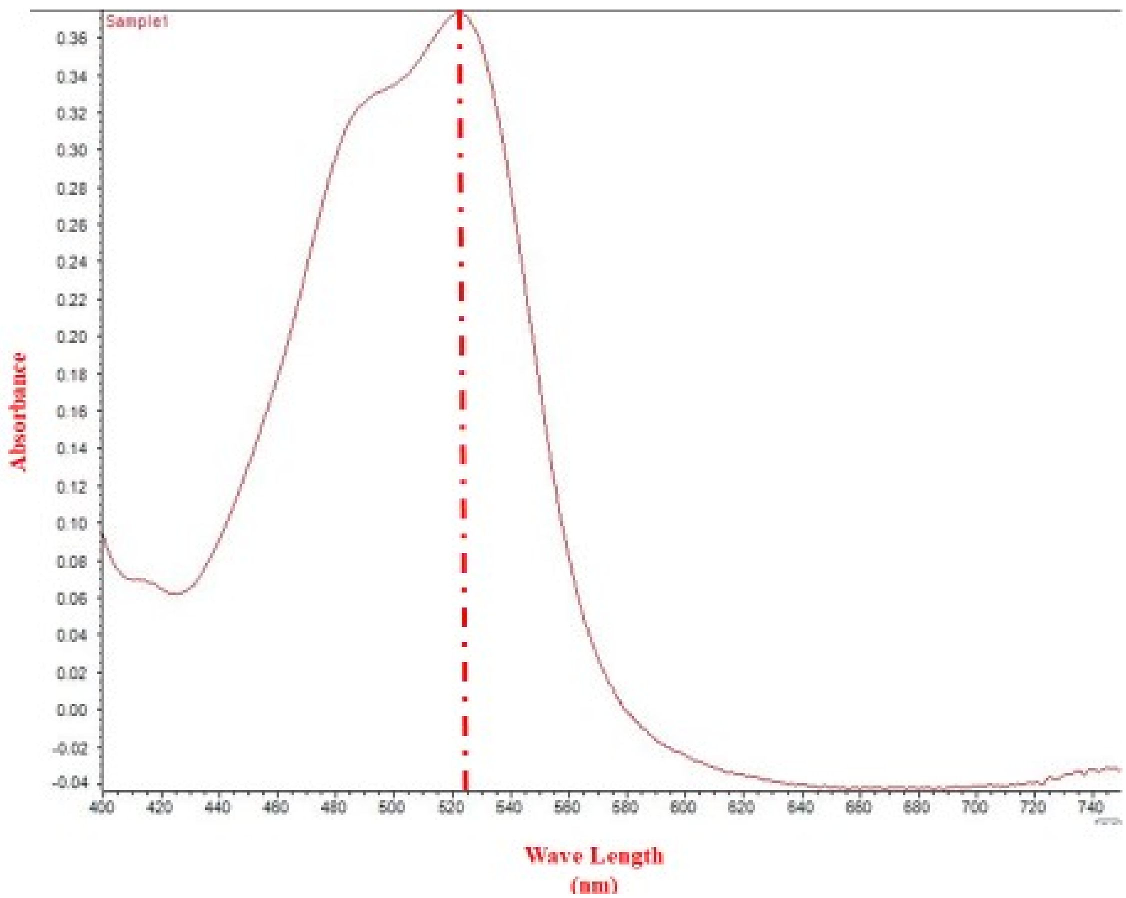


Figure 3.1 Bipy-ferrous absorbance in different wavelengths using UV-Vis spectrometry

For total iron measurement, 2 g of wet sludge sample was introduced into a heat resistant tube, then 10 ml of 1:1 HNO_3 was added and mixed with the sample. The sample was then heated in a water bath to a temperature of 95±5°C and maintained there for 10 minutes without boiling. After the sample cooled to room temperature, concentrated HNO_3 was added and the cover removed for 30 minutes. If a brown fume was generated, this indicated that organic material was being oxidized; in this case, the step (addition of 5ml of concentrate nitric acid) was repeated until no brown fume was observed, then the sample was heated again to 95±5°C for two hours. Thus, the sample was allowed to cool to room temperature, and 2 ml of DI water and 30% H_2O_2 was added. Additional

30% H_2O_2 was added until there were no further changes on appearance, then, the sample was heated again to 95±5°C for two hours. Finally, the sample was allowed to cool and diluted with DI water to 50 ml, then centrifuged to separate solids and liquids. Filtration was then used to collect the filtrate. The filtrate will eventually be analyzed by AAS (Atomic Absorption Spectrophotometer) for total iron concentration (US EPA 3050B).

Fecal coliform bacteria were used as the pathogenic indicator. The source of the bacteria was wastewater from a poultry farm (Quebec, Canada). The sample containing bacteria was enumerated in LB (Luria broth) medium overnight. Bioaugmentation was conducted to reach an adequate density of cells, and then 1 ml of prepared cultured LB was injected into 50 ml of fresh LB and incubated overnight to maintain a fecal coliform bacteria density around 10^9 cfu /100 ml. To identify the bacteria, M-FC agar (BD Difco™) was applied for a standard membrane filtration method with a serial dilution, then, counted on a grid filter (APHA/AWWA/WEF, 2012). In this study, to simulate the density of bacteria in sludge, 10 ml (per litter sludge) of cultured LB broth was injected into the reactor to achieve 10^9 cfu/100 ml for the initial bacterial concentration. Fecal coliform bacteria will grow on a 0.45 μm membrane (Fisher Scientific) with a tiny blue spot (Geldreich et al,. 1965), as in Figure 3.2.

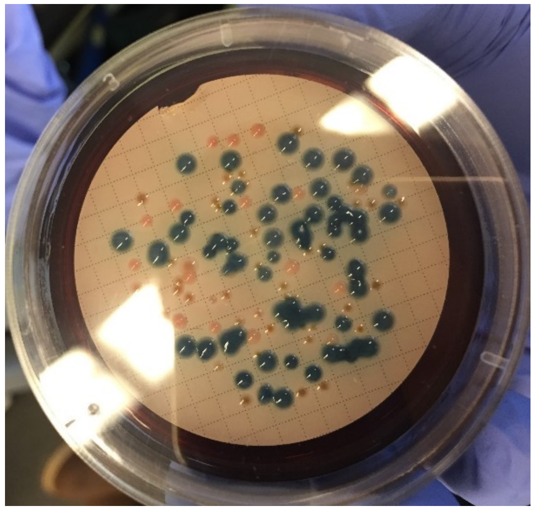


Figure 3.2 Fecal coliform growth on M-FC agar

3.2 Methodology for Investigation of Electro-Fenton Disinfection

3.2.1 Setup Configuration and Operation

The experiment was conducted in a batch reactor in a 12 cm × 7.5 cm × 10 cm rectangular PVC tank (Figure 3.3). One pair of stainless steel electrodes was used in this case. The contact area between electrode and the electrolyte is 6 cm × 7.5 cm. In this experiment, the values for the current were selected to be 0.2A, 0.4A and 0.8A, with the current density of 4mA/cm^2, 8mA/cm^2, 16mA/cm^2 , respectively. Electrodes were placed on two sides inside the reactor, separated by 12 cm. In this batch experiment, 1000 mL of sludge solution was used for each run. Extra DI water (Milli-Q® Integral) from Sigma Inc. was used for sludge dilution to a TS content of approximately 0.8–2.8%. Electrolysis was performed using a DC power supply (GENESYS™ 750W/1500W) from TDK-Lambda Americas, Inc., US. To keep the solution homogenous, a magnetic stirrer (Fisher Scientific) was placed in the bottom to mix the solution. A thermocouple was placed in the middle of the reactor for temperature recording using data logger. The tube was placed in the middle of the reactor for sample collection and connected with the peristaltic pump (Cole-Parmer Instrument. LLC).

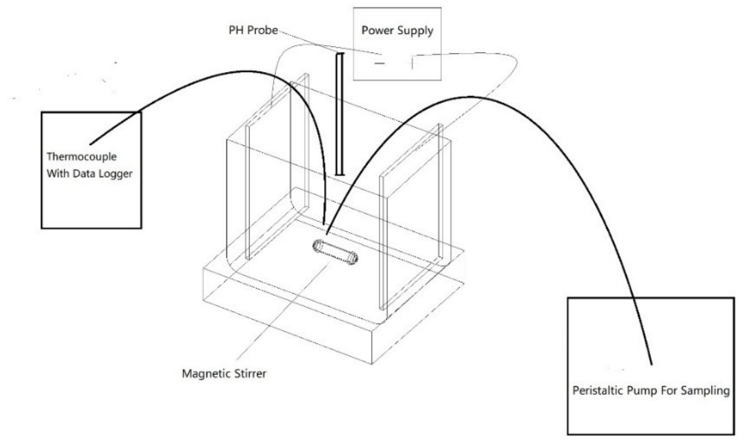


Figure 3.3 Schema of the experimental setup

Because the optimum operating pH for a Fenton reaction is between 3 and 4, in this experiment, most of the initial pH conditions were adjusted to a pH of around 4. After adjustment of the pH value, a certain amount of Fe^{2+} was introduced into the system. Then, 10 ml pre-cultured fecal coliform bacteria were injected into the system and rapidly mixed using a magnetic bar (during whole process). Finally, the fecal coliform density was brought to 5×10^6–5×10^7 cfu/ml so as to simulate the initial fecal coliform population condition. The experiment time started to count after the injection of hydrogen peroxide (30% hydrogen peroxide; Fisher Scientific). If the pH value was greater than 4, 1M hydrochloric acid was used to adjust it, but if the pH was lower than 4, no pH adjustment was implemented unless the pH was lower than 3, which was not in the range of optimum operating conditions. Additionally, for experiments 4.3-4 and 4.3-5 (Table 3.4), an absolute pH value of 4 was maintained by applying 1M hydrochloric acid and 1M sodium hydroxyl. During the whole process, pH was periodically measured when turning off the power supply. The duration of the measuring process was kept as short as possible to minimize the impact on the overall operation. Considering the difference of heat source for the experiments, external conductive heat was generated by using hot plate (Fisher Scientific), while heat generated by electric field was spontaneous occur inside the reactor.

3.2.2 Experiment Overview

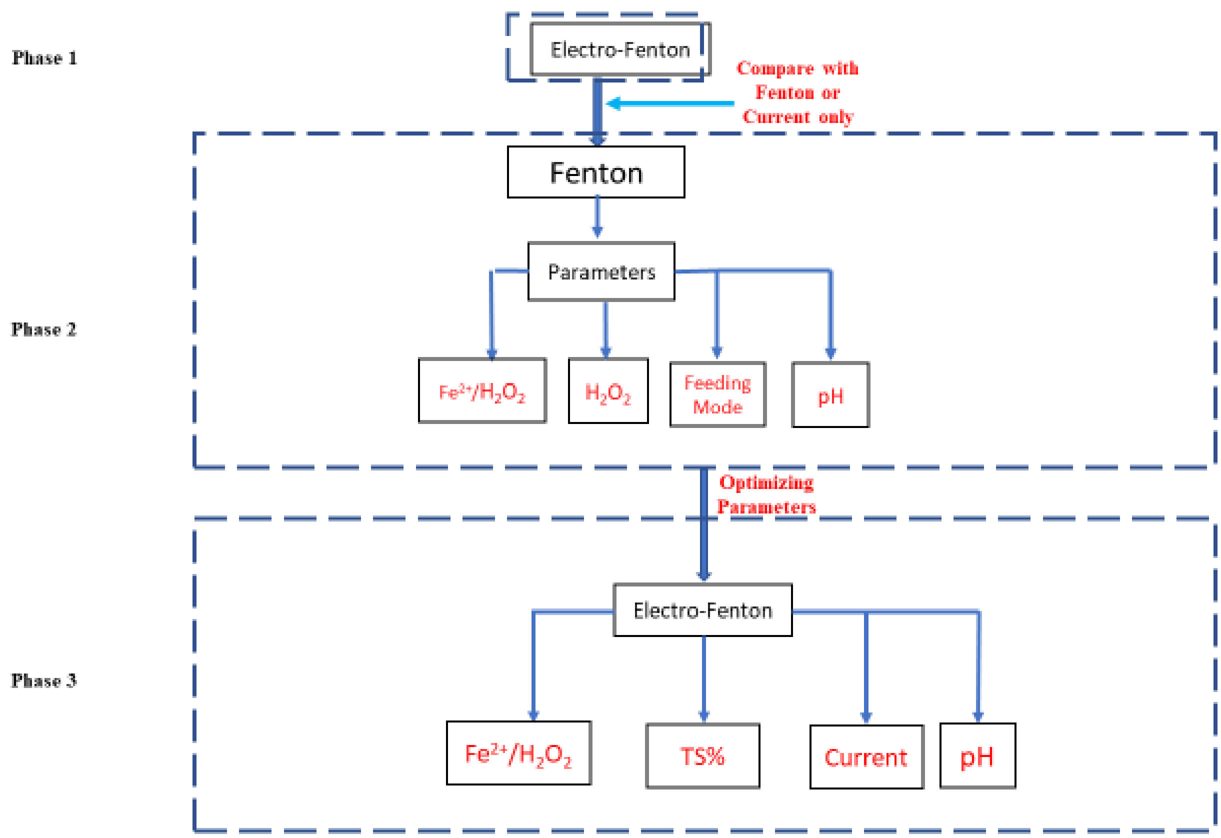


Figure 3.4 Schematic diagram for Electro-Fenton for Objective One

Figure 3.4 illustrates the experiment overview for approach one. Three phases of experiments were conducted.

Firstly, a determination of the effectiveness of multi-stressors on fecal coliform disinfection by Electro-Fenton reaction was done by comparing the experiments (Table 3.2) of Phase 1 to verify how much multi-stressors contribute to the fecal coliform disinfection

In Phase 2, the experiments focused on the individual stressors in a Fenton reaction to determine how much each of them contributed to the fecal coliform disinfection in terms of the seven different ratios of Fe^{2+}/H_2O_2 (Table 3.3), five H_2O_2 concentrations (Table 3.5), and various feeding mode and pH change (Table 3.4). Based on the determined parameters in Phase 2, the Phase 3 of the study investigated how individual stressors affect fecal coliform disinfection in Electro-Fenton like conditions regarding the 3 different ratios of Fe^{2+}/H_2O_2 (Table 3.7), four different TS% values (Table 3.6), two different pH values (Table 3.9), and three different current values (Table 3.8).

3.2.3 Tests Assessing Fecal Coliform Bacteria Content

Fecal coliform bacteria determination was according to different environmental parameters as defined in Tables 3.2 to Tables 3.9. Fecal coliform in experiments 4.3-1 to 4.3-3 (Table 3.4) and Table 3.5 were measured after 30 minutes of the treatment. However, the target value was log 3 reductions of fecal coliform.

Table 3.2 Phase 1: Determination of the Electro-Fenton effectiveness on fecal coliform disinfection

Experiment Parameters				Ref. Number	Related results
Current (A) / (current density)	Fe^{2+}/H_2O_2	H_2O_2 (ml)	Heat Source		
No	No	No	No	4.1-1	Figure 4.1A
No	No	No	Conductive heat	4.1-2	
No	1:6	0.5	No	4.1-3	
0.4(8mA/cm^2)	1:6	0.5	Electrical heat	4.1-4	
0.2(4mA/cm^2)	No	No	Electrical heat	4.1-5	Figure 4.1B
0.4(8mA/cm^2)	No	No	Electrical heat	4.1-6	
0.8(16mA/cm^2)	No	No	Electrical heat	4.1-7	

Table 3.3 Phase 2: Fenton reaction experiments for determination of the ratio of Fe^{2+}/H_2O_2 effect on the fecal coliform disinfection

Experiment Parameters				Ref. Number	Related results
Current (A)	Fe^{2+}/H_2O_2	H_2O_2 (ml)	Heat Source		
No	0	1	No	4.2-1	Figure 4.2A
No	1:12	1	No	4.2-2	
No	1:6	1	No	4.2-3	
No	1:3	1	No	4.2-4	
No	1:1.5	No	No	4.2-5	
No	2:1	No	No	4.2-6	
No	6:1	No	No	4.2-7	

Table 3.4 Phase 2: Fenton reaction experiments for determination of pH and feeding mode effects on the fecal coliform disinfection

Experiment Parameters					Ref. Number	Related results
Current (A)	Fe^{2+}/H_2O_2	H_2O_2 (ml)	Feeding mode	pH Control		
No	1:6	1	5	Yes	4.3-4	Figure 4.3B
No	1:6	1	1	Yes	4.3-5	
No	1:6	1	5	No	4.3-1	Figure 4.3A
No	1:6	1	3	No	4.3-2	
No	1:6	1	1	No	4.3-3	

Table 3.5 Phase 2: Fenton reaction experiments for determination of H_2O_2 concentration effect on fecal coliform disinfection

Experiment Parameters				Ref. Number	Related results
Current (A)	Fe^{2+}/H_2O_2	H_2O_2 (ml)	Feeding mode		
No	1:6	0.2	No	4.4-1	Figure 4.4
No	1:6	0.5	No	4.4-2	
No	1:6	1	No	4.4-3	
No	1:6	2.5	No	4.4-4	
No	1:6	5.9	No	4.4-5	

Table 3.6 Phase 3: Electro-Fenton experiments TS% effect on the fecal coliform disinfection

Experiment Parameters				Ref. Number	Related results
Current (A) /(current density)	Fe^{2+}/H_2O_2	H_2O_2 (ml)	TS%		
0.4(8mA/cm^2)	1:6	1	0.8	4.6-1	Figure 4.6
0.4(8mA/cm^2)	1:6	1	1.4	4.6-2	
0.4(8mA/cm^2)	1:6	1	2	4.6-3	
0.4(8mA/cm^2)	1:6	1	2.8	4.6-4	

Table 3.7 Phase 3: Electro-Fenton experiments for ratio of Fe^{2+}/H_2O_2 effect on fecal coliform disinfection

Experiment Parameters				Ref. Number	Related results
Current (A) / current density	Fe^{2+}/H_2O_2	H_2O_2 (ml)	Feeding mode		
0.4(8mA/cm^2)	0	0.5	1	4.9-1	Figure 4.9A
0.4(8mA/cm^2)	1:12	0.5	1	4.9-2	
0.4(8mA/cm^2)	1:6	0.5	1	4.9-3	

Table 3.8 Phase 3: Electro-Fenton experiments current effect on the fecal coliform disinfection

Experiment Parameters				Ref. Number	Related results
Current (A) / (Current density)	Fe^{2+}/H_2O_2	H_2O_2 (ml)	Heat Source		
0.2(4mA/cm^2)	1:6	1	Electrical heat	4.5-1	Figure 4.5
0.4(8mA/cm^2)	1:6	1	Electrical heat	4.5-2	
0.8(16mA/cm^2)	1:6	1	Electrical heat	4.5-3	

Table 3.9 Phase 3: Electro-Fenton experiments for pH effect on fecal coliform disinfection

Experiment Parameters				Ref. Number	Related results
Current (A) (current density)	Fe^{2+}/H_2O_2	H_2O_2 (ml)	pH		
0.8(16mA/cm^2)	1:6	1	4	4.8-1	Figure 4.8
0.8(16mA/cm^2)	1:6	1	7	4.8-2	

3.2.4 Test for pH in Electro-Fenton System

The pH value was measured when hydrogen peroxide injection divided into 5 times feeding in total of 1 ml H_2O_2; within 30 minutes with ratio of H_2O_2/ Fe^{2+}/H_2O_2=1:6 and pH=4, which was presented in section 4.3. Furthermore, in section 4.3, the pH value was also recorded 1 minute after the 1 ml H_2O_2 injection with a ratio of Fe^{2+}/ H_2O_2 of 0, 1:12, 1:6, 1:3 and 1:1, respectively.

3.3 Methodology for Investigation of the Bioxy^{TM}/Electrokinetic System

In approach two, configuration of the setup was similar to methodology one, but experiments in methodology two was conducted in two different reactors. The applied initial current values were based on a previous work (Safaei et al., 2012).

3.3.1 Setup Configuration and Operation

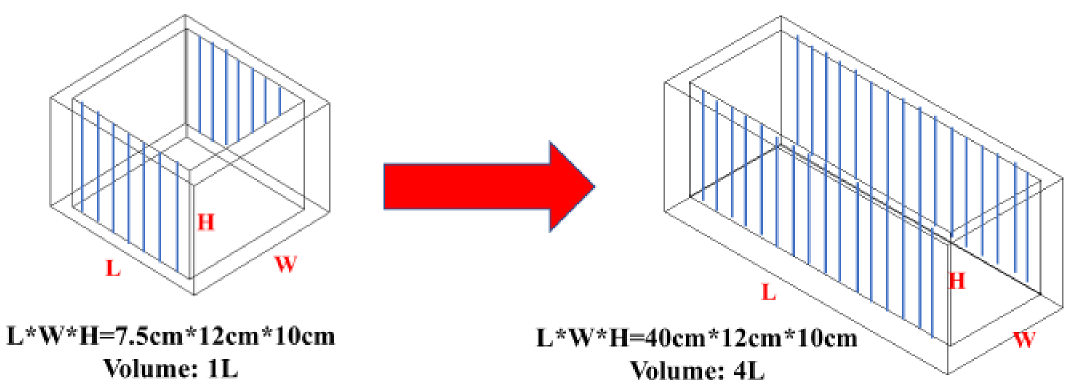


Figure 3.5 Two different configurations setup

The first stage was conducted at a 1 L scale reactor with the dimensions of L * W * H = 12 cm * 7.5 cm * 10 cm. The reactor was covered and insulated. Flat electrodes were placed on two sides inside the reactor, separated by 12 cm. Sludge was in the range of TS content between 1.2% and 3.5%, then ammonia salt was introduced into the sludge. Chemicals mixing with sludge was completed using a mechanical mixer. Then, 10 mL of pre-cultured fecal coliform medium were injected to the sludge matrix and mixed until totally homogenous. After that, pH adjustment was performed with hydrochloride acid to bring the pH to 4. Then, a certain amount of biocide was used and homogenized. In the last step, electricity was applied for treatment. Considering the difference of heat source for the experiments, external conductive heat was generated by using hot plate (Fisher Scientific), while heat generated by electric field was spontaneous occur inside the reactor.

In this experiment, a constant voltage gradient was used instead of constant current, because in this system, temperature was used for the very important factor of fecal coliform disinfection. Therefore, applying a constant voltage had the benefit of shortening the treatment time. In terms of temperature changes, a larger volume has a better thermal conservation ability; therefore, we also investigated the effect of volumetric changes on temperature and energy consumption as well as fecal coliform disinfection efficiency. For this reason, a 4 L insulated reactor of L*W*H = 40 cm * 12 cm * 10 cm was applied (Figure 3.5).

3.3.2 Experiment Overview

Figure 3.6 illustrated the overview of the approach 2. Firstly, the experiments determined the effectiveness of an electrokinetic system with ammonia salt and biocide for fecal coliform disinfection from experiments 5.1-1, 5.1-2, and 5.3-6. Furthermore, determination of temperature effect was conducted by applying different operational conditions (Exp. 5.1-1, 5.1-2, 5.3-2 and 5.3-6).

After proving the effectiveness of Electrokinetic/Bioxy^{TM} system in the first stage, continued experiments were conducted to optimize the system. Experiments 5.3-1, 5.3-2, 5.3-3, 5.3-4, 5.3-5 and 5.3-6 determined the optimal dosage for fecal coliform disinfection efficiency. Additionally, the experiments demonstrated the effect of pH on fecal coliform disinfection from experiment 5.4-1, 5.4-2 and 5.4-3.

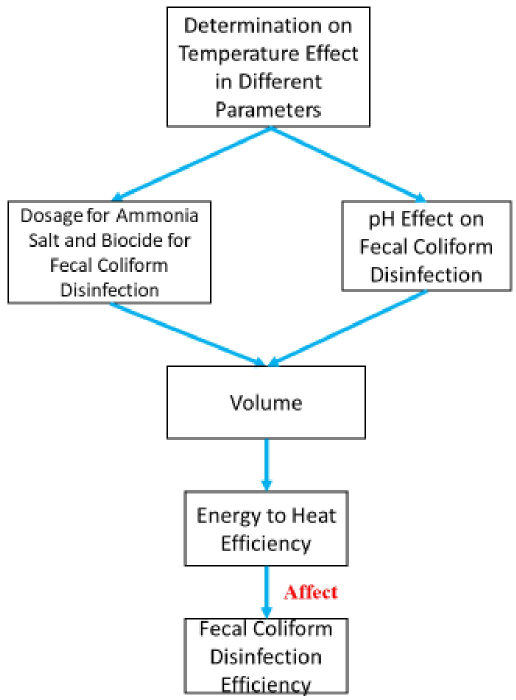


Figure 3.6 Schematic diagram for Objective Two

Finally, the experiments were conducted for different volumes to demonstrate the improvement of fecal coliform reduction efficiency regarding volume changes (Exp. 5.7-1, 5.7-2 and 5.7-3). An energy consumption evaluation was also included.

3.3.3 Test for Fecal Coliform Bacteria

Fecal coliform determinations were made according to the changes in different parameters following Tables 3.10, Tables 3.11, Tables 3.12 and Table 3.13. It should be noted that for the fecal coliform reduction tests as defined in Table 3.11, temperature was recorded as well according to the timing of the sample collection.

Table 3.10 Fecal coliform disinfection by conductive heat

Experiment Parameters				Reference Number	Related results
Voltage Gradient (V/cm)	Ammonia Salt (g/l)	Biocide (g/l)	Heat source		
No	No	No	Conductive heat	5.1-1	Figure 5.1
No	5	No	Conductive heat	5.1-2	

Table 3.11 Experiments for fecal coliform disinfection in different parameters combinations

Experiment Parameters				Reference Number	Related results
Voltage Gradient (V/cm)	Ammonia Salt (g/l)	Biocide (g/l)	TS%		
2.8	No	2.5	2.5	5.3-1	Figure 5.3
2	5	2.5	2.5	5.3-2	
2.8	1	2.5	2.5	5.3-3	
2.8	5	2.5	2.5	5.3-4	
2.8	5	1	2.5	5.3-5	
2.8	5	No	2.5	5.3-6	

Table 3.12 Experiments for fecal coliform disinfection under different pH values

Experiment Parameters				Ref. Number	Related results
Voltage Gradient (V/cm)	Ammonia Salt (g/l)	biocide (g/l)	pH		
2	1	2.5	4.2	5.4-1	Figure 5.4
2	1	2.5	6	5.4-2	
2	1	2.5	7.5	5.4-3	

Table 3.13 Experiments for fecal coliform disinfection for different parameters in 4L volume

Experiment Parameters				Ref. Number	Related results
Voltage Gradient (V/cm)	Ammonia Salt (g/l)	biocide (g/l)	Volume (L)		
2.8	5	2.5	4	5.9-1	Figure 5.9
2	5	2.5	4	5.9-2	
2.8	5	1	4	5.9-3	

3.3.4 Test for Energy Consumption and Temperature

In section 5.5, the results of the relationship between temperature and initial current are investigated. Temperature was recorded after 30 minutes of treatment, and initial current was recorded from the very beginning of the experiment.

The relationship between initial current and energy consumption is also presented in section 5.5. Energy consumption was calculated according to Eq. [5.2]. Voltage was chosen depending on the voltage gradient, and current was recorded every minute. Time was selected as the time when the temperature reached 30℃.

Chapter 4 Results and Discussion for Electro-Fenton Disinfection

A series of experiments (4.1-1, 4.1-2, 4.1-3, 4.1-4, 4.1-5, 4.1-6 and 4.1-7) as described in section 3.2 were conducted for determination of the effectiveness of the multi-stressors using Electro-Fenton system, and they are illustrated in section 4.1.

Ratio of Fe^{2+}/H_2O_2, feeding mode, pH and H_2O_2 concentrations are presented in section 4.2, 4.3 and 4.4 for investigating the Fenton reaction's impact on fecal coliform reduction.

Furthermore, the parameters such as ratio of Fe^{2+}/H_2O_2, TS%, pH, and current were evaluated for Electro-Fenton fecal coliform disinfection in section 4.5, 4.6, 4.7 and 4.8.

4.1 Comparison of Different Operational Conditions

Experiment 4.1-1 does not show a significant reduction of fecal coliform bacteria in the blank test, which confirms that the mechanical stress from the turbulence by magnetic bar and a pH =4 value did not affect on survival ability of the bacteria (0.2 log reduction after 30 minutes). This proves that fecal coliform bacteria can be used as a microbiological indicator under acidic conditions. Furthermore, sludge under an electric field can also generate heat in this system, this might also be a factor contributing to bacterial disinfection, as has been demonstrated previously by Daneshmand et al. (2012). Therefore, the effect of temperature was tested separately by using a hot plate to increase the temperature, as demonstrated in experiment. 4.1-2. The external conductive heat from the hot plate did not show any negative effects on the fecal coliform bacteria, proving that the temperature increase caused by heat from electric field in experiment 4.1-6 did not contribute to the fecal coliform reduction

Hydrogen peroxide is commonly considered a strong oxidant when added with ferrous iron. The reaction between Fe^{2+} and H_2O_2 can generate hydroxyl radicals $^{\bullet}OH$ (Eq. [2.3]). Therefore, an individual test was conducted to determine the fecal coliform disinfection efficiency of hydroxyl radicals. Experiment 4.1-3 achieved 1 log reduction in 10 minutes and almost 3 log reductions after 30 minutes of treatment. This demonstrates the effectiveness of the production of hydroxyl radicals from a Fenton-reaction on fecal coliform disinfection as an individual stressor, which is similar to the results of Ortega-Gómez et al. (2012). They achieved 3 log reductions after 60 minutes of photo-Fenton reaction treatment. However, they only achieved 1 log reduction in their experiment after 30 minutes. This might be due to an insufficient amount of ferrous iron in their matrix to interact with all of the available hydrogen peroxide, leading the hydrogen peroxide to dissociate. Besides, the disinfection mechanism in this study was based not only on the generation of hydroxyl radicals $^{\bullet}OH$ but also on the huge amount of ferric hydroxyo complex (Exp. 4.1-5 to 4.1-7 from Table 3.2) that contributed to the fecal coliform decrease (discussion below).

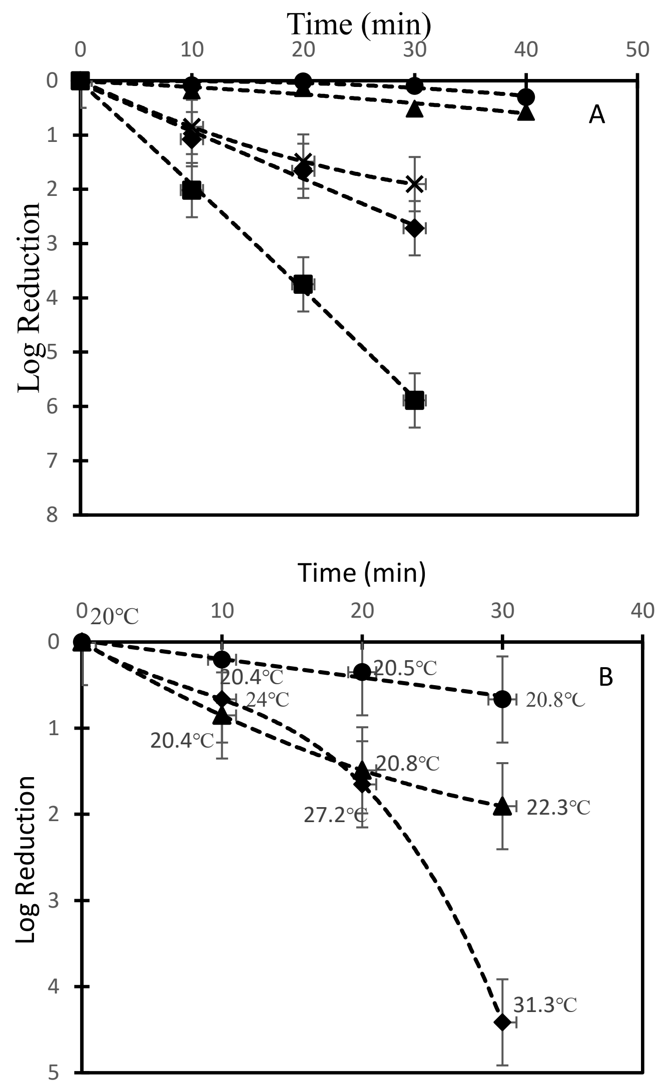


Figure 4.1 (A): Fecal coliform reduction vs. time in different operating conditions; (B): Fecal coliform vs. time for different currents

Note: (A): (●) 4.1-1 Blank condition; (▲) 4.1-2 Heat only (Temperature raise the same as when it only apply 0.4A); (×) 4.1-6 0.4A Current Only; (◆) 4.1-3 Fenton system with 0.5ml H_2O_2 only with Fe^{2+}/H_2O_2=1:6; (■) 4.1-4 Electro-Fenton system with 0.4A current with 0.5 ml H_2O_2 with Fe^{2+}/H_2O_2=1:6; Fecal coliform disinfection efficiency under different operating condition; All of the experiments above were operated under pH:4 by using HCl (B): (●) 4.1-5 0.2A Current only (▲) 4.1-6 0.4A Current only (◆) 4.1-7 0.8A Current only; All experiments operated under pH=4 condition.

Experiment 4.1-6 achieved 2 log reductions of fecal coliform after 30 minutes of treatment by electrical field only (0.4 A). Comparing the results of experiments 4.1-2 and 4.1-6, it can conclude that that heat is not a stressor causing fecal coliform disinfection in experiment 4.1-6. In some previous work, it has been demonstrated that electro-coagulation can also contribute to bacteria removal. This disinfection mechanism has been proven by Delaire et al. (2015); they stated declared that there are two main mechanisms contributing to electro-coagulation disinfection. The first contribution is from physical removal by the flocs formation process. The second is the generation of Fe^{2+} from the sacrificed stainless-steel anode; the Fe^{2+} is oxidized in the presence of oxygen in the medium, causing microbe disinfection. A similar approach was also demonstrated by Kim et al. (2011); they successfully utilized ferrous iron for MS2 coliphage disinfection.

As the current increased in experiments 4.1-5, 4.1-6 and 4.1-7, fecal coliform disinfection efficiency also increased. There was only a 0.5 log reduction observed for a current of 0.2 A, which increased to 4 log reductions at 0.8 A after 30 minutes of treatment. Comparing the results among 0.2 A, 0.4 A, 0.8 A, fecal coliform reduction increased significantly at 0.8 A, especially in the results after 20 minutes. Figure 4.1B demonstrates the relationship between temperature and time. It shows that at 0.2 and 0.4 A, the temperature remained the same (20-22℃) all along the treatment process, but at 0.8 A there were obvious differences from the very beginning, as the temperature increased to 24℃ at 10 minutes, 27℃ at 20 minutes and 31℃ at 30 minutes. It is interesting to find that there was no difference in fecal coliform reduction in the first 20 minutes between treatments applying 0.4 A and 0.8 However, when 0.8 A was used, 4 log reductions has been achieved in 30 minutes. For this reason, it was assumed that the extra reduction in fecal coliform bacteria was the effect of the temperature and the electrical field. This was also proven by Yin et al. (2018); they demonstrated that a temperature of 30℃ yields a 2 log reduction in *E. coli*.

Comparing the results of experiments 4.1-4, 4.1-3 and 4.1-6, a 6 log reduction was achieved in 4.1-4 after 30 minutes of treatment. The results of 4.1-6 indicated only a 1.9 log reduction, while applying only H_2O_2 in 4.1-3 achieved a 2.8 log reduction of fecal coliform bacteria. With these results, it has demonstrated the effectiveness of the Electro-Fenton system for fecal coliform disinfection, and that this system exerts extra negative stress on fecal coliform bacteria, leading to improved fecal coliform disinfection efficiency compared to the application of individual stressors.

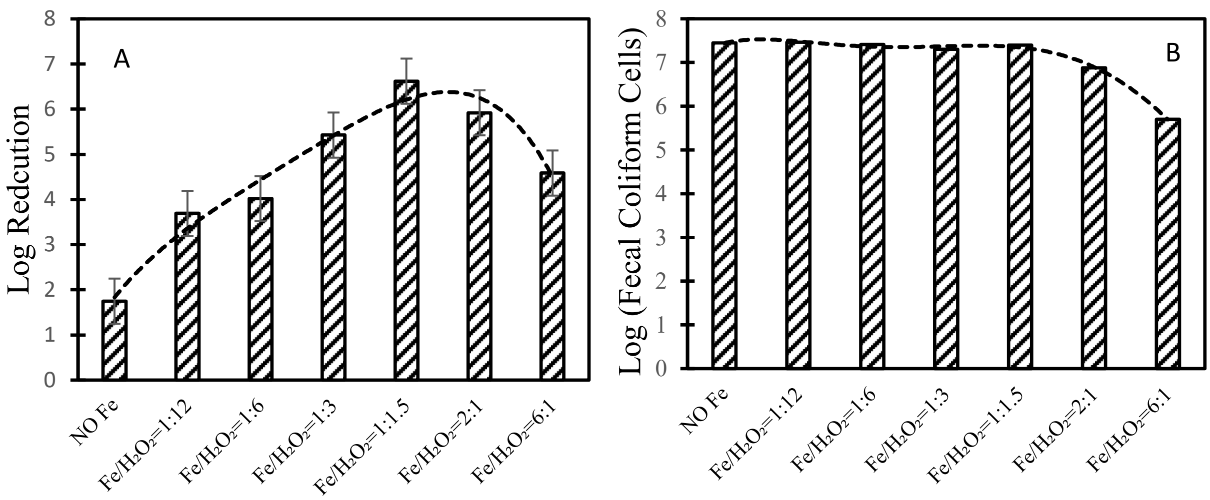


Figure 4.2 (A): Fecal coliform reduction after 30 minutes vs. different ratios of Fe^{2+}/H_2O_2 in Fenton system; (B): Initial population of fecal coliform vs. different ratios of Fe^{2+}/H_2O_2

4.2 Investigation of Fe^{2+}/H_2O_2 on Fenton System

Different ratios of Fe^{2+}/H_2O_2 were investigated in this section so as to identify the best ratio for fecal coliform disinfection. Without adding Fe^{2+} (Exp. 4.2-1), disinfection efficiency was not very significant, but a 1.8 log reduction was still achieved. This reduction was contributed by the presence of ferrous iron from our original sludge, which provided the necessary inputs for the Fenton-reaction. According to Selvakumar et al. (2009), under the condition of 1 mL/L of 30% H_2O_2, no obvious fecal coliform reduction was observed, which indicates that hydrogen peroxide did not function as a disinfectant in Figure 4.2A for fecal coliform disinfection.

At the same time, increased ferrous iron concentration resulted in a stronger fecal coliform reduction. A 3.8 log reduction was observed with a higher ferrous iron concentration in a 1:12 Fe^{2+}/H_2O_2 ratio. As Fe^{2+} increased, fecal coliform disinfection efficiency increased as well, indicating fecal coliform reduction proportionally increase as the amount of Fe^{2+} added. A 4 log reduction was observed for a Fe^{2+}/H_2O_2 ratio of 1:6 and a 5.2 log reduction for the ratio of 1:3. The maximum fecal coliform reduction was observed at the Fe^{2+}/H_2O_2 ratio of 1:1.5. This demonstrates the contribution of ferrous iron to fecal coliform disinfection, which is due to the increased production of hydroxyl radicals ($^\bullet OH$) with increasing Fe^{2+}. The results from previous research on the Fenton method (Juarez et al. 2010) has also shown that disinfection efficiency increases as the ratio of Fe^{2+}/H_2O_2 increases. It was assumed that the disinfection of MS2 bacteriophages was mediated by iron colloids. Furthermore, similar research conducted by Zhang et al. (2012) demonstrated that a higher Fe^{2+}/ H_2O_2 ratio could lead to low COD removal via hydroxyl radical ($^\bullet OH$) oxidation. They stated that the COD removal under a high Fe^{2+}/ H_2O_2 ratio occurs due to coagulation as the ferric iron concentration increases (Eq. [2.3]). However, their theory cannot be applied to fecal coliform disinfection in our case because the mechanism for disinfection in our system was based not only on hydroxyl radical ($^\bullet OH$) oxidation but also on coagulation disinfection (Delaire et al., 2015; Kim et al., 2011). Additionally, some research has focused on the effectiveness of ferric iron for bacterial reductions and has validated the application of ferric iron for bacterial disinfection (Reimers et al., 2015).

As shown in Figure 4.2B, an increasing Fe^{2+} concentration results in a reduction in initial fecal coliform density. It was observed that as the Fe^{2+}/ H_2O_2 ratio increased to 2:1, the initial population of fecal coliform bacteria decreased to $6.8*10^6$ cfu/mL, and it continued to decrease to $5.6*10^5$ cfu/mL at a ratio of 6:1. The reason for that relationship is that when excess Fe^{2+} is present in the sludge under air-saturated and low pH conditions, the ferrous iron will be oxidized by oxygen and generate disinfectants such as ($^{\bullet}OH$) and Fe(IV), contributing to *E. coli* reduction (Kim et al., 2011).

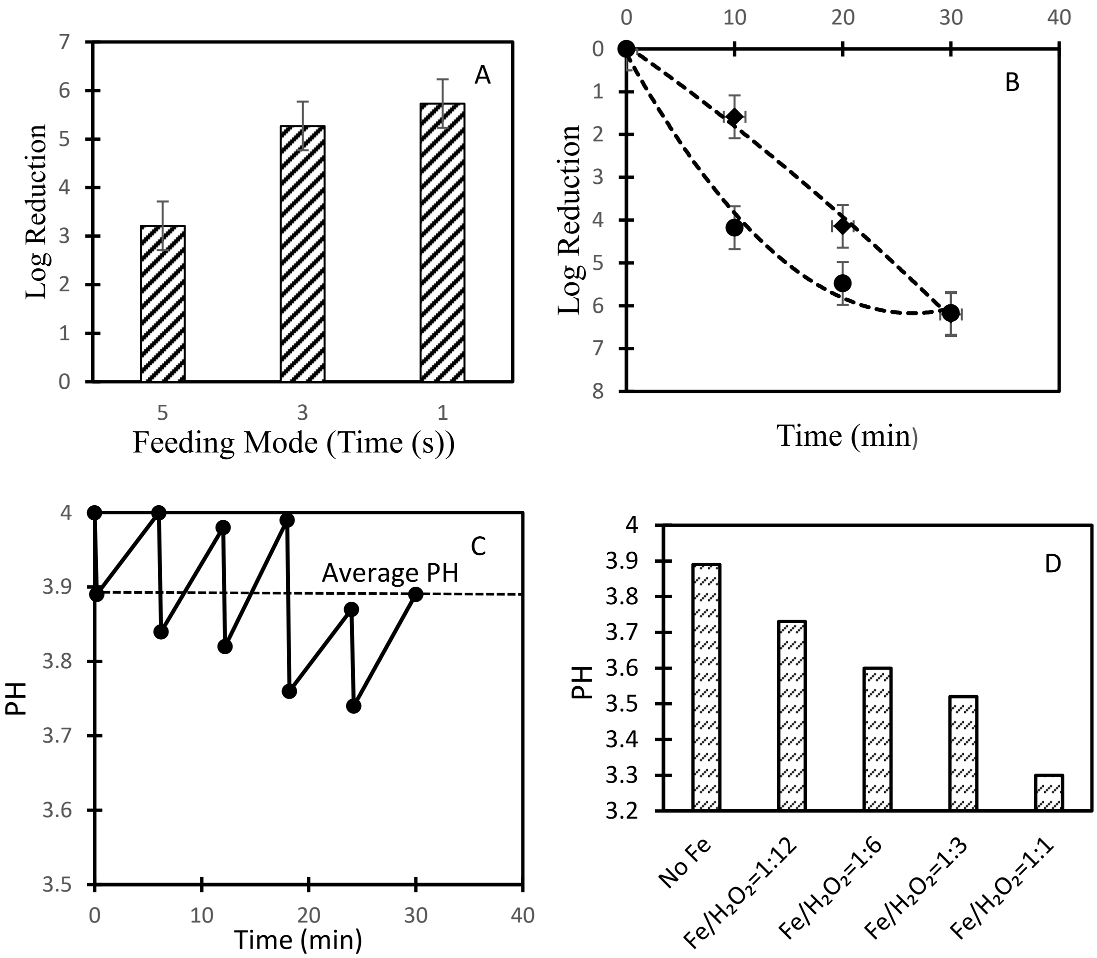


Figure 4.3 (A): Fecal coliform reduction vs. different feeding mode (i.e. frequency of H_2O_2 application); (B): Fecal coliform reduction vs. different feeding mode in constant pH; (C): pH vs. time; (D): pH vs. different ratio of Fe^{2+}/H_2O_2

Note: (A): Fecal coliform disinfection efficiency for the Fenton system after 30 minutes at different feeding modes 4.3-1, 4.3-2 and 4.3-3; (B) fecal coliform disinfection efficiency for Fenton system in two different feeding modes (maintained pH 4); (●) 4.3-5, once H_2O_2 application; (◆) 4.3-4, five times H_2O_2 application, while Fe^{2+}/H_2O_2 = 1:6 with 1ml H_2O_2; (C): the pH changes during the Fenton reaction for Fe^{2+}/H_2O_2 = 1:6 in 5 times H_2O_2 application ; (D): pH changes in the Fenton system when adding 1ml H_2O_2 at different Fe^{2+}/H_2O_2 ratios.

4.3 Study of Feeding Mode and pH on Fenton System

The method of feeding hydrogen peroxide H_2O_2 has a strong effect on production of hydroxyl radicals ($^{\bullet}OH$) according to Zhang et al. (2012). As shown in Figure 4.3A, experiments 4.3-1 4.3-2 and 4.3-3 were conducted under the condition of no pH control. As the times of feedings was increased, fecal coliform disinfection efficiency significantly decreased. Feeding five times only achieved a 3.1 log reduction in fecal coliforms, whereas 3-times feedings resulted in a 5.2 log reduction and 1-time feeding, a 5.8 log reduction. Figure 4.3B indicates that experiments 4.3-4 and 4.3-5 were conducted under the condition of maintaining an absolute pH value of 4. In the first 10 minutes, a 4.2 log reduction was achieved for 1-time feeding and only a 1.8 log reduction for 5-times feedings. However, the gap between the 2-times feeding modes became smaller over time. At 20 minutes, the treatment with 1-time feeding achieved a 5.5 log reduction and the 5-times feeding treatment achieved a 4 log reduction. It is interesting to see that eventually, the 5-times feeding treatment in experiment. 4.3-4 reached the same fecal coliform reduction as with 1-time feeding experiment. 4.3-5 after 30 minutes of treatment.

Therefore, it was concluded that the differences among different feeding modes were caused by the pH changes that occurred when applying hydrogen peroxide H_2O_2. The reason for that is that adding excess H_2O_2 will instantly initiate the Fenton reaction. A huge amount of ferrous iron is then converted into ferric iron according to Eq. [2.3], and then ferric iron is hydrolyzed into a form of ferric hydroxo complexes according to Lin and Lo (1997). Therefore, due to the sudden generation of a large amount of ferric iron, hydroxyl ions are converted into $Fe(OH)_n$, which results in the pH drop. That is why the results in Figure 4.3D show only slight changes in pH without adding ferrous iron (Exp. 4.3-1), but as increase the ferrous iron dosage, the pH gradually decreases. In particular, when the Fe^{2+}/H_2O_2 ratio was 1, the pH dropped to 3.3 after H_2O_2 injection. Also,

Figure 4.3C illustrates the changes in pH under the 5-times feeding mode. Each pH value drop was caused by a hydrogen peroxide injection, but because sludge is exposed to the atmosphere, absorption of carbon dioxide will slowly increase the pH, finally resulting in an average pH that remains around 3.9. This is the reason why feeding one time yields a higher fecal coliform reduction than feeding 5 times. Because the average pH value of 1-time feeding is 3.6 when $Fe^{2+}/H_2O_2 = 1:6$. According to previous work (Ghoneim et al., 2011; Wang et al., 2008; Zhou et al., 2007), the optimum pH for the Fenton reaction is near 3-3.5. Within this pH range, the production of hydroxyl radicals ($^\bullet$OH) in the solution is at its highest. That is why fecal coliform reduction has the highest efficiency under the 1-time feeding mode.

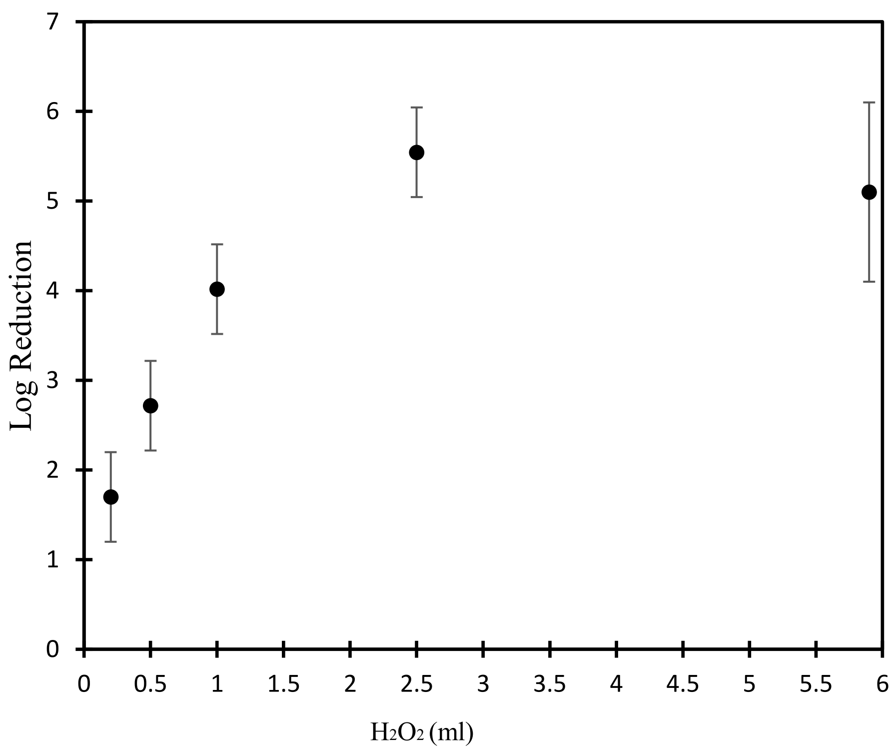


Figure 4.4 Fecal coliform reduction in 30 minutes vs. dosage of H_2O_2

Note: Fecal Coliform disinfection efficiency in Fenton reaction with different H_2O_2 dosage with Fe^{2+}/H_2O_2=1:6 under pH=4 operating conditions

4.4 Study of Dosage of H_2O_2 on Fenton System

From Figure 4.4, when applying 0.2 ml of hydrogen peroxide to the sludge matric, a 1.8 log reduction in fecal coliforms was observed. When the dosage was increased to 0.5 ml, a 2.5 log reduction achieved; finally, when the amount of hydrogen peroxide was increased to 5.9 ml, only a 5 log reduction was achieved after 30 minutes of treatment. Overall, in a particulate ratio of Fe^{2+}/H_2O_2, increasing the H_2O_2 dosage could achieve higher fecal coliform reduction; however, the significant changes only occur when the dosage of hydrogen peroxide is less than 1 ml. Further increases in the hydrogen peroxide dosage tend to result in lower disinfection efficiency, with a maximum achievement of around 5.5 log reduction in fecal coliforms. These results are similar to those of Deng et al. (2007); they used 6 ml of hydrogen peroxide to achieve maximum COD removal in leachate with a Fe^{2+}/H_2O_2 ratio of 1:6.

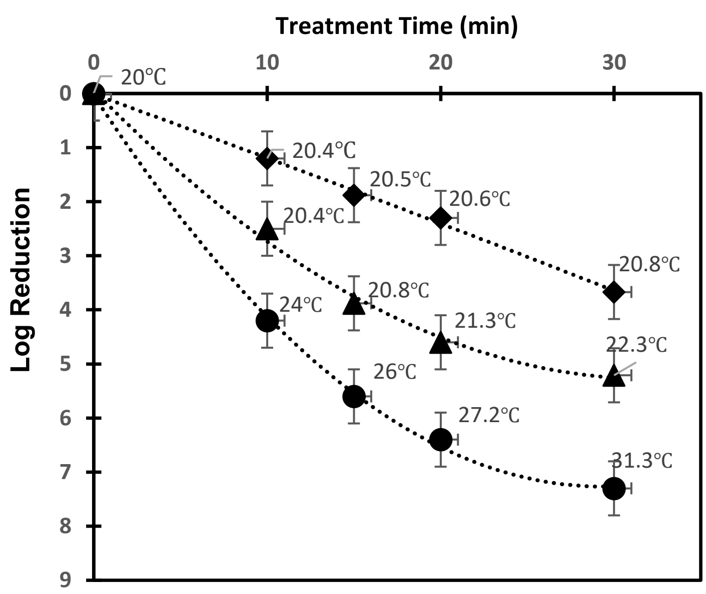


Figure 4.5 Fecal coliform reduction vs. time in different current generating Ohmic heat with time

Note: Experiments were under Electro-Fenton conditions of pH=4 with TS: 0.8%; additional ferrous iron and H_2O_2 (1ml) 5 times feeding mode; ratio of Fe/ H_2O_2 =1:6; current (◆) 4.5-1, 0.2A; (▲) 4.5-2, 0.4A; (●) 4.5-3, 0.8A; temperature was recorded along treatment time

4.5 Effect on Current Density for Electro-Fenton System

As shown in Figure 4.5, experiment 4.5-1 resulted in a 1 log reduction after 10 minutes of treatment and a 2 log reduction achieved in 20 minutes. Finally, after 30 minutes of treatment, a current of 0.2 A yielded a 3.6 log reduction in fecal coliforms. In experiment 4.5-2, a 2.5 log reduction was achieved in the first 10 minutes, which increased to a 5.2 log reduction in 30 minutes. For the experiment in 4.5-3, a higher fecal coliform reduction was observed, with a 7.1 log reduction achieved in 30 minutes.

The above results prove that under multi-stressor (Electro-Fenton) conditions, current is an important factor controlling fecal coliform disinfection efficiency. It is interesting to compare the results between experiments 4.1-6 and 4.5-2 and between experiment 4.1-7 and 4.5-3. In the first 20 minutes, no significant difference regarding fecal coliform reduction was observed between the 0.8 A and 0.4 A currents in experiments 4.1-7 and 4.1-6, while there was an obvious difference between experiment 4.5-3 and 4.5-2. This demonstrates that the effectiveness of the Electro-Fenton system can improve fecal coliform disinfection efficiency in the first 20 minutes of treatment.

These results also prove that heat generated by DC electric field was not the main stressor causing fecal coliform disinfection. However, some published articles have declared that heat generated from electric field can be used for disinfection (Yin et al. 2018). They demonstrated that by using heat generated by electric field, when temperature increased to 50 °C, a 4.5 log reduction could be achieved, while only a 0.68 log reduction was observed for a temperature of 28°C. Their results partially confirmed that the heat from electric field could cause negative stress on fecal coliform. Although increasing the current could produce a higher reduction rate, it also requires more energy to produce heat. Thus, it does not seem to be necessary for an adequate disinfection of fecal coliform in thickened WAS. Some investigations of cost-effectiveness should be conducted in the future.

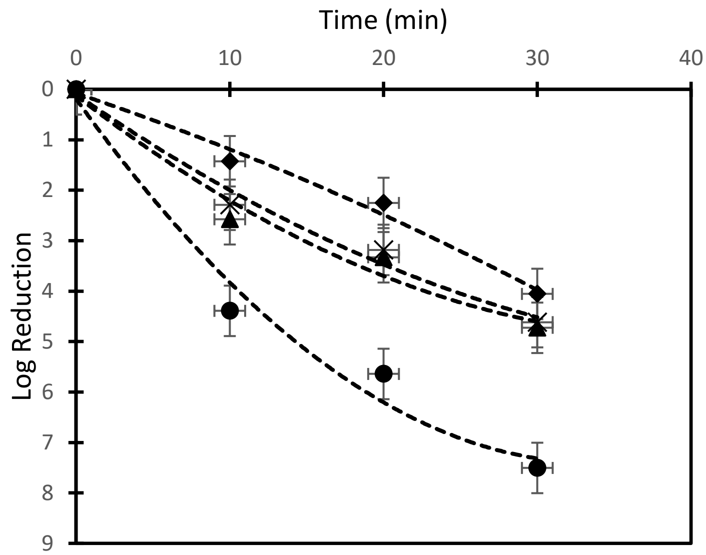


Figure 4.6 Fecal coliform reduction vs. time in different TS values

Note: Fecal Coliform disinfection efficiency in Electro-Fenton system with different TS% with Fe/H_2O_2=1:6 under pH:4 operating conditions; (●) 4.6-1, TS:0.8%; (▲) 4.6-2, TS:1.4%; (×) 4.6-3, TS:2% ; (◆) 4.6-4, TS:2.8% ;

4.6 Study of Total Solids (TS) for Electro-Fenton System

Another factor that considered in this paper is the thickness of the sludge. Comparisons were made for sludge samples with total solids (TS) ranging from 0.8 to 2.8%. The TS content of 0.8% corresponded with the highest fecal coliform disinfection efficiency after 30 minutes of treatment, with a 7.5 log reduction. Only a 4 log reduction was achieved in the 2.8% TS sludge. Interestingly, the results for TS contents of 1.4% and 2% were similar after 30 minutes of treatment, with a 4.6 log reduction for the 2% TS sludge and a 4.7 log reduction for 1.4% TS. This implies that with a high TS content, fecal coliform disinfection decreases. This is because of the presence of high organic loading, which consumes more hydroxyl radicals ($^{\bullet}OH$) than in the low TS medium.

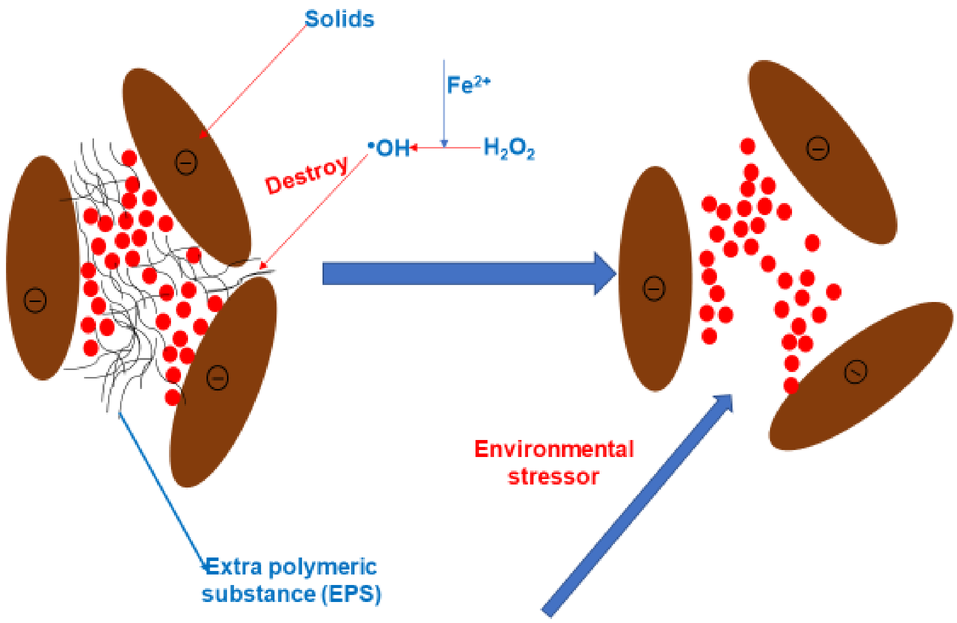


Figure 4.7 Schematic diagram of elimination of the protective EPS by developed process

Therefore, disinfection by oxidation decreases; instead, fecal coliform disinfection relies more on electrocoagulation in high TS sludge. Additionally, a high solid content also provides much more protection to bacteria because bacteria can be protected by the extra polymeric substance (EPS), which can act as a barrier, preventing contact between hydroxyl radicals and fecal coliforms (Xia et al., 2016), which shows in figure 4.7.

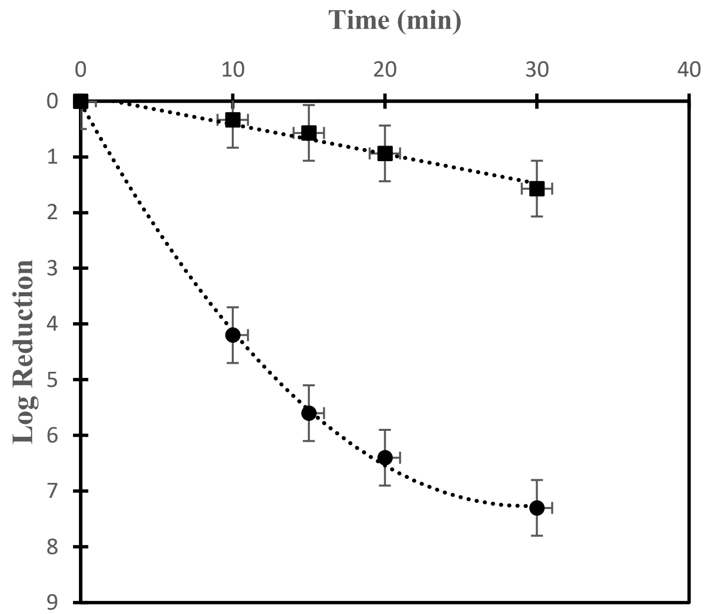


Figure 4.8 Fecal coliform reductions vs. time at pH 4 and pH 7

Note: (A): (●) 4.8-1 pH = 4; (■) 4.8-2 pH=7; experiments were under Electro-Fenton operation of 0.8 A; 1 ml H_2O_2 with Fe^{2+}/H_2O_2 = 1:6 and TS = 0.8%

4.7 Effect of pH for Electro-Fenton System

As demonstrated above in section 4.3, pH has a very important effect on fecal coliform disinfection in Fenton conditions, which assumed it was also important for Electro-Fenton as well. In experiment 4.8-1 and 4.8-2. A significant difference was observed regarding fecal coliform reduction between pH values of 4 and 7. The experiment at pH 4 achieved a 4 log reduction in the first 10 minutes, whereas only a 0.2 log reduction was achieved with a pH of 7 for test 4.8-2. No further significant change for the pH=7 treatment was observed even when the exposure time was extended to 30 minutes. This demonstrates that contact time in the system was not the main factor affecting disinfection. To the contrary, the amount of hydroxyl radicals (•OH) produced determined fecal coliform disinfection. This has also been shown by Diagne et al. (2007), who removed methyl parathion using hydroxyl radicals ($^{\bullet}OH$) at pH = 3-4 and at pH = 7. For this reason, it can be confirmed that the pH value is the critical factor for fecal coliform disinfection efficiency. According to Nidheesh and Gandhimathi (2012), an Electro-Fenton reaction can only occur under acidic conditions, so increasing pH leads to an increased reliance on the coagulation process rather than oxidation Eq.[2.2]. Additionally, the formation of ferrous iron in solution involves a series of reactions that depends on pH. In a neutral environment medium, ferrous iron will extensively hydrolyze, ending in the precipitation of $Fe(OH)^{2+}$ (Rodríguez-Chueca et al., 2014). Furthermore, another explanation (as has been suggested by Pang et al., 2011) may be attributed to the formation of surface bonding hydroxyl radicals. This assumes that hydroxyl radicals can be generated even in a neutral environment, but that they bond with the surface of insoluble metal species (e.g. ferric hydroxides). Subsequently, hydroxyl radicals cannot unselectively oxidize fecal coliforms.

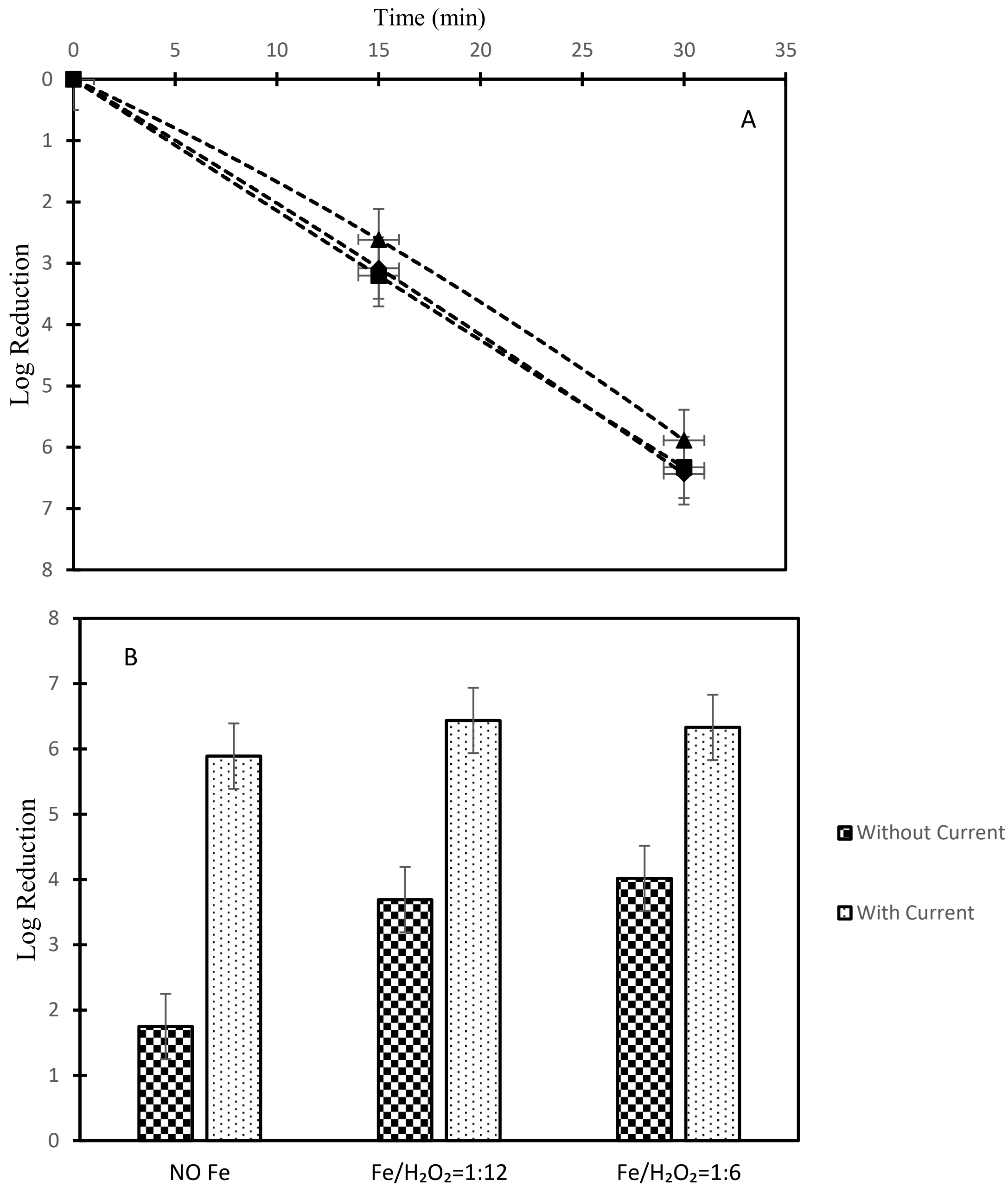


Figure 4.9 (A): Fecal coliform reduction vs. different ratios of Fe^{2+}/H_2O_2 with in Electro-Fenton system; (B): Fecal coliform reduction vs. different ratios of Fe^{2+}/H_2O_2 for the comparison between Electro-Fenton and Fenton system after 30 minutes

Note: (A): Experiment under the Electro-Fenton system of 0.4A current with 0.5 ml H_2O_2 in pH:4; (●) 4.9-3, Fe/H_2O_2 = 1:12; (▲) 4.9-2, Fe/H_2O_2 = 1:6; (◆) 4.9-1, No Fe addition;

4.8 Fe^{2+}/H_2O_2 effect on Electro-Fenton system

Figure 4.9A shows that all experiment 4.91, 4.9-2 and 4.9-3 achieved 3 log reduction after 15 minutes and 6 log reduction in 30 minutes, while no obvious change in fecal coliform reduction with changes to the Fe^{2+}/H_2O_2 ratio was observed. However, by comparing the results from experiments 4.2-1 4.2-2 and 4.2-3, it is apparent that the ratio of Fe^{2+}/H_2O_2 has a significant effect on Fenton disinfection, while it does not show such an effect on the Electro-Fenton system (Figure 4.9B). This demonstrates that the concentration of ferrous iron in the Electro-Fenton system is not as important as for a Fenton reaction. This could be due to the regeneration of ferrous iron at the cathode by Eq. [2.5], which reacts with hydrogen peroxide and produces sufficient hydroxyl radicals for the oxidation process. This can also explain why a Fenton reaction requires a Fe^{2+}/H_2O_2 ratio of 1:1.5 to achieve a 6.8 log reduction, whereas in an Electro-Fenton system, even a small amount of Fe^{2+} (from the original sample) can achieve a 6-log reduction in fecal coliforms. Finally, in this chapter, the experimental data error was around 7% for the fecal coliform disinfection (X axis), which based on ±0.5 log reduction deviation during the repetitive data. Error for the time (Y axis) is around ±2 minutes, which is due to the sample collection during the experiment.

Chapter 5 Results and Discussion for The Bioxy^{TM}/Electrokinetic System

A series of experiments were conducted (as defined in section 3.3) to compare Bioxy^{TM}/Electrokinetic results with a system without electrokinetic (external source of heat only) with respect to fecal coliform reduction (section 5.1). Then, a comparison of different parameters on fecal coliform reduction regarding ammonia salt, biocide and voltage gradient were presented in section 5.2.

Furthermore, the parameters such as pH, volume of sludge were evaluated regarding fecal coliform reduction in section 5.3 and 5.5, respectively. Besides, energy consumption was also considered in section 5.4.

5.1 Demonstration of the effectiveness of the Bioxy^{TM}/Electrokinetic system

The effect of temperature stress on fecal coliform disinfection efficiency is presented in Figure 5.1 and Figure 5.2. For the sludge in experiment 5.3-6, a 3.8 log reduction in fecal coliforms was achieved after 30 minutes, whereas at the same time in experiments 5.1-1 and 5.1-2, no significant fecal coliform reduction was observed. One notable observation is that after 30 minutes of treatment in experiment 5.3-6, the temperature reached 38.5 °C (heat providing by the electrical field); on the other side, the hot plate operation (providing external heat) also achieved this temperature. Therefore, it can be concluded that temperatures up to 38.5°C do not appear to cause any stress to the fecal coliform in sludge matrix (Figure 5.1).

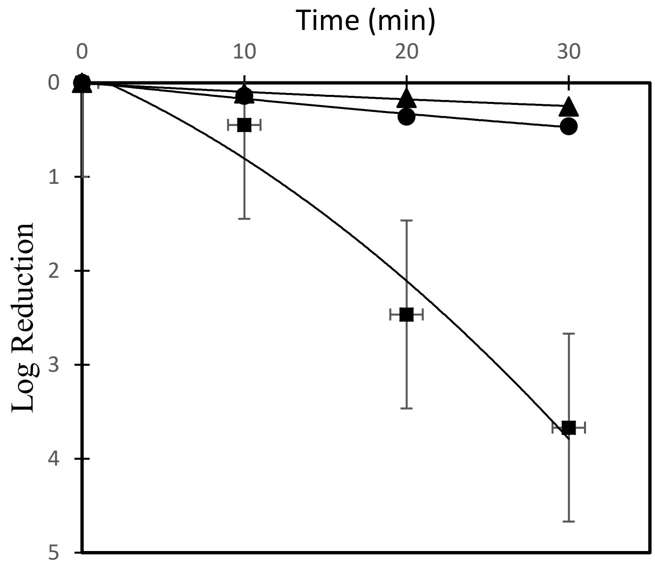


Figure 5.1 Fecal coliform reduction vs. time for different operation conditions

Note: (▲) 5.1-1,Without EK system (External source of heat only); (●) 5.1-2, Without EK system (External source of heat with 5g/L salt); (■) 5.3-6, 2.8V/cm + 5g/L Salt; Comparison on fecal coliform disinfection efficiency among different operation conditions; Temperature raising rate is the same for three of the experiments, for sludge with TS= 2.5% and pH=4.3

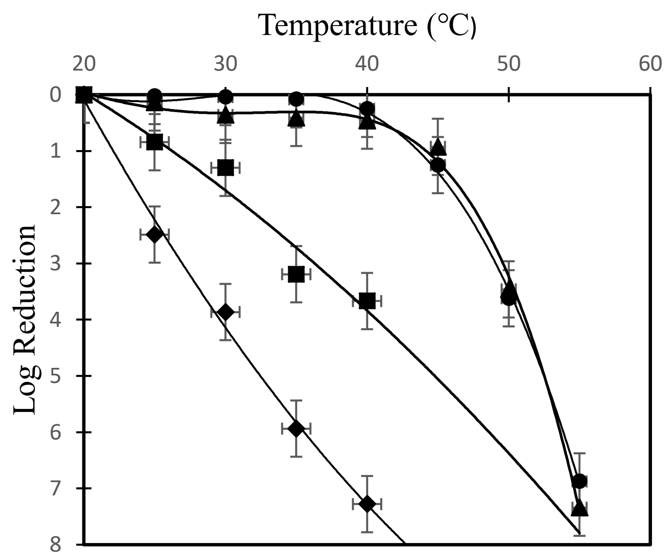


Figure 5.2 Fecal coliform reduction vs. time in different temperatures for different operation conditions

Note: (▲) Without EK system (External source of heat with 5g/L salt); (●) Without EK system (External source of heat only); (■) 2.8V/cm + 5g/L Salt; (◆) 2V/cm + 2.5g/L BS + 5g/L Salt; Fecal coliform disinfection results under different temperatures among four types of different operation conditions. Experiment was operated under TS=2.5% and pH=4.3 conditions

In experiments 5.1-1 and 5.1-2, there were no changes in fecal coliform density until the temperature reached 42℃. A 1 log reduction was achieved at 45℃, and a 7 log reduction at 55℃. This confirms the efficiency of thermal treatment for pathogen disinfection, as it mentioned above in the discussion on US EPA regulations. On the other hand, in experiment 5.3-6, a 1.5 log reduction in fecal coliforms was achieved at 30℃ and a 4 log reduction at 40℃, which is a huge difference compared to experiments 5.1-2. This could be attributable to the higher bacterial disinfection efficiency in the presence of an electrical field. It was speculated that electrokinetic might cause a charge imbalance between the inner and outer cell membranes, which leading to a compression on cell membrane and eventually causing irreversible cell damage (Zimmermann et al,. 1986). Application of multiple stressors (Electrokinetic/Bioxy™) to bacteria improves the fecal coliform disinfection efficiency (Safaei et al. 2013). Some published articles have also demonstrated that by using heat generated by DC electric field, when the temperature increases to 50 ℃, a 4.5 log reduction can be achieved, while a 0.68 log reduction was achieved for a temperature of 28℃ (Yin et al. 2018), which is very similar to our result in experiment 5.3-6.

In experiment 5.3-2, a 4 log reduction was observed at 30℃, which indicates much better fecal coliform disinfection efficiency compared to test 5.3-6. This increased to a 7 log reduction at 40 ℃, compared to only a 0.2 log reduction at 40℃ for conductive heat alone (Exp. 5.1-1). Therefore, we can assume that biocide combined with an electric field accelerates the overall fecal coliform disinfection. This might be also associated with in situ production of hydrogen peroxide when Peracetic acid (PAA) is generated from biocide (Eq. [5.1]- [5.2]). It might interact with the ferrous iron (Eq. [2.3]) to produce hydroxyl radicals, oxidizing the organic materials such as EPS (Extra polymer substrate). Meanwhile, because ferric iron is also produced by oxidation of ferrous iron, it could neutralize the charge on the surface of the solid particles and eventually decrease the thickness of the electrical double layer, weakening the protection for fecal coliforms provided by the electrical double layer (EDL) and EPS (Poortinga et al., 2002; Xia et al., 2016).

[5.1] $2NaCO_3 \cdot 3H_2O_2 \rightarrow 4Na + 2CO_3^{2-} + 3H_2O_2$ (Dagher et al., 2017)

[5.2] H_2O_2 + TAED →TriAED + DAED + 2 PAA (Dagher et al., 2017)

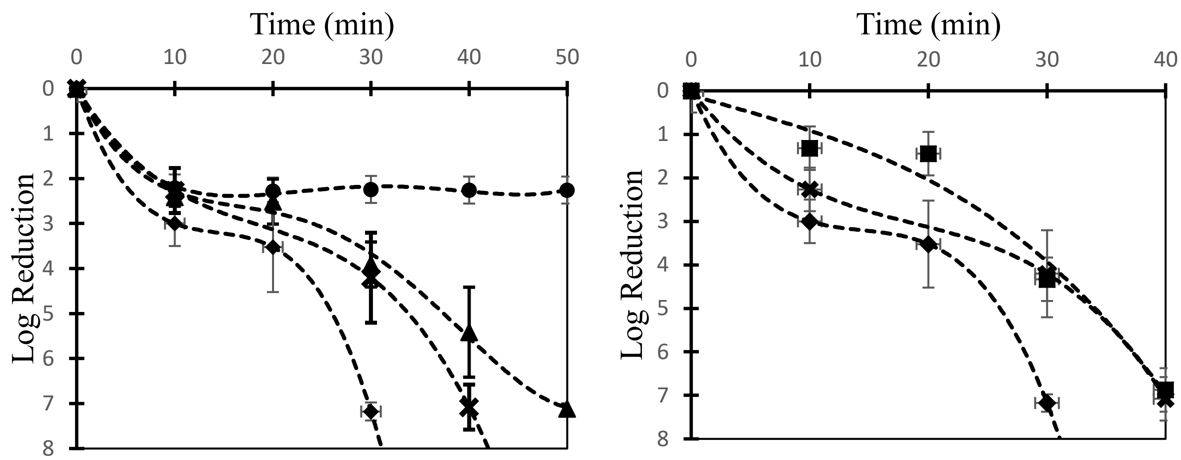


Figure 5.3 Fecal coliform reduction vs.time for different combinations of parameters

Note: (●) 5.3-1, 2.8V/cm (33.6V) + 2.5g/L BS; (▲) 5.3-2, 2V/cm (24V) + 5g/L Salt + 2.5g/L BS; (×) 5.3-3, 2.8V/cm + 1g/L Salt + 2.5g/L BS; (◆) 5.3-4, 2.8V/cm + 5g/L Salt + 2.5g/L BS; (■) 5.3-5, 2.8V/cm + 5g/L Salt+ 1g/L BS; Comparison fecal coliform disinfection for different dosages and voltage gradients; Experiments were conducted for sludge TS= 2.5% and pH=4 in 1L reactor volume

5.2 Optimization of Conditions for Fecal coliform Reduction

Figure 5.3 illustrates different combinations of parameters and indicates the optimum conditions for fecal coliform reduction. In experiment 5.3-1, a 2 log reduction was achieved after 10 minutes of treatment, and no further disinfection was observed within 50 minutes. Therefore, it was assumed that the 2 log reduction is contributed by the dissociation of the biocide reagent and the generation of hydroxyl radicals (Eq. [2.1]). However, the provided amount of biocide (BS) can only generate less than approximately 0.2 mg/L of hydrogen peroxide (Dagher et al., 2017). Therefore, in such medium reach in organics such amount of produced hydroxyl radicals cannot fully oxidize the sludge medium. For this reason, only a 2 log reduction of fecal coliforms was achieved. Therefore, without the support of ammonia salt and electrical field, BS seems to be the only factor affecting fecal coliform disinfection, and a voltage gradient of 2 V/cm (24V) is too low for these conditions. A significant reduction of fecal coliform at the first 10 minutes was observed due to initial application of biocide generating H_2O_2 and PAA in-situ, which permitted to advance an oxidation of fecal coliform. However, since the medium was operated under a high organic carbon load, a rise of temperature was necessary to proceed with subsequent inactivation of microorganisms.

In experiment 5.3-4, a 7.5 log reduction of fecal coliform was observed within 30 minutes. In this experiment, ammonia salt acted as another important factor contributing to fecal coliform disinfection. However, as the voltage gradient decreased to 2 V/cm in 5.3-2, fecal coliform disinfection efficiency decreased to a 4 log reduction after 30 minutes, but a 7 log reduction was not achieved until 50 minutes. In contrast, in experiment 5.3-5, a 4.2 log reduction was achieved within 20 minutes and a 7 log reduction in 40 minutes. It is assuming that a combination of stressors: imbalance of charge on cell membranes, and electrokinetic heat rise due to conductivity increase and higher voltage gradient. Increasing the voltage is much more efficient for generating more heat in the presence of increased amounts of additives. Others have attributed this difference to the exothermic redox reaction within an DC electrical field (Safaei et al,. 2013), which shows in figure 5.4.

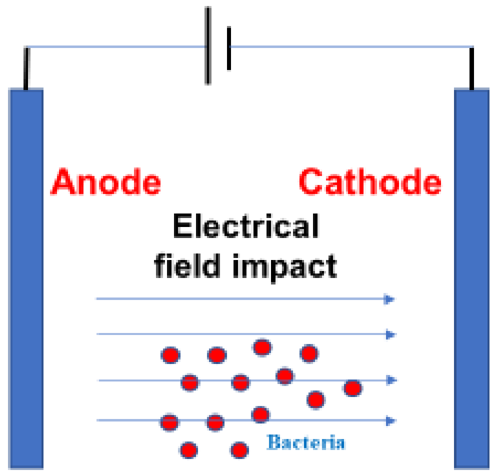


Figure 5.4 Potential mechanisms of the fecal coliform inactivation

The results of experiments 5.3-3, 5.3-4 and 5.3-5 are compared in Figure 5.3B. In experiment 5.3-5, only a 1 log reduction was achieved in the first 10 minutes, whereas a 2 log reduction was achieved in experiment 5.3-4. This demonstrates that the amount of biocide affects the disinfection efficiency by controlling the amount of hydroxyl radicals generated. However, in experiment 5.3-5, a 4.5 log reduction was achieved in 30 minutes, eventually achieving a 7 log reduction within 40 minutes.

Comparing experiment 5.3-3 and 5.3-5, there was a small difference in fecal coliform disinfection in the early stage, but both achieved 7 log reductions in 40 minutes. This is because a higher amount of ammonia salt produces a higher temperature in the same amount of treatment time, and this slowly compensated for the initial difference in fecal coliform disinfection as the treatment went on.

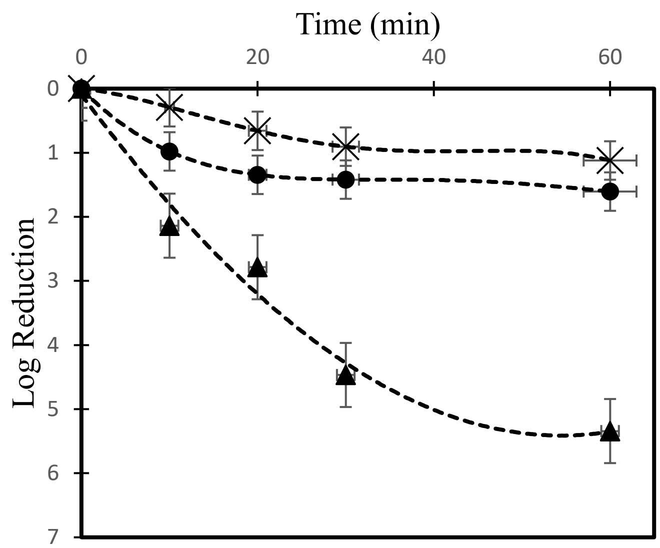
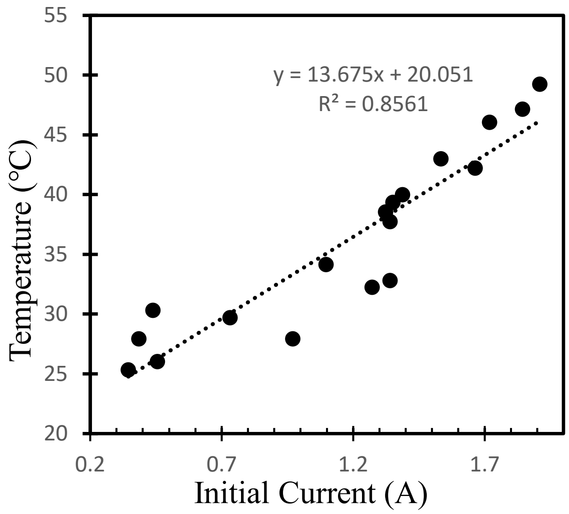


Figure 5.5 Fecal coliform reduction vs. time for different pH values

Note: (×) 5.5-3, 2V/cm + 2.5g/L BS + 1g/L Salt pH:7.5; (●) 5.5-2, 2V/cm + 2.5g/L BS + 1g/L Salt pH:6; (▲) 5.5-1, 2V/cm + 2.5g/L BS + 1g/L Salt pH:4.2; Comparison on Fecal coliform disinfection efficiency under pH value in 4.2, 6, 7.5; Experiments were conducted under sludge TS:2.5%

5.3 Effect of pH in Electrokinetic/Bioxy^{TM} System

Figure 5.5 shows the fecal coliform disinfection efficiency at different pH values. When the pH was 7.5 in experiment 5.5-3, fecal coliform disinfection only achieved a 0.2 log reduction, with just a 1 log reduction after one hour of treatment. When the pH value was decreased to 6, a 1 log reduction was achieved after 10 minutes, with no further disinfection of fecal coliforms. This demonstrates that in an acidic environment, the system has higher efficiency for fecal coliforms disinfection. It was assumed that biocide is acting as an on-site hydrogen peroxide generator, with the present of ferrous iron in the sludge matrix. When Fenton reaction takes place pH value is the most important factor. Therefore, when the pH value was lowered to 4.2 in experiment 5.5-1, a 2 log reduction was achieved in 10 minutes and a 5.5 log reduction in fecal coliforms was observed within 1 hour. This verified the hypothesis we mentioned above in section 4.7.



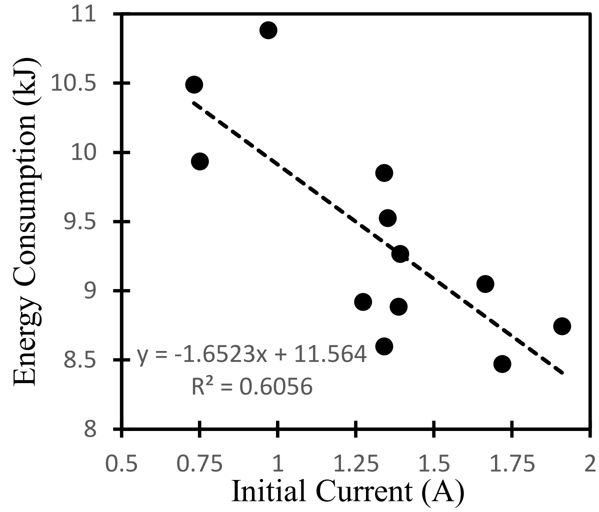


Figure 5.6 (A) Temperature vs. initial current; (B) Energy consumption vs. initial current

Note: "Initial current" indicates current value right after turning on the power supply (A) Temperature after 30 minutes treatment; (B) Energy consumption used to reach 30℃; Operational configuration was under electrode distance of 12cm, and intercept surface area of 7.5cm×10cm.

5.4 Temperature and Energy Consumption Evaluation

Figure 5.6A shows the relationship between initial current and expected temperature after 30 minutes in 1L electro-reactor. As the initial current increased, the temperature also similarly increased. An initial current of 0.3 A reached 25℃ in 30 minutes, whereas a 1.7 A initial current reached 45℃. The factors that affect temperature are the electrolyte conductivity and applied voltage. Table 5.1 shows that with the salt concentration increase shorter time which is required to achieve 70℃.

Table 5.1 Time to 70℃ vs. different operation conditions

Time (min)	Energy (kW/h)	Salt (g/L)	Biocide (g/L)	TS (%)	Voltage (V/cm)
35	0.090	1	0.5	5.5	2.8
59	0.1	2	2.5	1.8	2.8
55	0.099	3	2.5	1.8	2.8
52	0.098	4	2.5	1.8	2.8

At a TS of 5.5%, it takes 35 minutes only to reach 70℃, whereas when at TS = 1.8% it requires 59 minutes, even at a higher salt concentration. However, Kaur and Singh (2016) demonstrated that a relatively higher particle content achieved lower heat generation and required a longer time to reach a given temperature. The explanation for this contradiction can be attributed to the higher ferrous iron concentration in thickened sludge compared to more diluted sludge. Because heat is generated by DC electrical field, the electricity consumption can be directly calculated as the product of time, voltage and current based on Eq. [5.2].

[5.2] E=UIT

E----Energy consumption (kJ)

U----Voltage (V)

I----Current (A)

T----Time (s)

From Figure 5.6B, energy consumption is lower when the initial current is high. For a 1.75 A initial current, 8.5 kJ was required to achieve 30℃, whereas for 0.75 A, 10.5 kJ was required. That could indicate that a higher initial current lead to higher energy efficiency for the conversion of electricity to temperature.

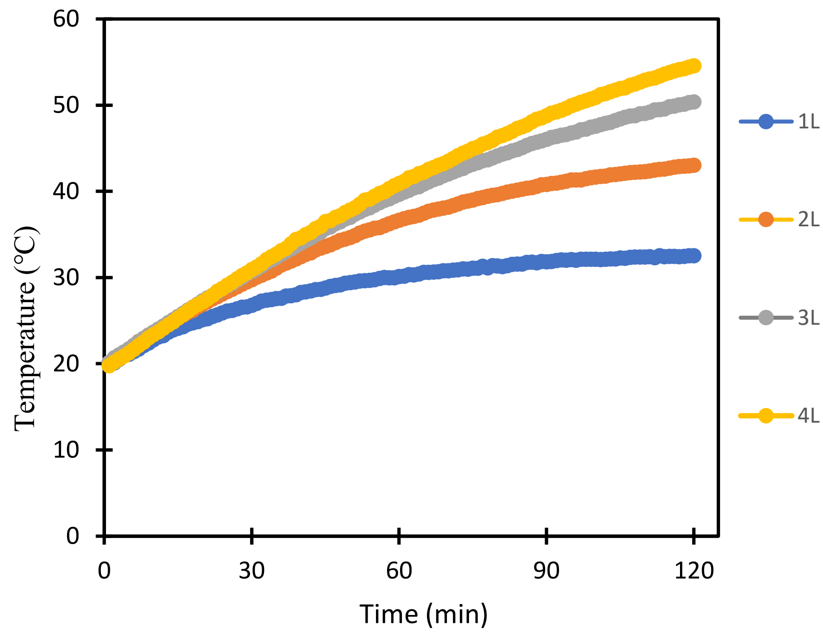


Figure 5.7 Temperature vs. time for different rector volumes

Note: Operation was conducted in reactor with 4L volume, with 2V/cm + 5 g/L Salt + 1 g/L BS and TS = 0.8%, pH=4

5.5 Effect of Volume for Electrokintic/Bioxy^{TM} System

Figure 5.7 demonstrates the relationship between temperature and volume of sludge. Experiments were operated in the same voltage gradient 2V/cm (24V) with 5g/L salt when sludge had TS = 0.8% and pH = 4. The 1 L-volume reactor could achieve 30°C after 120 minutes only, and the maximum temperature was around 30°C. When the volume was increased to a 2 L, significant changes occurred. A temperature increase to 40°C was achieved after just 90 minutes of treatment, while at the same time, 45°C was achieved for 3L, and 50°C for 4 L - reactors. That means that as the treatment volume increases, a higher temperature can be reached, and a shorter time is needed to reach the same temperature. That could show that a higher volume is more efficient for converting electricity to heat. Table 5.2 shows that the energy consumption to reach 33°C in a 1 L (2V/cm) reactor is 402 kJ/L, whereas a 2 L reactor (2V/cm) requires one fourth of that energy (106 kJ/L) only. Thus, Fig. 5.7 show value of temperature that can be achieved after 30 minutes based on the reactor volume.

Table 5.2 Energy consumption efficiency (kJ/L) for different volumes to achieve different temperatures

	1L	**2L**	**3L**	**4L**
33°C	402	106	87	86
40°C	*	223	158	150
50°C	*	*	337	276

Legend: (*) means cannot achieve the target temperature

It was shown as volume increases, the energy consumption efficiency is greatly improved (87kJ/L for 3L and 86 kJ/L for 4L). It is also interesting to underline as the target temperature increases, the difference in energy efficiency between two volumes increases as well. The difference for a target temperature of 33°C is around 1 kJ/L (equal to 87 - 86 kJ/L), for 40°C it increases to 8 kJ/L (158-150 kJ/L), and for 50°C it increases further to 61 kJ/L (337-276 kJ/L).

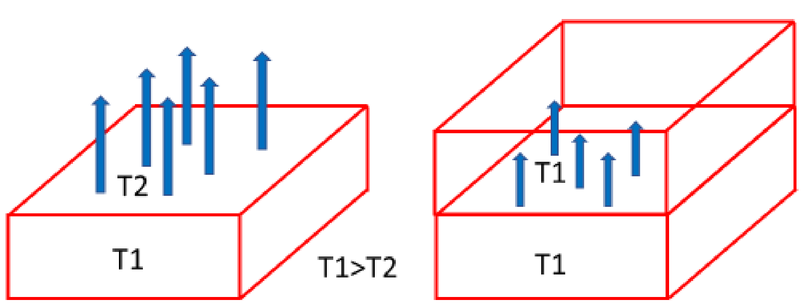


Figure 5.8 Schematic diagram for the heat loss during the volume increase

Because energy input is constant, Eq. [5.3] shows that the temperature gap between different volumes can be explained by the loss of energy to the environment. In Eq. [2.12], the heat loss rate is inversely proportional to dimensions Δx . Therefore, heat loss will decrease as the dimensions increase. Besides, temperature gradients between two mediums is highly affecting the heat loss rate (Eq. [2.2]), figure 5.8 demonstrated the effect on the change of ΔT from the change of volume, which verified the results from figure 5.7.

[5.3] $\Delta T = E_{in} - E_{out}$

ΔT---- Temperature changes (°C)

E_{in}---- Energy input (kJ)

E_{out}---- Energy output (kJ)

Consequently, increased volume results in lower volumetric heat loss in the system; therefore, the system is more efficient in the temperature generation.

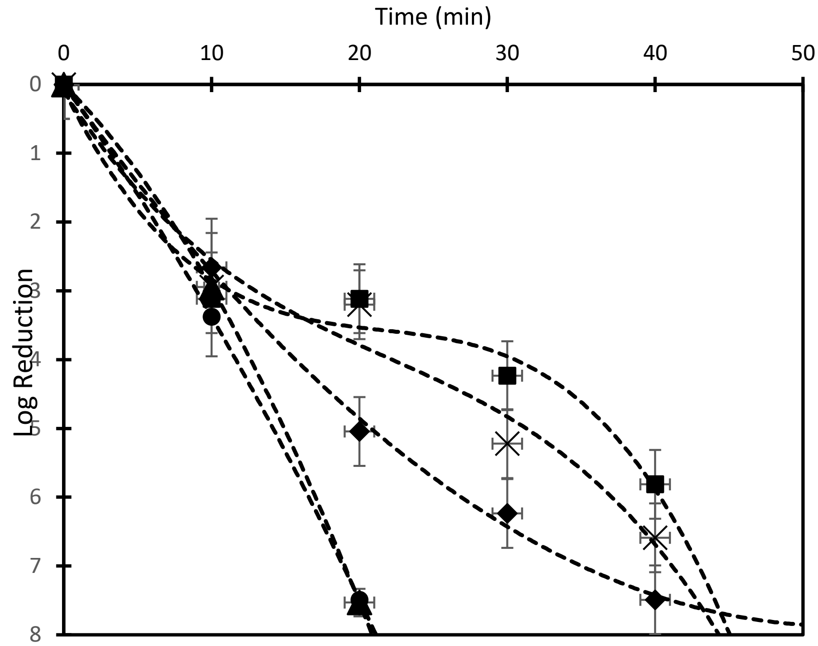


Figure 5.9 Fecal coliform reduction vs. time for different combinations of the operational conditions in different volumes

Note: (●)5.3-4 (2.8V/cm + 5g/L Salt + 2.5g/L BS in 1L); (▲)5.9-1 (2.8V/cm + 5g/L Salt + 2.5g/L BS) in 4L ; (×)5.9-2 (2V/cm + 5g/L Salt + 2.5g/L BS) in 4L; (◆)5.9-3 (2.8V/cm + 5g/L Salt + 1g/L BS) in 4L; (■)5.3-2 (2V/cm + 5g/L Salt + 2.5g/L BS) in 1L; Fecal coliform disinfection efficiency under different conditions at a large scale for sludge having pH=4, TS=3.5%

Recall that in section 5.1, it was concluded that temperature is one of the most important determinants of fecal coliform disinfection. As it is obvious in Figure 5.1, higher temperature has better fecal coliform reduction efficiency. The results from experiment 5.9-1 and 5.3-4 show that both achieved 7 log reductions within 20 minutes, with no significant difference between the 1L scale and 4L scale (Figure 5.9). Because of the high voltage and high dosage of ammonia salt in the system, a 38.5°C temperature was easy to achieve regardless of the volume difference, which is satisfactory for fecal coliform disinfection according to the assumption in section 5.1. However, under the lower voltage gradient, used in experiments 5.3-2 and 5.9-2, fecal coliform disinfection efficiency was slightly different between the 4 L and 1 L reactor volumes. For the 1 L scale, only a 4 log reduction was achieved after 30 minutes, whereas for the 4 L scale, a 5.2 log reduction in fecal coliforms could be achieved. Therefore, volume can affect fecal coliform disinfection efficiency because a larger volume can retain more heat in the reactor so that temperature increases faster. In this chapter, the experimental data error was around 7% for the fecal coliform disinfection (X axis), which based on ±0.5 log for fecal coliform reduction during the repetition of analysis. An error for the time (Y axis) was around ±2 minutes, which was due to the sample collection during the experiment.

5.6 Cost estimation

Electrokinetic/Bioxy™ cost estimation was based on Exp 5.3-2, when Eq. [5.2] was selected for the electricity calculation; selected time was related to achieving the temperature of 40°C. Experimental time was around 53 minutes, while constant voltage gradient of 2V/cm (24V), current within a range of 1.3 – 1.6 A were applied. Table 5.4 shows cost estimation for 1L and projection for 1000L using Eq [5.4] for scale up process.

[5.4] Large scale cost = small scale cost * $\left(\frac{Large\ scale\ capacity}{small\ scale\ capacity}\right)^{0.7}$ (Rashmi, 2011)

0.7---When the scale change from 1L scale to 1000L scale

Table 5.3 Cost estimation for Electrokinetic/Bioxy™ system

Utilization	Consumption	Price	Operation costs (CAD/dry tone) for small (1L) scale	Operation costs (CAD/dry ton) for larger scale (1 1000L)
Electricity	~1.5 (kWh/ L dry sludge)	$0.035/kWh *	0.05	6.5
Ammonia salt	5g/ L for 2.5% TS (200g / 1 ton dry sludge)	$200 / 1000kg (Ammonia salt)	0.04	5.2

Legend: * cost of energy in Quebec province for industrial units

Cost estimation for Electro-Fenton system were compared in this section. Electro-Fenton system cost estimation was based on Exp 4.1-4 and 4.6-4. Eq [5.2] was selected for energy calculation, where time was related to this achieving 3 log reductions for fecal coliform disinfection. Therefore, Exp 4.1-4 used 20 minutes and 4.6-4 used 30 minutes because of different TS values. In both cases, current was maintained constant at 0.4A; voltage was in the range of 20 – 30 V. The cost estimation for Electro-Fenton disinfection is illustrated in Table 5.4. To assess a feasibility of the electrical field disinfection system, the cost comparison to conventional sludge disinfection treatment was showed in Table 5.5.

Table 5.4 Cost estimation for Electro-Fenton system

Utilization	Consumption	Unit price	Operation costs (CAD/dry ton) for small (1L) scale	Operation costs (CAD/dry ton) for larger scale (1000L)
Electricity	0.16-0.33 (kWh/ L dry sludge)	\$0.035/kWh	0.011 – 0.056	1.38 – 7.04
H_2O_2	62.5 – 125 mL/ L dry sludge	\$500 / 1000L	0.015 – 0.03	1.89 – 3.7

Comparison of the costs of both Electro-Fenton and Electrokinetic/biocide to conventional treatment methods, shows that electricity and main reagents' costs are much lower. However, to get realistic data, it is necessary to investigate the system at a larger (pilot) scale, where the costs of electrodes, energy for mechanical operation, etc. would be included.

Table 5.5 Cost estimation for conventional methods for sludge disinfection (US. EPA 2000a)

Processes	Price USD/dry ton
Alkalinity stabilization	67-481
Incineration	341 - 344

Chapter 6 Conclusions, Applications, Future Work, Contributions

6.1. Conclusions

The study showed a new approach to sludge management leading to decrease operation costs and improvement the final product quality. Both proposed methods were able to transform sludge into Class A biosolids.

6.1.1 Conclusions with respect to the technology applying Electro-Fenton method:

(1) Experiments successfully demonstrated the effectiveness of the Electro-Fenton system for fecal coliform disinfection. The system achieved at least a 3 log reduction in fecal coliforms within 30 minutes (8 $mA/cm^2/L$, 1 ml H_2O_2 (30%) /L), which meets the requirement (log 3) for Class A quality biosolids. The multi-stressor conditions resulting from Electro-Fenton reaction proved to attain a better fecal coliform reduction compared to individual stressors, for which only a 1.9 log reduction for electrical field only (8 $mA/cm^2/L$) and a 2.7 log reduction for the Fenton reaction (1 ml H_2O_2 (30%) /L) were achieved.

(2) All parameters such as pH, Fe^{2+}/H_2O_2 ratio, H_2O_2 concentration and H_2O_2 feeding mode affect fecal coliform disinfection efficiency, particularly pH, which is the dominant factor. A pH value of 7 does not show any disinfection ability for achieving Class A biosolids (>3 log reduction) even under high current (16 mA/cm^2) conditions, while operated under pH value of 4 can successfully achieve 3 log reductions within 15 minutes in lower current (8mA/cm^2) sludge.

(3) DC current strongly affects the Electro-Fenton system; a fecal coliform log reduction proportionally increased as current increase. A required 3.6 log reduction was achieved at 0.2 A (4 mA/cm^2) only while a 5.2 log reduction at 0.4 A (8 mA/cm^2) and a 7.3 log reduction at 0.8 A (16 mA/cm^2) were reached under the following conditions, Fe^{2+}/H_2O_2 = 1:6 with 5 times feedings (over 30 min) of 1 ml H_2O_2 (30%) in 1L volume of sludge.

(4) Furthermore, the TS value has a significant impact on fecal coliform disinfection efficiency using Electro-Fenton; the log reduction (within 30 min) was 3.5 greater for a TS value of 0.8% than for 2.8% TS in 1L volume. The results show that Fe^{2+}/H_2O_2 ratio does not have a significant impact on fecal coliform disinfection in the Electro-Fenton system, which can be assumed that electrical regeneration of ferrous iron for improving the Fenton reaction's efficiency takes place. Ratio of Fe^{2+}/H_2O_2 = 1: 1.5 achieved 7 log reductions for Fenton disinfection, while for Electro-Fenton, 7 log reductions can be reach without adding of Fe^{2+}. Besides, the cost evaluation regarding electricity consumption was estimated in the range of 0.011 – 0.056 CAD / dry ton sludge.

6.1.2 Conclusions with respect to technology which applying Electrokinetics enhanced with biocide

(1) The study demonstrated that heat generated by electrokinetic achieved a much better disinfection efficiency compared to external conductive heat, which considers as a conventional method for sludge disinfection. Moreover, with the assistance of the in-situ hydrogen peroxide generator BioxyS™, the BioxyS™/Electrokinetic system exhibited greater efficiency at fecal coliform disinfection. External conductive heat at 40°C did not cause a reduction of fecal coliforms, whereas for the same temperature, a 3.5 log reduction was achieved with the heat generated by electrokinetic system and a 7.2 log reduction by the BioxyS™/Electrokinetic system.

(2) A higher dosage of BioxyS™ (BS) achieved Class A biosolid quality at a faster rate. Voltage value as well as BS and ammonia salt concentrations are proportionally related to

the temperature rise. However, the BS application alone does not lead to a significant sludge disinfection.

(3) The pH value is the dominant factor affecting performance. Under the condition of 2 V/cm with 2.5 g/L BS and 5 g/L ammonia salt, the experiment, conducted at pH = 4, achieved a 5.3 log reduction, whereas a 1.3 log reduction was obtained for pH = 6 and a 1 log reduction for pH = 7.

(4) Higher initial current (higher conductivity, voltage gradient), a larger reactor volume can increase temperature faster during the operation process. To achieve 50℃, 4L volume of sludge spends 276 kJ/L, while it took 337 kJ/L for 3 L. Subsequently, fecal coliform disinfection can be slightly improved in a larger reactor volume.

6.2 Application

The Electro-Fenton system exhibits a strong capacity for fecal coliform disinfection. Compared to normal Fenton reaction disinfection, the system could require much less Fenton reagent (hydrogen peroxide and ferrous iron) to achieve Class A Biosolids. This system can be applied to waste-activated sludge, particularly to thickened sludge with ferrous coagulant where additional dosage of iron might not be necessary. Electrokinetic\Bioxy™ system could also be implemented for sludge having a high content of TS. It is a novel technology for rapid municipal sludge stabilization and satisfying the safety requirements for its subsequent use. This approach can be applied by small and medium size municipal wastewater treatment plants for sludge disinfection due to low capital costs and a good quality sludge production. Overview of application of two novel disinfection technologies, which use electrical field for disinfection process. This study showed that both novel technologies namely Electro-Fenton and Electrokinetic/Biocide, can successfully achieve Class A biosolids. It is suggested that, under low TS content condition (<2.8% in sludge), it would be better to choose the Electro-Fenton disinfection system due to lower operation costs. However, when TS content is higher than 2.8%, mixing of sludge become much difficult, which can affect an adequate dispersion of additives and hydroxyl radicals within the reactor. Therefore, it is suggested to use Electrokinetic/BioxyS™ system for disinfection of sludge with a high TS content. Application of electrical field and generation of heat can be used in a large range of TS contents and achieve the required sludge disinfection having a lower dependence to water content. Electro-Fenton application to sludge disinfection was declared as a new invention (Luo and Elektorowicz, 2018).

6.3. Future Work

- Electro-Fenton system requires to investigate disinfection with respect to other pathogens such as viruses, helminth ova and spores, which have higher resistance to stressors compared to bacteria. Some regulations also require such checking.
- Further work could focus on the sludge dewaterability, which based on the primary analysis during the experiment from observation and particle size analysis results.
- Further investigation should be conducted on continuous flow reactors regarding fecal coliform disinfection.
- Study on the particle size distribution change over the time, which might affect the dewater ability of the sludge
- Optimization and modeling of disinfection kinetics should be conducted with respect to of current, ferrous iron as well as hydrogen peroxide, while minimizing the exposure time.
- In terms of approach two, the disinfection efficiency of a large-scale reactor should be investigated since larger volumes.

6.4. Contribution

First, this study developed a new approach to fecal coliform inactivation in municipal sludge by using Electro-Fenton system to satisfy the requirement for Class A biosolids. It also proved the effectiveness of Electro-Fenton system has greater efficiency of fecal coliform reduction than Fenton reaction. It demonstrated the feasibility by using lower dosage of Fenton reagent to achieve better fecal coliform disinfection efficiency in thickened sludge.

Second, this study demonstrated the effectiveness of BioxyS^{TM}/Electrokineitc system regarding fecal coliform disinfection of municipal sludge. A relationship between temperature and fecal coliform reduction was demonstrated. Furthermore, it proved an impact of the rector volume on temperature rise, which showed a feasibility to achieve a better fecal coliform reduction efficiency at larger scale.

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