RADIO FREQUENCY SLUDGE HYDROLYSIS AS AN ENERGY EFFICIENT ALTERNATIVE TO MICROWAVE AND CONDUCTIVE HEATING FOR ADVANCED ANAEROBIC DIGESTION

by

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The following individuals certify that they have read, and recommend to the College of Graduate Studies for acceptance, a thesis/dissertation entitled:

RADIO FREQUENCY SLUDGE HYDROLYSIS AS AN ENERGY EFFICIENT ALTERNATIVE TO MICROWAVE AND CONDUCTIVE HEATING FOR ADVANCED ANAEROBIC DIGESTION

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ABSTRACT

The slow degradation of complex organics such as waste activated sludge (WAS) is a well-known limitation that impacts the process rate of conventional anaerobic digestion (AD). Thermal pretreatment can accelerate the digestion process by disrupting the structure of WAS before AD. The present research was initiated by comparing the two commonly used thermal pretreatment methods, conductive (conventional) heating (CH) and microwave (MW) hydrolysis, for enhanced sludge disintegration and AD performance. A bench-scale programmable MW oven operated at a frequency of 2.45 GHz was used for MW pretreatment. The CH was performed using a custom-built pressure sealed vessel which could simulate the MW pretreatment under any arbitrary heating profiles. After comparing the CH and MW pretreatments, a novel and highly efficient radio frequency (RF) pretreatment system at a frequency of 13.56 MHz was designed, manufactured, and tested for the first time. The RF system was custom-designed based on the dielectric characteristics of thickened WAS (TWAS) to achieve very efficient as well as uniform heating. The effects of the novel RF pretreatment system on sludge solubilization and AD performance were compared with those of the commercially available MW ovens.

Considering the obtained results and analyses, under identical thermal profiles, the thermal pretreatment methods (CH, MW at 2.45 GHz, and RF at 13.56 MHz) achieved similar sludge disintegration as well as AD performance (p-value>0.05). However, the pretreatment temperature, heating rate, and holding time were significant factors in determining the sludge

solubilization ratio and AD performance. Ohmic heating was found as the primary heating mechanism at a frequency of 13.56 MHz. It causes the ionic conduction flow to dominate the heating mechanism in the custom-designed RF pretreatment system by contributing to more than 99% of the total dissipated power. Considering the impedance measurement results, the power transfer efficiency of the RF heating system was above 88% throughout the operation. The overall energy efficiency of the RF pretreatment system was measured between 67.3 to 95.5% for the temperature range of 25 to 120°C which was significantly higher than the MW system efficiency which varied from 37 to 43%.

PREFACE

The research performed included the design of a RF sludge pretreatment system, experimental design, design and operation of bioreactors, characterization testing, data analysis, and writing research articles. This work was conducted by myself with the assistance of Dr. Cigdem Eskicioglu as the principal investigator of the project and Dr. Thomas Johnson as the co-investigator. The electrical design of the RF pretreatment system was done by Mr. Md. Saimoom Ferdous, a former electrical engineering master student.

A version of Chapter 3 was published in the journal of *Bioresource Technology* (*http://dx.doi.org/10.1016/j.biortech.2015.03.113*). A portion of this chapter was published as part of the proceedings of the *Water Environment Federation (WEF) Annual Technical Exhibition and Conference* (September 26-30, 2015, Chicago, Illinois, USA.). A portion of this chapter was also presented at the *43rd British Columbia Water and Waste Association (BCWWA) Annual Conference* (May 27-30, 2015, Kelowna, BC, Canada.). All the data analysis and the writing of the papers were performed by myself. Dr. Cigdem Eskicioglu provided feedback and review of the paper.

A version of Chapter 4 was published in the journal of *Waste Management* (*http://dx.doi.org/10.1016/j.wasman.2016.04.014*). A portion of this chapter was presented at the *8rd Canadian Biosolids and Residuals Conference* (May 17-19, 2016, Edmonton, AB, Canada.). All the data analysis and writing of the papers performed by myself. Dr. Cigdem Eskicioglu provided feedback and review of the manuscript.

A portion of chapter 5 was presented at the *44rd BCWWA Annual Conference* (May 1-3, 2016, Whistler, BC, Canada.). The conference presentation was done by myself. Dr. Cigdem Eskicioglu and Dr. Thomas Johnson provided feedback and review of the abstract. A portion of this chapter was also presented at the *University of British Columbia's 2nd Engineering Graduate Symposium* (June 8, 2015, UBC Okanagan, Kelowna, BC, Canada). The conference presentation was done by myself. The presentation received the "Best Presentation Award".

A version of Chapter 6 was published in the Journal of *Water Research*. All the data analysis and the manuscript writing were performed by myself. Dr. Cigdem Eskicioglu and Dr. Thomas Johnson provided feedback and review of the manuscript. A portion of this chapter was also presented at the *2nd International Water Association (IWA) Conference on Holistic Sludge Management (HSM)* and published as part of the conference proceedings (June 7-9, 2016, Malmö, Sweden.). All the data analysis and the paper writing were performed by myself. Dr. Cigdem Eskicioglu and Dr. Thomas Johnson provided feedback and review of the paper.

An extended abstract prepared based on the results of Chapter 7 was submitted to the *15th IWA World Conference on Anaerobic Digestion* (October 17-20, 2017, Beijing, China.) and accepted for oral presentation. A portion of this chapter was also presented at the *4th Water and Environment Student Talks (WEST) Conference* (June 11-13, Vancouver, BC, Canada.). All the data analysis and writing the papers were performed by myself. Dr. Cigdem Eskicioglu and Dr. Thomas Johnson provided feedback and review of the paper and presentation. A version of Chapter 7 is being prepared for the submission to the journal of *Water Research*.

The full citation of the research papers and/or presentations published based on the results of this research are as follows:

1. **Hosseini Koupaie, E.**, Eskicioglu, C. (**2015**) "*Below and above boiling point comparison of microwave irradiation and conductive heating for municipal sludge digestion under identical heating/cooling profiles*". Bioresource Technology 187, 235-245.

2. **Hosseini Koupaie, E.**, Eskicioglu, C. (**2015**). "*Microwave vs. conventional thermal sludge pretreatment technique: A comprehensive comparison under identical thermal profile*". WEFTEC 2015, September 26-30, Chicago, Illinois, USA. 2507-2522.

3. **Hosseini Koupaie, E.**, Eskicioglu, C. (**2015**). "*Application of thermal hydrolysis for advanced anaerobic digestion of Kelowna's biosolids*". 43rd BCWWA Annual Conference, May 27-30, Kelowna, BC, Canada.

4. **Hosseini Koupaie, E.**, Eskicioglu, C. (**2015**). "*Enhanced anaerobic digestion of municipal biosolids via thermal hydrolysis*" 2nd Engineering Graduate Symposium, June 8, UBC Okanagan, Kelowna, BC, Canada.

5. **Hosseini Koupaie, E.**, Eskicioglu, C. (**2016**) *"Conventional heating vs. microwave sludge pretreatment comparison under identical heating/cooling profiles for thermophilic advanced anaerobic digestion".* Waste Management 53, 182-195.

6. **Hosseini Koupaie, E.**, Eskicioglu, C., Johnson, T. (**2016**). "*Novel application of radio frequency heating in sludge pretreatment for advanced anaerobic digestion*". 2nd IWA Conference on Holistic Sludge Management, June 7-9, Malmö, Sweden.

7. **Hosseini Koupaie, E.**, Eskicioglu, C. (**2016**). "*Enhanced thermophilic anaerobic digestibility via thermal pretreatment: A case study on the City of Kelowna's biosolids*". 8th Canadian Biosolids and Residuals Conference, May 17-19, Edmonton, AB, Canada.

8. **Hosseini Koupaie, E.**, Eskicioglu, C., Johnson, T. (**2016**). "*Introducing a novel highly efficient electromagnetic sludge pretreatment system to enhance the City of Kelowna's proposed anaerobic digestion process*". 44th BCWWA Annual Conference, May 1-3, Whistler, BC, Canada.

9. **Hosseini Koupaie, E.**, Johnson, T., Eskicioglu, C. (**2017**) *"Advanced anaerobic digestion of municipal sludge using a novel and energy-efficient radio frequency pretreatment system".* Water Research 118, 70-81.

10. **Hosseini Koupaie, E.**, Eskicioglu, C., Johnson, T. (**2017**). "*Assessment of overall energy recovery from the City of Kelowna's raw and thermally-hydrolyzed sewage sludge via anaerobic digestion*". 4th Water and Environment Student Talks (WEST) Conference, June 11-13, Vancouver, BC, Canada.

11. **Hosseini Koupaie, E.**, Eskicioglu, C., Johnson, T. (**2017**). "*Electromagnetic ohmic heating at a frequency of 13.56 MHz as an energy-efficient alternative to the microwave sludge hydrolysis at 2.45 GHz: A study under semi-continuous flow regime*". 15th IWA World Conference on Anaerobic Digestion, October 17-20, 2017, Beijing, China. (Accepted for oral presentation).

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LIST OF ACRONYMS AND ABBREVIATIONS

AAFC	Agriculture and Agri-Food Canada
AD	Anaerobic digestion
ANOVA	Analysis of variance
AOP	Advanced oxidation process
Bardenpho	Barnard denitrification phosphate
BC	British Columbia
BCWWA	British Columbia Water and Waste Association
BMP	Biochemical methane potential
BNR	Biological nutrient removal
BOD	Biochemical oxygen demand
CH	Conductive (conventional) heating
COD	Chemical oxygen demand
DAF	Dissolved air flotation
DWSC	Dewatered sludge cake
EC	Environment Canada
EM	Electromagnetic
EPS	Extracellular polymeric substances
EU	European Union
F/M	Food to microorganism ratio
FPS	Fermented primary sludge
GC	Gas chromatography
HA	Humic acid
HPH	High pressure homogenization
HRT	Hydraulic retention time

HSM	Holistic sludge management
IC	Industry Canada
IR	Infrared
ISM	Industrial scientific and medical
IWA	International Water Association
KWWTP	Kelowna's wastewater treatment plant
LAP	Land application plan
LCA	Life-cycle assessment
MCDM	Multi-criteria decision making
MLSS	Mixed liquor suspended solids
MLVSS	Mixed liquor volatile suspended solids
MPN	Most probable number
MW	Microwave
NRC	National Research Council
NSERC	Natural Sciences and Engineering Research Council of Canada
OLR	Organic loading rate
OMRR	Organic matter recycling regulation
ORP	Oxidation-reduction potential
PAOs	Phosphorus accumulating organisms
PCC	Pearson correlation coefficient
PS	Primary sludge
R^2	Coefficient of determination
RBC	Rotating biological contactor
RF	Radio frequency
SBR	Sequencing batch reactor

SCOD	Soluble chemical oxygen demand
SRT	Sludge retention time
TCOD	Total chemical oxygen demand
TPAD	Temperature-phased anaerobic digestion
TS	Total solids
TWAS	Thickened waste activated sludge
UASB	Upflow anaerobic sludge blanket
UBC	University of British Columbia
UK	United Kingdom
UV	Ultraviolet
VFA	Volatile fatty acid
VS	Volatile solids
VSC	Volatile sulfur compound
WAS	Waste activated sludge
WEF	Water Environment Federation
WHO	World Health Organization
WWTP	Wastewater treatment plant

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To my parents, sister, brothers

and

To my wife

CHAPTER 1. INTRODUCTION

This chapter provides general information about the production and management of organic-rich sludge which is the main by-product of biological wastewater treatment processes. In this context, the situation of the Okanagan Valley and the City of Kelowna (BC, Canada) is defined, which serves as the motivation for the research. Furthermore, the research objectives, hypotheses, and a summary of the applied methodology are described.

1.1. BACKGROUND

Municipalities rely on physical, chemical, and biological treatment processes to treat their municipal and industrial wastewater. As a result of these treatment processes, municipal sludge, a by-product of treatment, is generated in wastewater treatment plants (WWTP). Currently, about 0.7 million tons of dry municipal sludge are produced annually in Canada. In comparison, the United States and Europe see more than 7 and 10 million tons of dry municipal sludge produced annually respectively (Pérez-Elvira et al., 2006; USEPA, 1999). To service the demands of growing cities and respond to the increasingly stringent sludge reuse and disposal regulations, existing treatment plants are expanding, resulting in an increased production of municipal sludge. On the other hand, the sludge handling and disposal costs more than 50% of the total operating expenses of a municipal WWTP (Appels et al., 2008). Additionally, the quality of waste sludge is decreasing as a result of anthropogenic (human-made) macro- and micro-pollutants such as heavy metals, pharmaceuticals, and personal care products (i.e. hormones), limiting agricultural and land reuse applications (Hamid & Eskicioglu, 2012). Considering all these facts, the management of wastewater residual sludge has now become one of the world's largest and most critical management challenges.

A conventional municipal WWTP consists of primary and secondary (biological) treatment processes. As a result of these treatment processes, two separate sludge streams, primary and secondary, are formed. The primary sludge contains readily biodegradable organic matter captured through the primary sedimentation stage. The secondary sludge, also known as waste activated sludge (WAS), contains excess microbial cells, which are a by-product of the biological treatment process. If sludge streams are further stabilized via biological, chemical, and physical means, the sludge cake is known as biosolids and have beneficial uses in land applications as a fertilizer. In the province of British Columbia (BC), the Organic Matter Recycling Regulation (OMMR), established by the BC Ministry of Environment, should be followed for all biosolids land application (OMRR, 2008). Under the OMRR, biosolids are classified as Class A or Class B based on the concentration of the fecal coliforms and heavy metals in the biosolids. For biosolids to be classified as Class A, the fecal coliforms content should be less than 1000 most probable number (MPN)/g of dry total solids (TS). Class B biosolids are, however; allowed to contain up to 2,000,000 MPN fecal coliform per gram of dry TS and have more restrictions placed on them such as fencing and extra storage on site before land application. The regulation does not currently distinguish heavy metal limits for Class A and B biosolids.

The City of Kelowna is situated in the Okanagan Valley in the southern interior of the Province of BC, Canada. The Kelowna's WWTP (KWWTP) has a design capacity of 70,000 m^3/d, and currently serves a population of approximately 115,000 people. At the WWTP, the wastewater undergoes physical treatment processes (screening, grit removal, and primary sedimentation) followed by a biological nutrient removal (BNR) system. The biologically treated wastewater is then filtered and disinfected via ultraviolet light before being released into the Okanagan Lake. The employed BNR system is a modified Barnard denitrification phosphate process (Bardenpho®). The purpose of the BNR process is to provide simultaneous removal of carbon and nutrients (nitrogen and phosphorus) to prevent eutrophication (excess algal growth) in the receiving environment. The waste activated sludge generated through the BNR process is settled in secondary sedimentation tanks and thickened via dissolved air flotation (DAF) units. The fermented primary sludge (FPS) and thickened waste activated sludge (TWAS) streams are mixed at a ratio of 33:67% by volume. The mixed sludge is then sent to a centrifuge unit and dewatered to produce dewatered sludge cake (DWSC) with a TS content of 18 to 22% by weight. Figure 1-1 shows the overall wastewater treatment process and fate of the generated biosolids at KWWTP.

Currently, the KWWTP biosolids are trucked 46 km to a composting facility in Vernon, BC where it is mixed with wood chips and aerated for a period of approximately 55 days. The final product is then screened to remove deleterious materials and aged for approximately 2 months to produce a commercial fertilizer called “OgoGrow”. Recycling the biosolids by using them as a soil amendment can greatly improve soil stability, porosity, and water retention (Ramulu, 2002; Singh & Agrawal, 2008). It is also a beneficial way to recycle organic matter, valuable nutrients, and essential trace elements while reducing the use of chemical fertilizers (Lu et al., 2012; Pascual et al., 2004; Passuello et al., 2010; Wang et al., 2008). From BNR point of view, the land application practice also eliminates the need for side stream treatment in order to remove phosphorus. However, this biosolids management practice has disadvantages including high transportation cost and high energy consumption of the air blowers in the composting facility. Additionally, there have been numerous complaints from nearby residents regarding the odor emanating from the facility (Rolke, 2013). Unfortunately, due to the limited capacity of the composting sites, large quantities of OgoGrow are given away for free each year to make room for further processing of biosolids. Considering all the explanations, investigation of new biosolids management options is essential.

In recent years, the City of Kelowna has begun to investigate ways to further reduce the amount of the biosolids generated at a rate of approximately 60 dry tonnes/day at KWWTP. One option (which was evaluated as a part of this research) is the implementation of an anaerobic digester at KWWTP. Anaerobic digestion (AD) is a biological process in which the organic waste (i.e. biosolids, animal manure, and kitchen waste) is converted into methane-rich biogas and fertilizer in an oxygen-free environment (Chen et al., 2008; Metcalf & Eddy, 2013). As a result of this process, the methane recovered from the biosolids digestion can be further utilized for electricity generation and the digester remaining materials (digestate) can be used as fertilizer after dewatering, if it meets regulatory limits for land application. In addition to the methane generation, the AD process causes the reduction of pathogens and odor of the digestate making it a favorable fertilizer (Moody et al., 2009). Furthermore, simultaneous co-digestion of the biosolids with other locally produced organic wastes (i.e., wine/fruit-juice production waste or municipal landfill leachate) can provide better process performance and financial advantages

(Barrantes Leiva et al., 2014). These features make AD appealing both from an environmental and energy perspective compared to other disposal options (Appels et al., 2008; Sanscartier et al., 2012).

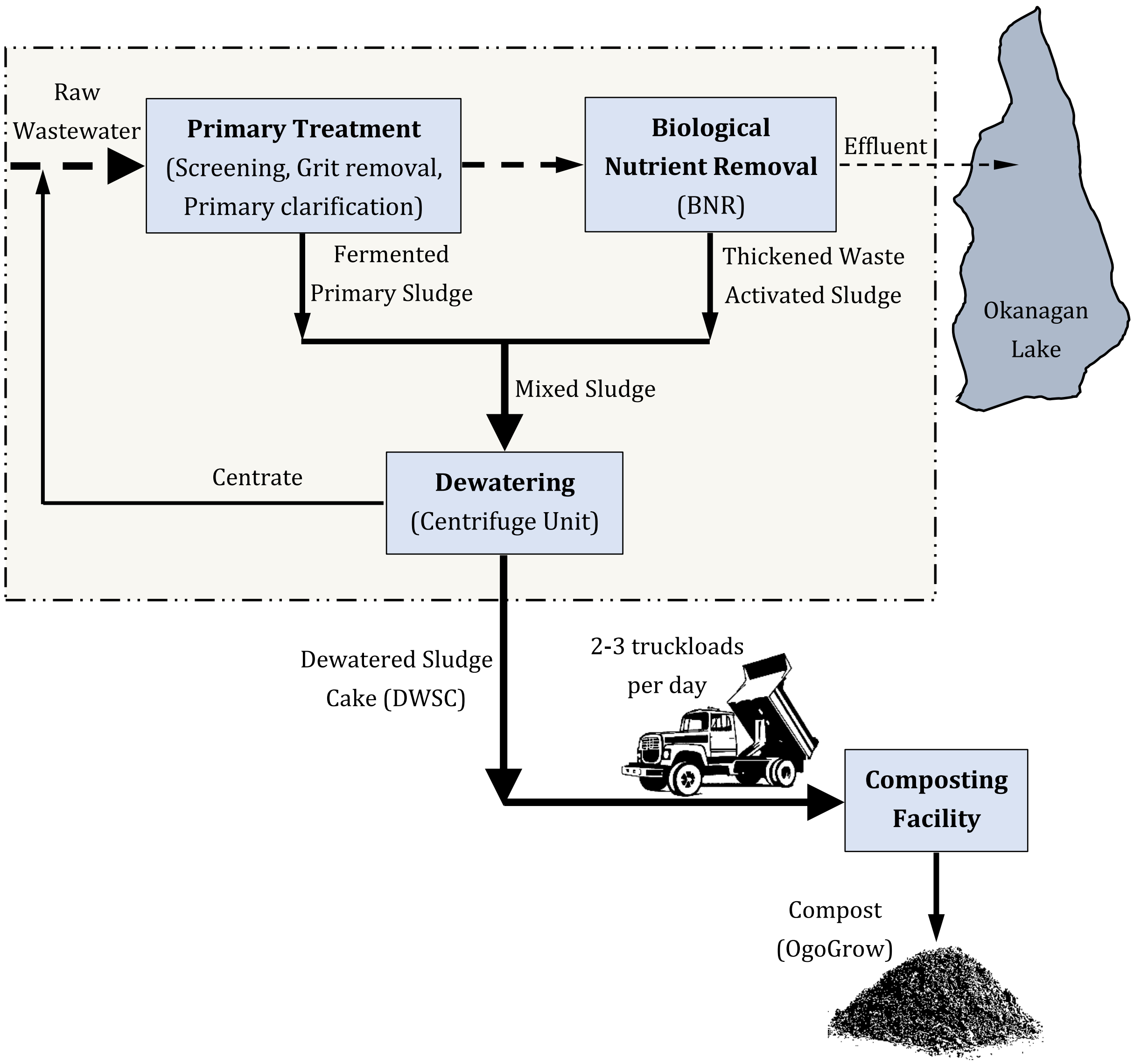


Figure 1-1. The overall treatment process and the fate of biosolids at the City of Kelowna's wastewater treatment plant

A limitation of conventional AD is the slow degradation of complex organic waste such as WAS containing extracellular polymeric substance (EPS) and microbial cells resisting anaerobic biodegradation. As a result, the required sludge retention time (SRT) is long, digester capacity is large, and consequently capital and operating costs for conventional AD are high (Tyagi & Lo, 2013). For example, mesophilic AD of municipal sludge requires a minimum SRT of 20-25 days with only partial (45-50%) volatile solids (VS) destruction and deactivation of pathogens, therefore, producing Class B biosolids with restricted land application. As a way of reducing SRT in AD, along with increasing biogas production, pretreatment methods can be used to disrupt the structure of WAS. Examples of pretreatment include thermal, mechanical, and biological processes that increase the availability of intracellular and extracellular biopolymers in the liquid phase (Bordeleau & Droste, 2011; Zhen et al., 2017). The objective of pretreatment methods is to convert large and complex organic structures into smaller more soluble organic structures, thus, accelerating the subsequent AD process. A system that uses pretreatment and AD is known as advanced AD.

Thermal hydrolysis is one of the most common methods of pretreatment in advanced AD. Thermal energy disintegrates sludge structures releasing intracellular materials. The enhanced availability of intracellular material reduces SRT and increases the rate of biogas generation (Tyagi & Lo, 2013). Thermal hydrolysis was first applied in 1978 and was used to improve sludge digestibility and dewaterability (Haug et al., 1978). Since then, the method has been widely used and reported to be effective in sludge disintegration while improving the performance of AD. The thermal pretreatment was applied initially via conductive (conventional) heating (CH). In CH, the heat is transferred by conduction, which is governed by the temperature gradient as well as the material's thermal conductivity. During the last decade, however, there have been many studies evaluating the application of microwave (MW) heating in WAS hydrolysis. According to the literature, the annual published research papers on the application of MW heating for pretreating municipal sludge increased significantly from one paper in 1991 to more than 40 papers in 2011 (Tyagi & Lo, 2013).

1.2. RESEARCH HYPOTHESES

Numerous studies have been done so far to compare CH and MW pretreatments. However, due to the contradictory findings of the published research (Climent et al., 2007; Eskicioglu et al., 2006; Park & Ahn, 2011a; Pino-Jelcic et al., 2006; Sólyom et al., 2011), the literature is currently inadequate to properly answer whether there are any advantages of choosing one of these thermal pretreatment methods over another (MW or CH) for disintegration of waste sludge. It is postulated that the main reason behind the contradictory results of the previous research is the inability to maintain identical thermal profiles between the MW and CH pretreatments. Considering the results from previous studies, it is hypothesized that the final temperature, the heating rate, and the holding (stationary) time are important factors in determining sludge disintegration and subsequent digestion performance (*Hypothesis I*). It is believed that validating this hypothesis will allow for explanation of the contradictory results of previous studies in which only the final temperature (not the heating rate or holding time) was kept identical between CH and MW pretreatments.

The second hypothesis proposed in this research is that, as long as the aforementioned factors (pretreatment temperature, heating rate, and holding time) are controlled, the method of pretreatment (MW vs. CH) is not a significant factor (*Hypothesis II*). The second hypothesis, if validated, rejects the non-thermal (athermal) effect of MW irradiation for enhanced sludge pretreatment compared to the CH. The MW athermal effect postulated by some of the researchers was believed to be due to the polarization of macromolecules and their alignment with the EM field poles, that may cause breakage of hydrogen bonds and therefore additional disintegration of sludge flocs compared to CH (Eskicioglu et al., 2007b).

Another important observation made from literature review is that all the MW pretreatment studies have been constrained to a heating frequency of 2.45 GHz which is due to the availability of the commercial equipment (such as kitchen or bench-scale MW ovens) (Chi et al., 2011; Eskicioglu et al., 2009; Hu et al., 2012; Ji Park et al., 2010; Kuglarz et al., 2013; Mehdizadeh et al., 2013). A frequency of 2.45 GHz is used in these heating systems because MW power is absorbed by the water in the food (Metaxas, 1996). However, a frequency of 2.45 GHz may not necessarily be the most optimal frequency for pretreatment of WAS. There are several disadvantages associated with pretreating sludge at a frequency of 2.45 GHz. First, since the penetration depth decreases significantly at high frequencies (MW range), it is difficult to heat large volumes at 2.45 GHz (Kingston & Jassie, 1988). Second, it is difficult to obtain uniform heating throughout the load volume. Third, the power efficiency of high power MW sources (generators) are limited and the design of high power MW system can be expensive. A recently published comprehensive review article reports that almost all of the thermal pretreatments consuming electricity (i.e. MW) have a negative overall energy balance which means that they do not satisfy their input energy demand, although high sludge disintegration and/or improved biogas production are achieved (Cano et al., 2015). Therefore, it is hypothesized in this research that more optimal thermal disintegration can be achieved by implementing high power radio frequency (RF) heating at an optimum frequency (selected based on electrical characteristics of Kelowna's WAS) (*Hypothesis III*).

1.3. RESEARCH OBJECTIVES

To validate the hypotheses stated in section 1.2, the short term, 5-year, objectives of this research were defined as follows:

I. Compare the effect of CH and MW pretreatments on sludge disintegration (solubilization) over a wide range of final heating temperatures and ramp rates under identical thermal profiles (*Hypotheses I and II)*.

II. Study the simultaneous effects of the pretreatment method (CH vs. MW), final temperature, and heating ramp rate on performance of mesophilic and thermophilic batch AD (*Hypotheses I and II)*.

III. Design and fabrication of a novel and efficient RF sludge pretreatment system for advanced sludge AD (*Hypothesis III)*.

IV. Compare the effects of the RF (at 13.56 MHz) and MW (at 2.45 GHz) pretreatments on sludge solubilization and mesophilic batch AD under identical heating profiles (*Hypothesis III).*

V. Performance evaluation and energy assessment of the RF and MW pretreatment systems for advanced sludge AD under semi-continuous flow regime (*Hypothesis III)*.

1.4. RESEARCH TASKS AND METHODOLOGY

To achieve the objectives of the research stated in section 1.3, the following tasks/ methodology were adapted:

I. The DWSC samples were pretreated using both a CH and MW pretreatment system. For CH, a custom-built CH system simulating MW hydrolysis under identical heating and cooling profiles was used. The MW pretreatment was applied using a high pressure lab-scale MW station operating at a frequency of 2.45 GHz. The effects of three main pretreatment parameters including pretreatment method (CH and MW), heating ramp rate (3, 6 and 11°C/min) and final temperature (80, 120 and 160°C) on sludge solubilization were evaluated simultaneously. A statistical analysis was performed to evaluate the significance of the obtained results.

II. Next to the solubilization study (objective I), 45 mesophilic and 33 thermophilic batch anaerobic digesters were set-up to compare the effect of MW and CH on the performance of digesters. Before the start of the batch study, the inocula were acclimatized to both thermally-pretreated and non-pretreated sludge samples.

III. Design of the RF sludge pretreatment system was started by evaluating several possible configurations for the general geometry and physical aspects of the system (conceptual design). The conceptual design step was followed by the electrical analysis and simulation of the system which was conducted by Saimoom Ferdous, a former master student in electrical engineering supervised by Dr. Thomas Johnson. Material selection and purchasing was the next step. Aluminum was chosen for all metal parts of the system.

Teflon was used for the heating vessel as it has a very low dielectric loss factor. A detailed mechanical design was then created using Solidworks software and was used in the manufacturing process. Lastly, different parts of the system were built and assembled at the UBC School of Engineering machine shop. A close-loop LabView program was developed to control the thermal profile during RF heating. It should be noted that the RF system was designed based on the dielectric characteristics of TWAS to achieve a very efficient and uniform heating.

IV. The effects of three parameters including pretreatment method (RF vs. MW), final temperature (60, 90 and 120°C), and stationary (holding) time (0, 1 and 2 h) on sludge solubilization and performance of mesophilic batch AD were evaluated simultaneously. Energy measurements were also made to compare the efficiency of the RF heating system with that of the MW system operating at 2.45 GHz.

V. The pretreated TWAS samples were prepared using the RF and MW systems at the final temperatures of 80 and 120°C. Four lab-scale semi-continuous flow mesophilic digesters fed with pretreated TWAS plus one mesophilic digester fed with non-pretreated sludge (control) were set up. The same number of digesters were also set up under thermophilic condition. After a five-month acclimation period, the digesters were operated for 80 days at a SRT of 20 d. The operation was then continued at a SRT of 10 d for 40 days.

1.5. DISSERTATION STRUCTURE

This dissertation is divided into eight chapters. The current introduction chapter provides background information about wastewater treatment, municipal sludge production, and the technology of AD, followed by stating the research hypotheses, objectives, and the general methodology applied. The second chapter contains a detailed literature review providing more in-depth information about municipal wastewater compositions and treatment, sludge management methods, principals and kinetics of AD, and sludge pretreatment. The advantages, disadvantages, and limitations of a variety of sludge pretreatment methods are also discussed. The literature review forms the foundation which is necessary to address the current research gap in the field. In Chapter 3, the experimental results regarding the comparison of CH and MW pretreatments for DWSC solubilization are discussed. The findings of a comparison study between the CH and MW pretreatments to enhance mesophilic and thermophilic batch AD are presented in Chapter 4. Chapter 5 provides detailed information about the novel RF sludge pretreatment system which was designed as part of this project. In Chapter 6, the effects of the custom-designed RF pretreatment system at 13.56 MHz and a MW oven operating at 2.45 GHz on TWAS solubilization and mesophilic batch AD are presented. The results of the last experimental phase which was conducted under semi-continuous flow regime, both at mesophilic and thermophilic conditions are described in Chapter 7. The conclusions of this research and recommendations for future work are provided in Chapter 8.

CHAPTER 2. LITERATURE REVIEW

This chapter provides a literature review about municipal wastewater composition and its treatment, municipal sludge characteristics and its management options, principals of AD, kinetics of AD, and sludge pretreatment. Among the pretreatment techniques, the focus is on thermal pretreatment techniques for advanced sludge anaerobic digestion. In this regards, the current state of knowledge and research in the field is reviewed. The advantages, disadvantages, and limitations of the existing thermal pretreatment methods, along with the current research gap in the field are discussed.

2.1. MUNICIPAL (DOMESTIC) WASTEWATER

The large-scale urbanization and growth of the cities' population have resulted in a significant amount of municipal (domestic) wastewater which needs to be treated prior to being discharged back to the environment. The typical composition of municipal wastewater is listed in Table 2-1. As seen in Table 2-1, the higher the water usage (per capita) for domestic purposes is, the lower the concentration of the contaminants in the wastewater will be. As a result, the wastewater strength is lower in developed countries like Canada with higher domestic water consumption compared to that of developing countries such as Bangladesh. Municipal wastewater is mainly composed of dissolved and suspended organic matter. In addition to the organic matter, there are some other constituents present in the municipal wastewater including inorganic compounds, sediments, oil and grease, nutrients (nitrogen and phosphorus), and pathogenic microorganisms. It may also contain trace amount of some macro- and micro-pollutants such as heavy metals, pharmaceuticals, and personal care products (i.e. hormones).

Table 2-1. The typical composition of municipal (domestic) wastewater [a]

Description	Concentration in municipal wastewater		
	Low (750 L/capita)	**Medium (460 L/capita)**	**High (240 L/capita)**
Biochemical oxygen demand (mg/L)	110	190	350
Chemical oxygen demand (mg/L)	250	430	800
Total solids (mg/L)	390	720	1230
Dissolved solids (mg/L)	270	500	860
Suspended solids (mg/L)	120	210	400
Total nitrogen (mg-N/L)	20	40	70
Total phosphorus (mg-P/L)	4	7	12
Organic phosphate (mg-P/L)	1	2	4
Inorganic phosphate (mg-P/L)	3	5	10
Chlorides (mg/L)	30	50	90
Volatile organic compounds (mg/L)	<0.1	0.1-0.4	>400
Oil and grease (mg/L)	50	90	100
Sulfate (mg/L)	20	30	50
Total coliform (MPN/100 mL)	10^{6}-10^{8}	10^{7}-10^{9}	10^{7}-10^{10}
Fecal coliform (MPN/100 mL)	10^{3}-10^{5}	10^{4}-10^{6}	10^{6}-10^{8}

[a] The data were adapted from Metcalf & Eddy (2013).

The biochemical oxygen demand (BOD) and chemical oxygen demand (COD) are used to address the degree of the organic pollution in wastewater (Table 2-1). The BOD is equivalent to the amount of oxygen that is consumed by the aerobic microorganisms to degrade the organic matter in the wastewater. Therefore, this parameter only represents the concentration of the biodegradable organic compounds. The BOD measurement is also time-consuming and can be challenging in the presence of toxic compounds in wastewater inhibiting the microbial activity during the test. Thus, the COD test has been widely used because it can be measured much faster (in 3-5 hours compared to 5 days for the BOD analysis) and also indicates the amount of oxygen that would be required to oxidize all of the organic compounds (including non-biodegradable organics).

2.2. MUNICIPAL WASTEWATER TREATMENT

2.2.1. Primary treatment

Wastewater treatment processes are designed in order to reduce the adverse effects of the pollutants present in the wastewater. Figure 2-1 shows the process flow diagram of a conventional municipal WWTP containing primary and secondary treatment stages followed by anaerobic sludge digestion process. The primary treatment typically includes all or some of the following processes: screening, grit removal, equalization, and primary clarification. The screening process is used to remove large materials such as rags, sticks, and debris from the stream to prevent the damage and blockages of the equipment. After screening, the wastewater flows through the grit chambers where the dense materials such as sand, silt, coffee ground, and glass pieces are settled out and collected at the bottom of the tank. The grit chambers are designed in a way to prevent the organic materials from settling and keep them suspended in the liquid stream. Both the screened debris and the settled grits are normally sent to municipal landfills for disposal. Due to the fluctuation of the produced wastewater, the grit chambers are usually followed by equalization tanks to prevent the hydraulic and organics shocks, providing an almost constant flowrate.

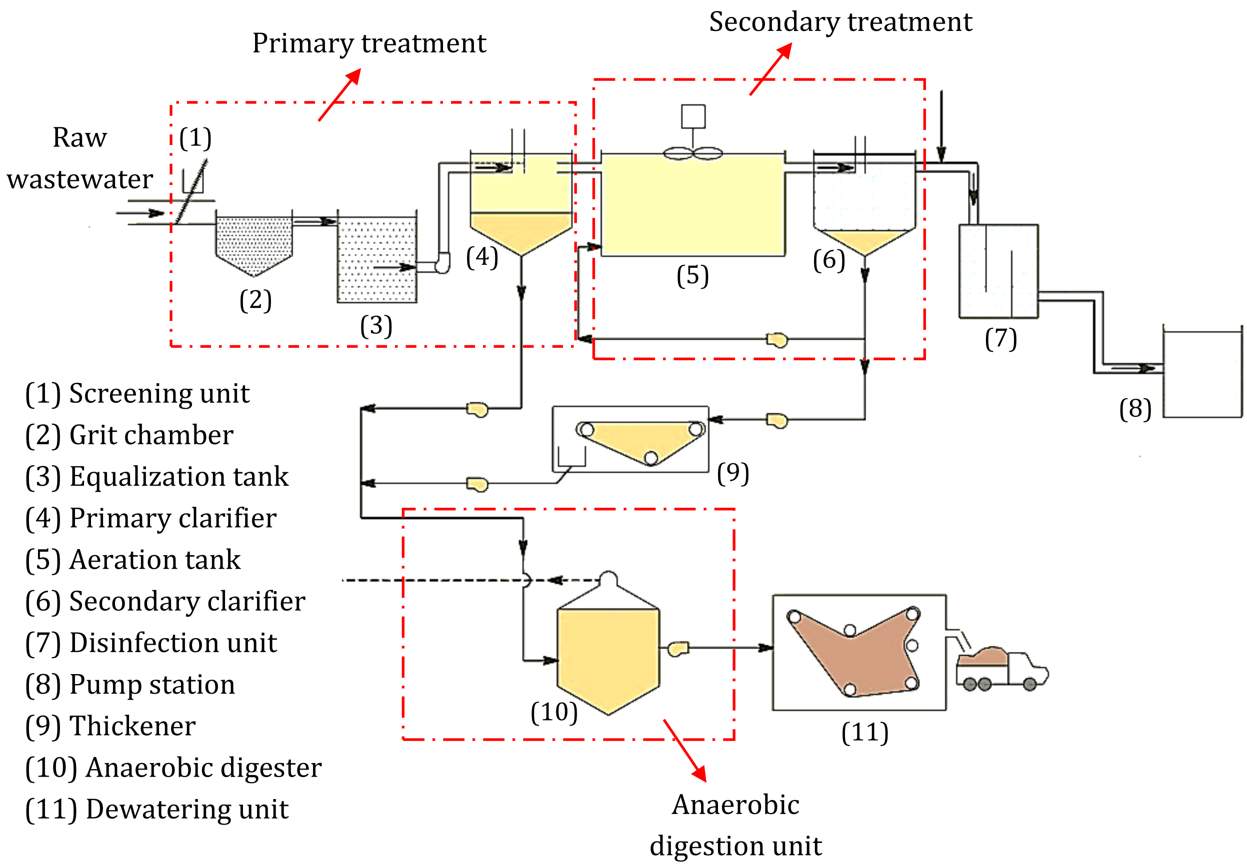


Figure 2-1. The process flow diagram of a conventional municipal wastewater treatment plant followed by anaerobic sludge digestion process (Modified from *http://www.globalspec.com*)

After removing the coarse and settleable materials, the wastewater enters primary clarifiers where the majority of the suspended matter is settled out. Although removing organic matter is not the goal of the primary treatment, a portion of the organics is also removed from the wastewater stream during the settling process. Typically, 55-70% of the total suspended solids (including 25-35% of organics) are removed via primary sedimentation (Metcalf & Eddy, 2013).

2.2.2. Secondary treatment

After primary treatment, the wastewater undergoes secondary treatment process where biological removal of the organics (mostly in dissolved and colloidal forms) is achieved. There are a variety of biological treatment technologies which can be used for this purpose such as aerated lagoons, rotating biological contactors (RBC), plug flow tanks, sequencing batch reactors (SBR), oxidation ponds, extended aeration systems, attached-growth biofilm reactors, and upflow anaerobic sludge blanket (UASB). For municipal wastewater treatment purposes, a biological method called activated sludge process is commonly used. In this method, the treatment process is achieved using a mixed culture of aerobic microorganisms in an aeration tank followed by secondary clarifier. In this process, the microorganisms uses the organic matter (oxygen demanding compounds) as a carbon source to grow and perform their metabolic activities. This process results in the production of water, carbon dioxide (CO_2) and new microbial cells.

The mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) are usually used as the index of the microorganism population in the aeration tank. The biological treatment tank is followed by secondary clarifiers where the MLSS is separated from the liquid part of the treated wastewater (supernatant) by gravitational settling. As seen in Figure 2-1, a portion of the collected sludge at the bottom of the secondary clarifiers is returned to the aeration tank to maintain the concentration of the microorganisms at a constant and optimum level.

2.2.3. Tertiary treatment

Nowadays, the stringent regulations on the quality of the treated wastewater necessitate further processing of wastewater after the secondary treatment. The purpose of the advanced or tertiary treatment processes is mainly to reduce the concentration of nutrients (nitrogen and phosphorus) in the effluent of WWTP prior to being released to the environment. The nutrient-containing wastewater can cause eutrophication if discharged untreated. Sequential aerobic-anoxic treatment processes is one of the commonly used techniques for nitrogen removal. Attached-growth biofilm processes have been also received significant attentions for nitrogen removal, since simultaneous nitrification-denitrification reactions can occur within one reactor. Phosphorus reduction can also be achieved using phosphorus accumulating organisms (PAOs) or addition of metal salts (i.e., iron or aluminum salts) (Metcalf & Eddy, 2013).

2.3. MUNICIPAL SLUDGE CHARACTERISTICS

Municipal WWTPs generate sludge as a by-product of the physical, chemical, and biological treatment processes. The daily average amount of the produced sludge ranges from 60 to 90 g dry-weight per capita per day (Appels et al., 2008). The characteristics of wastewater sludge depend on various parameters such as wastewater composition, type of the applied treatment systems, and the sludge age, etc. The understanding of the sludge characteristics is necessary for choosing the optimum sludge treatment or disposal method. Primary sludge (PS) and secondary sludge which is also called waste activated sludge are two main sludge streams produced in large quantities at the municipal WWTPs. The typical characteristics of the PS and WAS are summarized in Table 2-2.

The PS generated during primary settling of wastewater is composed of settleable organic and inorganic matter. On average, 100-300 g of PS is produced per 1 m^3 of the municipal wastewater treated, assuming 50-65% removal of the wastewater total suspended solids during primary sedimentation (Wang et al., 2007). The PS is generally grayish in color, has a slimy structure, produces offensive odor, and has high content of pathogens (Metcalf & Eddy, 2013). Due to the high content of readily biodegradable organic matter, the PS is a good candidate for being anaerobically digested. It has also better dewatering properties compared to the WAS (Turovskiy & Mathai, 2006).

The WAS is the by-product of biological treatment processes which is collected during the secondary sedimentation. Microbial cells and EPS are the primary components of the WAS. The EPS are high-molecular weight polymeric compounds which are comprised of biopolymers (polysaccharides, proteins and lipids), nucleic acids, and humic substances (Frolund et al., 1995; Park, 2002). The biodegradability of WAS is much lower than that of the PS, and for that reason it requires longer retention time during the biological transforming processes like AD. In WWTPs, the WAS is often mixed with the PS and sent for further processing.

Table 2-2. The typical characteristics of municipal sludge [a]

Description	Primary sludge (PS)	Waste activated sludge (WAS)
Specific gravity (-)	1.02	1.005
pH	5.0 - 8.0	6.5-8.0
Total solids (%)	5 - 9	0.8-1.2
Volatile solids/Total solids (%)	60 - 80	60-90
Total chemical oxygen demand (g-TCOD/g-VS)	2.0	1.4
Alkalinity (mg/L as $CaCO_3$)	500 - 1500	600 - 1200
Nitrogen (N, % of TS)	1.5 – 4	2.4 – 5.0
Phosphorus (P_2O_5, % of TS)	0.8 – 2.8	2.8 – 11.0
Energy content (kJ/kg TS)	23,000 - 29,000	19,000 - 23,000

[a] The data were adapted from multiple sources (Aldin, 2010; Metcalf & Eddy, 2013; Parker, 2005; Turovskiy & Mathai, 2006).

2.4. SLUDGE MANAGEMENT

In a conventional WWTP which employs primary clarification followed by aerobic biological processes, up to 60% of the initially diluted organic matter ends up as sludge (Garrido et al., 2013). The produced sludge (PS and WAS) represents only 1−2% of the wastewater volume. However, due to the large quantity of the wastewater being treated, a significant amount of sludge is produced every day. Figure 2-2 illustrates the annual production of sewage sludge (in dry weight) in some of the developed countries. As seen in Figure 2-2, in Canada, approximately 0.7 million tons of dry municipal sludge are produced every year. This represents the production of 50-55 g of dry sludge per capita per day. Currently, the sludge handling and disposal comprise more than half of the land and carbon footprint of the municipal WWTP (Koh et al., 2010). From a financial perspective, sludge management is also an expensive stage of wastewater treatment life cycle receiving 40 to 60% of the operating cost of municipal (Appels et al., 2008). Therefore, searching for safe, environmental-friendly, and cost-effective sludge management options is one of the important challenges in the waste management field.

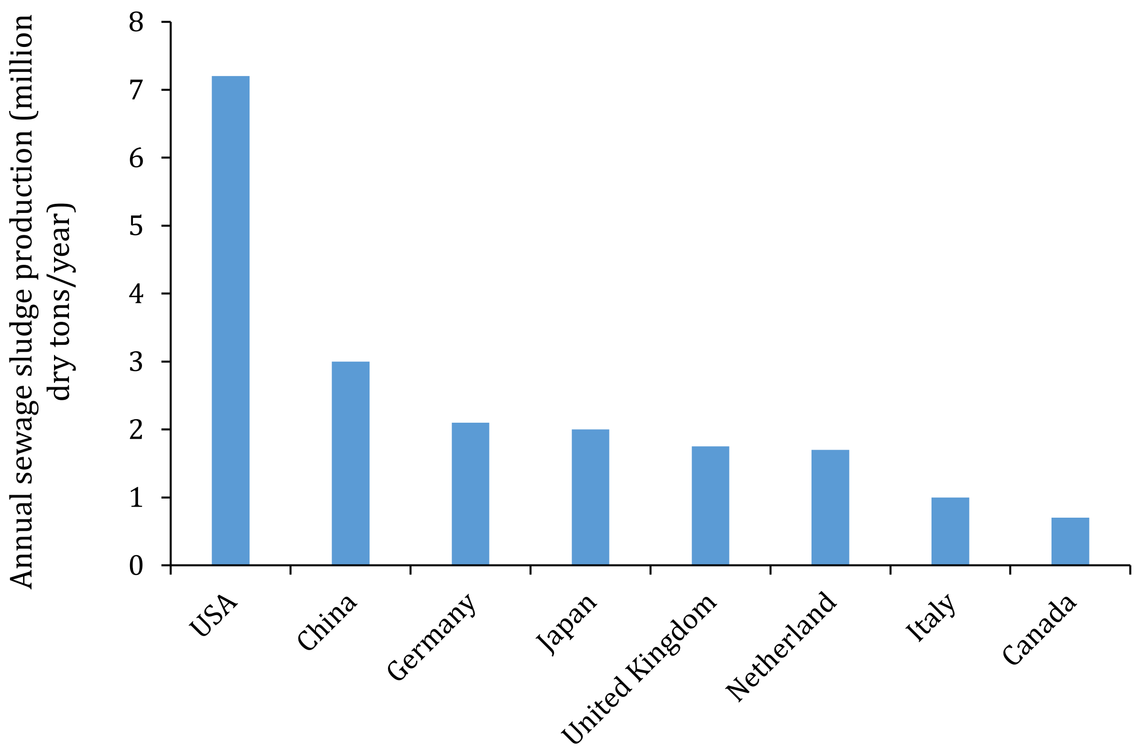


Figure 2-2. The annual sewage sludge production in Canada and some other developed countries (the data were adapted from multiple sources (Pérez-Elvira et al., 2006; USEPA, 1999; Zhen et al., 2017))

Traditionally, a variety of methods have been used for sludge management and disposal such as ocean disposal, incineration, landfilling, and land application. However, these methods are facing strong opposition from environmental authorities and also from the public domain. Figure 2-3 compares the primary sludge management/disposal methods used in the United States, China, Japan, and Canada. A few countries are still using the ocean as a location for dumping their municipal sludge. However, in most of the countries the disposal of the sludge to ocean are legally prohibited (Bastian, 1995). Incineration is a commonly used sludge reduction method in the countries that have limited land. This method achieves significant volume reduction since almost all of the organic matter present in the sludge are mineralized during the process. Incineration is however associated with high capital and operating costs and has the risk of producing hazardous volatile gases, threatening both health and environment (Metcalf & Eddy, 2013). The risk of air pollution has forced many regions to either prohibit the incineration or to enact more strict standards in order to reduce the level of hazardous compounds in the effluent gas. Landfilling is another common practice for sewage sludge disposal. However, the application of this method is becoming more and more challenging due to limited landfill space, poor economics, and production of uncontrolled leachate causing groundwater contamination (Mahmood & Elliott, 2006).

As seen in Figure 2-3, the land application of sludge as a soil fertilizer is a common practice in the US and Canada due to its beneficial aspects. Previous lab studies and also the full-scale practices have proven that the land application of biosolids improves soil fertility due to its high nutrient (nitrogen, phosphorus) content reducing the need for chemical fertilizers (Andrew et al., 1997; Lu et al., 2012; Moussavi et al., 2010; Pascual et al., 2004; Passuello et al., 2010; Singh & Agrawal, 2010; Wang et al., 2008). However, there has been a concern that land application of biosolids may impose a risk to the environment and human health due to the fact that different contaminants (i.e., pathogens, heavy metals, endocrine disrupting chemicals, etc.) are concentrated into the sludge during wastewater treatment processes (Cano et al., 2015; Fourier, 1955; Incropera et al., 2007; Turner, 1976; Turovskiy & Mathai, 2006). Therefore, the land application of biosolids is regulated by increasingly stringent regulations in most of the countries (i.e., in the US by 40 CFR Part 503 (Ho, 2003) and in European countries by European Union (EU) Directive 86/278/EEC (WHO, 2002)). The regulations require that the municipal sludge undergoes stabilization treatment (i.e., long-term storage, biological, chemical or thermal processes, etc.) before it is land applied.

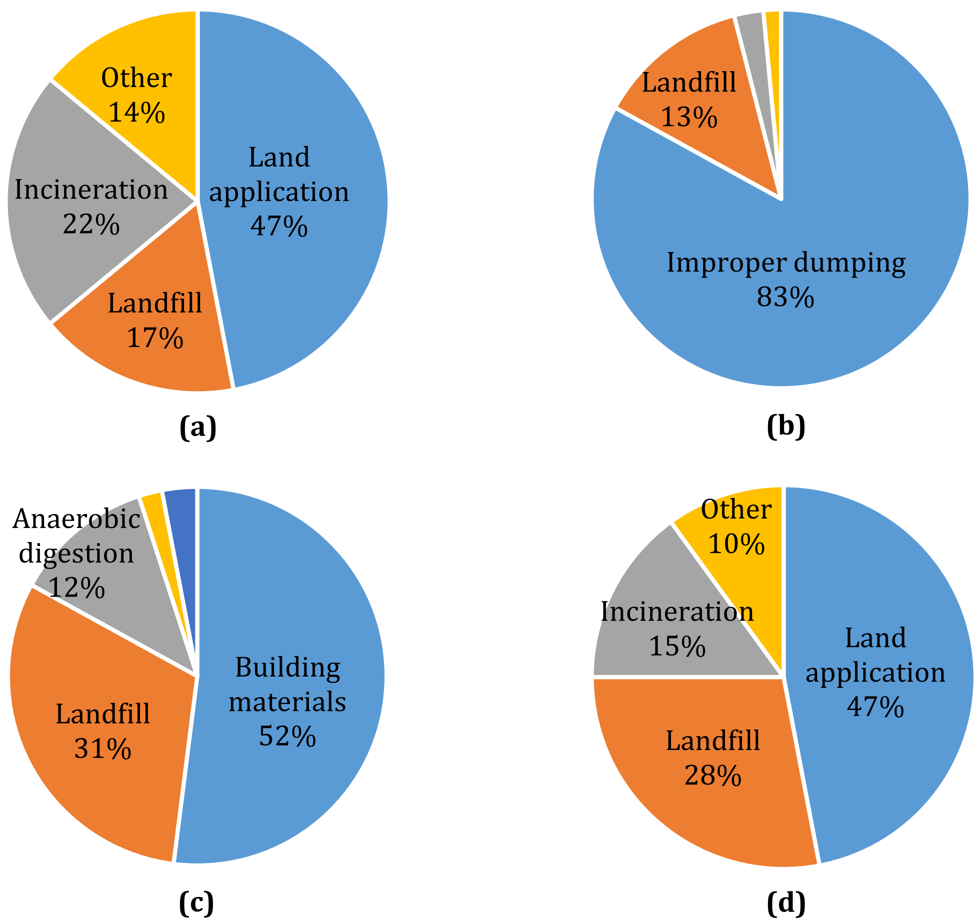


Figure 2-3. Municipal sludge handling and disposal in (a) United States, (b) China, (c) Japan, and (d) Canada (the data were adapted from Zhen et al. (2017))

In BC, biosolids recycling is regulated under the OMRR (OMRR, 2008). The OMRR is enabled under the Environmental Management Act (administered by the BC Ministry of Environment) and the Health Act (administered by the Ministry of Health). Under the OMRR, the biosolids are classified either as Class A or Class B. Table 2-3 listed the OMRR criteria for a biosolids to be classified as Class A or Class B. As seen in Table 2-3, Class A biosolids contain lower fecal coliform densities (< 1,000 MPN/g dry weight or dw) than Class B biosolids (< 2×10^6 MPN/g dw). The stringent criteria allow for more liberal distribution and application of Class A biosolids. For biosolids land application, a Land Application Plan (LAP) has to be prepared by a qualified professional, as prescribed in the Environmental Management Act and OMRR. A LAP is prepared for each site and for each occurrence of land application and is approved for one year. A LAP is not required for Class A biosolids (CCME, 2010).

Table 2-3. The OMRR[a] criteria for Class A and Class B biosolids (OMRR, 2008)

Criteria	**Class A Biosolids**	**Class B Biosolids**
	Process Criteria	
Pathogen Reduction	√	√
Vector Attraction Reduction	√	√
Fecal Coliform (MPN/g dw [b])	<1000	<2,000,000
	Quality Criteria (µg/g dw)	
Arsenic	75	75
Cadmium	20	20
Chromium [c]	-	1,060
Cobalt	150	150
Copper [c]	-	2,200
Lead	500	500
Mercury	5	15
Molybdenum	20	20
Nickel	180	180
Selenium	14	14
Zinc	1,850	1,850
Foreign Matter	≤ 1%	≤ 1%
	Monitoring	
Sampling and Analysis	Required	Required
Record Keeping	Required	Required

[a] OMRR: Organic Matter Recycling Regulations

[b] dw: dried waste

[c] The Ministry of Environment proposed to add chromium and copper maximum allowable standards to Class A biosolids as 1060 µg/g dw and 2200 µg/g dw, respectively (BC, 2017a).

2.5. SLUDGE STABILIZATION

Pathogen removal, odor reduction, and elimination of the putrefaction potential are the primary goals of sludge stabilization. These goals are achieved by reducing the easily biodegradable organics of the sludge, so that the remaining fraction would be unsuitable for microbial activity (Metcalf & Eddy, 2013). In addition, the volume reduction and improved dewaterability of the sludge are often achieved through sludge stabilization. Stabilization processes can be also used as method for energy and/or nutrients recovery from sludge. The sludge stabilization has been initiated more than 20 years ago and is now practiced all around the world, especially in Europe, United States, Canada, Japan and other developed countries (Anjum et al., 2016). The stabilized sludge is called biosolids which is low in pathogens, organic matter, heavy metals, and other harmful compounds compared to the raw sludge. The biosolids can be land applied for different purposes such as site remediation, and agricultural and/or domestic use as a fertilizer if they meet specific standards. A variety of including lime (or alkaline) stabilization, composting, thermal stabilization, and biological stabilization (aerobic and anaerobic digestion) can be used for sludge stabilization depending on the specific objectives that need to be achieved.

2.5.1. Alkaline stabilization

Alkaline stabilization and conditioning involves the addition of alkaline chemicals such as burnt lime (calcium oxide), slaked lime (calcium hydroxide), carbide lime, fly ash, and cement kiln dust (Werther & Ogada, 1999). For alkaline stabilization, sufficient quantity of lime is added to the raw (untreated) sludge to increase the pH to 12 or higher. Different chemical reactions occur at the high pH altering the chemical composition of the sludge. This creates an undesirable environment for the survival of the microorganisms present in the sludge. Alkaline stabilization prevents putrefaction and odor formation as long as the pH remains at the high level. However, the concentration of the residual lime reduces as a result of lime consumption through the chemical reactions, decreasing the pH. Therefore, excess lime should be added to maintain the pH above 12, hindering subsequent microbial activity. The addition of some forms of lime such as calcium oxide (CaO) to the sludge generates large amount of heat due to the hydration reactions. This increases the sludge temperature which results in even more pathogen removal in the stabilized sludge (Metcalf & Eddy, 2013).

2.5.2. Thermal stabilization

Thermal stabilization involves increasing the sludge temperature (up to 200°C) under the pressure of 20-30 bar. By controlling the temperature, contact time, and amount of the added air (oxygen), the process can achieve a variety of purposes (Werther & Ogada, 1999). Thermal process creates significant changes in the composition of the sludge by breaking down the gel structure of the sludge and coagulating the solids. This results in the release of the bond water reducing the water affinity of the sludge. Therefore, along with pathogen removal, improved dewaterability is also achieved through thermal stabilization. The disadvantages of thermal stabilization include higher mechanical complexity than most of the systems that WWTP operators are familiar with, more maintenance, production of odorous off-gas which needs to be deodorized before release, and increasing the concentration of soluble organics in the released liquid (Ewing et al., 1978).

2.5.3. Biological stabilization

2.5.3.1. Composting

Composting is a biological-based method of sludge stabilization that is used widely around the word. The composting may be an appropriate stabilization method if the land availability is not an issue and when the final composted materials can be used as a fertilizer. The composting is often achieved under aerobic condition where the production of hydrolytic enzymes and increased growth rate facilitate the hydrolysis of the large-molecular compounds into more simple-structure materials. Sludge compositing is typically achieved during three sequential temperate-based steps. It starts with the growth of the mesophilic bacteria where their metabolisms elevate the sludge temperature. The continuous increase in temperature starts the second step of the process in which the thermophiles become active. The pathogen removal is achieved as a result of the high temperature formed during this step. The maturation phase is the last step in which the temperature is reduced resulting in the reactivation of mesophiles. Large space and moisture requirement, odor issues, and high energy expenditure due to the aerobic nature of the process are the common drawbacks of composting.

2.5.3.2. Digestion

A large portion of the municipal sludge is composed of organic matter. Therefore, it can be stabilized through biological treatment (digestion). The sludge is often thickened to a concentration of 3% TS or higher before the digestion process (Anjum et al., 2016). The sludge thickening can be achieved using a variety of methods including gravity thickening, rotatory drum filtration, centrifugation, and dissolved air flotation. After thickening, the sludge is digested where the conversion of organic matter occurs. Pathogen removal and odor reduction are also achieved through digestion process (Semblante et al., 2015). In the case of WAS, the degradation of macromolecules such as carbohydrates, proteins, and humic substances is the key stage of the digestion process and greatly influences the overall performance of the system. Therefore, the efficiency of the process highly depends on the composition of the sludge being digested.

The sludge digestion can be applied in the presence or absence of oxygen, which is called aerobic and anaerobic digestion, respectively. The digestion process can also be performed under the mesophilic (35-37°C) or thermophilic (50-55°C) temperature. Aerobic digestion proceeds under continued aeration of sludge. The process starts with the depletion of the soluble organic substrates and is followed by the endogenous respiration of the microbial cells which exist in the sludge structure. Despite the high removal efficiency of the organic matter, the aerobic sludge digestion has several disadvantages. The aerobic digestion needs continuous aeration and constant mixing, and therefore it is an energy intensive method. Another drawback of the aerobic digestion is the production of high amounts of ammonium nitrogen as a result of the protein degradation. The concentration of the ammonium nitrogen is even higher at operating temperatures above 35°C due to the inhibition of nitrification-denitrification reactions (Anjum et al., 2016).

As previously explained, the larger portion of the organic matter present in municipal wastewater ends up as PS and/or WAS. Therefore, the produced sludge contains more than 60% of the initial energy of the wastewater (3.2 $\frac{MJ}{kg\ TS}$) which has the potential to be recovered

(Werther & Ogada, 1999). Sludge stabilization under the anaerobic condition is an alternative technique to the aerobic digestion providing the opportunity of producing stabilized sludge and at the same time recovering energy as biogas. The detailed discussion of AD is provided in section 2.6.

2.6. ANAEROBIC DIGESTION

2.6.1. Current status of anaerobic digestion

As previously mentioned in section 2.4, municipal sludge is required to be stabilized for further applications. AD consists of a series of biochemical reactions in an oxygen-free reactor which can be used for sludge stabilization. During AD, a portion of organic matter is converted to methane-rich biogas, while the remaining residual can be classified as biosolids. The type (class) of biosolids is typically defined based on its heavy metal and pathogen concentration.

AD is a common practice for sludge stabilization and bioenergy production in the Europe due the financial incentives promoting the use of the renewable energy. However, the statistics show that the AD technology has not received the same attention for energy recovery in North America. According to Lewis et al. (2008), 22% of the approximately 16,000 WWTPs operated at the time of the study in United States used AD for sludge stabilization, and of that, only 2% benefitted from the bioenergy production capability of the AD for electricity or heat generation. This has been due to the historically low investment return rate, and relatively low price of fossil fuels (Lewis et al., 2008). The availability of enough land to be used for other sludge management/disposal methods such as composting or landfilling has also encouraged about

80% of the WWTPs to implement other methods of sludge stabilization other than AD (Scanlan et al., 2010). In recent years, environmental-related issues such as global warming, climate change, and air pollution along with the rise of the energy cost have shifted the conventional trend of the sludge management towards more environmentally-friendly and energy-efficient technologies such as AD. The EU is a world leader in the “organics to bioenergy” field generating about 60% of the world’s biogas from organic matter. Among the EU countries, Germany has made the greatest progress in the field with a production of approximately $60 \times 10^3\ MWh/year$, of which more than $50 \times 10^3\ MWh/year$ results from anaerobic digestion or co-digestion processes. United Kingdom (UK), Italy, and France are the next largest producers in the list with a production of more than $20.5 \times 10^3, 12.7 \times 10^3$, and $4.1 \times 10^3\ MWh/year$, respectively (Raboni & Urbini, 2014). Although the contribution of AD processes in the total biogas production in these countries is much lower than that of the Germany, the trend of the biogas production is in the direction of a progressive increase from AD of organic matters and a decrease from landfills (Beurskens & Hekkenberg, 2011). Beside the full-scale application, AD has increasingly received a lot of attention in research. Figure 2-4 shows the ascending trend in the number of published research papers in the field of AD during a 20-year period of 1996-2016.

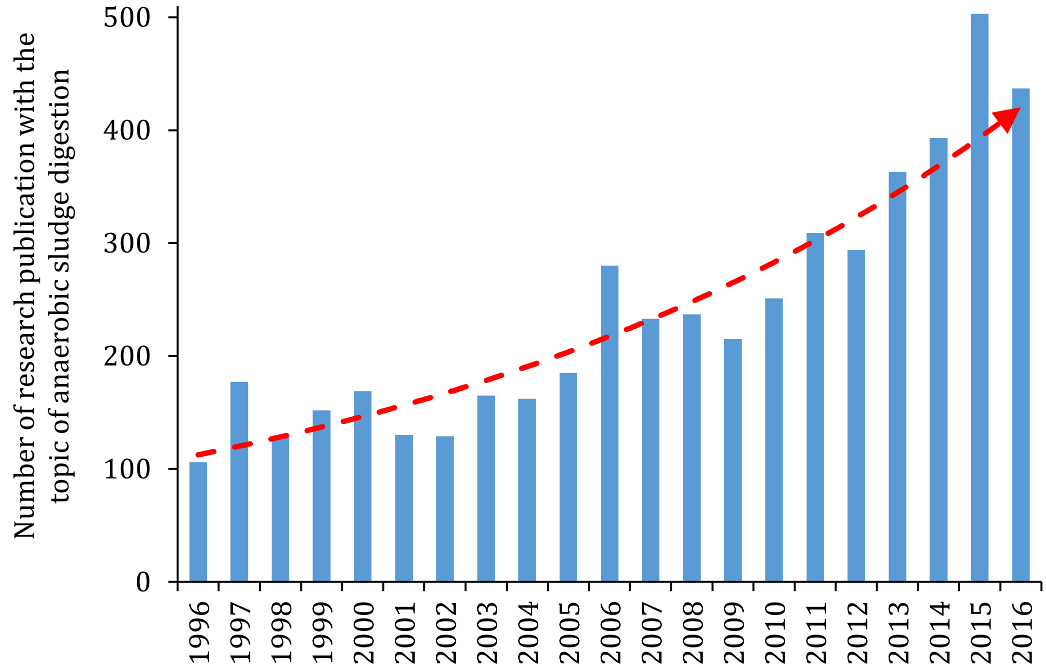


Figure 2-4. The number of research paper published with the topic of sludge anaerobic digestion from 1996 to 2016 (data generated from *https://www.scopus.com/*)

Along with the global movement towards more sustainable energy production methods, the use of AD for bioenergy generation are actively promoted by many Federal Government Departments in Canada including NSERC, National Research Council (NRC), Environment Canada (EC), Agriculture and Agri-Food Canada (AAFC), and Industry Canada (IC). Towards this goal, the Province of BC has committed to obtain a minimum of 93% of its electricity from clean renewable sources (BC, 2017b). In response to this commitment and also due to the challenges of the composting scenario which is currently practiced for the municipal sludge management at the City of Kelowna, the implementation of an anaerobic digester at KWWTP as an alternative way of biosolids management has been under consideration by the City of Kelowna. Figure 2-5 illustrates the process flow diagram of the proposed AD scenario at KWWTP.

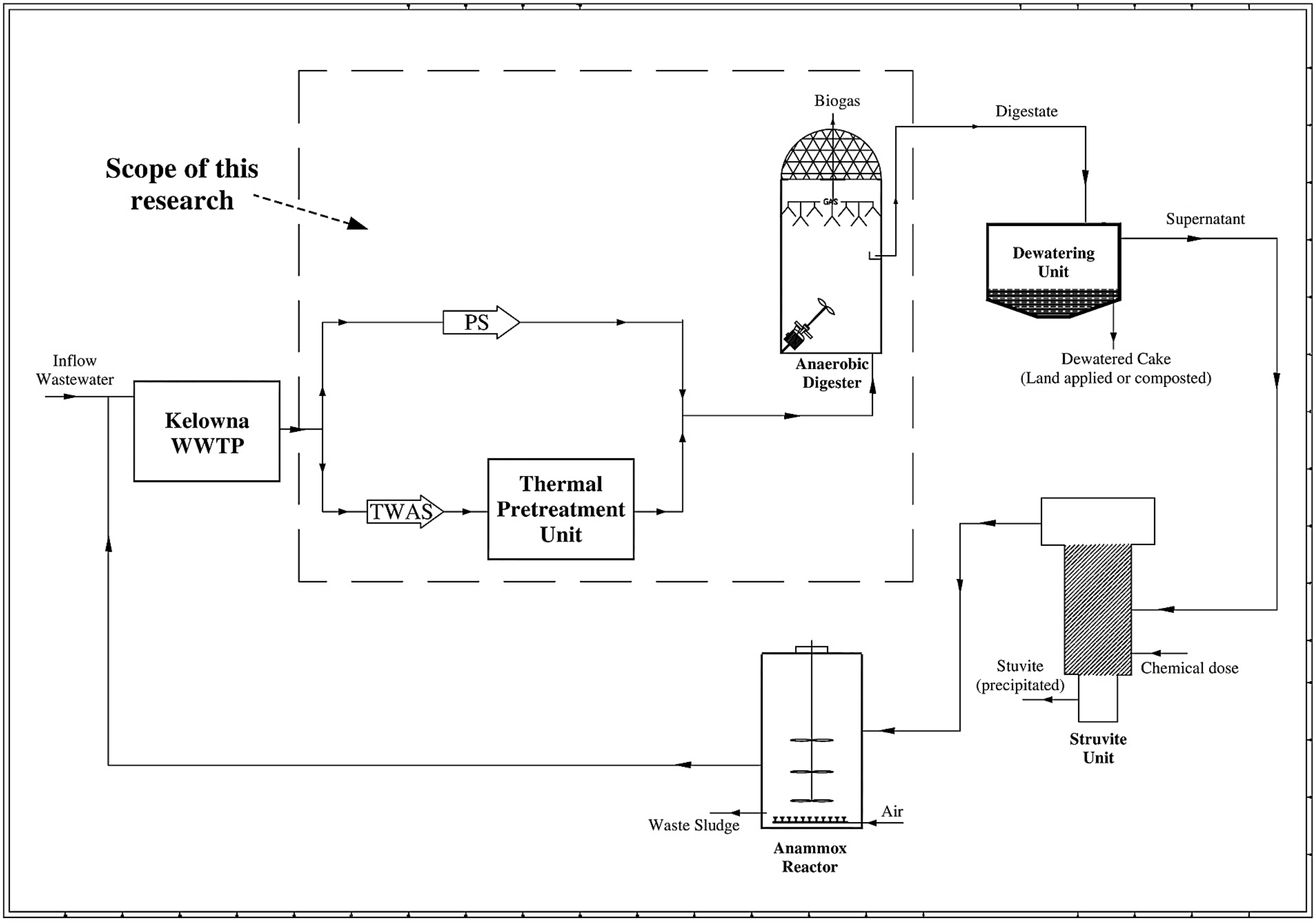


Figure 2-5. The schematic of the proposed anaerobic digestion scenario for Kelowna's wastewater treatment plant

2.6.2. Principles of anaerobic digestion

AD is a complex biochemical process requiring strict anaerobic (oxygen-free) conditions with significantly low oxidation-reduction potential (ORP) (less than -200 mV) to proceed effectively. As shown in Figure 2-6, AD consists of four main stages including hydrolysis, acidogenesis, acetogenesis and methanogenesis. AD starts with the hydrolysis stage where the insoluble and high molecular weight organic compounds in waste such as lipids, polysaccharides, proteins, and nucleic acids are broken down into soluble or low molecular-weight compounds (i.e. sugar, amino acids). The major microbial group involved in the hydrolysis stage is hydrolytic fermentative bacteria. These bacteria are able to break down the polymeric chains of organic matter using their extracellular enzymes (Vavilin et al., 2003). During the second stage (acidogenesis), the components formed via hydrolysis are further split resulting in the production of volatile fatty acids (VFA), ammonia (NH_3), carbon dioxide (CO_2), hydrogen sulfide (H_2S) and other by-products (Appels et al., 2008). Acidogenesis stage is followed by acetogenesis where the VFAs and alcohols produced in the previous stage are converted to acetic acid, CO_2 and H_2 by acetogens (Batstone et al., 2002). Methanogens are the group of microorganisms running the last stage of AD. The terminal electron acceptor in methanogenesis is not oxygen, but carbon. The final stage of methanogenesis produces methane mainly by two groups of methanogens. The first group splits acetate into methane (CH_4) and CO_2 (Eq. (2-1)). The second group uses hydrogen as an electron donor and CO_2 as an acceptor to produce CH_4 (Eq. (2-2)).

$$CH_3COOH \rightarrow CH_4 + CO_2 \qquad (2\text{-}1)$$

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \qquad (2\text{-}2)$$

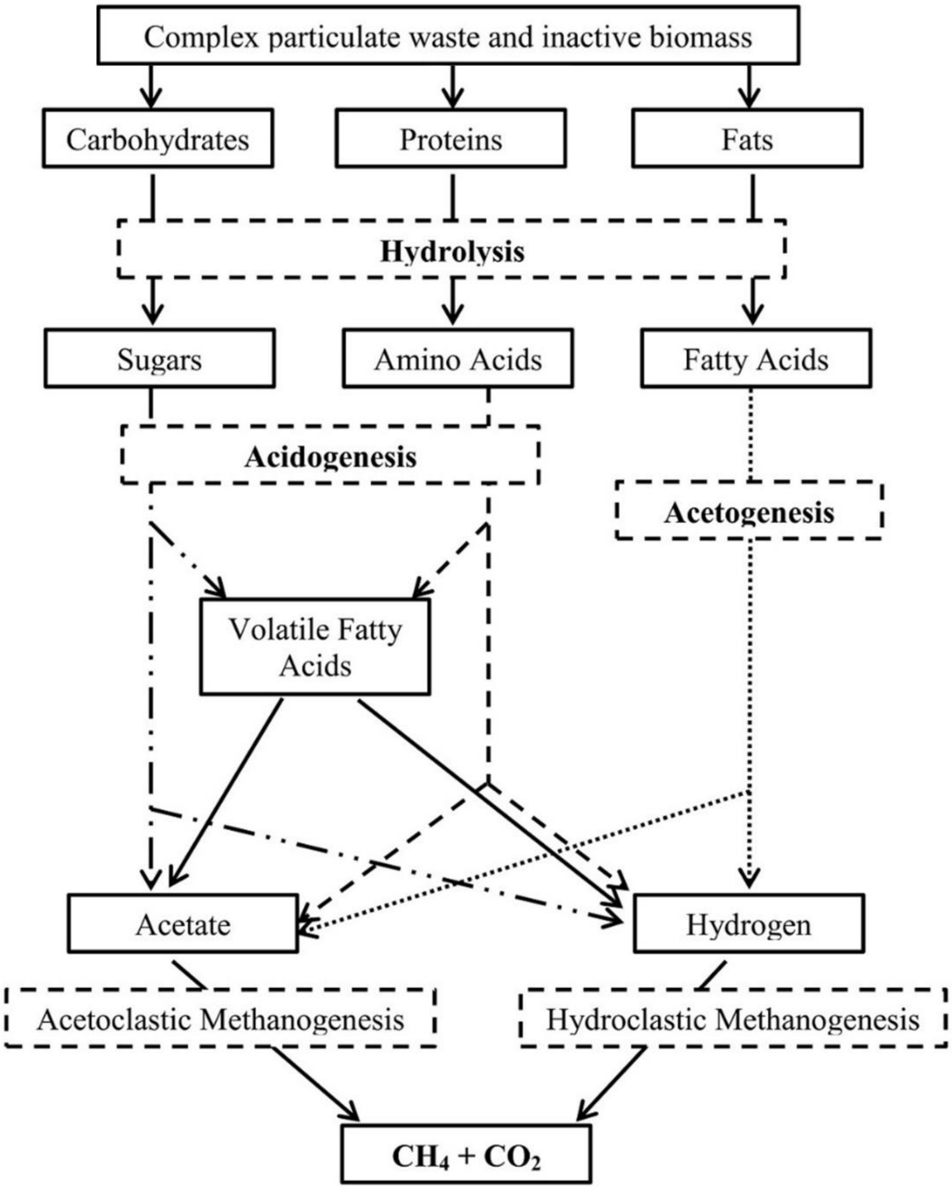


Figure 2-6. The main stages of sludge anaerobic digestion process (adapted from Batstone et al. (2002))

In conventional AD, all four stages occur simultaneously in a single, heated, and completely-mixed vessel (i.e. single-stage digestion). Dual-stage AD is another process where hydrolysis/acid formation and methane formation occur in two separate vessels. In single-stage AD, the methanogens are at the risk of being inhibited due to the presence of toxic compounds or because of the unfavorable environmental conditions, which leads to the accumulation of VFAs in the digester. This can decrease the digester pH and further reduce the microbial activity of bacterial communities especially methanogens. Therefore, AD operation requires a contact monitoring and controlled environmental conditions.

2.6.3. Affecting factors

The rate and stability of the AD process depend on the activity of different microbial groups which necessitates a delicate balance among their population for a successful AD. There are several physical and chemical factors which affect the population and activity of the microorganisms involved in AD. The key parameters affecting AD include temperature, sludge retention time, concentration of VFAs, alkalinity, pH, organic loading rate (OLR), and ammonia.

2.6.3.1. Temperature

The temperature significantly affects the physicochemical properties of the substrate being digested as well as the population and metabolism of different microbial groups involved in the digestion process. According to thermodynamics, while the endergonic reactions, under standard conditions, are more favoured at higher temperatures, the exergonic reactions such as hydrogenotrophic methanogenesis are adversely affected at elevated temperatures (Rehm et al., 2000). Figure 2-7 shows a schematic of the effect of temperature on the anaerobic digestibility of primary sludge. Figure 2-7 can be divided into four distinguishable parts with respect to the digestion temperature. Both the laboratory and full-scale results have shown that the digestion rate is rather slow at a temperature below 16°C (Figure 2-7, part (a)). Increasing the temperature above 16°C improves the activity of the mesophiles, resulting in a higher digestion rate (Figure

2-7, part (b)). Between 28°C and 42°C, the digestion proceeds more rapidly with the optimal performance at 35-37°C (Figure 2-7, part (c)). Above 37°C, a decrease in the digestion rate is often observed until the temperature reaches around 42°C. At this temperature the thermophilic organisms in the digestion process begin to start working. The digestion rate at the thermophilic temperature range (42-65°C) is often higher than that of the mesophilic due to the faster hydrolysis reaction (Casey, 2006). The thermophiles reach their optimum performance at around 55°C (Figure 2-7, part (d)).

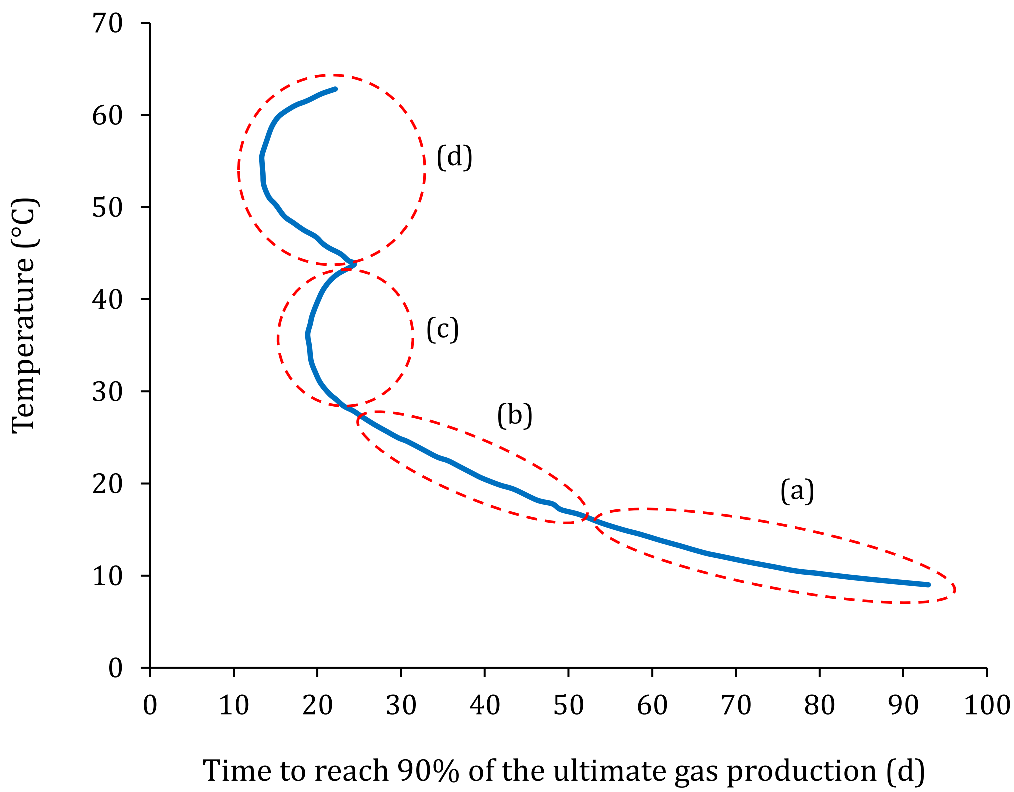


Figure 2-7. The effect of temperature on the anaerobic digestion time of primary sludge for the temperature ranges of (a) below 16°C, (b) 16 to 28°C, (c) 28-42°C, and (d) 42 to 65°C (the data used to plot the graph were adapted from Casey (2006))

Increasing the digestion temperature has several advantages. It increases the concentration of the soluble organics, thus, enhancing their utilization rate. It also improves the bacterial growth rate and their metabolisms. Higher pathogen removal can be also achieved at high temperatures. Beside these benefits, the elevated temperatures can have a variety of adverse effects. It increases the ammonia concentration which is known as an inhibitory compound to the microorganisms. Thermophiles are more sensitive to the change in the environmental parameters, and also show less stability than mesophiles in response to the variation of the operating conditions (Appels et al., 2008). Higher energy requirement is another drawback of thermophilic AD when compared to mesophilic AD.

2.6.3.2. Sludge retention time

Another important factor affecting the overall performance, pathogen destruction capability, and the solids removal efficiency of a digester is the SRT. The SRT is defined as the average residence time of solid particles in the digester. The proper SRT of a digester depends on the growth and reproduction rate of the different groups of microorganism present in the system. In another words, the fraction of microorganisms lost from the digester through the effluent (digestate) should be replaced by the newly grown cells. Therefore, in the case of a too short SRT, the bacteria do not have sufficient time to reproduce themselves which will eventually cause the process failure. A SRT of 10 d is usually suggested as the shortest SRT for AD (Appels et al., 2008). The suggested 10 d SRT is coming from the fact that the slow growing methanogens require at least five days to prevent them from being withdrawn from the digester. Accumulation of VFAs has also been observed for a SRT less than 8 d (Appels et al., 2008). The typical SRT of a full-scale mesophilic anaerobic digester is 20 to 25 d. In general, the longer the SRT of a digester, the higher the pathogen and solids removal would be. However, in order to achieve a longer SRT, a larger digester volume is required which results in greater capital cost, higher land footprint, and more energy expenditures.

2.6.3.3. Volatile fatty acids, alkalinity and pH

The concentration of VFAs, the levels of alkalinity, and the pH of the digestion mixture are three interdependent parameters which highly affect the digester performance. Different groups of microorganisms involved in AD process have their own optimum pH range. The acidogenic and methanogenic bacteria are the least and the most sensitive groups to pH fluctuations, respectively. Fermentative bacteria can perform at the pH rage of 4 to 8.5, while the optimum performance of methanogens is achieved within a much narrower pH range (6.5 and 7.2) (Hwang et al., 2004). At high pH values, the propionic and acetic acids are the primary products of the acidogenesis stage, while at low pH, the butyric and acetic acids are mainly generated (Hwang et al., 2004).

The concentration of VFAs and alkalinity determine the digester pH. Through the digestion process, the pH tends to decrease as a result of the VFAs production during the acidogenesis stage. The pH drop is however prevented by the production of alkalinity in the form of carbon dioxide, bicarbonate, and ammonia during the methanogenesis stage (Turovskiy & Mathai, 2006). Haandel and Lubbe (2007) reported that the complete acidogenesis-methanogenesis reactions result in a net alkalinity production of 0.44 g as $CaCO_3$ per g of the sludge being digested (Haandel & Lubbe, 2007). It was stated that AD can be operated at a VFA concentration of less than 100 mg/L to above 5000 mg/L, if the digester has enough buffering capacity to maintain the neutral pH (Droste, 1996).

The accumulation of VFAs has been reported as one of the main causes of AD instability and/or failure. The VFA accumulation can significantly drop the pH, interrupting the activity of the microorganisms, especially the methanogens. Addition of external alkalinity to the digester is common practice to avoid a drop in pH due to VFA accumulation. The literature suggested a VFA to alkalinity (as $CaCO_3$) ratio of 0.1-0.35 for a stable digester (Boe, 2006). It was also stated that a molar ratio of above 0.7 between the concentration of VFA and bicarbonate is required for a well buffered AD (Appels et al., 2008). Recycling a portion of the digester effluent to the influent is another technique to maintain the buffering capacity of the digester (Droste, 1996).

2.6.3.4. Organic loading rate

The rate at which the organic matter enters the digester is called organic loading rate (OLR). More organics are provided for the fermentative microorganisms at high OLRs, increasing the rate of VFA production in the digester. In the case of sufficient alkalinity and buffering capacity, the digester stability will be maintained under high OLR. This provides the opportunity for the methanogens to utilize the produced VFAs, converting them into biogas. However, due to the slower growth rate of methanogens compared to that of the acidogens, the VFA accumulation can be occurred due to a sudden increase in the OLR. The VFA accumulation can drop the digester pH causing the disruption of the methanogenesis stage. This will further decrease the pH, leading to more inhibition to the methanogens which will eventually cause system failure. The implementation of two-phased anaerobic digesters provide more control over the methanogens by separating them from the acid formers preventing their intervention. This technique has gained a lot of attention in recent years as a solution to increase the digestion performance and also decrease the risk of VFA accumulation.

2.6.3.5. Ammonia

The transformation of nitrogenous compounds during AD results in the production of free ammonia (NH_3). Compared to the ammonium (NH_4^+), free ammonia is known to be more toxic as it can penetrate through cell membranes (Chen et al., 2008). The concentration of free ammonia increases by increasing the pH and temperature (Metcalf & Eddy, 2013). Therefore, the risk of ammonia inhibition is higher for the thermophilic digesters compared to that of the mesophilic ones. The methanogenic bacteria are the most susceptible group of microorganisms to the toxicity of the NH_3. However, acclimation to the ammonia toxicity has been reported to significantly increase the tolerance threshold of the methanogens. At a pH greater than 7.4, the ammonium-N has been observed to have the inhibition effect at a concentration range of 1,500-3,000 mg/L. The concentration of more than 3,000 $\frac{mg\ NH_3}{L}$ was however reported to be toxic at any pH (Metcalf & Eddy, 2013). Due to the relatively low protein content found in municipal sludge, the ammonia toxicity is unlikely to occur during sludge AD.

2.6.4. Kinetics of anaerobic sludge digestion

The hydrolysis is usually known as the rate limiting step in AD process utilizing complex organic waste (Miron et al., 2000; Tiehm et al., 2001; Vavilin et al., 2003; Xie et al., 2011). In the case of WAS digestion, the hydrolysis stage is particularly important since the major fraction of sludge structures are microbial cells confined with layers of EPS that are known to be a relatively resistant substrate to the microbial degradation. The microbial cell wall itself has a semi-rigid structure containing glycan strands that are cross-linked by peptide chains. This structure provides sufficient strength to protect the cell from osmotic lysis. Several studies have shown that the overall sludge AD rate is governed by the hydrolysis of the cell walls and EPS (Eskicioglu et al., 2006; Tiehm et al., 2001; Weemaes & Verstraete, 1998a; Weemaes & Verstraete, 1998b).

According to the literature, the hydrolysis can be considered as a first-order reaction with respect to the concentration of the substrate (organic matter). The organic matter is often quantified by the VS or total chemical oxygen demand (TCOD) of the sludge (Coelho et al., 2011; Puchajda & Oleszkiewicz, 2006; Xie et al., 2011). It is well proven that first-order reaction rates can represent biogas-methane production as well as substrate (VS, TCOD) utilization during AD (Alkaya & Demirer, 2011; Eskicioglu & Ghorbani, 2011; Eskicioglu et al., 2006; Hosseini Koupaie et al., 2014). The first-order reaction model is shown in Eq. (2-3)

$$r_{su} = \frac{dC}{dt} = -kC_t \tag{2-3}$$

in which r_{su}, C and k are the substrate biodegradation rate $(\frac{mg}{L.d})$, substrate concentration (mg/L), and first-order specific biodegradation rate constant $(1/d)$, respectively. Eq. (2-4) is derived after integration and rearrangement of Eq. (2-3)

$$C_t = C_u e^{-kt} \tag{2-4}$$

in which C_t, C_u, and t are substrate concentration in the digester at time t (mg/L), ultimate biodegradable substrate (mg/L), and digestion time, respectively.

2.7. Sludge pretreatment

The bottleneck of conventional sludge AD is slow degradation of the municipal secondary sludge or WAS. This necessitates long SRT (20-30 days) requiring a large digester volume which eventually leads to high capital cost (Appels et al., 2008). According to the literature, the structure of WAS, is a mixture of different microbial cells, organic and inorganic compounds that adhere together with EPS, resisting AD (Park & Ahn, 2011b). Since hydrolysis is known as the rate-limiting stage of sludge AD, any attempts to increase the rate of hydrolysis will eventually improve the overall performance of the AD. For this purpose, a variety of pretreatment methods are used to break down (disintegrate) the polymeric network of WAS resulting in the release of intra- and extra-cellular biopolymers into the sludge liquid phase (solubilization), accelerating the digestion process (Bordeleau & Droste, 2011). Previous studies have proven that the sludge becomes more bio-available after pretreatment, and biogas production increases. Accelerated degradation rate of digestion results in reduced volume requirement for the digester (for a given organic load) which is directly linked to the capital cost (Carrere et al., 2010). The most common sludge pretreatment methods include biological, chemical, mechanical, and thermal processes. These methods are discussed in the sections 2.7.1 to 2.7.4, putting the emphasis on thermal hydrolysis as it is linked to the main objectives and deliverables of this research.

2.7.1. Biological pretreatment

2.7.1.1. Dual-stage anaerobic digestion

An enhanced hydrolysis process with biological pretreatment under aerobic or anaerobic conditions is achieved via an additional stage of bioreactor placed prior to the main AD process. The total process is known as dual-stage AD. One of the most commonly used dual-stage AD techniques is temperature-phased anaerobic digestion (TPAD) in which the feed (sludge) undergoes anaerobic thermophilic (55°C) or hyper-thermophilic (60-70°C) acid-phase digestion followed by a mesophilic (35°C) methane-phase digestion. There have been different configurations of TPAD which have been studied in sludge digestion, such as short thermophilic stage digestion (4, 8 and 12 h SRT) which occurs prior to mesophilic digestion (10 d SRT) (Roberts et al., 1999), two-stage thermophilic and mesophilic digestion (7, 14 and 20 d SRT) (Wahidunnabi & Eskicioglu, 2014), and a co-phase digestion process with internal circulation between thermophilic and mesophilic stages (Song et al., 2004). Figure 2-8 illustrates the schematic of a temperature co-phase anaerobic sludge digestion system. Studies have shown that including a thermophilic stage prior to the mesophilic digester increases the rate of organics removal and biogas production. According to a modeling study carried out by Ge et al. (2010), the enhanced performance of TPAD is mainly due to an improved hydrolysis coefficient rather than the increase of the substrate biodegradability (Ge et al., 2010).

The implementation of an aerobic system prior to the AD process is an alternative to the anaerobic pretreatment technique. The aerobic stage has been observed to improve the degradation of some of the compounds which are recalcitrant to the anaerobic biodegradation

(Subramanian et al., 2007). Hasegawa et al. (2000) observed a 50% increase in the biogas production through the use of a thermophilic aerobic sludge digestion system prior to the mesophilic AD (Hasegawa et al., 2000). Besides the enhancement in AD performance, previous studies have demonstrated the potential of TPAD in producing Class A biosolids for land application, although the energy requirement is much smaller than a fully thermophilic single- or dual-stage AD process (Han & Dague, 1997; Riau et al., 2010).

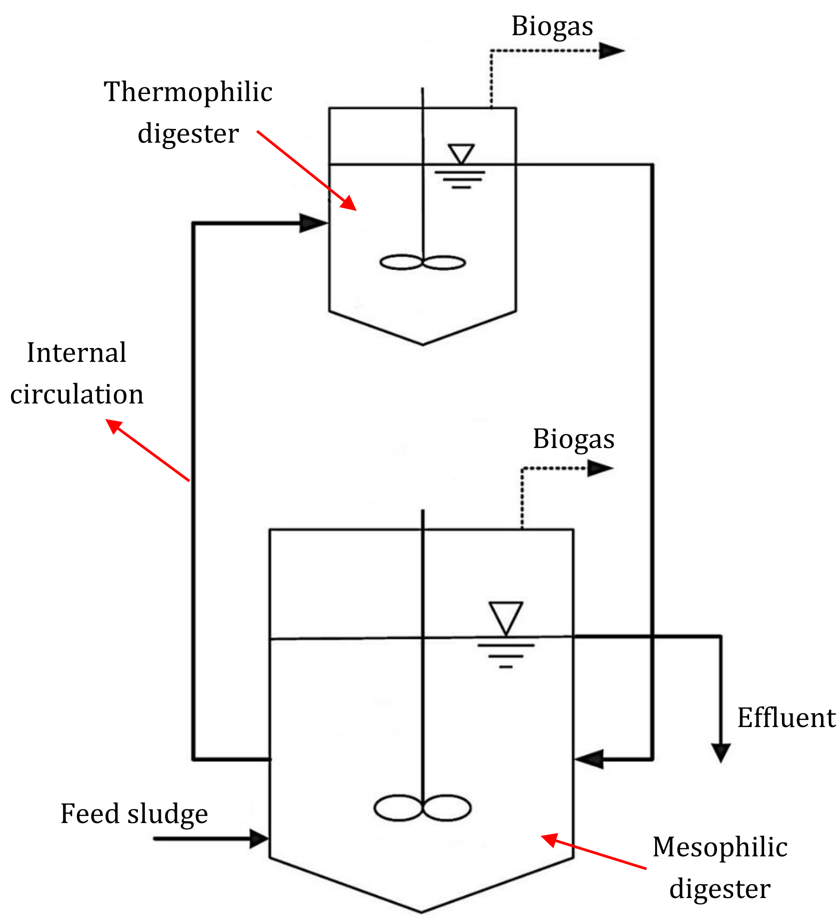


Figure 2-8. The schematic of a temperature co-phase anaerobic sludge digestion system with an internal circulation line between the digesters (adapted from Song et al. (2004))

2.7.1.2. Enzymatic pretreatment

The addition of specific bacteria strains or external enzymes to the feed sludge stream is another biological pretreatment technique which is called enzymatic pretreatment. The literature contains some studies which prove the positive effect of these pretreatment methods to accelerate the hydrolysis process, thus, improving the overall performance of the AD. For example, in a study carried out by Miah et al. (2004), a 200% improvement was observed in thermophilic sludge AD by adding *Geobacillus sp.* strain AT1 Culture (Miah et al., 2004). Enhanced hydrolysis was also achieved by directly adding the external enzymes (extracellular proteolytic enzymes) to sludge to facilitate the bacterial enzymatic catalyzed reaction (Guellil et al., 2001). However, compared to other sludge pretreatment (hydrolysis) methods, these pretreatments have not been very practical mainly due to the high operational cost of enzymes needed for large volumes of municipal sludge.

2.7.2. Chemical pretreatment

In chemical pretreatments, chemical compounds are externally added to the sludge stream. In general, chemical pretreatment requires low energy input and in most of the cases results in effective solubilization along with enhanced dewaterability of the digested sludge (Bordeleau & Droste, 2011). However, this process may result in the harming of beneficial microorganisms in the AD. Moreover, chemical addition can speed up equipment corrosion. Also, formation of some non-biodegradable intermediate compounds has been reported as a result of chemical hydrolysis especially with an increase in chemical dose (Neyens et al., 2003). Alkaline and oxidative techniques are the two common chemical pretreatment methods used for municipal sludge hydrolysis before AD (Neyens et al., 2003).

2.7.2.1. Alkaline hydrolysis

The addition of alkali, or an ionic salt of an alkali metal (water soluble base), has been shown to be an effective method for sludge hydrolysis. The order of effectiveness of alkaline pretreatment was reported as follows; NaOH > KOH > $Mg(OH)_2$ >$Ca(OH)_2$ (Kim et al., 2003). However, high doses of alkaline agents have been shown to have an adverse effect on the activity of the anaerobic microorganisms (Mouneimne et al., 2003). One of the techniques to prevent the overdose of the alkali agents is the combination of thermal and low-dose alkaline pretreatments called thermal-alkaline processes. The combined method also eliminates the necessity of reaching high temperatures for the same solubilization ratio compared to the sole thermal pretreatment (Carrere et al., 2010). The application of combined thermal-alkaline pretreatment (addition of 7 $\frac{g\ NaOH}{L\ of\ sludge}$ at a temperature of 121°C for a duration of 30 min) before two-stage acidogenic (HRT:6 d, 37°C)-methanogenic (HRT:12 d, 41°C) AD was evaluated by Park et al. (2005). The authors` proposed method achieved more than 520 $\frac{mL\ CH_4}{g\ VS{-}added}$ which was around 80% more than that of the control (non-pretreated) digester (Park et al., 2005). Increasing the pH of the sludge solution to 10 by adding 1.65 $\frac{g\ KOH}{L}$ and then increasing the temperature to 130°C for 60 min was also reported to increase the biogas production by 30% in mesophilic batch AD (Valo et al., 2004). In another study, Dogan and Sanin (2009) investigated the effect of thermal-alkaline pretreatment on performance of both batch and continuous-flow mesophilic AD. The pretreatment condition was the addition of NaOH to the sludge medium to maintain the pH above

12, then increasing the medium temperature using MW until 160°C and then holding for 16 min. The applied pretreatment resulted in 20% and 75% improvement in biogas production in batch and continuous-flow digesters respectively. In addition to the enhanced biogas production, more than 20% improvement in digestate dewaterability was obtained in the digester which was continuously-fed with pretreated sludge compared to the non-pretreated digester (Dogan & Sanin, 2009).

2.7.2.2. Oxidative hydrolysis

Peroxidation and ozonation are the two most common advanced oxidation processes (AOPs) used for sludge pretreatment. The oxidative hydrolysis methods disintegrate the sludge structure by the generation of hydroxyl radicals (OH^0), an extremely powerful oxidant (Appels et al., 2008). Hydrogen peroxide (H_2O_2) is one of the most intense oxidizers known. An enhanced oxidation power has been reported using the combined application of H_2O_2 with other treatment options such as ultraviolet (UV), ultrasound, and thermal processes. As an example, Eskicioglu et al. (2008) reported 11-34% removal of TS, COD, and total biopolymers by adding 1 g $\frac{H_2O_2}{g\ TS}$ to the TWAS. The increased solubilization ratio and also COD oxidation were observed when peroxidation was applied in combination with MW-elevated temperatures (>80°C). The authors found that the biogas production rates from H_2O_2-pretreated sludge samples were lower than that of non-pretreated and MW pretreated samples due to the formation of refractory compounds during the chemical reaction, resisting anaerobic biodegradation (Eskicioglu et al., 2008b). Ozone (O_3), another commonly used oxidant in drinking water disinfection, is a chemical commonly used for sludge disintegration. Although improved solubilization of sludge has been observed with increasing the dose of ozone (up to 0.2 $\frac{g\ O_3}{g\ TS}$), it was reported that above a critical dose, it diminishes the solubilization and methane potential due to the oxidation of the solubilized organics particles to CO_2 rather than facilitating the methane conversion (Carrere et al., 2010).

2.7.3. Mechanical pretreatment

Mechanical pretreatment involves the techniques that physically disintegrate and solubilize the sludge structure releasing the intracellular materials into the liquid phase. Ultrasonic (sonication) and high pressure homogenization are the two widely used mechanical pretreatment methods for disintegration of municipal sludge.

2.7.3.1. Ultrasound (sonication)

The term “ultrasound” is used to describe the cyclic sound energy (compression and expansion) with a frequency greater than 20 kHz which is outside the audible range. Typically, depending on the frequency range, ultrasound is divided into three regions, power ultrasound (20-100 kHz), high frequency ultrasound (100 kHz-1 MHz), and diagnostic ultrasound (1-500 MHz). The development and collapse of the cavitation bubbles is the most acceptable mechanism which has been proposed for the ultrasound sludge disintegration. When ultrasound waves propagate in sludge medium, it induces a cyclic succession of compression and expansion (rarefaction) phases (Wu et al., 2013). The compression cycle imposes a positive pressure by squeezing the molecules and cells together and the rarefaction cycle exerts a negative pressure by pulling the molecules apart. When the negative pressure (during the rarefaction cycle) exceeds the tensile strength of liquid, small micro bubbles (cavitation bubbles) are formed. The cavitation bubbles grow in successive cycles until they reach to an unstable diameter that collapse violently (Pilli et al., 2011). Upon collapsing, each of the bubble creates a hotspot and increases the local temperature and pressure up to 5,000 K and 2000 atm, respectively (Wu et al., 2013). Additionally, cooling rates have been observed to be as fast as 10^9 K/s (Wu et al., 2013).

Most of the studies which investigated the effect of ultrasound sludge pretreatment before AD have experienced an increase in sludge solubilization ratio, VS removal, and biogas production depending on the input pretreatment variables. The pretreatment variables include power level, duration, sludge type, sludge TS and VS content, and the frequency used. In lab-scale, the applied specific energies in the range of 1000 to 16000 $\frac{KJ}{kg\ TS}$ resulted in a positive net energy balance. The biogas improvement in the range of 25-140% and 10-45% were reported as a result of pretreatment under batch and semi-continuous AD, respectively (Salsabil et al., 2009). The full-scale application of ultrasound sludge pretreatment also turned out to be a successful practice. For example, Xie et al. (2007) evaluated the energy ratio between net energy generation and input energy for a full-scale ultrasound pretreatment before municipal sludge AD. The authors reported a 45% improvement in biogas production with an energy ratio of 2.5 (Xie et al., 2007). In another study, Barber (2005) investigated the performance of several full-scale ultrasound plants in Germany, Austria, Switzerland, Italy and Japan. It was concluded by the author that on average, 20-50% improved VS reduction and biogas production have been achieved using full-scale ultrasound sludge pretreatment system (Barber, 2005).

2.7.3.2. High pressure homogenization

High pressure homogenization (HPH) is a relatively new mechanical technique used for sludge disintegration, although this technique is not completely original as it has been used for lysing cells in microbiology. This technique has aroused attentions during the last decade because of its low investment and operational costs, low energy consumption, high lysis efficiency, and positive effect on AD performance (Rai & Rao, 2009; Stephenson et al., 2005). The HPH sludge pretreatment mechanism involves applying extremely high pressure (100-1500 bar) on the sludge structure and then creating a strong depressurization (Sutradhar et al., 2013). Figure 2-9 shows the schematic of the main components involved in HPH. Using this system, the sludge is first pressurized through a narrow orifice via a positive displacement air pump (Sutradhar et al., 2013). This creates a shear force on the cell membranes in the sludge. When the external pressure exceeds the cells membrane resistance, the cell membrane ruptures. The sludge sample then goes under an sudden pressure drop by an instant expansion releasing the intracellular substances in to the liquid phase (Zhang et al., 2012).

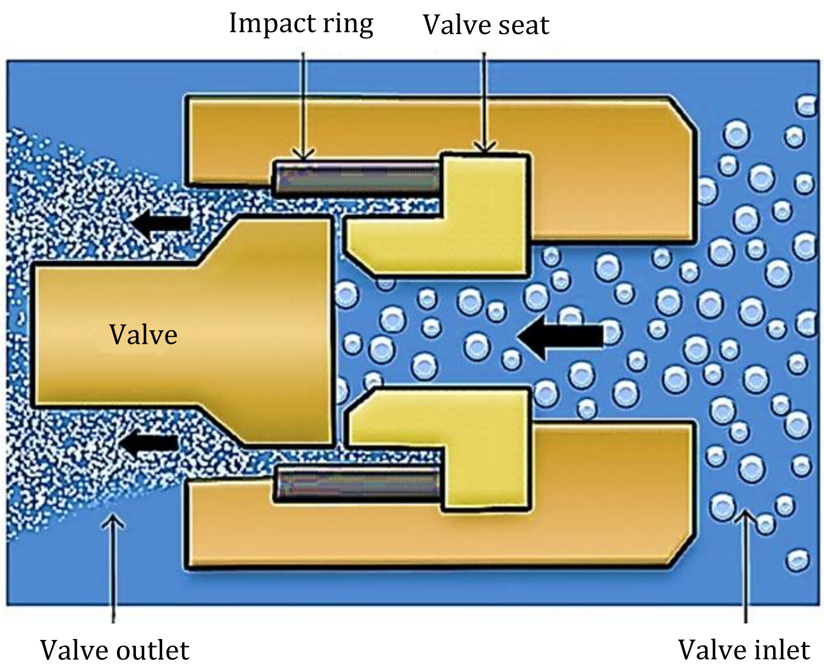


Figure 2-9. The schematic of the high-pressure homogenization process (adapted from Sutradhar et al. (2013))

Beside the lab-scale studies, the HPH has been successfully implemented in full-scale. For example, in an industrial application, more than 30% and 23% improvement in biogas production and TS removal was achieved when a fraction of the digested sludge was pressurized under 150 bar and re-enter to the digester (Onyeche, 2007). MicroSludge is a modified version of HPH technology patented by Paradigm Environmental Technologies Inc. (Vancouver, BC). In MicroSludge, the sludge is first treated with chemicals (i.e. NaOH) to increase the pH above 10. The chemical treatment softens the sludge structure and protects the homogenizing valve from mechanical damages. Chemical addition also weakens the cells walls, making them more susceptible to be ruptured under HPH. In a municipal WWTP in Los Angeles where the primary and secondary sludge are mixed with a ratio of 32/68 (%, w/w) before entering the digester, the application of HPH on WAS improved the solids reduction by 7% (Stephenson et al., 2007).

2.7.4. Thermal pretreatment

Thermal hydrolysis is one of the most common pretreatment methods applied for advanced AD at the full-scale. Thermal energy causes the disintegration of sludge structure which results in the release of intracellular materials for acid and methane forming bacteria to utilize at a faster rate. Thermal hydrolysis was first used in 1978 to enhance the sludge dewaterability (Haug et al., 1978). Since then, it has been widely used and reported to be effective in sludge disintegration and also improving the performance of sludge AD in numerous studies (Eskicioglu et al., 2009; Li & Noike, 1992; Park et al., 2004; Park & Ahn, 2011a; Toreci et al., 2011). Thermal hydrolysis can be achieved via CH, MW, or RF heating techniques. The detailed discussion about different thermal pretreatment techniques is provided in the sections 2.7.4.1 to 2.7.4.4.

2.7.4.1. Conductive (conventional) heating

In CH, the heat transfer is occurred by the movement of free electrons and microscopic collision of particles (atoms and molecules). The conduction of heat takes place in all phases of matter including solids, liquids, and gases, however, it is often the dominant heat transfer mechanism in liquids and solids. Through conduction, the energy is transferred from more energetic to less energetic particles due to the energy gradient (Incropera et al., 2007). A kitchen oven is a simple example of CH where the heating coils are the source of energy. As shown in Eq. (2-5), the one-dimensional conductive heat transfer rate (q) through a layer is proportional to the layer surface area (A) whereas the temperature difference across the layer (ΔT) is inversely proportional to the thickness of the layer (Δx). Figure 2-10 illustrates the schematic of the conductive heat transfer.

$$Conductive\ heat\ transfer\ rate \propto \frac{(surface\ area)(temperature\ difference)}{(thickness)} \tag{2-5}$$

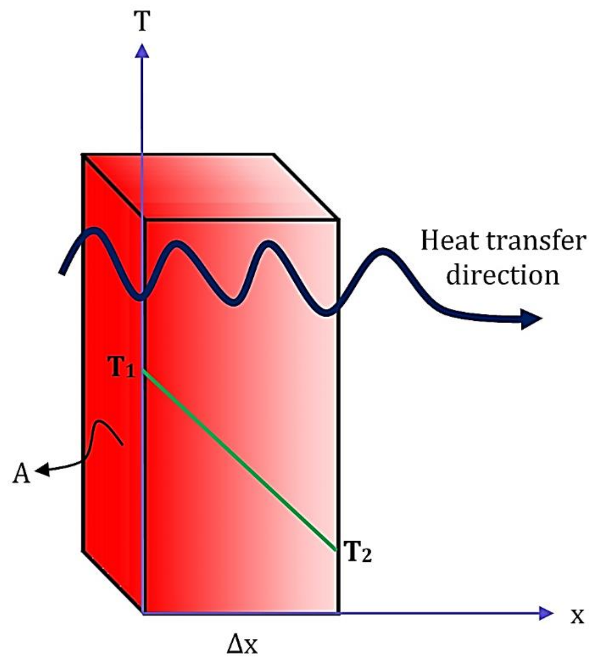


Figure 2-10. The schematic of the conductive heat transfer

The mathematical form of the Eq. (2-5), presented in the Eq. (2-6), is known as Fourier's law (Fourier, 1955). In Eq. (2-6), q is the conductive heat transfer rate (J/sec or W), dT/dx is the temperature gradient in the direction of heat flow (°C/m or K/m), A is the area perpendicular to the heat flow direction (m^2), and k is the thermal conductivity of the material through which the heat is transferred (W/m.°C or W/m.K).

$$q = -kA\frac{dT}{dx} \tag{2-6}$$

Thermal conductivity is a unique characteristic of a material and is a function of temperature. Under steady-state conditions (shown in Figure 2-10) and under a linear temperature gradient, Eq. (2-6) can be simplified as follows:

$$q = -kA\frac{T_2 - T_1}{\Delta x} \tag{2-7}$$

In general, as a result of enhanced molecular and atomic movements, the thermal gradient of a medium increases as the temperature increases. Figure 2-11 compares the change in the thermal conductivity of pure water and DWSC as a function of temperature. As per Figure 2-11, for a given temperature, the thermal conductivity of DWSC is much lower than that of water especially for the temperatures below 150°C. Therefore, between 0-200°C, the application of conductive heating for DWSC is not as effective as for water (Song et al., 2014).

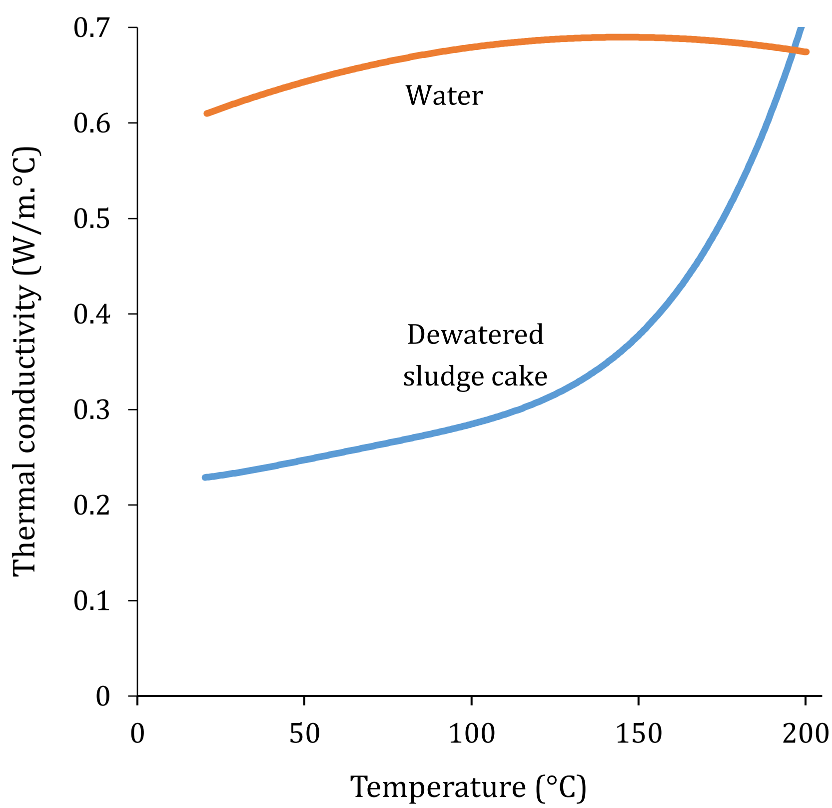


Figure 2-11. The thermal conductivity of water and dewatered sludge cake (DWSC) as a function of temperature (adapted from Song et al. (2014))

Figure 2-12 shows the change in the thermal conductivity of WAS and digested PS as a function of the sludge TS concentration. The higher the TS concentration of the sludge is (the thicker the sludge is), the lower the thermal conductivity will be (Vesilind & Martel, 1989). On the other hand, in order to reduce the specific energy consumption during pretreatment (per g TS-added), it is desirable to thicken the sludge (dewater) as much as possible before the pretreatment. This however decreases the thermal conductivity requiring more energy for increasing the temperature of the thickened sludge.

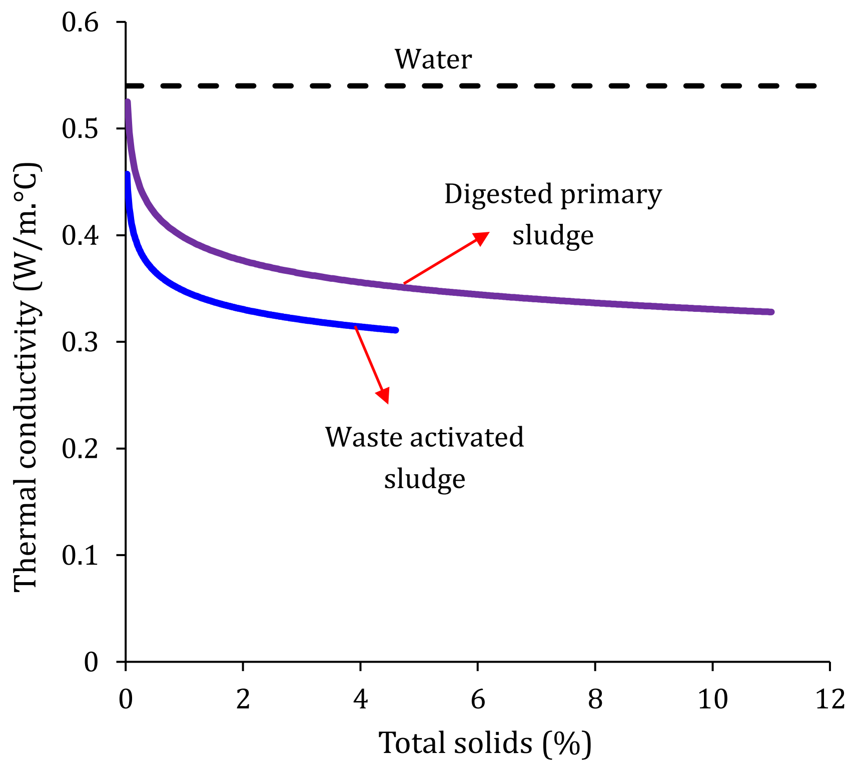


Figure 2-12. The change in the thermal conductivity of sludge with respect to the total solids concentration (adapted from Vesilind & Martel (1989))

Besides the higher energy consumption of the CH at elevated TS concentrations which is due to the decreased thermal conductivity, obtaining uniform heating throughout the load is another challenge in CH methods. This is because significant thermal gradients can be created by the distance between the heating source and the internal parts of the load (Tyagi & Lo, 2013). Decreasing the heating rate is a strategy to reduce the thermal gradient. However, it increases the energy loss due to the longer pretreatment time. CH methods also suffer from a transient thermal lag between the applied source temperature and the temperature inside the medium being heated (Hosseini Koupaie & Eskicioglu, 2015a). The lag-phase can be considerably high at full-scale applications by using large volume heating vessels which limits the maximum effective ramp rate of heating.

2.7.4.2. Microwave heating (dielectric heating)

As illustrated in Figure 2-13, unlike CH in which the heat is transferred through conduction and as a result of the microscopic movement and/or collision of the particles inside a medium, in electromagnetic (EM) heating, the electric field directly interacts with the medium's particles, increasing its molecular kinetic energy (Figure 2-13 (b)).

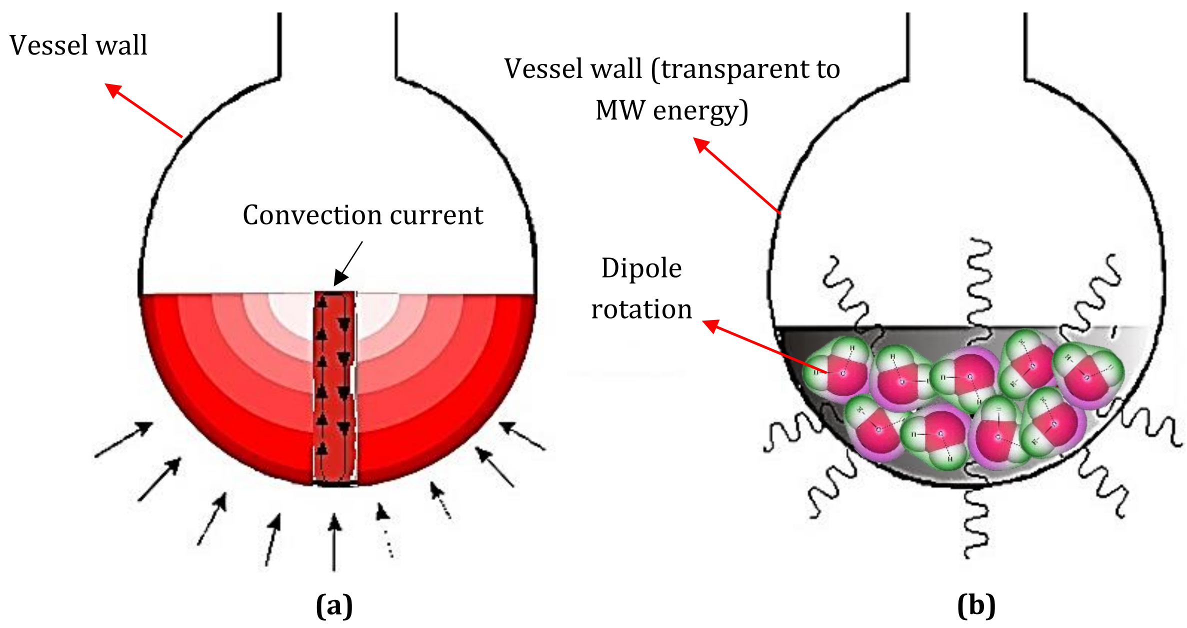


Figure 2-13. The schematic of (a) conductive heating, and (b) microwave heating (redrawn from Kingston & Jassie (1988))

In the electromagnetic spectrum, the MW irradiation occurs at a frequency of 300 MHz to 300 GHz corresponding to the wavelengths of 1 m to 1 mm, respectively (Figure 2-14). A frequency of 2.45 GHz is commonly used in domestic and industrial MW ovens because they are designed for processing food (Tyagi & Lo, 2013). Moreover, the water found in foods is a good absorbent of MW energy at this frequency (Tyagi & Lo, 2013).

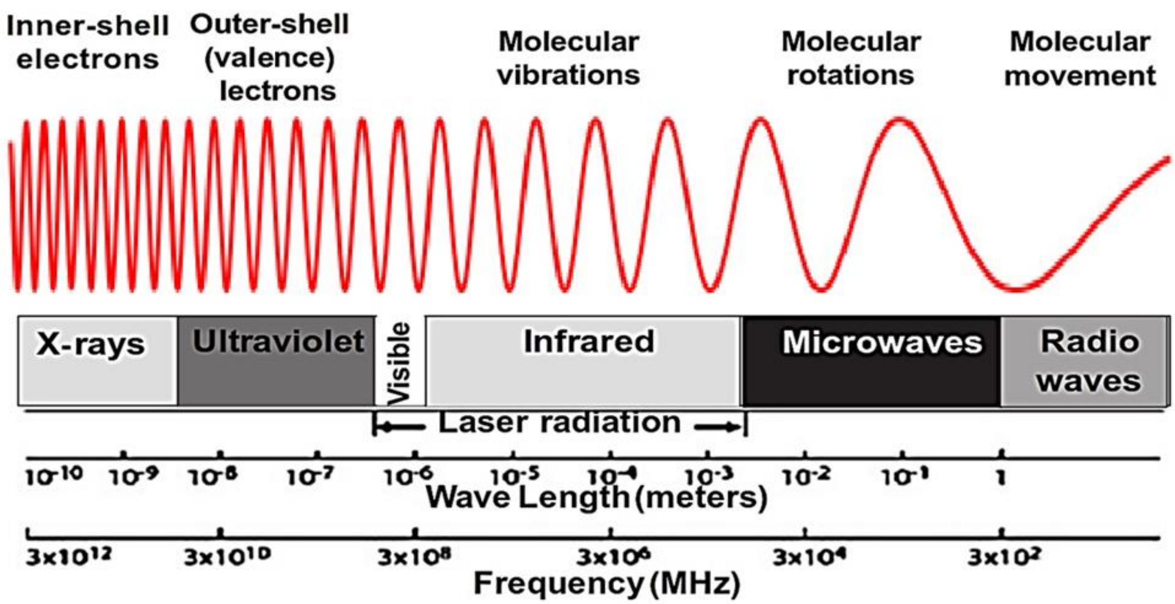


Figure 2-14. The electromagnetic spectrum (redrawn from Kingston & Jassie (1988))

In general, the EM heating results from the interaction of the chemical constituents of the load with the EM field. The two major mechanisms involved in EM heating are molecular friction which is primarily due to the disruption of weak hydrogen bonds associated to the dipole rotation of the free water molecules (dipolar loss) and collision of free ions due to their movement as a result of interaction with EM field (conduction loss) (Kingston & Jassie, 1988). As shown in Figure 2-15 (a), when a dielectric material (such as water or WAS) is exposed to an electric field at a low frequency, the dipoles have enough time to follow the external electric field oscillation which results in the storage of the external source energy in the dielectric (polarization effect). The permittivity (ε) indicates the ability of a dielectric to resist an electric field. The higher the permittivity of a medium, the more it is polarized in the presence of an electric field. The relative static permittivity (ε_r) known also as dielectric constant is defined as the ratio of the capacitance of a capacitor made of a dielectric to the capacitance of an identical capacitor which has vacuum as a dielectric. The dielectric constant is measured under a zero frequency electric field (static electric field). The ε_r can be calculated as follows:

$$\varepsilon_r = \frac{\varepsilon}{\varepsilon_0} \tag{2-8}$$

where, ε_r and ε are the relative and absolute static permittivity (F/m), respectively and ε_0 is the vacuum permittivity which has a value of $8.854187817 \times 10^{-12}$ F/m.

In a more general form (under a time-varying EM field), the permittivity of a material is a frequency-dependent complex number defined as follows:

$$\varepsilon_r(\omega) = \varepsilon_r'(\omega) - j\varepsilon_r''(\omega) \tag{2-9}$$

where, $\varepsilon_r(\omega)$ is the relative complex permittivity (at a frequency of ω), $\varepsilon_r'(\omega)$ is the real part of the permittivity representing the ability of the medium to store external energy (polarization capability), $\varepsilon_r''(\omega)$ is the imaginary part of the permittivity linked to the dissipation of energy through the medium in the presence of an electric field, and j is the imaginary unit.

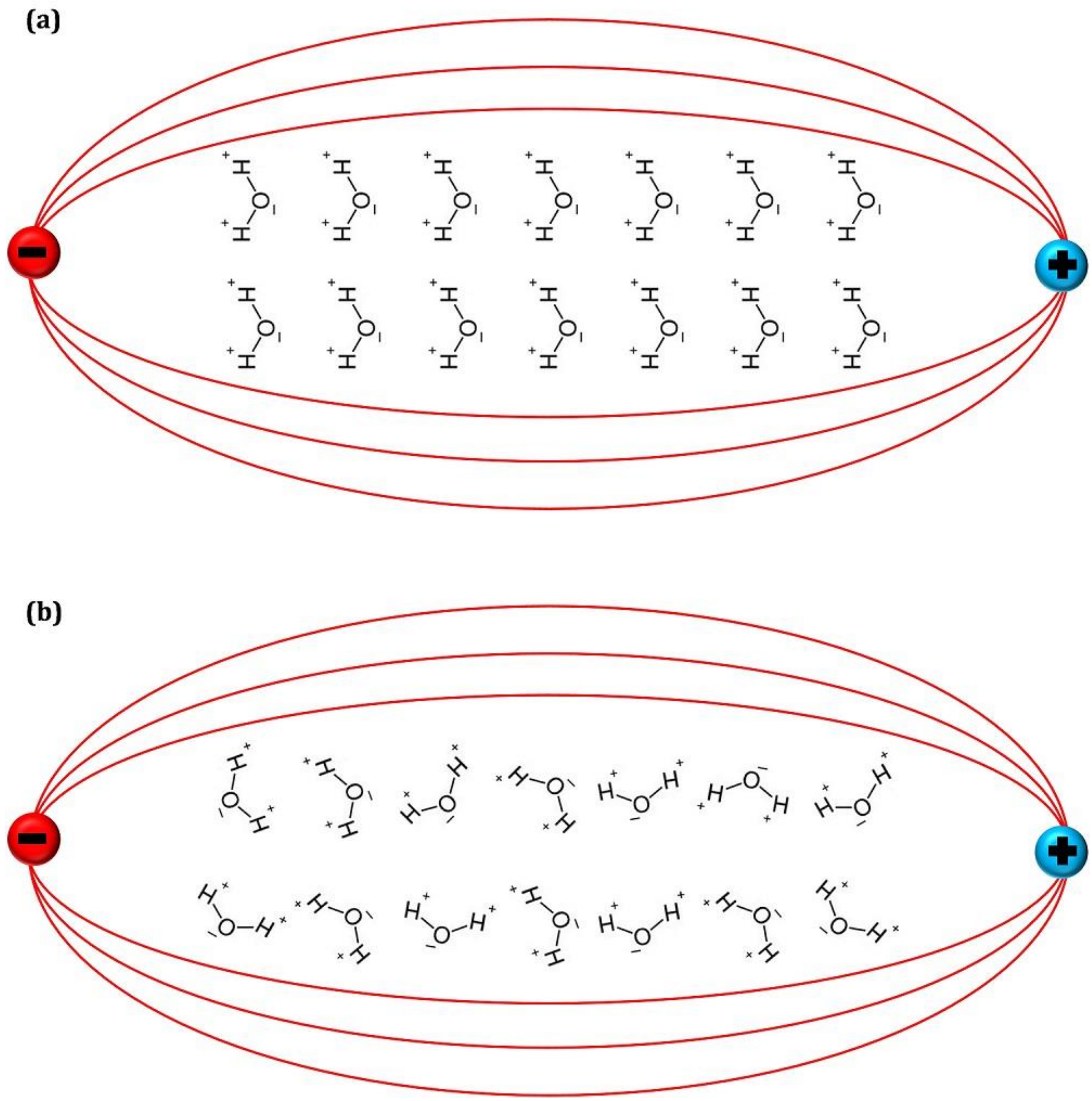


Figure 2-15. Response of the water molecules to the electromagnetic field under (a) low frequencies, and (b) high frequencies

While at low frequencies, the dipoles freely follow the field direction. When a high-frequency electric field is applied (i.e., MW>1 GHz), the dipoles cannot maintain their original positions during the field oscillation and therefore the dipolar polarization lags the external electric field (Figure 2-15 (b)). The term "lag" means the energy from the applied electric field is absorbed by the dielectric and dissipated as heat. If the frequency increases further, the reorientation polarization charge starts to significantly lag behind the phase of the electric field, resulting in a significant decrease of the dielectric constant and a rise in the loss factor (ε''). This causes even faster dissipation of the energy as heat through the dielectric (relaxation loss) (Metaxas, 1996). The loss factor (ε'') represents the amount of the energy lost (absorbed) by the medium and is dissipated as heat. The higher the loss factor of a dielectric, the more effective it can be heated via an electric field (Kingston & Jassie, 1988). The rest of the energy (which is not absorbed), penetrates to deeper parts of the dielectric. Because the EM waves propagate through the entire dielectric volume, the term volumetric heating is also used for EM heating.

Considering the above explanations, at MW range, the EM heating is mainly dominated by the dipole rotation mechanism (dipolar loss mechanism). For many polar materials, the loss factor (ε'') peak is broad and occurs at around 800-3000 MHz. This explains the reason why two industrial, scientific and medical (ISM) frequencies for heating are located in this band (915 and 2450 MHz) (Metaxas, 1996). It is worth noting that the bulk (free) water has a loss factor peak (relaxation peak) at 18-22 GHz, however, the bound water in most of materials has a peak shifted towards the lower frequencies. The materials can be classified into the following three categories depending on their behavior in response to an EM field:

- Absorbers: which absorb the majority of the EM energy and have a high loss factor (i.e., water).
- Transparent: which pass the EM waves and have a very low loss factor (i.e., Teflon).
- Reflectors: which reflect the EM waves (i.e., metal).

There are several limitations of the EM heating at high frequencies (i.e., MW range). First of all, the penetration depth is short at high frequencies. Therefore, it is difficult to achieve effective in-depth heating. It is worth noting that, the concept of half-power depth (for a specific material at a given frequency) is one of the commonly used methods to determine the penetration depth. The half-power depth is defined as the distance from the surface of the medium at which the EM power is reduced to one-half compared to the power at the surface (Kingston & Jassie, 1988). Non-uniform heating as well as the creation of hot and cold spots throughout the medium (due to the short wavelength at high frequencies) are the other drawbacks of MW heating. Moreover, design and manufacturing of an efficient high power EM generator at high frequencies can be very costly.

2.7.4.3. Conductive heating vs. microwave pretreatment

Table 2-4 summarizes the previous studies that attempted to compare CH and MW irradiation for sludge pretreatment with limited success. Despite the numerous studies done in the field, there is still no clear answer to whether there are any advantages of choosing one of the two commonly used thermal pretreatment methods over another (MW or CH) for disintegration of waste sludge. One of the biggest challenges in comparing these thermal pretreatments has been the lack of a controlled system allowing to apply CH and MW pretreatments under identical thermal profiles (maintaining both final sample temperature and heating/cooling rate identical between MW and CH throughout the pretreatment). The lack of such a controlled pretreatment condition can explain the contradictory findings of published research that have dealt with comparing the effects of CH and MW pretreatments on sludge disintegration and AD performance

(Climent et al., 2007; Eskicioglu et al., 2006; Park & Ahn, 2011a; Pino-Jelcic et al., 2006; Sólyom et al., 2011).

In a study, no significant differences in ultimate biogas production between MW-pretreated and non-pretreated sludge was observed, while CH at 70°C improved biogas production by 58% (Climent et al., 2007). In the same study, the high temperature (110-130°C) CH pretreatment was applied using an autoclave for the duration of 20-30 min. However, low temperature (70°C) CH was done using a 500 ml beaker placed in a thermostatic bath for significantly higher duration (9-72 h) that resulted in higher solubilization and AD enhancement than the MW pretreatment performed up to the final temperature of 100°C for a much shorter period of 3 to 10 min (Climent et al., 2007). Another study conducted by Eskicioglu et al. (2006) reported higher improvement in mesophilic batch biogas production when the sludge was pretreated at 96°C using a water bath (simulating CH) compared to the MW-pretreated sludge with a kitchen-type of MW oven at 2.45 GHz. Similarly, in this study, although the final temperature reached was the same by both thermal pretreatments, the total duration of heating was a lot longer for CH (80 min) compared to MW irradiation (5 min) which resulted in significantly higher solubilization and digestion performances. One year later, in another study, the authors reported improved biogas production from MW-pretreated samples with an industrial MW oven over CH-pretreated samples in a water bath under identical heating profile (Eskicioglu et al., 2007b). However, due to the limitation of the CH system used (water bath), this study was constrained to the temperatures of under boiling point of water (50-96°C) as well as uncontrolled heating rates (Eskicioglu et al., 2007b). In a study carried out by Park & Ahn (2011a), 53% and 33%

improvement in biogas production was achieved when MW and CH pretreatments were used, respectively. A 6.3% biogas production enhancement was reported by Pino-Jelcic et al. (2006) for microwaved/digested sludge over the conventionally heated/digested sludge. Both aforementioned studies implemented CH and MW pretreatments with different heating profiles although same final desired temperatures were reached. More recently, other researchers (Kuglarz et al., 2013) used a laboratory MW unit and a closed vessel equipped with a heating coil as CH for pretreatment of secondary sludge. According to the results of this study, the authors finally concluded that MW is superior over CH in terms of both sludge solubilization and biogas production. The authors' conclusion is however questionable, as the MW and CH pretreatments were not performed under identical heating rates. Although, in all these studies, one of the thermal pretreatment methods (MW or CH) was reported to be superior over another, similar methane yield was achieved using MW-pretreated and CH-pretreated secondary sludge in a study carried out by Sólyom et al. (2011).

Considering these results from previous studies, it is postulated in the current research that if all the pretreatment parameters are controlled and kept identical, both the MW (at 2.45 GHz) and CH pretreatment methods achieve similar results (*Hypothesis II*), which is going to be evaluated as part of this research.

Table 2-4. The studies comparing conductive heating and microwave sludge pretreatment [a]

Pretreatment systems used	Sludge type	Pretreatment conditions/ Digester type	Main results	Source
MW: 1250 W/2.45 GHz household oven CH: Concentric ring water bath	TWAS (TS=5.9%)	Final temperature: 96°C MW heating time: 5 min CH heating time: 80 min Digester type: batch mesophilic anaerobic digester	Soluble concentration of COD and sugar increased 361% and 308%, respectively after CH. After MW, their concentration however increased only 143% and 15%, respectively. CH and MW pretreatments resulted in 475% and 211% higher ultimate biogas productions, respectively compared to control digester. Overall k values were 0.37, 0.27 and 0.22 d^{-1} for control, MW-pretreated and CH-pretreated digesters, respectively. The average of substrate utilization rate (mg $COD_{removed}$/L.d) increased 136% and 75% as a result of CH and MW pretreatments, respectively.	(Eskicioglu et al., 2006)
MW: 1000 W/ 2.45 GHz household oven CH: 500 ml glass beaker warmed using a hotplate	Mixed sludge	Final temperature: 65°C MW heating time: 110 s CH heating time: 960 s Digester type: continuous mesophilic anaerobic digester	Up to 45°C temperature difference between the surface (95°C) and the bottom (50°C) of the MW-pretreated samples. Application of MW pretreatment resulted in 16.4% and 6.3% increase in biogas production compared to the control and CH/digested sludge, respectively. Volatile solids removal efficiency of 53.9, 51.3 and 49.0 was achieved in MW-pretreated, CH-pretreated and control digesters, respectively. The fecal coliforms log inactivation for MW/digested, CH/digested and control sludge was 4.2, 2.9 and 1.5, respectively. The log inactivation of *Salmonella* spp. was greater than 2, 1.9 and 1.1 for MW/digested, CH/digested and control samples. MW pretreatment was found to be more effective than CH pretreatment in improving the digestate dewaterability.	(Pino-Jelcic et al., 2006)

Table 2-4. The studies comparing conductive heating and microwave sludge pretreatment a (cont'd)

Pretreatment systems used	Sludge type	Pretreatment conditions/ Digester type	Main results	Source
MW: 1250 W/ 2.45 GHz programmable high pressure lab station CH: 230 mL concentric ring water bath with manual mixing	TWAS (TS=4.6-5.5%)	Final temperature: 50, 75 and 96°C MW-1 heating time: 3 min MW-2 and CH heating time: 20, 40 and 60 min Digester type: batch mesophilic anaerobic digester	Similar effect of MW and CH pretreatments on COD and biopolymers solubilization of waste activated sludge. Application of MW pretreatment at final temperatures of 50, 75 and 96°C resulted in 6%, 6% and 13% higher ultimate biogas production than similarly CH-pretreated samples, respectively indicating MW athermal effects. MW pretreatment resulted in greater level of COD solubilization due to longer pretreatment times. Linear relation between MW final temperature and level of hydrolysis was observed.	(Eskicioglu et al., 2007b)
MW: Modified 1000 W/ 2.45 GHz household oven CH: 500 W electric mantle	Centrifuged secondary sludge (TS=7-8%)	Final temperature: 85°C MW heating time: 1.2 min CH heating time: 10 min Digester type: batch mesophilic anaerobic digester	The COD solubilization results were 3.27 and 5.26 after MW and CH pretreatments, respectively. MW and CH pretreatments resulted in 208 and 203 $\frac{mL}{g\ VS\text{−}added}$ methane yield respectively. No evidence of MW athermal effect on sludge anaerobic biodegradability was found as similar methane yield was measured using MW and CH pretreatments.	(Sólyom et al., 2011)
MW: A cavity resonator equipped with a 700 W magnetron at 2.45 GHz CH: Laboratory heater	Dairy-industry (TS=31.8%) and meat-industry (TS=27.2%) primary sludge	Final temperature: 95°C CH heating time: 10 min CH holding time: 60 min MW holding time: 10-40 min Digester type: batch mesophilic anaerobic digester	Enhancement of the maximum and the initial rate of MW-pretreated sludge degradation in comparison with the CH-pretreated sludge. Significant effect of the applied MW power level on sludge volatilization as the increased power level transformed the solids into a volatile form more effectively. Relative to control in biogas yield of the MW-pretreated/digested samples was significantly higher than that of the CH-pretreated/digested samples.	(Beszédes et al., 2011)

Table 2-4. The studies comparing conductive heating and microwave sludge pretreatment a (cont'd)

MW: 1600 W high pressure lab station CH: 500 mL Erlenmeyer on a magnetic hot plate stirrer	Mixture of primary (TS=13.7%) and secondary sludge (TS=19.4%) at 1:1 volumetric ratio	Final temperature: 80°C MW and CH heating time: 4 min CH and MW holding time: 5 min Digester type: semi-continuous mesophilic anaerobic digester	The COD solubilization ratio of CH-pretreated and MW-pretreated samples were achieved 10.7 and 12.4, resulting in 2.7 and 3.2 times higher than that of the control, respectively. The VS and COD removal efficiencies in the CH-pretreated digesters were 37.4 and 47.9, respectively and 39.8 and 50.7 in the MW-pretreated digesters, respectively. The dewaterability of the CH-pretreated sludge was better than that MW-pretreated samples. 33% and 53% relative (to control) improvement in daily biogas production was obtained using CH and MW pretreatments, respectively.	(Park & Ahn, 2011a)
MW: 1000 W/ 2.45 GHz household oven CH: Electrical hot plate equipped with a mixer	Primary (TS=2.35) and TWAS (TS=2.2%)	Final temperature: 72-93°C Heating time: 7.5-13 min Heating rate was almost identical (but limited) between MW and CH Digester type: batch mesophilic	The COD solubilization ratio of primary and waste activated sludge was achieved 14.3% and 3.0%, respectively with negligible differences between MW and CH pretreatments. MH and CH revealed similar effects on the anaerobic biodegradability of primary and waste activated sludge as the differences between anaerobic methane yield of MW-pretreated/digested and CH-pretreated/digested samples were not statistically significant.	(Vergine et al., 2014)
MW: 800 W/ 2.45 GHz household oven CH-1: thermostatic water bath CH-2: autoclave	Secondary sludge (TS=3.8-4.9%)	MW temperature: 100°C CH temperature: 70 and 110-134°C MW heating time: 180-600 s Digester type: batch thermophilic anaerobic digester	The ratio of soluble to total VS in the sludge samples pretreated with MW and high-temperature CH was 3 and 11 times more than that of non-pretreated sludge samples, respectively. No difference in thermophilic biogas production was observed between the digesters fed with high-temperature CH-pretreated and MW-pretreated sludge samples. Thermophilic biogas production increased 50% when low temperature (70°C) CH pretreatment was applied.	(Climent et al., 2007)

Table 2-4. The studies comparing conductive heating and microwave sludge pretreatment a (cont'd)

MW: 1000 W/ 2.45 GHz household oven CH: Water batch	1- PS (TS=6%), WAS (TS=4%) and anaerobic digester sludge (TS=3%) 2- Isolated fecal coliforms from primary and secondary sludge	Final temperature: 20-100°C Sludge heating time with MW: 30-120 s Sludge heating time with CH: 1.9-4.8 min Isolated fecal coliforms heating time with MW: 7-23 s Isolated fecal coliforms heating time with CH: 58-866 s	MW irradiation for 90 s at 65°C led to complete destruction of coliforms, however CH required about 4.8 min at 100°C The CH led to increased ETS and β-Galactosidase activities of fecal coliforms as temperature increased from 20°C to 57°C, due to fecal coliform growth instead of inactivation. However, MW irradiation resulted in decreased activity. Between 57-68°C, MW irradiation resulted in a greater decrease in bacterial activity than CH. Optical densities of MW-irradiated samples diminished more at 260nm compared to that of samples exposed to CH indicating that MW disrupted DNA in fecal coliform cells at lower temperatures than CH.	(Hong et al., 2004)
MW: 900 W/ 2.45 GHz oven CH: Closed steel vessels equipped with a 900 W heating coil	TWAS (TS=5.3%)	Final temperature: 100°C MW heating time: 75-429 s (for 700 W) and 58-311 s (for 900 W) CH heating time: 95-493 s Anaerobic digester type: batch mesophilic	CH sludge pretreatment exhibited almost half solubilization for the same range of temperature compared to the MW irradiation. The concentrations of NH_4^+ and PO_4^{-3} in the MW-pretreated sludge were 2-3 times higher than that of CH-pretreated. CH pretreatment resulted in 50% less methane yield compared to that of MW pretreatment.	(Kuglarz et al., 2013)
MW: 700 W/ 2.45 GHz monomode unit CH: Laboratory heater	Dairy-industry (TS=42.8%) primary sludge	Final temperature: 95°C MW heating time: 10-40 min	MW pretreatment increased the sludge BOD_5/COD ratio 2 times more than that of CH pretreatment. MW pretreatment enhanced more effectively the anaerobic biogas production than CH pretreatment.	(Beszédes et al., 2009)

[a] MW: microwave; CH: conductive heating; TWAS: thickened waste activated sludge; COD: chemical oxygen demand; BOD: biochemical oxygen demand; TS: total solids; VS: volatile solids

2.7.4.4. Radio frequency (ohmic) heating

Since 1940 when the RF heating was first introduced for industrial applications (Wang et al., 2011), it has been widely applied in wood drying, food industries and agricultural processes (Moyer & Stotz, 1947), baking processes (Rice, 1993) and meat production (Laycock et al., 2003). RF heating has also been used as a sterilization technique. For example, successful application of RF heating was reported in killing navel orange worms in walnuts (Wang et al., 2001) and reducing the microbial activity in fruit juices (Geveke et al., 2007). A frequency of 27.12 MHz as one of the ISM (Industrial scientific and medical) allocated frequencies has been used in most of the RF heating applications, so far. However, the RF heating can also be applied using the two other ISM allocated frequencies, 40.68 MHz and 13.56 MHz (Domdouzis et al., 2007).

For non-magnetic materials, permittivity (ε) and conductivity (σ) are two fundamental parameters used when determining the dominant heating mechanism. At high frequencies, the atomic bonds in the molecular structure of the materials obstruct free movement of charges (electrons) or ions (atoms or molecules with net positive or negative charge). Therefore, the polarization occurs due to the local realignment of bound electrons (i.e., bound electron in water molecules). Now, if an EM field at high enough frequency is applied, the polarization charge cannot follow the field direction resulting in dissipation of energy as heat (dipole loss).

At low frequencies (RF band), the conductivity of the medium plays a significant role in the heating mechanism. The conductivity represents how freely the charges or ions can move (drift) when an electric field is applied to a material. The higher the conductivity of a material, the easier the charges (or ions) can move in response to an electric field. When a material is exposed to an electric field, the electric field applies a force on free charges (and ions in case the medium is electrolyte) resulting in their drift. The force applied to a particle of charge "q" can be calculated per Eq. (2-10). In this equation, F is the applied force in newton (N), q is the charge in coulomb (C) and E is the electric field with the unit of N/C or V/m.

$$F = qE \tag{2-10}$$

The movements of charges and ions create a conduction flow which is called electronic current flow and ionic current flow depending on whether the electric field is applied to a conductor or an electrolyte (i.e., salt water), respectively. Figure 2-16 schematically shows the ionic current flow through salt water electrolyte under a static (DC) electric field. As illustrated in Figure 2-16, in the case of an ionic current flow, electrochemical reactions occur at the electrodes along with a net movement of mass (Na^+ and Cl^-) across the medium (Metaxas, 1996). Therefore, as time passes, more ions are involved in electrochemical reactions (in the presence of enough DC power) which results in a decrease in the current flow. Finally, if there is no way to regenerate the ions, the current flow is stopped. The electrochemical reactions which result in a change in the potential of the electrodes is called electrode polarization.

The collision of atoms and molecules inside a conductive medium in response to an electric field generates heat. Because the source of the generated heat is the energy that is lost through the internal (molecular) resistance of the medium, this method of heating is called ohmic heating. The rate of the heating depends on both conductivity and the magnitude of the electric field. Figure 2-17 schematically illustrates how ohmic heating occurs in salt water solution under a DC electric field (Metaxas, 1996).

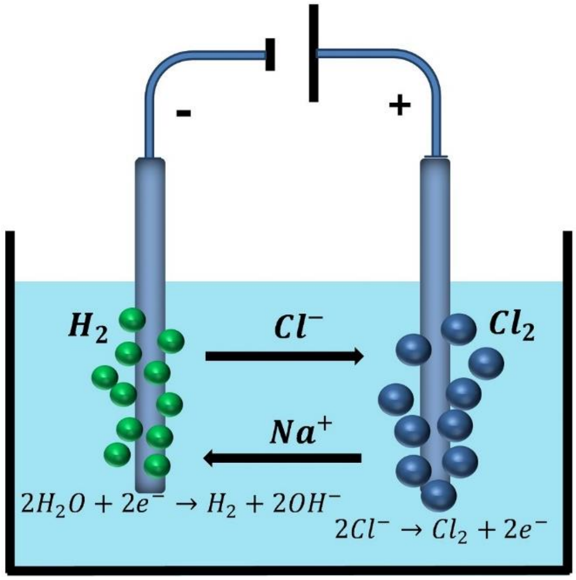


Figure 2-16. The schematic of ionic current flow through salt water electrolyte under static (DC) electric field

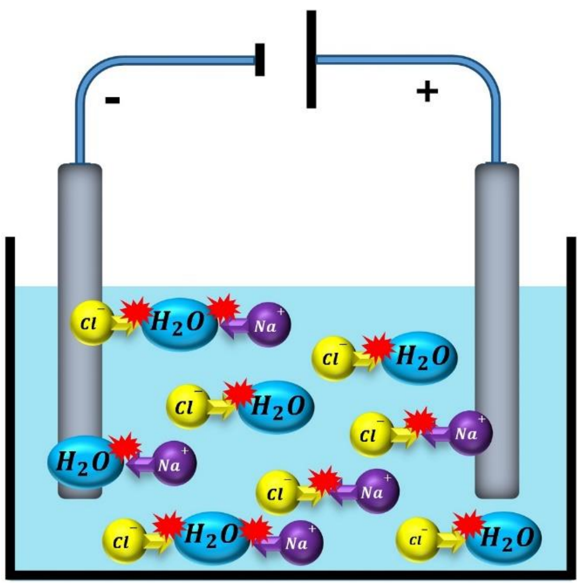


Figure 2-17. Collision of atoms and molecules during ionic current flow (ohmic heating) under static (DC) electric field

2.8. ELECTRICAL CHARACTERISTICS OF WASTE ACTIVATED SLUDGE

Figure 2-18 (a) shows the schematic of WAS floc structure. The scanning electron microscopy images of the WAS adapted from Leppard et al. (2003) are also shown in Figure 2-18 (b). As shown in Figure 2-18, WAS is a heterogeneous mixture of different microorganisms (i.e., bacteria), organic and inorganic compounds that adhere together with three-dimensional gel-like matrix called EPS. The EPS is negatively charged due to its carboxyl functional groups. Cations are other parts of sludge floc which are essential in binding the EPS to the microbial cells and forming a unified network (Park, 2002). All these compounds are immersed in bulk (free) water. Bulk water occupies the largest fraction of the WAS sludge.

Understanding of the electrical properties of a material which represent its behavior in the presence of an electric field is a fundamental stage in designing an efficient EM heating system. As one of the critical parameters affecting the heating performance, determining the frequency (or the frequency range) at which the optimum heating is achieved highly depends on the material electrical properties. Regarding this requirement, a study was carried out by Bobowski et al. (2012) at UBC which laid the foundations of determination of an optimum frequency for sludge heating and therefore designing a new heating apparatus for municipal sludge heating for enhanced AD. In the study carried out by Bobowski et al. (2012), the complex permittivity of two waste sludge streams; TWAS and DWSC, sampled from KWWTP were measured at a wide range of frequency (3 MHz to 40 GHz). At the time of the study, the TS concentration of the TWAS and DWSC was measured 4.5% and 18%, respectively (Bobowski et al., 2012).

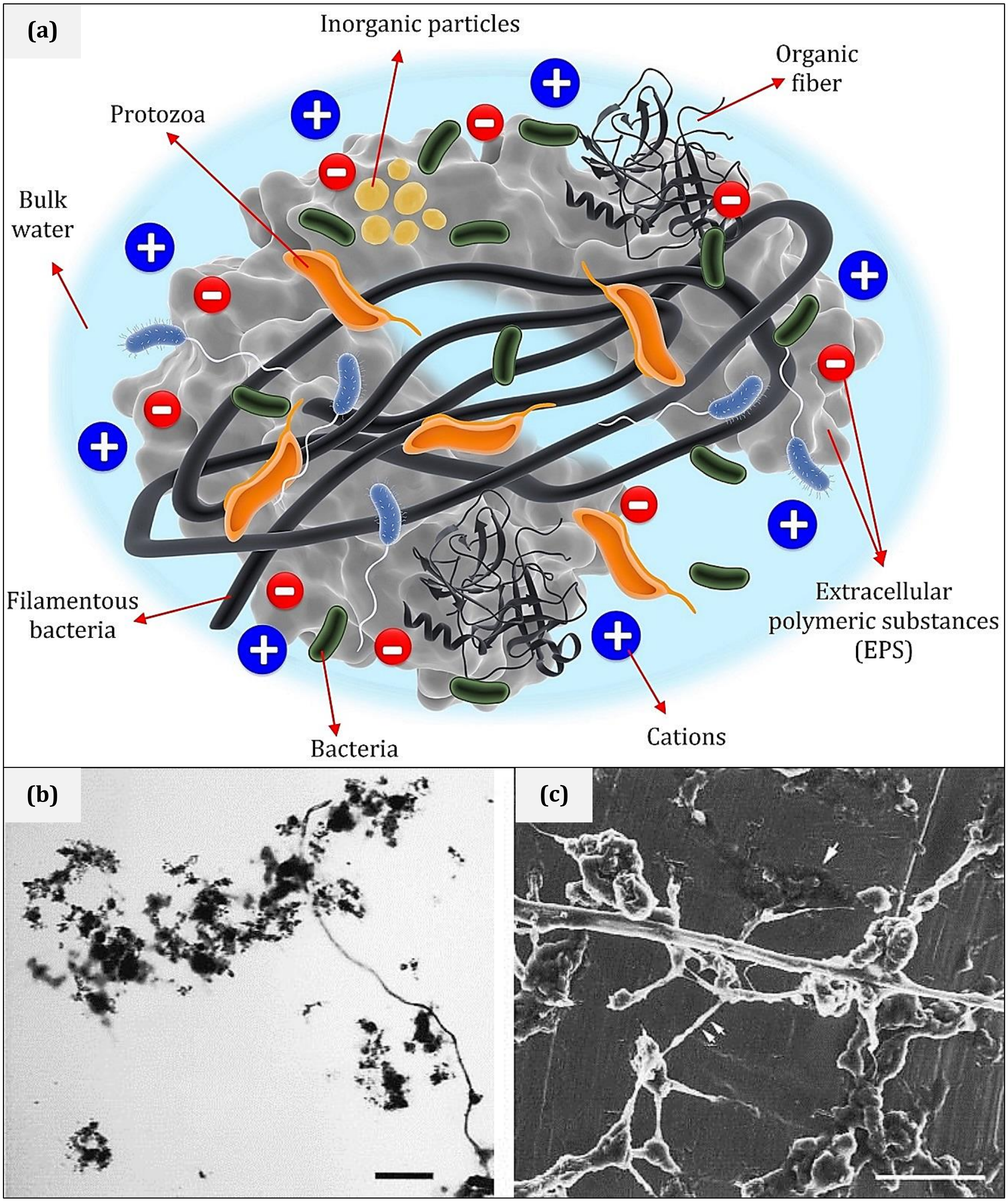


Figure 2-18. Waste activated sludge floc; (a) schematic of sludge structure and its main constituents, (b) conventional optical microscopy image; (c) scanning electron microscopy image. Length of scale bars is 100 μm (adapted from multiple sources (Leppard et al., 2003; Nielsen et al., 2012))

Figure 2-19 compares the change in the real and imaginary parts of the complex permittivity of KWWTP sludge (TWAS and DWSC) with that of pure water at the frequency of 3 MHz to 40 GHz. The data used to plot Figure 2-19 was adapted from Bobowski et al. (2012). As seen in Figure 2-19 (a), at the frequencies greater than 100 MHz, the change in ε' of TWAS and pure water follows similar pattern, while below 100 MHz, their pattern is clearly distinguished. Similar behavior called β-dispersion has been reported for biological samples such as cells and tissues at low frequencies (100 KHz to 10 MHz) (Pethig, 1984). The β-dispersion is generally explained by Maxwell-Wagner effect in which when an electric current passes across the interface of materials with different permittivity and conductivity, the charges are accumulated at their interface (Iwamoto, 2012). Now, taking into account the microstructure of WAS floc (Figure 2-18), the observed β-dispersion (Figure 2-19 (a)) is most likely related to the charging of the cell membranes which act as the interface between extra- and intra-cellular compounds. This interpretation also explains the improved β-dispersion observed for DWSC at 18% TS which is much thicker than TWAS at 4.5%, containing more cellular compounds. The sludge electrical characterization also reveals that at lower ranges of frequency (below 1 GHz), the ionic conductivity dominates the energy loss mechanisms. This corresponds to the term $\frac{\sigma_{dc}}{\omega\varepsilon_0}$ resulting in a straight line slope below 1 GHz in Figure 2-19 (b).

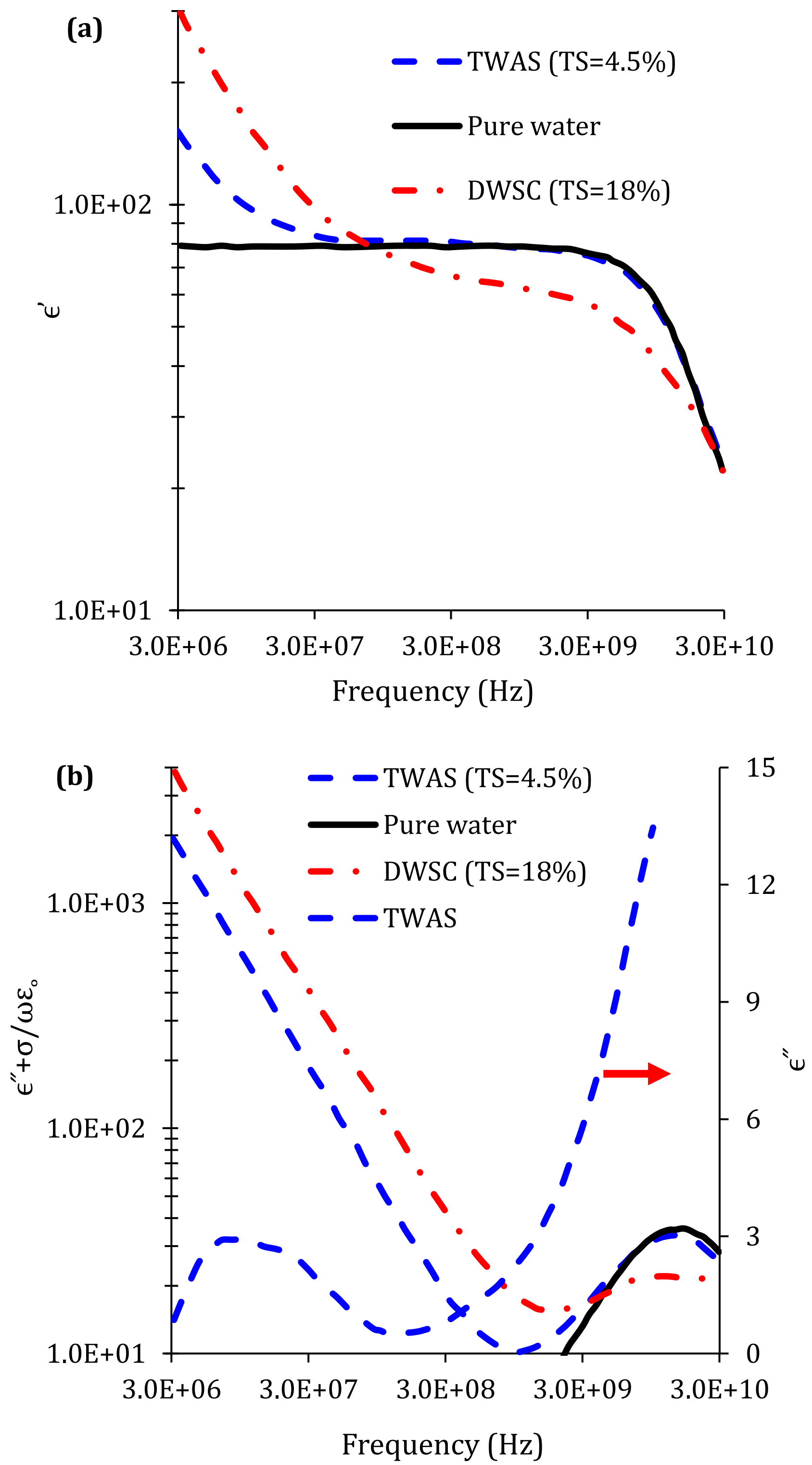


Figure 2-19. The complex permittivity of TWAS, DWSC and pure water; (a) real part of complex permittivity ($\boldsymbol{\varepsilon}'$), (b) loss factor (plotted based on the data reported by Bobowski et al. (2012))

2.9. SUMMARY

The structure of WAS floc includes a heterogeneous mixture of different microorganisms, organic and inorganic matter attached together with EPS. This forms a complex 3D polymeric network immersed in bulk water which resists AD. The slow degradation of WAS known as the bottleneck of conventional AD is due to the rate limiting hydrolysis step, essential to disintegrate the WAS structure at the beginning of AD. As a result, a long retention time is required to convert the organic fraction of WAS in conventional AD. This leads to the large digester volume and high capital cost. In an attempt to overcome the bottleneck of conventional AD, the WAS structure can be disrupted via different (i.e. thermal, mechanical, biological methods and combination of them) methods called "*pretreatment*" resulting in the release of intracellular and extracellular biopolymers of sludge in to liquid phase, accelerating the digestion process. These pretreatments, with the common goal of converting large, complex organics into small organics, are applied to WAS externally (outside the digester vessel) before sludge is fed to the digester. The overall process (pretreatment followed by AD) is known as an "*advanced AD*". Thermal hydrolysis using either CH or MW irradiation has been one of the leading topics in the field of sludge pretreatment during the last two decades. The literature review on the topic revealed the following two research gapes; 1) the lack of a comprehensive study to compare the two thermal sludge pretreatment techniques (CH and MW irradiation) under controlled experimental conditions (under identical thermal profile) and 2) the limitation of almost all the previous MW pretreatment studies to a single frequency of 2.45 GHz. To fill the first literature gap, in this research, a study was conducted using a pressure-sealed vessel controlled by a computer as a CH system that mimics the MW heating under identical thermal profile. Using the custom-designed CH system, the comparison between the CH and MW (at 2.45 GHz) pretreatments was performed in a much more controlled experimental condition than that of previous studies. The methodology and results of the comparison studies are discussed in Chapters 3 and 4. To address the second knowledge gap stated above, fundamental research was performed to design, build, and assess a novel EM sludge pretreatment system at a frequency of 13.56 MHz. The design steps followed along with the findings of the conducted research are described in Chapters 5, 6, and 7.

CHAPTER 3. CONDUCTIVE HEATING VS. MICROWAVE IRRADIATION FOR ENHANCED SLUDGE DISINTEGRATION UNDER IDENTICAL THERMAL PROFILES

3.1. INTRODUCTION

The primary goal of this research (*Objectives III, IV and V*) was to design, implement and evaluate a novel energy-efficient sludge pretreatment system that goes beyond the limitations of the currently available thermal hydrolysis methods. However, this goal could not be achieved without understanding the effects of the existing thermal pretreatment systems (CH and MW irradiation) on sludge solubilization and AD performance. As previously discussed (Section 2.7.4.3), due to the significant inconsistency among the results of the published research, it is hard to derive a solid conclusion about any possible differences between CH and MW pretreatment method on sludge disintegration. Therefore, the objective of the first phase of this research was to compare CH and MW (at 2.45 GHz) pretreatment systems under identical thermal profile for enhanced sludge solubilization. It is worth noting that to achieve an identical thermal profile, the final temperature reached as well as the heating/cooling ramp rate should be identical between CH and MW pretreatments.

There are two hypotheses tested in this chapter. First, not only the final temperature reached but also the heating/cooling profiles are important factors determining sludge solubilization for using MW and CH pretreatment methods. Validating this hypothesis with an extensive data set will allow for explanation of the contradictory results of previous studies in which only the final temperature (not the heating/cooling rates) was kept identical. The second hypothesis proposed is that, under identical thermal profile (identical pretreatment temperature and the heating/cooling rate), the pretreatment method (MW vs. CH) is not a significant factor determining the sludge disintegration ratio.

The key research infrastructure needed for successful validation of the aforementioned hypotheses was a computer controlled CH system, because commercial units are limited to certain temperature ranges and heating rates. Recent research within our UBC Bioreactor Technology Group was successful in custom-building a CH system which could simulate MW heating under identical heating profile. The first study (Mehdizadeh et al., 2013) that utilized this CH applicator (for comparison with MW heating) was however limited to a single heating rate of 7.5°C/min which did not allow for arriving at any conclusion simultaneously for the effect of pretreatment method, temperature and heating rate. Moreover, for the current research, the CH applicator was modified from its previous version to follow an identical cooling rate as MW, in addition to the identical heating rate. Therefore, the purpose of this phase was to provide a comprehensive comparison between MW and CH on sludge solubilization over a wide range of temperature (80, 120 and 160°C) and heating rate (3, 6 and 11°C/min) under identical heating/cooling profiles.

3.2. MATERIALS AND METHODS

3.2.1. Sludge characteristics

In this chapter, the thermal pretreatment was performed on the DWSC generated at KWWTP. The KWWTP has the design capacity of 70,000 m^3/d, and currently serves a population of approximately 100,000 people. The overall treatment process at the treatment plant includes screening, grit removal, primary sedimentation, fermentation, biological nutrient removal (occurred in the bioreactors), secondary sedimentation, filtration (by disc filters), and ultraviolet disinfection. The treatment process in the bioreactors is a modified Barnard denitrification phosphate process (Bardenpho®) for simultaneous removal of carbon, nitrogen and phosphorus. The biological treatment starts with anaerobic fermentation of wastewater. The wastewater then undergoes sequential nitrification-denitrification processes in anoxic-aerobic tanks during which the majority of the ammonia and nitrate are converted to the nitrogen gas (N_2). The VFAs generated from the fermentation of the PS provide a carbon source for the denitrifying bacteria. The phosphorus is also absorbed by the phosphorus accumulating organisms. The overall hydraulic retention time (HRT) of the biological treatment processes is approximately 9.3 h. The SRT however varies between 6 to 13 d during the summer and winter seasons, respectively. The effluent from the bioreactors flows into the secondary clarifiers where WAS is generated by settling. The WAS is then pumped to a DAF unit and further thickened. The fermented PS and thickened secondary sludge streams are mixed at a ratio of 33:67 by volume. The mixed sludge is then sent to a centrifuge unit and dewatered to produce DWSC. It is worth noting that conducting thermal pretreatment on DWSC with high solids concentration reduces the heat dissipation in water resulting in minimizing the energy consumption per sludge dry weight. The characteristics of the DWSC used in this study is listed in Table 3-1.

Table 3-1. The characteristics of the KWWTP dewatered sludge cake

Description [a]	**Value**
pH	5.8 ± 0.2
TS (% w/w)	19.2 ± 0.34
VS (% w/w)	16.8 ± 0.35
VS/TS (%)	87.6 ± 0.24
Ammonia (mg/L)	678 ± 82
TCOD (mg/L)	265,702 ± 9,422
SCOD (mg/L)	11,991 ± 591
Volatile fatty acids (VFAs)	
Acetic acid (mg/L)	939.3 ± 27.9
Propionic acid (mg/L)	855.0 ± 0.7
Butyric acid (mg/L)	63.0 ± 1.4

[a] TS: total solids; VS: volatile solids; TCOD: total chemical oxygen demand; SCOD: soluble chemical oxygen demand

3.2.2. Thermal pretreatment systems

3.2.2.1. Conductive heating system

The operating configuration and the major components of the CH system used in this research are illustrated in Figure 3-1. The custom-built CH system consisted of the following components: pressure-sealed vessel, pressure gauge and safety valve, external fiberglass insulator, DC power supply, digital multimeter, thermocouple, control software and safety shield. The pressure-sealed vessel was made of a copper cylinder (height, internal diameter and thickness of 92 mm, 38 mm and 3.2 mm, respectively) wrapped with 150 cm of 0.3 mm-diameter insulated nichrome wire (#80/20) with the total electrical resistance of 500 Ω. The wire insulation allowed the temperature to increase beyond 200°C. A high-temperature epoxy was used to secure the wrapped wire to the body of the vessel. It should be mentioned that the pressure-sealed vessel was designed in a way to have the same internal dimensions as MW unit Teflon vessels in order to achieve similarity between MW and CH pretreatments. The temperature of the sample was directly measured using a Type-K thermocouple located at the center of the copper vessel. A pressure gauge (Winters PEM Series) and also a pressure safety valve connected to the cap of the vessel were used to control the pressure during the operation. The pressure-sealed vessel was connected to a programmable DC power supply (Sorensen, Ametek) and a multimeter (Agilent, 34401A) attached to the thermocouple. The voltage of the DC power supply was controlled with a computer equipped with a custom-developed LabVIEW program. Control over the DC voltage (which was directly linked to the power delivered to the sample) allowed for heating of the DWSC samples at any arbitrary heating ramp rates. Using an external fiberglass insulator surrounding the pressure-sealed vessel, the MW cooling rate was also simulated (in addition to the heating rate controlled by the computer) to achieve completely identical thermal (heating/cooling) profiles between CH and MW pretreatments. Figure 3-2 shows the infrared (IR) digital photo of the CH pressured-sealed vessel confirming that the heat was evenly distributed throughout the load (sludge sample) during the pretreatment.

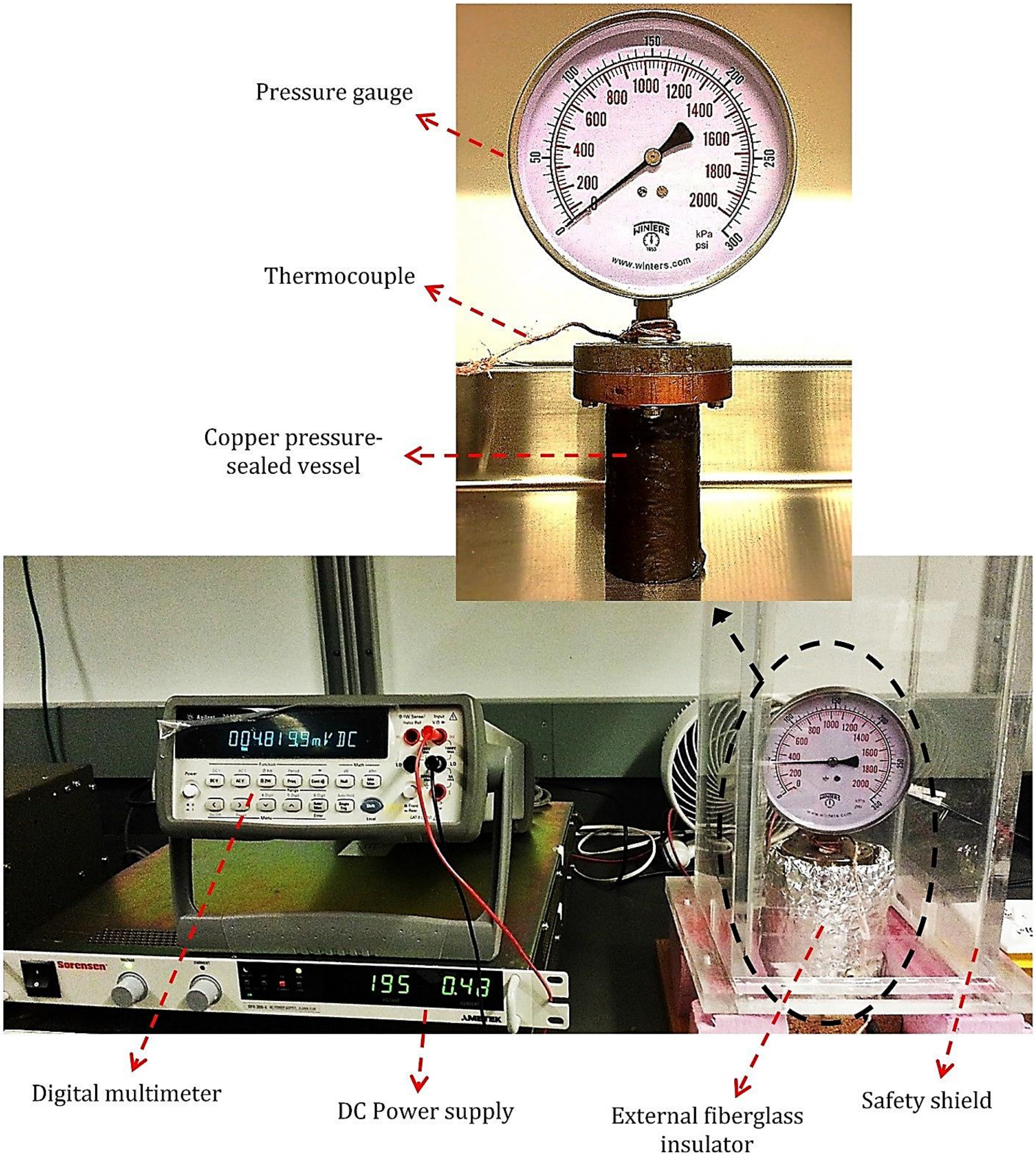


Figure 3-1. The operating configuration and major components of the conductive heating system

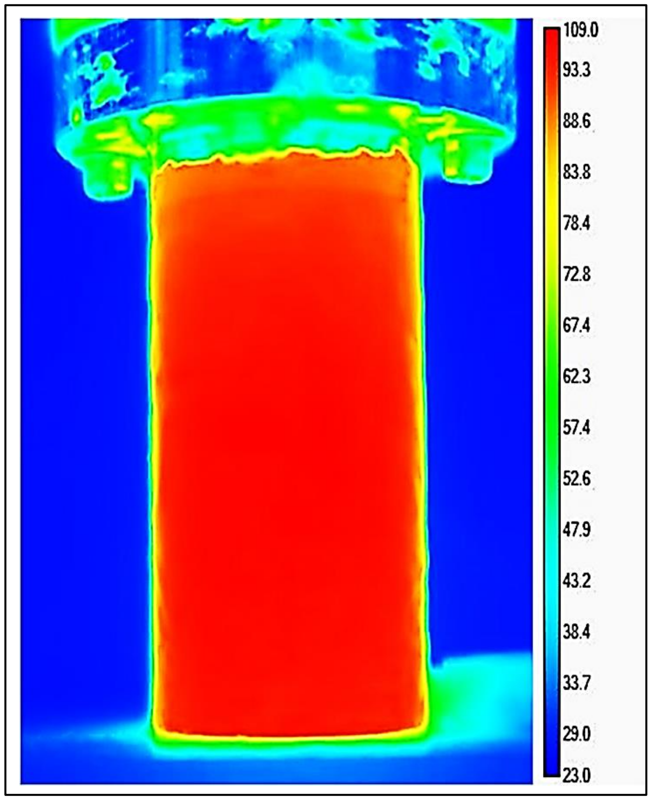


Figure 3-2. The infrared digital photo of the CH pressured-sealed vessel (the values in the picture show the temperature range from 23 to 109°C)

3.2.2.2. Bench-scale 2.45 GHz microwave system

The bench-scale MW heating system used in this research was an ETHOS-EZ model manufactured by Milestone Inc., Connecticut, USA (Figure 3-3). The MW oven is rated at 1200 W and operates at a frequency of 2.45 GHz. The ETHOS-EZ has twelve 100 mL Teflon pressure-sealed vessels rotating on a carousel. The system can heat loads up to a maximum temperature and pressure of 300°C and 35 bar, respectively. The temperature of the TWAS samples was measured continuously using an ATC-400-CE thermocouple. An automated controller monitored and controlled the heating ramp rate throughout the operation.

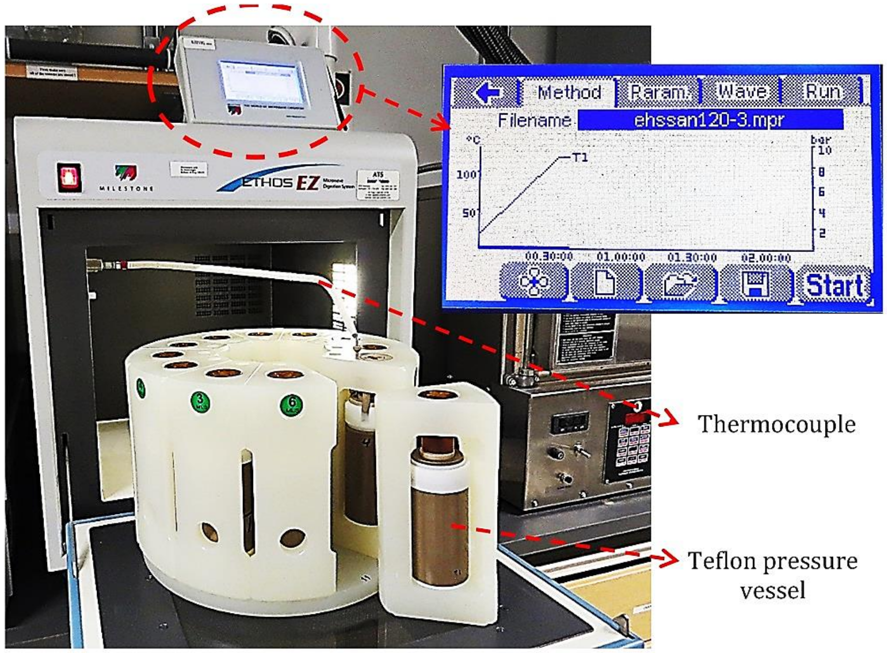


Figure 3-3. The bench-scale 2.45 GHz microwave oven and its major components

3.2.3. Thermal pretreatment parameters studied

A wide range of final pretreatment temperature below and above boiling point (80, 120 and 160°C) and heating ramp rate (3, 6 and 11°C/min) was considered. Table 3-2 shows the experimental design used in this study including nine different combinations of the input parameters (pretreatment method, heating ramp rate and final temperature) and one control scenario (without pretreatment). Before starting the pretreatment, the pressure-sealed vessel (shown in Figure 3-1) was calibrated for three selected heating rates (3, 6 and 11°C/min) using an open-loop trial and error method for DWSC samples. Calibration of MW station was also done via its close-loop controller. Therefore, both MW and CH pretreatments could be performed at identical final temperatures and heating/cooling rates. Figure 3-4 compares the thermal profiles during CH and MW pretreatment of DWSC which proves almost completely identical heating and cooling rates between CH and MW.

Table 3-2. The experimental design used

Pretreatment No.	Final Temperature (°C)	Heating rate (°C/min)	Pretreatment Method [a]
1	80	3	MW
2	80	6	MW
3	80	6	CH
4	80	11	CH
5	120	3	MW
6	120	3	CH
7	120	6	MW
8	120	6	CH
9	120	11	MW
10	120	11	CH
11	160	3	CH
12	160	6	MW
13	160	6	CH
14	160	11	MW
Control			

[a] CH: Conductive heating; MW: Microwave

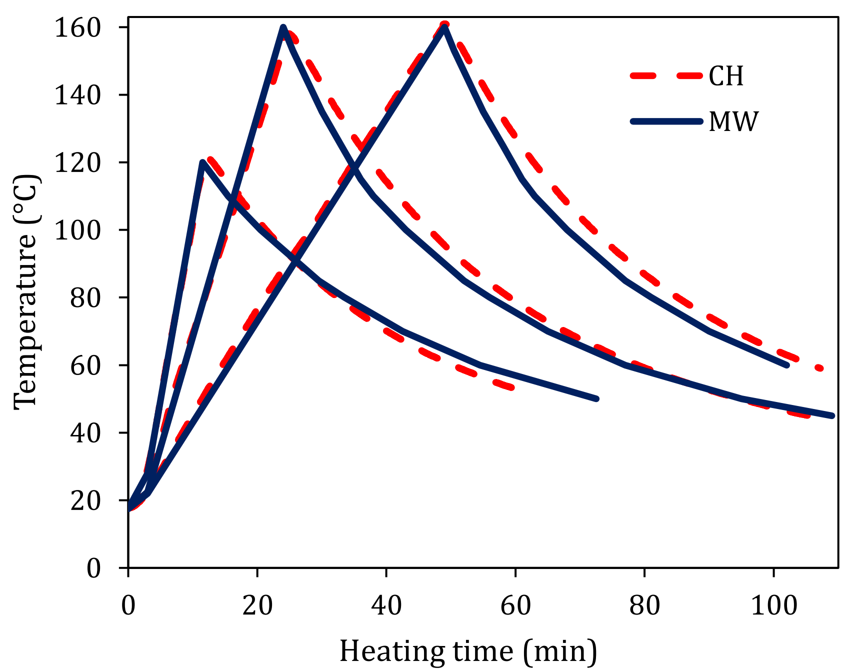


Figure 3-4. The thermal profiles of the conductive heating and microwave pretreatment

3.2.4. Analytical procedures

The TS and VS analyses were done according to the Standard Methods, sections 2540 B and 2540 E, respectively (APHA, 2005). The TS analysis was performed by drying the samples at a temperature of 105°C for a minimum duration of 12 hr. For VS determination, the dried samples were placed in a furnace and kept at a temperature of 550°C for a minimum of 1 hour. The analysis of COD, sugar, protein and humic acid (HA) were conducted on both total and soluble fraction of DWSC samples. The separation of soluble fraction of the DWSC was as follows: the samples were diluted and then centrifuged at 10,000 rpm for 20 min via a Fisher Scientific Sorvall Legend XT automatic centrifuge. The supernatant (liquid fraction) of the centrifuged samples was then filtered via 0.45 μm membrane filters. The filtered liquid (through 0.45 μm) was considered as the soluble fraction. Quantification of COD concentration was performed according to the closed reflux colorimetric method outlined by Standard Methods (APHA, 2005). The procedure of sugar analysis proposed by Dubois et al. (1956) was followed for sugar measurement. An Evolution 60S UV-Vis spectrophotometer (Thermo Fisher Scientific, Inc.) at the wavelengths of 600 nm and 490 nm was used for COD and sugar analyses, respectively. The modified Lowry protein assay was also applied for the analysis of proteins and HA (Frolund et al., 1995). Protein and HA measurement was performed at the wavelength of 750 nm using a microplate reader/spectrophotometer (BioTek Synergy HT Multi-detection). The protein and HA concentrations were adjusted according to the following equations.

$$A_{total} = A_{protein} + A_{HA} \quad (3\text{-}1)$$

$$A_{blanc} = 0.2A_{protein} + A_{HA} \quad (3\text{-}2)$$

$$A_{protein} = 1.25\ (A_{total} - A_{blanc}) \tag{3-3}$$

$$A_{HA} = A_{blanc} - 0.2A_{protein} \tag{3-4}$$

where, A_{total}, A_{blanc}, $A_{protein}$, and A_{HA} are the total absorbance with $CuSO_4$, the total absorbance without $CuSO_4$, the protein absorbance, and the HA absorbance, respectively. The representative calibration curves for the COD, sugar, protein, and HA determination are provided in Appendix A (Figures A-1, A-2, A-3, and A-4).

3.2.5. Statistical analysis

Statistics including mean and standard deviation were calculated for the collected data using Microsoft Excel 2010. The experimental data were analyzed by multifactor analysis of variance (ANOVA) at 95% confidence level ($\alpha = 0.05$) using Minitab Software 17 to evaluate the statistically significant effects of independent variables (pretreatment method, heating ramp rate and final temperature). Since the ANOVA method does not specify exactly which means are significantly different from which other means, the Fisher's least significant difference test was applied for comparing all pairs of means. The Anderson-Darling test was used to judge if the data follow normality distributions. The sample preparation (using the pretreatment scenarios listed in Table 3-2) was done randomly following a randomized experimental order determined by Design-Expert 9 software.

3.3. Results and discussion

Considering several previous studies addressing the solubilization effects of thermal pretreatment using MW and/or CH methods, the focus of the first part of this research was to assess the effects of pretreatment method, heating rate (3, 6 and 11°C/min) and final temperature (80, 120 and 160°C) on DWSC solubilization. Thermal pretreatment is expected to disrupt the sludge structure resulting in the release of intracellular biopolymers (i.e., proteins, sugars, lipids and nucleic acids) and extracellular biopolymers (i.e., EPS) from the sludge floc structure into the soluble phase. Therefore, measurement of soluble biopolymers has been known to be a good estimate of organic matters solubilization in biosolids (Eskicioglu et al., 2006). In this study, the effects of thermal pretreatment using MW and CH under identical heating/cooling profile on solubilization of COD and three biopolymers including protein, sugar and humic acid were evaluated.

3.3.1. Effect of pretreatment parameters on COD solubilization

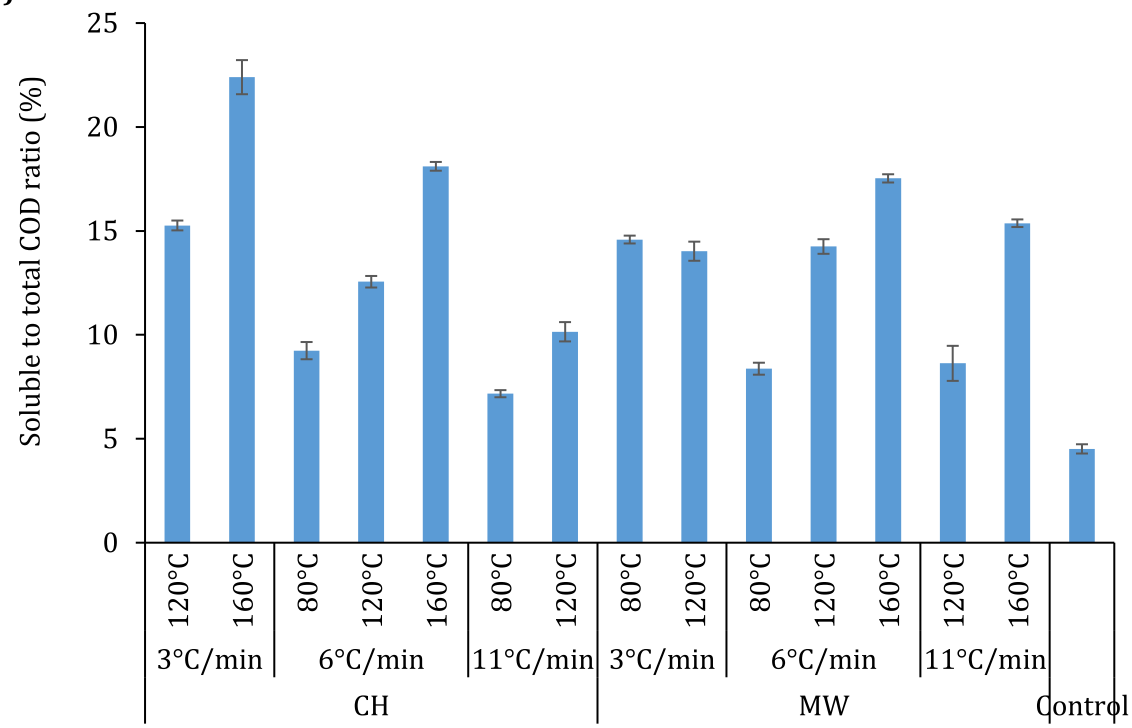
The ratio of the soluble COD (SCOD) to the total COD (TCOD) of the pretreated DWSC samples and their relative (to control) improvement in COD solubilization are shown in Figure 3-5 (a) and (b), respectively. The solubilization ratio (SR) and relative (to control) improvement in solubilization ratio (SR_r) were determined using Eq. (3-5) and Eq. (3-6), respectively.

$$SR\ (\%) = \frac{Soluble\ fraction\ concentrion}{Total\ fraction\ concentration} \times 100 \tag{3-5}$$

$$SR_r = \frac{SR\ of\ the\ pretreated\ DWSC}{SR\ of\ the\ control\ DWSC} \tag{3-6}$$

As seen in Figure 3-5 and also proven by the statistical analysis, increasing the final temperature increased the COD solubilization ratio (p-value = 0000<0.05). The statistically significant differences in COD solubilization ratio were also observed among the DWSC samples exposed to different heating ramp rates (p-value = 0.000<0.05). According to the results, increasing the heating rate (which is directly linked to the decrease of the heating time) had an adverse effect on the COD solubilization ratio (Hosseini Koupaie & Eskicioglu, 2015b). The positive effect of increasing pretreatment temperature on COD solubilization has been previously reported by other researchers (Appels et al., 2010; Eskicioglu et al., 2008a; Ji Park et al., 2010). There are also other studies that address the effects of heating rate on sludge solubilization (Park et al., 2009; Toreci et al., 2011). In agreement with the results of this study, higher improvement in the solubilization of MW-pretreated sludge samples at the rate of 3.75°C/min was observed compared to that of 7.5°C/min (Toreci et al., 2011). Similarly, the greatest solubilization ratio was achieved at the lowest applied MW heating rate (2.9°C/min) in another study (Park et al., 2009). Both of these studies which investigated the effects of heating rate on sludge solubilization were however limited to the application of MW pretreatment, and did not compare the results with a CH system. In the current study, despite the effects of both heating ramp rate and final temperature, no statistically significant effect of pretreatment method (MW and CH) on COD solubilization was observed under identical heating/cooling profile (p-value = 0.668>0.05). Among the applied pretreatment conditions, the maximum COD solubilization ratio of 22.4 ± 0.8% (5 times improvement over the control sample) was obtained at 160°C and 3°C/min (highest temperature and lowest heating rate), while the minimum solubilization ratio of 7.2 ± 0.2% was achieved at 80°C and 11°C/min (lowest temperature and highest heating rate). It is worth noting that the solubilization ratio of the control DWSC sample (without pretreatment) was only 4.5 ± 0.2%. The main effect test revealed that the average COD solubilization ratio was 9.8, 12.5, and 18.4% for the final temperatures of 80, 120 and 160°C, respectively and 16.6, 13.3, and 10.3% for the heating rates of 3, 6 and 11°C/min, respectively. The representative Anderson-Darling test normality plot for the COD solubilization data is presented in Appendix B (Figure B- 1). The representative constant variance test results for COD solubilization data are also illustrated in Appendix C (Figure C- 1).

(a)



(b)

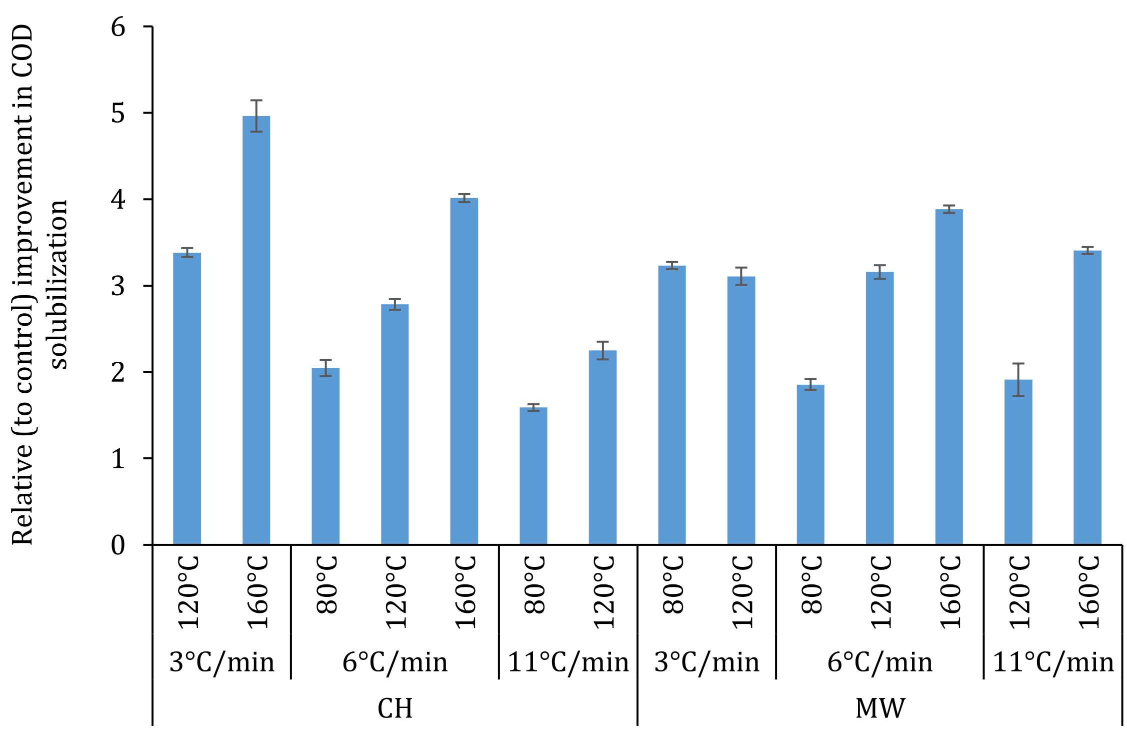
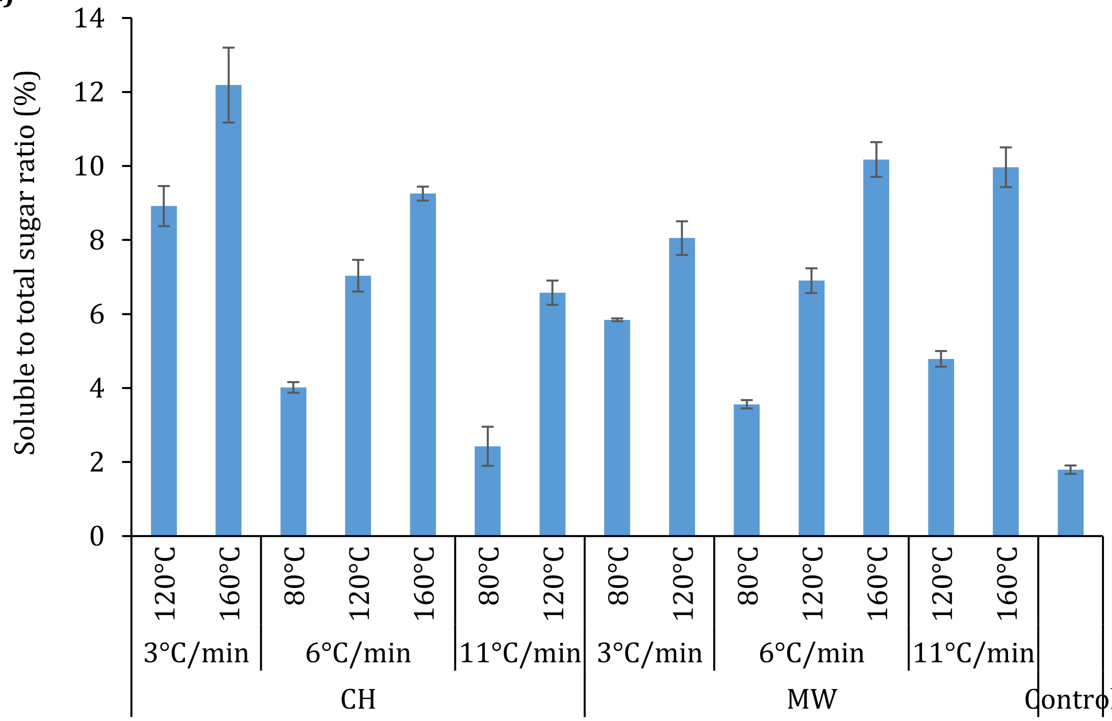


Figure 3-5. The effect of thermal pretreatment on COD solubilization: (a) soluble to total ratio; (b) relative (to control) improvement in solubilization

3.3.2. Effect of pretreatment parameters on biopolymers solubilization

Figure 3-6 (a) and (b) show the soluble to total sugar ratio of DWSC samples and their relative (to control) improvement in sugar solubilization, respectively. The analysis of variance proved statistically significant effect of the pretreatment temperature on sugar solubilization (p-value = 0.000<0.05). As seen in Figure 3-6, increasing pretreatment final temperature increased the concentration of soluble sugar. The average of sugar solubilization ratio increased from 1.8% (for control sample) to 4.0, 7.0, and 10.4% for the DWSC samples exposed to the final temperatures of 80, 120, and 160°C, respectively. The effect of the heating ramp rate on the sugar solubilization was also found to be statistically meaningful (p-value = 0.001<0.05). Considering the results of the main effect test, the heating rates of 3, 6 and 11°C/min resulted in an average sugar solubilization ratio of 8.7, 6.8, and 5.9%, respectively. This confirms that increasing the heating time (decreasing the heating rate) increased the concentration of soluble sugar. The highest and the lowest relative (to control) improvement ratio in sugar solubilization were 12.2 and 2.4, respectively (Figure 3-6 (b)). Similar to the results obtained from the COD solubilization experiments, using MW or CH pretreatment method had no statistically significant effect on sugar solubilization (p-value = 0.334>0.05).

(a)



(b)

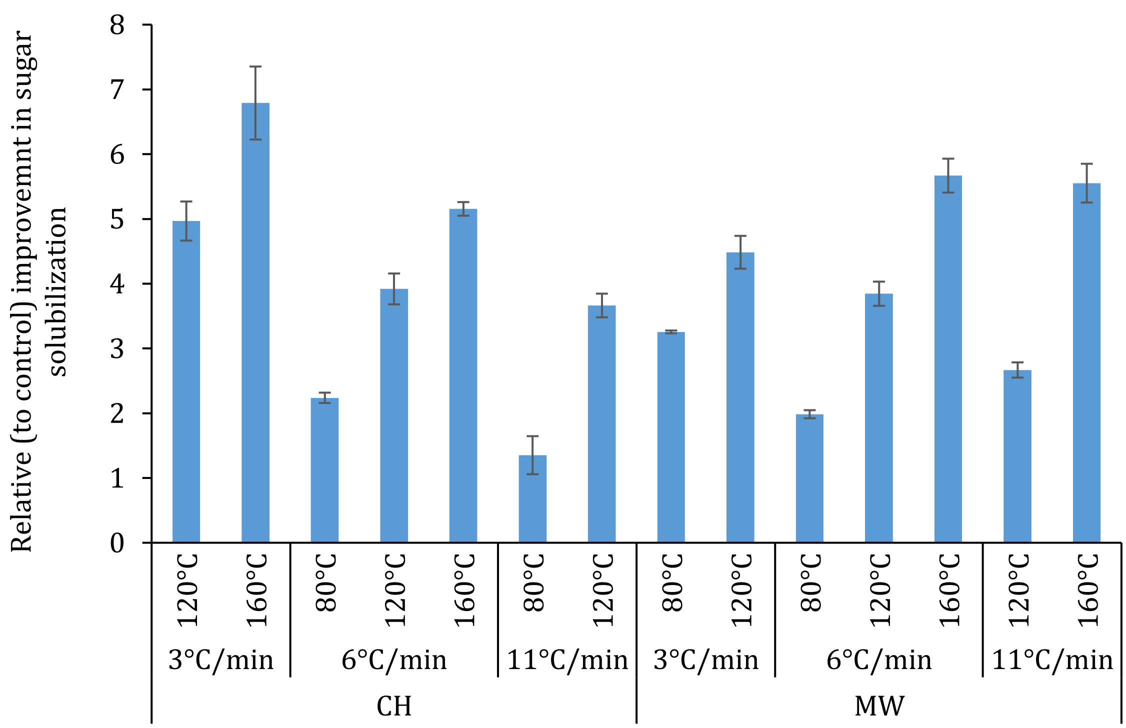


Figure 3-6. The effect of thermal pretreatment on sugar solubilization: (a) soluble to total ratio; (b) relative (to control) improvement in solubilization

The solubilization results of protein and HA are illustrated in Figure 3-7. As expected and consistent with the COD and sugar solubilization results, no statistically significant difference was observed between the MW and CH pretreatments in terms of protein (p-value = 0.112>0.05) and HA (p-value = 0.780>0.05) solubilization. However, the statistically significant effect of the pretreatment temperature on protein (p-value = 0.000<0.05) and HA (p-value = 0.000<0.05) was proven. The analysis of variance also showed that heating rate had statistically significant effect on both protein (p-value = 0.000<0.05) and HA (p-value = 0.001<0.05) solubilization. The change in the solubilization ratio of protein and HA with respect to the final temperature and heating rate demonstrated the same pattern as that of the COD and sugar. Increasing pretreatment final temperature and ramp rate had positive and negative effect on soluble concentration of these compounds, respectively.

The contour plot of the solubilization ratio vs. the final temperature and heating rate for COD, sugar, protein, and HA is shown in Figure 3-8 (a), (b), (c), and (d), respectively. As seen in Figure 3-8, the solubilization ratio increases in the direction of increasing the final temperature and decreasing the heating rate. It can be also inferred from the slope of the contour lines that, in general, the effect of the final temperature on solubilization was higher than that of the heating rate, although both of these parameters were proven to have statistically significant effects on the solubilization of DWSC. This inference is in agreement with the results of the statistical analysis in which the p-values corresponding to the final temperature were lower than those corresponding to the heating rate.

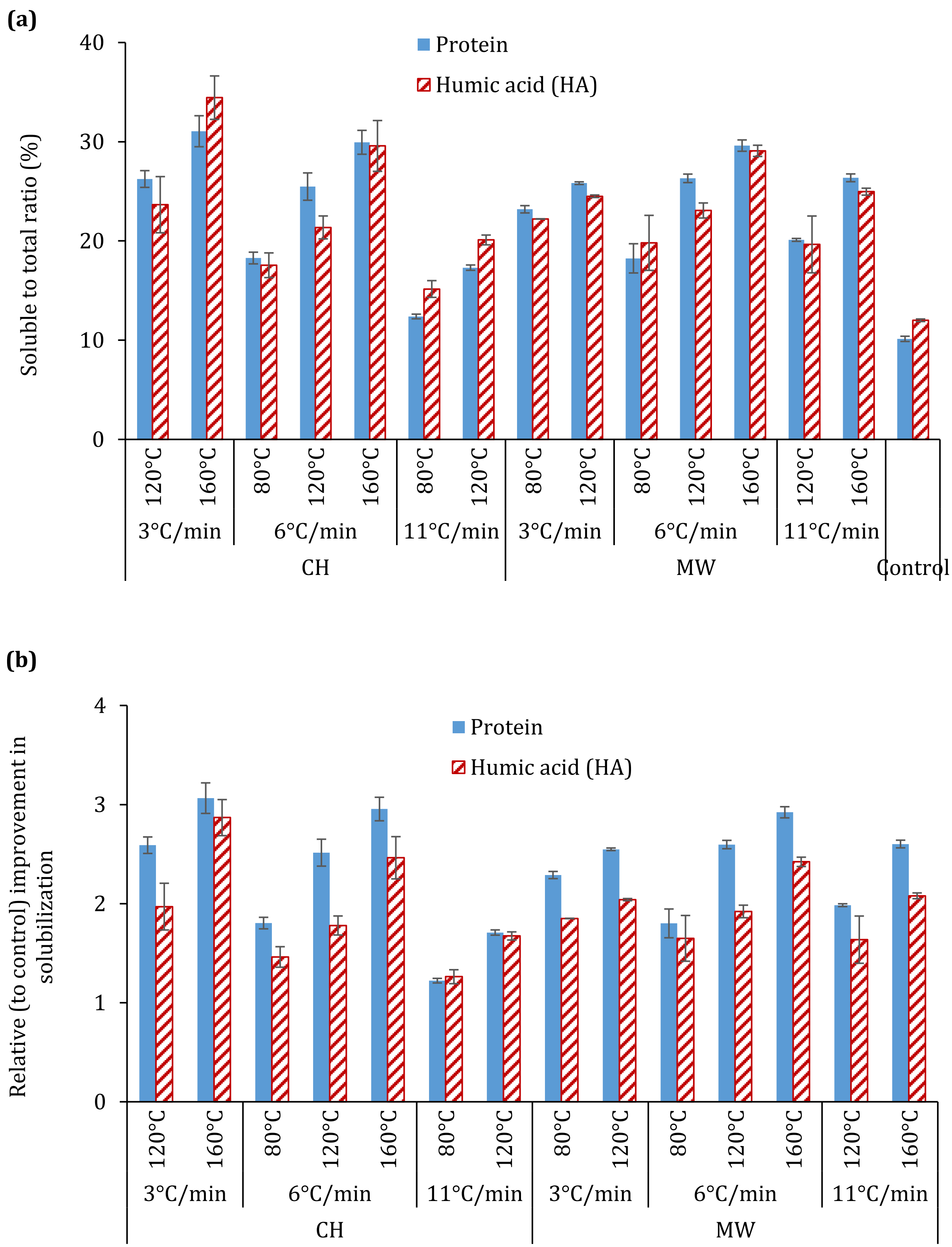
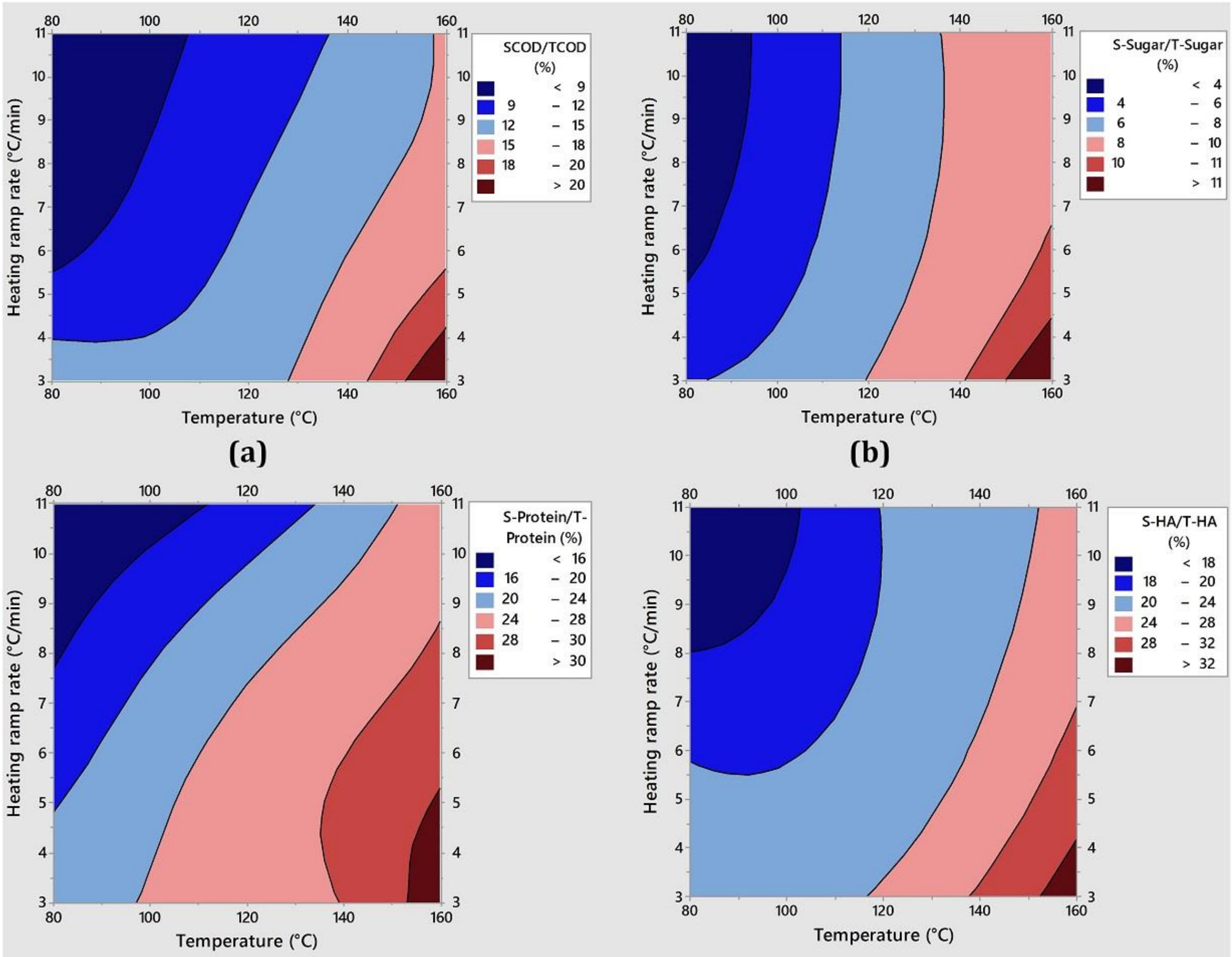


Figure 3-7. The effect of thermal pretreatment on protein and humic acid solubilization: (a) soluble to total ratio; (b) relative (to control) improvement in solubilization



(c) **(d)**

Figure 3-8. The contour plot of solubilization ratio vs. final temperature and heating rate for (a) COD; (b) sugar; (c) protein and (d) humic acid (HA)

3.4. SUMMARY

In this chapter, the effect of the CH and MW (at 2.45 GHz) pretreatment techniques on municipal sludge disintegration were compared under identical thermal profiles (identical pretreatment temperature and heating/cooling rate) and over a wide range of pretreatment temperature (below and above boiling point) and heating ramp rate. It was statistically proven in this study that not only the pretreatment temperature, but also the heating ramp rate have a significant effect on sludge disintegration (COD, sugar, protein and HA solubilization), which validated the solubilization related part of the *Hypothesis I*. Increasing pretreatment temperature improved COD and biopolymer solubilization ratios, while increasing heating ramp rate had an adverse effect on sludge solubilization. Considering the effects of both final temperature and heating rate, any comparison between MW and CH pretreatment methods should be conducted under identical thermal pretreatment profiles, otherwise it results in unreliable and contradictory conclusions (as observed in the literature). No statistically significant difference was observed between the two pretreatment methods in terms of COD, sugar, protein and HA solubilization when CH and MW hydrolysis were applied under identical thermal profiles, which validated the solubilization related part of the *Hypothesis II*. This observation rejects the assumption of enhanced sludge disintegration using MW pretreatment compared to that of the CH as a result of MW non-thermal (athermal) effects.

CHAPTER 4. CONDUCTIVE HEATING VS. MICROWAVE IRRADIATION FOR ENHANCED MESOPHILIC AND THERMOPHILIC BATCH ANAEROBIC SLUDGE DIGESTION

4.1. INTRODUCTION

It was statistically proven in Chapter 3 that not only the final temperature reached but also the heating rate has significant effects on municipal sludge disintegration (COD, sugar, protein, and HA solubilization). It was also observed and statistically confirmed that under the identical thermal profile (identical pretreatment temperature and heating rate), the CH and MW pretreatments achieve similar sludge solubilization ratios. Considering the findings of the previous chapter, the present chapter aims to study the simultaneous effects of the pretreatment method (CH vs. MW), final temperature, and heating ramp rate on performance of mesophilic and thermophilic AD under batch-flow regime. It will be also examined in this chapter that whether the results of the solubilization study (discussed in Chapter 3) will be in agreement with the findings of a comparison study between CH and MW pretreatment for enhanced mesophilic and thermophilic AD. The following sections of the chapter provide detailed discussions about the methodology as well as the findings of the batch AD study. Due to the large number of reactors operated simultaneously, for data management, the results are presented and discussed within mesophilic and thermophilic clusters for clarity.

4.2. MATERIALS AND METHODS

4.2.1. Mesophilic and thermophilic inocula

The mesophilic inoculum was taken from a pilot-scale anaerobic digester which has been utilizing KWWTP mixed sludge for more than 2 years. The pilot digester was fed two times a day and operated under a SRT of 20 d. Thermophilic inoculum was taken from a full-scale digester located at the Annacis Island WWTP in Vancouver (BC, Canada), which utilizes a mixture of PS and WAS. In order to acclimatize the mesophilic inoculum to the pretreated sludge, a 4 L side-armed Erlenmeyer flask with an effective volume 3 L was set-up as semi-continuous anaerobic digester. The digester was semi-continuously fed with pretreated DWSC for approximately 4.5 months at a SRT of 20 d, under the OLR of 1.68 ± 0.04 $\frac{g\ VS}{L.d}$. For the acclimation of thermophilic inoculum, two Erlenmeyer flasks (each with total and effective volume of 2 L and 1.2 L, respectively) were semi-continuously operated for the same period of time and under similar SRT and OLR.

The configuration of the semi-continuous digesters is illustrated in Figure 4-1. As per Figure 4-1, the Erlenmeyer flasks were sealed with two-port rubber stoppers. The ports in the rubber stopper were used to collect biogas as well as to withdraw the digester effluent (digestate) daily. Before sealing, the dissolved and headspace oxygen of the digesters was removed using N_2 purging. The digesters were fed semi-continuously once a day (7 days/week) through the side arm of the flask. The produced biogas was collected in 2 L Tedlar® bags. The biogas volume was measured daily with a manometer connected to a suction pump. The biogas volume was adjusted to the standard temperature and pressure (STP) condition (0°C and 1 atm).

The mesophilic and thermophilic digesters were placed in the incubators set at 90 rpm and average temperatures of 35 ± 1°C and 55 ± 1°C, respectively. The representative calibration curve for the biogas volume measurement with manometer is provided in Appendix A (Figure A- 5).

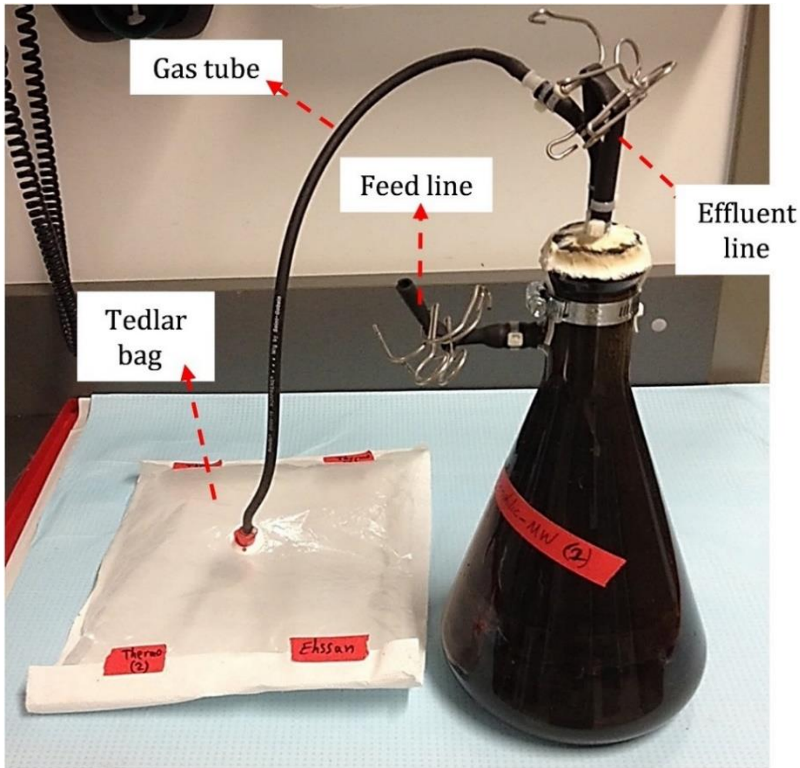


Figure 4-1. The configuration of the semi-continuous anaerobic digesters

To minimize the possible chronic or acute inhibition to the acid/methane formers, the most intense thermal pretreatment condition (pretreatment temperature and heating ramp rate of 160°C and 11°C/min, respectively) was used for feed during the acclimation period. The TS and VS concentration of the mesophilic inoculum were 2.05% and 1.35% (by weight) at the end of the acclimation period, respectively. The thermophilic inoculum also had a TS and VS concentration of 2.1% and 1.53% (by weight) at the end of the acclimation period, respectively.

4.2.2. Sludge characteristics

The characterization of DWSC samples was presented earlier in Table 3-1. The mesophilic and thermophilic batch anaerobic digesters were set-up using the sludge samples pretreated according to the experimental design of the solubilization test (Chapter 3). After pretreatment, the DWSC samples were diluted 5 times and stored at 4°C in the fridge to be used in further experiments. All the pretreated samples stored in the fridge were used within the next 48 h. Since the thermal pretreatment, especially at elevated pretreatment temperatures (>80°C), reduces the microbial activity of the samples, storing the pretreated samples in the fridge for less than 48 h should have negligible effects on the results of the subsequent experiments. Also, because both MW- and CH-pretreated sludge samples were stored in the fridge for the same period of time, any possible bias to prevent direct comparison should be negligible.

4.2.3. Batch anaerobic digestion set-up

Figure 4-2 shows the experimental design used to study the simultaneous effects of pretreatment method, temperature, and heating rate on performance of mesophilic and thermophilic batch AD. The experimental design included three heating rates (3, 6, and 11°C/min), three pretreatment temperatures (80, 120, and 160°C), two pretreatment methods (CH and MW), and two digestion temperatures (35 and 55°C). As per Figure 4-2, twenty seven batch anaerobic digesters including triplicates fed with acclimated mesophilic inoculum and pretreated DWSC were operated. The same number of batch digesters (27) were set-up with the acclimated thermophilic inoculum. In addition to the digesters (corresponding to the experimental design in Figure 4-2), two sets of digesters (in triplicate) fed with non-acclimated mesophilic inoculum and the least (3°C/min and 80°C) and the most (11°C/min and 160°C) intense thermally pretreated substrate were set up. The purpose of operating non-acclimated digesters were to assess whether the impact of pretreatment is the same for acclimated and non-acclimated inocula and also to investigate any potential inhibitory (or stimulatory) effects of thermal pretreatment on non-acclimated inoculum. Finally, for both acclimated and non-acclimated scenarios, one set of control (without pretreatment) and one set of blank digesters (no sludge sample, only inoculum) were included in the experiment. Therefore, a total of 78 batch anaerobic digesters including triplicates (45 mesophilic and 33 thermophilic digesters) were set up and operated simultaneously. The total and effective volume of each digester were 160 and 80 mL, respectively. The photo of the batch anaerobic digesters is shown in Figure 4-3.

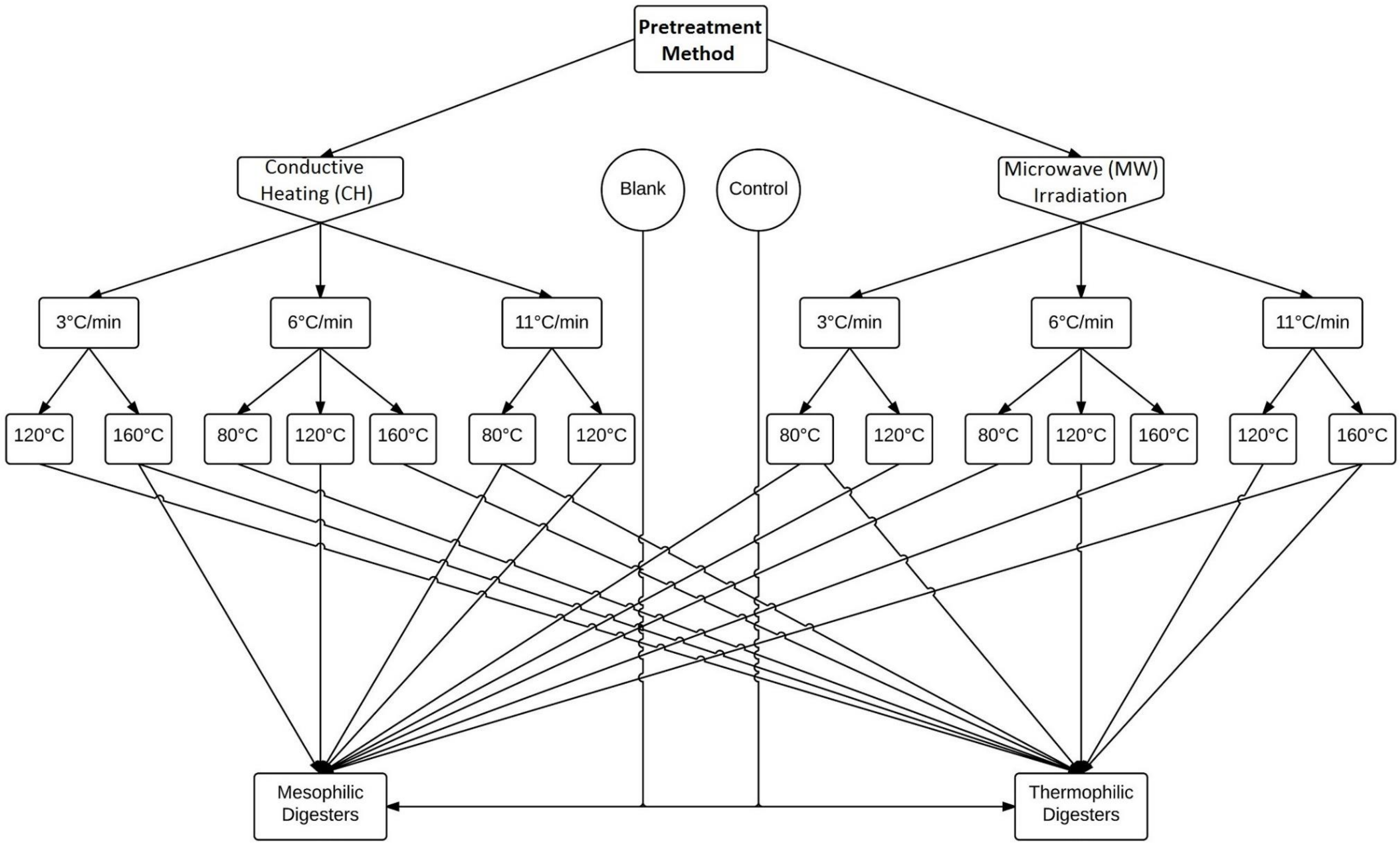


Figure 4-2. The experimental design of the batch anaerobic digestion study

Figure 4-3. The photo of the batch anaerobic digesters

The batch AD experiments were initiated by placing the pretreated or non-pretreated substrate and acclimated or non-acclimated inocula into the bottles. One of the main parameters affecting the performance of batch anaerobic digesters is the food to microorganism ratio (F/M) which is calculated by dividing the g of VS in the feed substrate to the g of VS in the inoculum (Angelidaki et al., 2009; Neves et al., 2004). In this study, the F/M ratio was kept 2.1 ± 0.2 for all the digesters. The AD requires a pH within the range of 6.5-7.5 for optimum performance (Droste, 1996). Therefore, external alkalinity was added into each digester in form of potassium bicarbonate and sodium bicarbonate (each equivalent to 2000 mg/L of $CaCO_3$) to keep the pH of the digesters within the acceptable range. As suggested in the literature and in order to minimize the residual organic compounds already present in the inocula, the inocula was starved and degassed prior to the start of the assays (Angelidaki et al., 2009). After adding the inoculum and substrate to the digesters and before sealing the bottles, the digesters were purged with nitrogen gas for 2 minutes to remove the residual oxygen. The digesters were then sealed immediately with rubber septa and aluminum crimp caps. The excess pressure was then released by puncturing the septa with a needle connected to a manometer. Finally, the mesophilic and thermophilic digesters were placed in two separate incubators/shakers (Innova 44R) set at 90 rpm and a temperature of 35°C and 55°C, respectively.

4.2.4. Analytical procedures

4.2.4.1. Biogas analysis

The biogas production was measured by puncturing the rubber septum with a syringe connected to a manometer. The biogas determination was done every 24 h initially, but when its production slowed down, the measurements were taken once every two to three days until. Finally, the volume of the biogas generated in the blank digesters (containing only inoculum) was subtracted from the biogas produced by main digesters in order to calculate the biogas yields from the substrates only. The volume of produced biogas was also corrected to the STP condition (0°C and 1 atm). Quantifying the biogas composition was conducted by measuring the percentage of methane, oxygen, nitrogen and carbon dioxide in the digester headspace using an Agilent 7820A Gas Chromatograph (GC) equipped with a thermal conductivity detector (oven, inlet and outlet temperatures: 70, 100 and 150°C, respectively) and a three meter Agilent G3591-8003/80002 packed column (2 mm internal diameter (ID), divinylbenzene stationary phase). The GC utilized helium as the carrier gas (flow rate: 25 mL/min) provided by Air Liquide Co. (Kelowna, BC.). The composition of the gas mixture used to calibrate the GC was as follows: 6.82% N_2, 19.90% CO_2, and 73.28% CH_4. For the analysis, 0.5 mL of the biogas was withdrawn from the gas line attached to the Tedlar® bag with an Agilent gas-tight syringe. The collected gas was then injected into the GC inlet septum and the punctured section of tubing was cut-off after sampling (to prevent biogas leakage). The composition of biogas was determined assuming that the percentage of gases other than methane, oxygen, nitrogen and carbon dioxide is negligible in the digester headspace.

4.2.4.2. Volatile fatty acid analysis

Total VFAs (summation of acetic, propionic and butyric acids) were measured by injecting filtered supernatant samples into an Agilent 7890A GC equipped with a capillary column (Agilent 19091F-112, HP-FFAP polyethylene glycol TPA column length x ID: 25 m, 320 μm) and a flame ionization detector (carrier gas flow rate: 25 mL He/min, oven, inlet and outlet temperatures: 200, 220 and 300°C, respectively). Following the procedure proposed by Ackman (1972), iso-butyric acid was used as the internal standard (Ackman, 1972). During sample preparation for VFA analysis, the filtration of the pretreated or un-pretreated sludge samples was done using 0.22 μm membrane filter after filtering with 0.45 μm membrane filter. To monitor the accumulation/consumption of VFAs within the digesters, 1.5 mL of the digester content was sampled using a 2 mL syringe. After centrifuging the samples, they were filtered through 0.22 μm centrifuge tube filters (Costar™ Centrifugal, Spin-X, Fisher Scientific, Inc.).

4.2.4.3. Other analyses

The TS and VS concentrations were determined following the procedure described by the Standard Methods, sections 2540 B and 2540 E, respectively (APHA, 2005). Alkalinity measurement was done using the procedure described by the Standard Methods, section 2320B. The concentration of the ammonia nitrogen (NH_3 (aq) and NH_4^+) was determined using an ammonium selective electrode connected to a pH/ion meter (Accumet Excel XL25 dual channel). In order to reduce the biological activity in the sample and also preventing the color interference, the ammonia and alkalinity analyses were performed on the supernatant of the samples centrifuged at 10,000 rpm for 20 min. The representative calibration curve for the ammonia ($NH_3 - N$) determination is provided in Appendix A (Figure A- 6).

4.2.5. Statistical analysis

Statistics including mean and standard deviation were calculated for the collected data using Microsoft Excel 2010. The statistically significant effects of the independent variables (pretreatment method, final temperature and heating ramp rate) were evaluated via multi-factor ANOVA considering 95% confidence level ($\alpha = 0.05$) using Minitab Software 17. The Pearson correlation coefficient (PCC) at 95% confidence level ($\alpha = 0.05$) was also used as a measure of the linear dependence degree between the variables whenever correlation (dependence) analysis was required.

4.3. RESULTS AND DISCUSSION

4.3.1. Mesophilic batch anaerobic digestion study

4.3.1.1. Effect of pretreatment parameters on mesophilic biogas production

Operation of the batch digesters was continued until no biogas production was observed. Figure 4-4 compares the cumulative biogas yields (at STP of 0°C, 1 atm) of all mesophilic batch anaerobic digesters during the total 59 days operation period. The biogas production rate of the non-acclimated mesophilic digesters at the first 25 days was significantly lower than that of the acclimated ones (Figure 4-4) confirming that inoculum acclimation prevents and/or lessens acute inhibition and lag phase at the beginning of the digester operation. Monitoring the VFAs concentration and pH of the digesters also confirmed the inhibition of methanogens and accumulation of VFAs in non-acclimated digesters (Figure 4-5). As seen in Figure 4-5 (a), the maximum VFAs concentration in acclimated and non-acclimated digesters was achieved at 3rd and 9th days of operation, respectively. It is also discernible that after 25 days, the concentration of the VFAs in acclimated digesters was significantly low (less than 50 mg/L in most of the acclimated digesters) confirming that the majority of VFAs in acclimated digesters were consumed during the first 25 days. However, in the case of non-acclimated digesters, the results showed that after 25 days, the concentration of VFAs was more than 1400 mg/L. The VFAs concentration results are in agreement with the results of pH as the minimum of pH was observed at 3rd and 9th days of operation in acclimated and non-acclimated digesters, respectively (Figure 4-5 (b)). It should be mentioned that due to the availability of enough alkalinity in all digesters, the pH remained above 6.8 throughout the digestion.

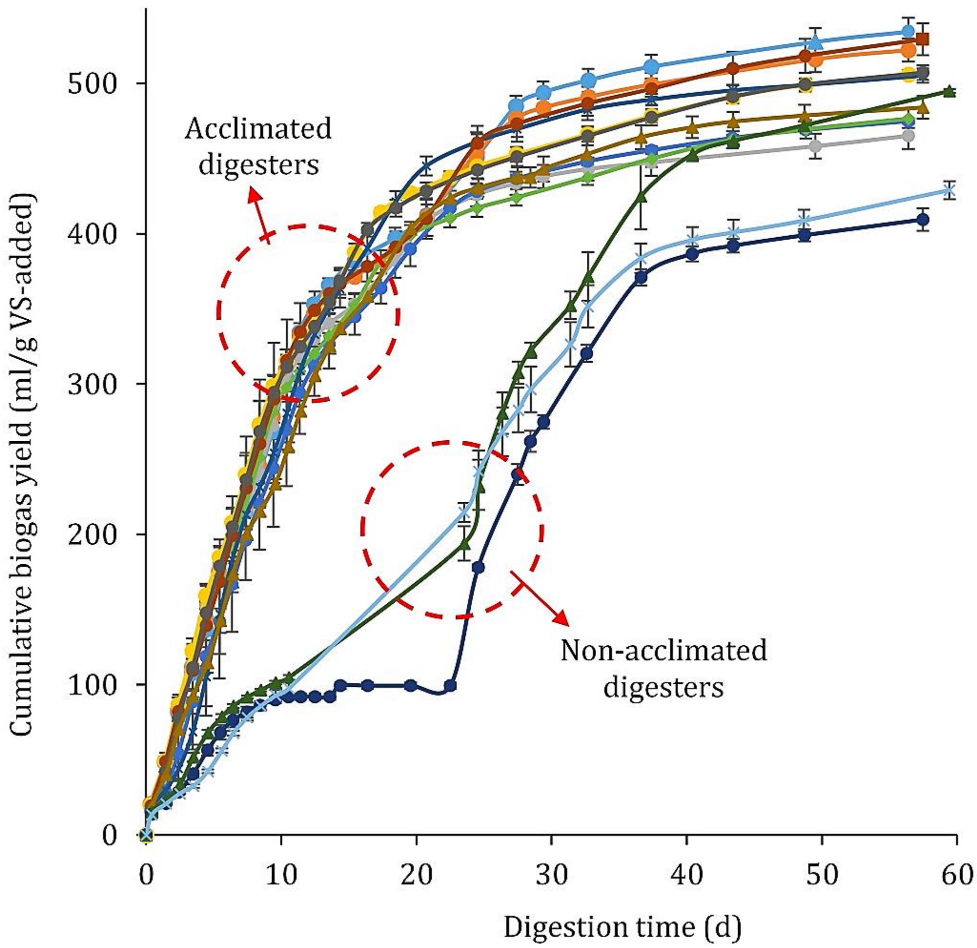
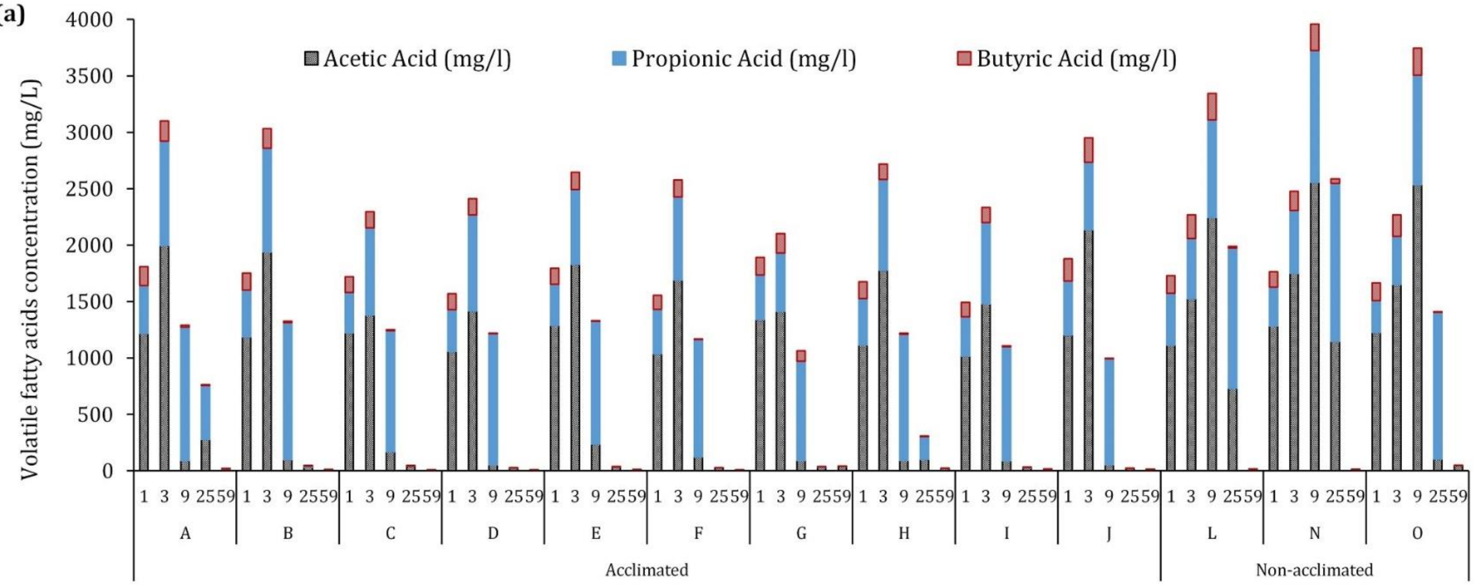
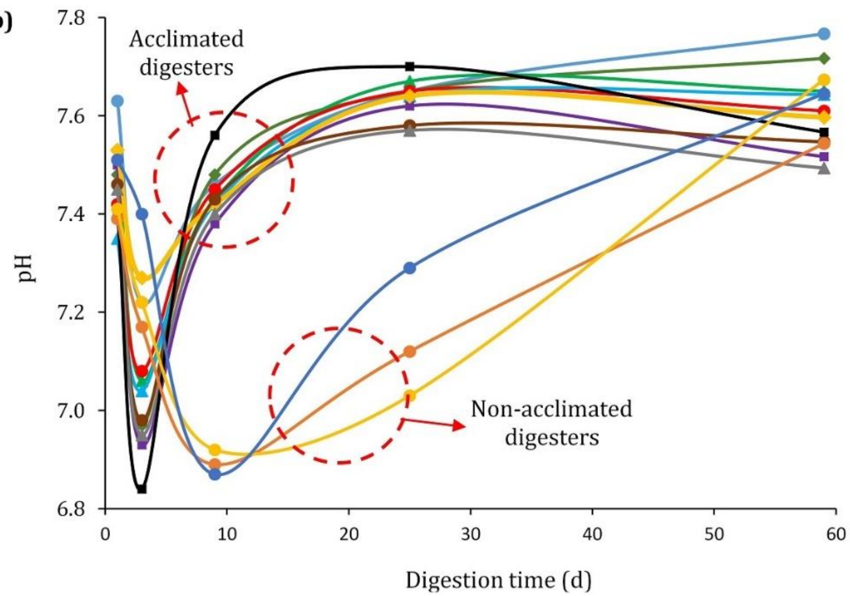


Figure 4-4. The cumulative biogas yield in mesophilic digesters at the STP of 0°C, 1 atm

(a)



(b)



Inoculum	Label	Method	Ramp Rate (^oC/min)	Temperature (^oC)
Acclimated	A	MW	3	80
	B	MW	6	80
	C	MW	6	160
	D	MW	3	120
	E	MW	11	160
	F	CH	11	120
	G	CH	3	160
	H	CH	11	80
	I	CH	6	120
	J	Control		
Non-acclimated	L	Control		
	N	MW	3	80
	O	MW	11	160

Figure 4-5. The changes in (a) volatile fatty acid concentration, and (b) pH of the mesophilic digesters during the operation period

The 59-d (ultimate) and 25-d biogas yields (per g VS-added) and their relative (to control) improvement in both acclimated and non-acclimated mesophilic digesters are illustrated in Figure 4-6. Considering each acclimated digester with positive biogas yield improvement (A, B, D, G, H and I), the improvement in ultimate biogas yield was higher than that of 25-d biogas yield except for the digester “G” which was pretreated at 160°C and 3°C/min (Figure 4-6 (b)). According to the solubilization results, the DWSC samples pretreated at 160°C and 3°C/min resulted in the maximum COD, sugar, protein and HA solubilization ratios (Figure 3-5, Figure 3-6, and Figure 3-7). Therefore, the higher improvement in 25-d biogas yield of this digester (compared to its improvement in ultimate biogas yield) can be explained by the availability of high concentration of soluble organic matters which were consumed during the first 25 days of the digester operation.

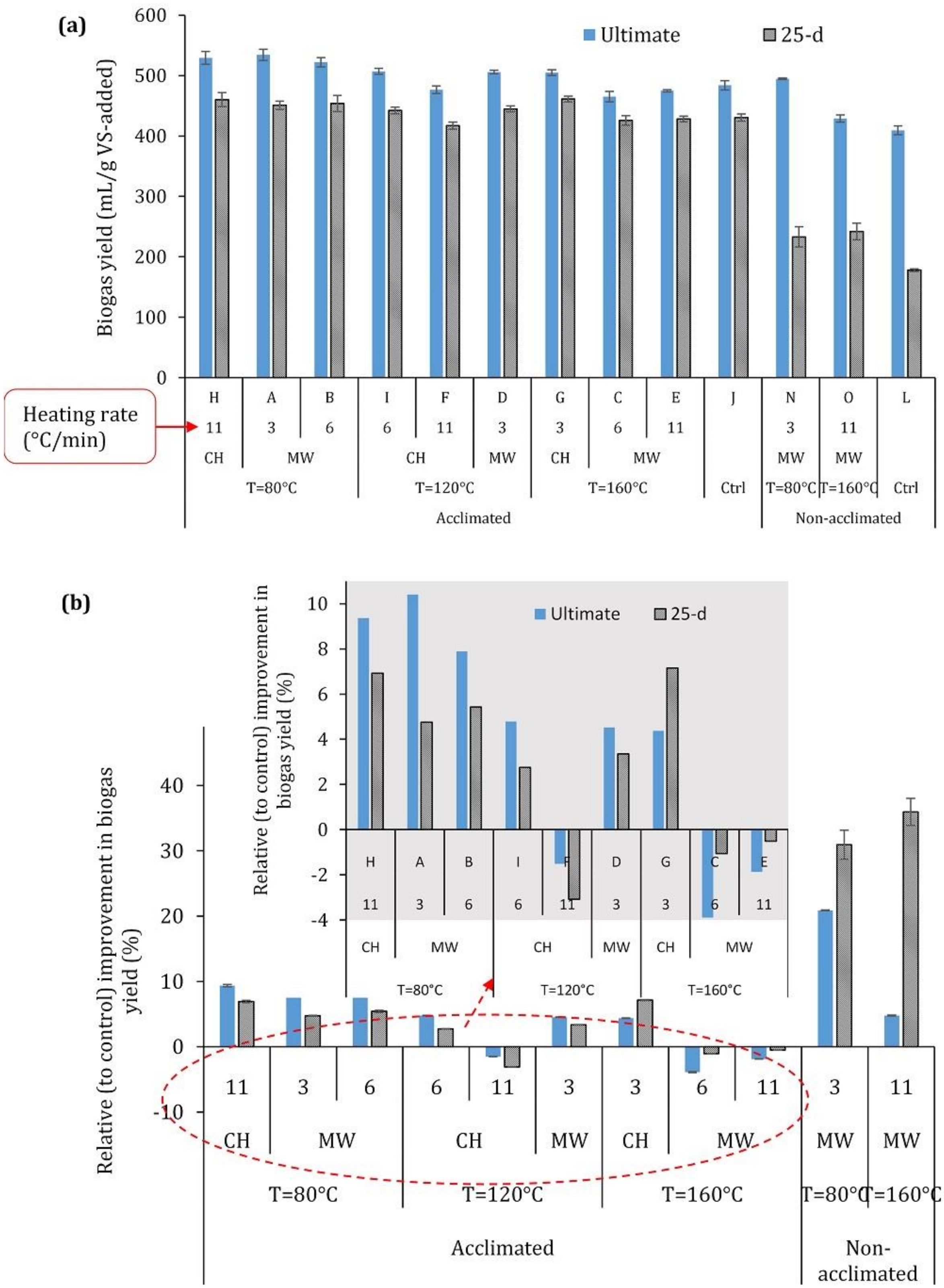


Figure 4-6. The mesophilic biogas yield and its relative (to control) improvement (adapted from Hosseini Koupaie & Eskicioglu (2016))

Interestingly, among the acclimated digesters, the improvement (over the acclimated control digester) in ultimate biogas yield was greater for those with the pretreatment temperature of 80°C (A, B and H). The ultimate biogas yield in the digesters “A”, “B” and “H” was 10.4, 7.9 and 9.4% higher than that of the acclimated control digester. On the other hand, the biogas production of the digesters exposed to the highest pretreatment temperature and/or heating rate (C, E and F) was less than that of the acclimated control digester resulted in the negative improvement values (Figure 4-6 (b)). The results of the analysis of variance also proved the statistically significant effects of final temperature (p-value = 0.000<0.05) and heating ramp rate (p-value = 0.000<0.05) on ultimate biogas production. According to the obtained results, increasing the intensity of the thermal pretreatment (final temperature and heating rate) had an adverse effect on biogas production, as the lowest temperature (80°C) and heating rate (3°C/min) was found to be the optimum thermal pretreatment condition. These results are more remarkable when taking into account that no statistically significant effect of pretreatment method, CH vs. MW, (p-value = 0.322>0.05) on biogas yield was observed in this study. In agreement with these results, Climent et al. (2007) reported the positive effects of low-temperature (70°C) sludge thermal pretreatment on thermophilic biogas production, while no effect of high temperature (134°C) pretreatment was observed. The authors` conclusion regarding the comparison between MW and CH methods is however arguable since their experiments were not conducted under identical thermal profiles for MW and CH (Climent et al., 2007).

As shown in Figure 4-6, among the non-acclimated digesters (L, N and O), both 25-d and ultimate biogas yields of those fed with pretreated DWSC (N and O) was higher than that of the non-acclimated control digester (L) which proves the positive effect of thermal pretreatment on performance of non-acclimated anaerobic digesters. It is also clear that the 25-d biogas yield improvement (over the non-acclimated control) was higher for the digester with the most intense thermal pretreatment temperature and heating rate (O). According to the solubilization study, the pretreatment condition of 160°C and 11°C/min (used as substrate for the digesters "O") resulted in higher soluble organics (Figure 3-5, Figure 3-6, and Figure 3-7 in Chapter 3) compared to the pretreatment condition of 80°C and 3°C/min (used as substrate for the digesters "N"). Therefore, the higher biogas production observed after 25 days in the digester "O" compared to that of the digester "N" can be explained by the availability of more readily biodegradable compounds released after pretreatment. However, the digester with the lowest pretreatment temperature and heating rate (N) showed significantly higher ultimate biogas yield improvement (20.9%) compared to only 4.8% improvement achieved in the digester with highest pretreatment temperature and heating rate (O). These observations suggest that initially (at the first 25 days), increasing the pretreatment intensity had a higher positive effect on the biogas production, however greater improvement in ultimate biogas production was achieved when lower pretreatment intensity was applied. These findings (obtained from the non-acclimated digesters) are in agreement with the results of acclimated digesters in which less intense thermal pretreatment (80°C and 3°C/min) was found to be more effective in terms of increasing ultimate biogas production.

Figure 4-7 (a) shows the contour plot of ultimate biogas yield as a function of pretreatment temperature and heating rate. The main effect plots of ultimate biogas yield vs. pretreatment method, final temperature and heating ramp rate are also presented in Figure 4-7 (b). As seen in Figure 4-7, increasing the final temperature and heating rate resulted in a decrease in the ultimate biogas yield. The average ultimate biogas yield obtained was 515.1, 498.2 and 493.6 $\frac{mL}{g\ VS-added}$ for the heating rate of 3, 6 and 11°C/min, respectively and 528.6, 496.6 and 481.7 $\frac{mL}{g\ VS-added}$ for the final temperature of 80, 120 and 160°C, respectively (Figure 4-7 (b)). The findings of this research suggest that as the intensity of CH or MW pretreatment increase, some of the organic compounds that were denatured during thermal pretreatment lose their biogas (methane) potential even under acclimated inoculum conditions. As reported in the literature, there are high molecular weight recalcitrant compounds such as melanoidin which are produced as a result of polymerization of low molecular weight intermediates, such as carbohydrates and amino compounds during Maillard reaction at elevated temperatures (Dwyer et al., 2008; Nursten, 2005; Penaud et al., 2000). In agreement with this finding, Shahriari et al. (2010) reported the formation of refractory compounds during MW pretreatment of whole organic fraction of municipal solid waste at 175°C. The authors observed a decrease in anaerobic biogas production at high temperature MW pretreatment compared to waste pretreatment at lower temperatures. It was also reported by the authors that the MW-pretreated samples at 175°C displayed a dark brown color (an indicator for melanoidins production), opposed to the lighter color of the controls and other pretreatment samples (Shahriari et al., 2010). Similarly, in the present research, a significant color difference between the non-pretreated DWSC samples and those pretreated at 160°C was observed (Figure 4-8). The representative Anderson-Darling test normality plot and the representative constant variance test results for the mesophilic ultimate biogas yield data are presented in Appendix B (Figure B- 2) and Appendix C (Figure C- 2), respectively.

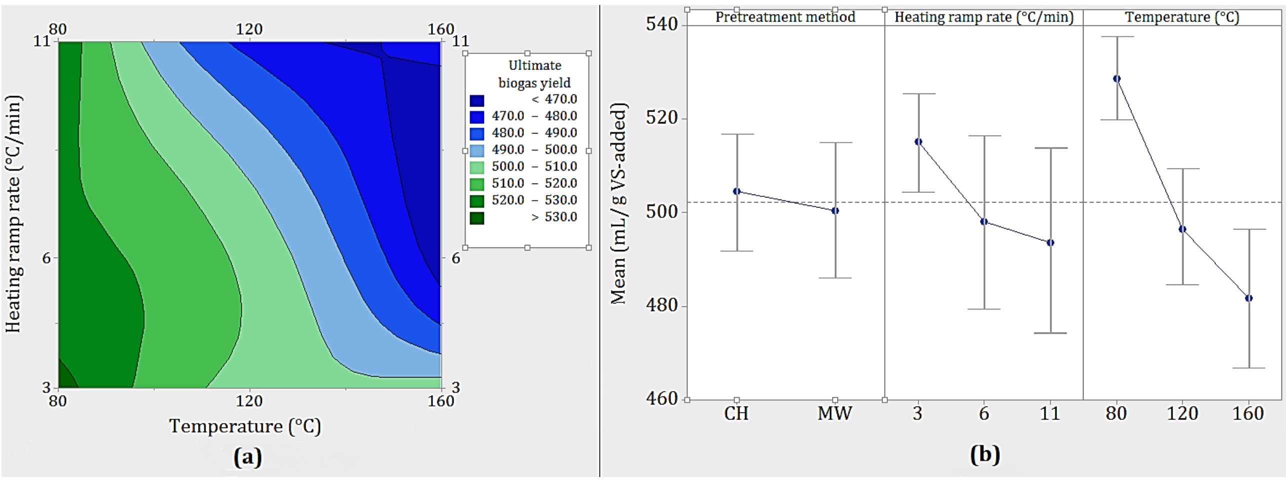


Figure 4-7. The effect of pretreatment parameters on the ultimate mesophilic biogas yield, (a) the contour plot and (b) the main effect plot (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

Figure 4-8. The color difference between the control (non-pretreated) and thermally pretreated DWSC samples, (a) control sample; (b) MW-pretreated sample at 160°C

4.3.1.2. Effect of pretreatment parameters on biodegradation kinetic rate

The results of cumulative biogas production (Figure 4-4) show that more than 84% of ultimate mesophilic biogas production (after 59 days) in the acclimated digesters was generated within first 25 days. Monitoring the VFAs concentration (Figure 4-5) also shows that the majority of the VFAs were utilized during 25 days after setting up the acclimated digesters. Therefore, the first 25 days of the operation period was considered for biodegradation kinetics study. As discussed in Chapter 2 (Section 2.6.4), previous studies have proven that first-order kinetic reaction can represent the biogas production as well as the organics (VS, TCOD) biodegradation during AD process (Eskicioglu & Ghorbani, 2011; Eskicioglu et al., 2006; Hosseini Koupaie et al., 2014). According to the calculated coefficient of determination (R^2) values (0.9885 ± 0.0053), the first-order reaction model well describes the kinetics of VS biodegradation (biogas production) in all acclimated mesophilic digesters.

Figure 4-9 compares the specific biodegradation rate constant obtained from fitting first-order reaction model to the VS time-course profiles (during 25 days of digesters operation) in the acclimated digesters. As seen in Figure 4-9, increasing pretreatment final temperature improves the rate of VS biodegradation. This observation was also proven to be statistically significant (p-value = 0.000<0.05). The analysis of variance also confirmed the significant effect of heating rate on k values (p-value = 0.028<0.05), as increasing the heating rate (decreasing pretreatment time) resulted in a decrease in VS biodegradation rate. No statistically significant effect of pretreatment method on VS biodegradation rate constant was however found (p-value = 0.142>0.05). As obvious from Figure 4-9, the digesters with the pretreatment conditions of

“80°C and 11°C/min” and “160°C and 3°C/min” demonstrated the highest and lowest biodegradation rates, respectively. These results were also proven by the contour plot of VS specific biodegradation rate constant as a function of pretreatment temperature and heating rate (Figure 4-10 (a)). According to the results of the main effect test (Figure 4-10 (b)), the average of k values in the digesters fed with DWSC pretreated at the temperatures of 80, 120 and 160°C were 0.080, 0.091 and 0.100 d^{-1}, respectively. The heating rates of 3, 6 and 11°C/min also resulted in an average k values of 0.092, 0.091 and 0.088 d^{-1}.

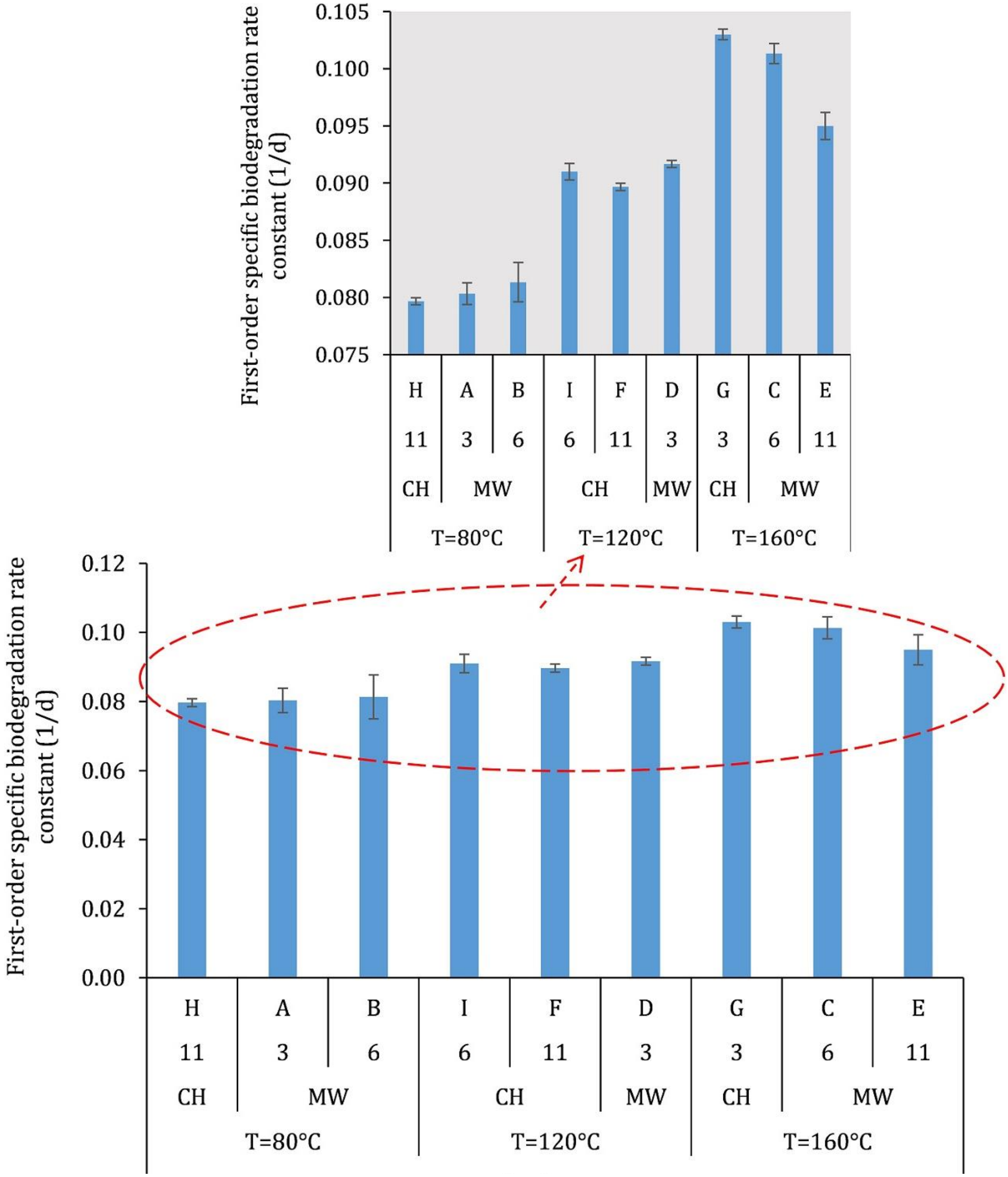


Figure 4-9. The effect of thermal pretreatment on first-order specific biodegradation rate constant in mesophilic digesters

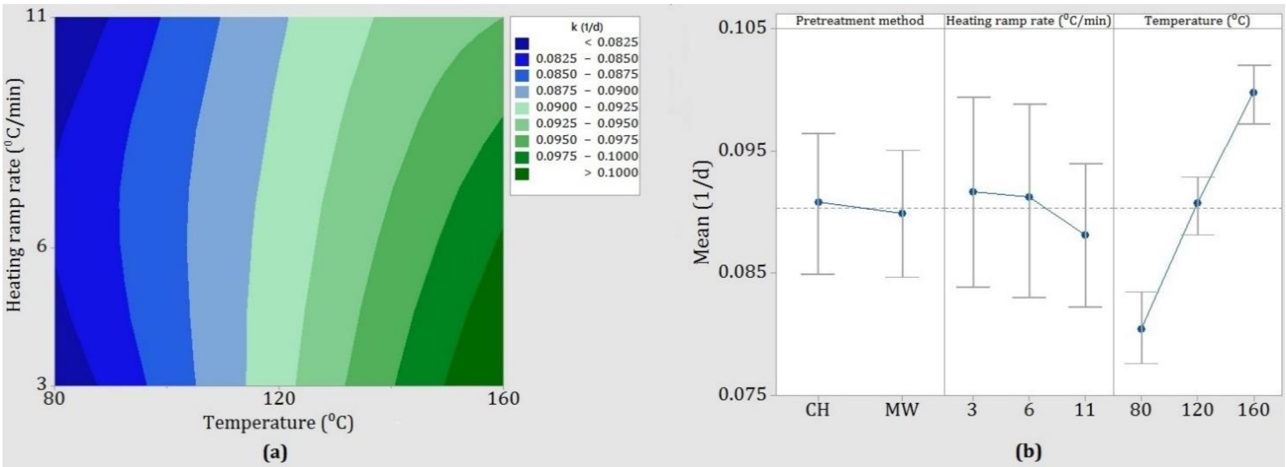


Figure 4-10. The effect of pretreatment parameters on specific biodegradation rate constant in mesophilic digesters, (a) the contour plot and (b) the main effect plot (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

Taking into account that the trend of temperature and heating rate effects on sludge solubilization (discussed in Chapter 3) and on the VS biodegradation rate (presented in the current chapter) are similar (increasing pretreatment temperature and heating rate had positive and negative effect, respectively), a correlation analysis was performed to evaluate whether there is any statistically proven correlation between the sludge solubilization and VS biodegradation rate. Figure 4-11 demonstrates the correlation between the *k* values determined from the acclimated digesters and sludge solubilization ratios (COD and biopolymers). As shown in Figure 4-11, the PCC determined for *k* vs. COD, sugar, protein and HA solubilization ratios were all above 0.8 at the corresponding p-value below 0.05. These results suggest a highly significant correlation between the VS biodegradation rate and COD/biopolymer solubilization ratio.

The overall findings from solubilization tests and anaerobic digestion studies (biogas production and organics biodegradation kinetic results) can be explained by the fact that increasing pretreatment temperature increases the amount of soluble organics which can be utilized faster during anaerobic digestion and then results in an increase in biodegradation rate. Increasing pretreatment heating rate which has negative effects on organics solubilization however decreases the organics biodegradation rate. In addition, increasing pretreatment intensity (increasing temperature and heating rate) most likely generates some refractory compounds with less biogas potential and therefore decreases the ultimate biogas production. On the other hand, the application of either MW or CH methods under identical thermal profiles yields similar results in terms of sludge solubilization, biogas production and VS biodegradation rate, revealing no statistically significant difference between MW and CH.

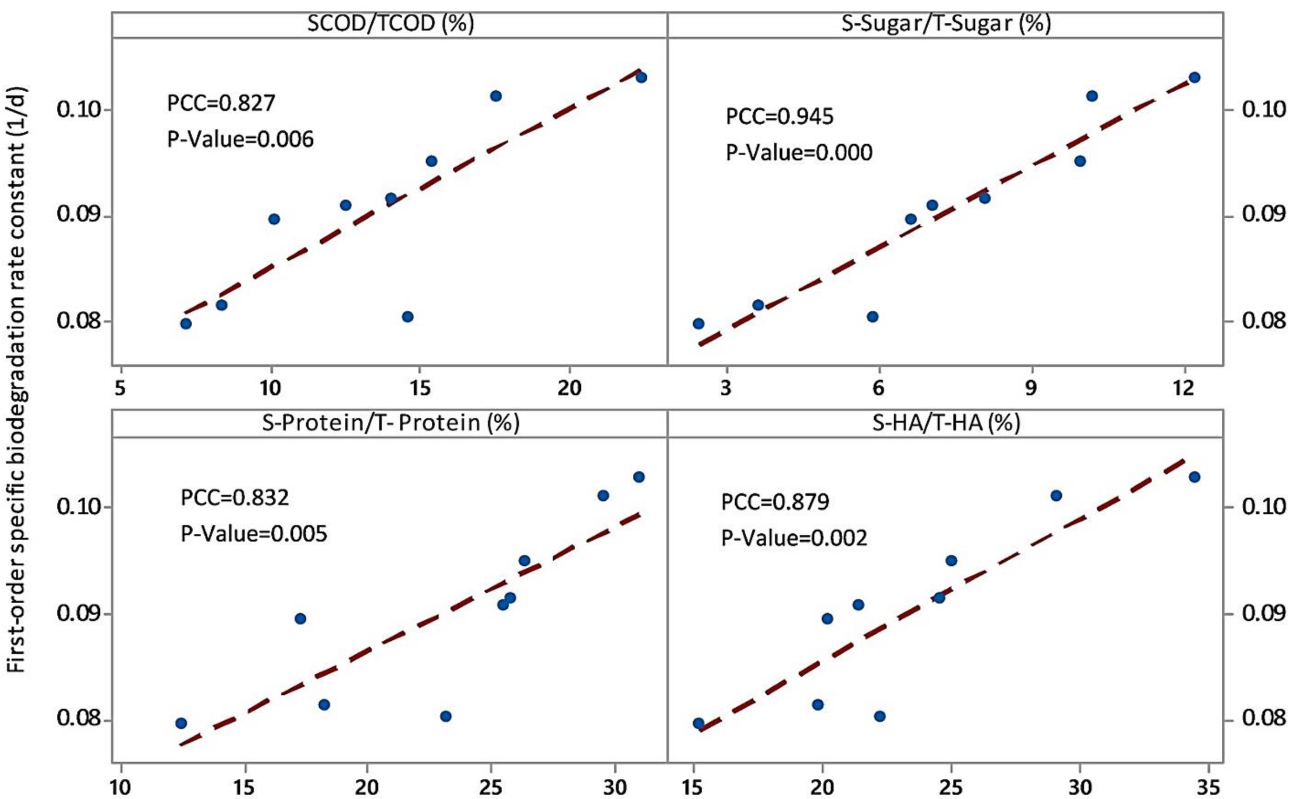


Figure 4-11. The correlation between specific biodegradation rate constant in mesophilic digesters (k) and sludge solubilization ratio (PCC: Pearson correlation coefficient)

4.3.2. Thermophilic batch anaerobic digestion study

4.3.2.1. Effect of pretreatment parameters on thermophilic biogas production

The thermophilic digesters were operated for the same period as that of mesophilic digesters (approximately 2 months). Figure 4-12 shows the daily cumulative biogas yields (at STP) of the thermophilic digesters during the entire operation period. As per Figure 4-12, among the pretreated thermophilic digesters, the digesters fed with DWSC samples pretreated at 160°C achieved the lowest initial (during the first week) cumulative biogas yield.

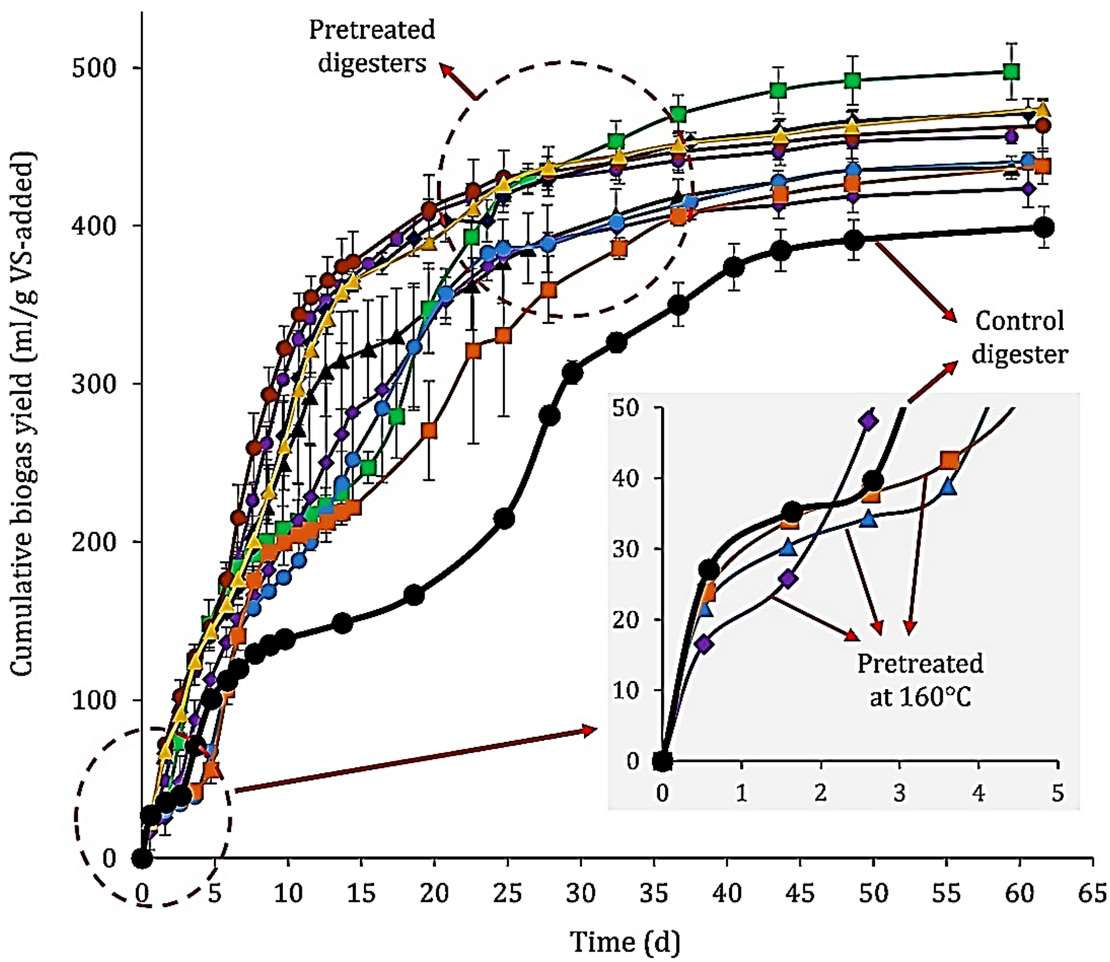


Figure 4-12. The cumulative biogas yield in thermophilic digesters at the STP of 0°C, 1 atm

Figure 4-13 compares the ultimate biogas yield of the thermophilic digesters. As per Figure 4-13, the ultimate biogas yield of all the thermophilic digesters fed with pretreated DWSC was greater than that of the control (non-pretreated) digester. Among the pretreated digesters, the digesters fed with pretreated DWSC at 160°C demonstrated the lowest ultimate biogas yield (Figure 4-13).

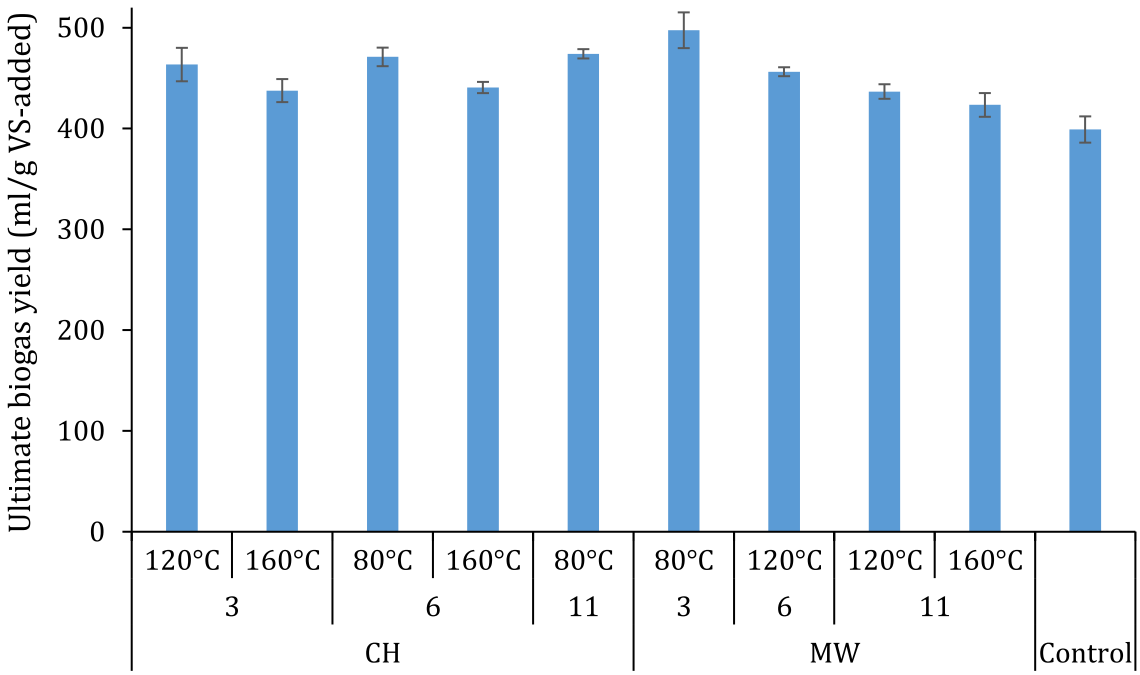


Figure 4-13. The ultimate biogas yield of the thermophilic digesters at the STP of 0°C, 1 atm

Figure 4-14 illustrates the relative (to control) improvement in ultimate biogas yield (per g VS-added) and ultimate methane yield (per g COD-added) in the thermophilic digesters fed with pretreated sludge. Among the pretreated digesters, the highest ultimate biogas yield (497.5 ± 17.7 $\frac{mL}{g\ VS-added}$) was achieved at the lowest heating ramp rate and final temperature (3°C/min and 80°C). This pretreatment scenario resulted in 24.7% and 22.6% relative (to control) improvement in ultimate biogas and methane yields, respectively (Figure 4-14). However, the lowest ultimate biogas yield (among the pretreated digesters) was 423.5 ± 11.8 $\frac{mL}{g\ VS-added}$ obtained under the most intensive pretreatment condition (maximum heating ramp rate and final temperature of 11°C/min and 160°C). These results again suggest that as the temperature of thermal pretreatment increases above a certain level, some of the organic compounds lose their biogas/methane potential even under acclimated-inoculum conditions.

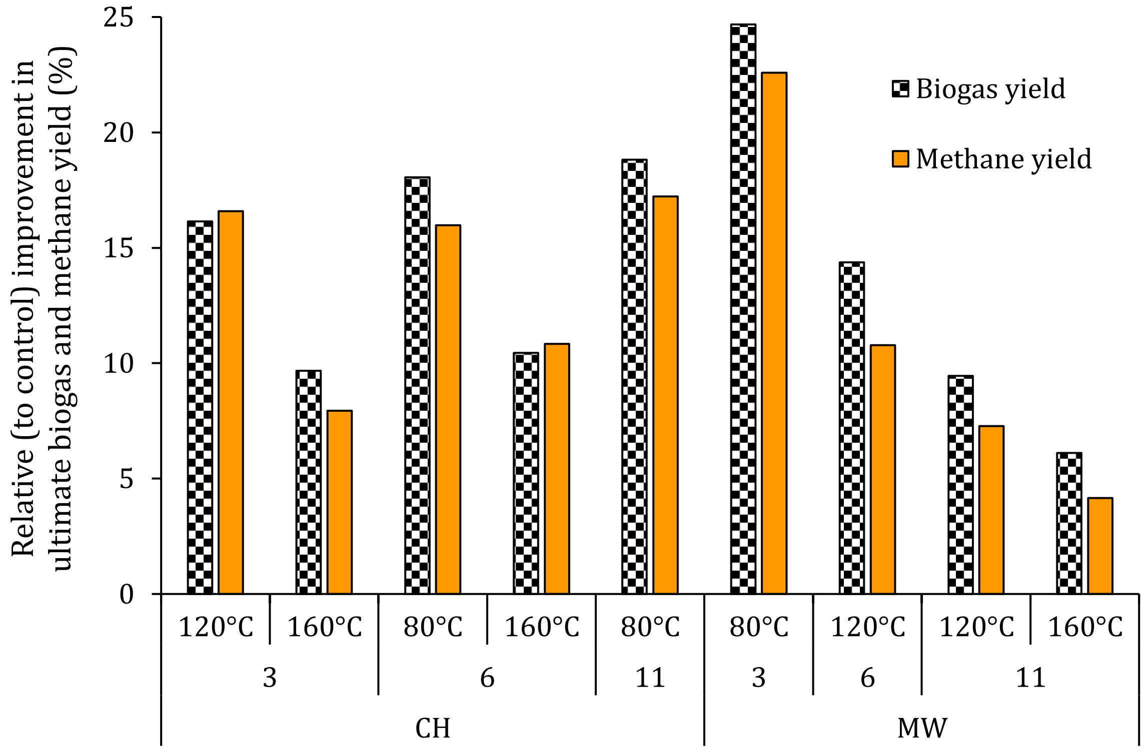


Figure 4-14. The relative (to control) improvement in ultimate biogas and methane yield

The changes in the VFA concentration and the pH of the thermophilic digesters during the operation period are shown in Figure 4-15 and Figure 4-16, respectively. It is worth noting that the VFA analysis was performed by measuring the concentration of acetic acid, propionic acid, and butyric acid since they are the major VFAs produced during the AD process. As seen in Figure 4-15, the maximum VFA concentration was achieved at the 3rd day of operation except for the control and 160°C-pretreated digesters. The results of monitoring the VFA concentration are compatible with the results of pH analysis (Figure 4-16). As observed from Figure 4-16, the minimum pH of the 160°C-pretreated digesters was observed at the 9th day of operation, while the minimum pH for the remaining digesters occurred at the 3rd day of operation. These observations coincide with the biogas production results (Figure 4-12) in which the initial biogas production (during the first week of operation) of the control and the 160°C-pretreated digesters were lower than that of the other digesters. These results can be explained by the fact that the activity of the acidogenic bacteria (that are responsible for VFA production leading to the pH drop) in 160°C-pretreated digesters was most likely affected by the inhibitory compounds generated at an elevated temperature of 160°C. In terms of the control digester however, the lack of high concentration of readily biodegradable (hydrolyzed) compounds (that are generated during sludge pretreatment) could be the reason for its low initial biogas production..

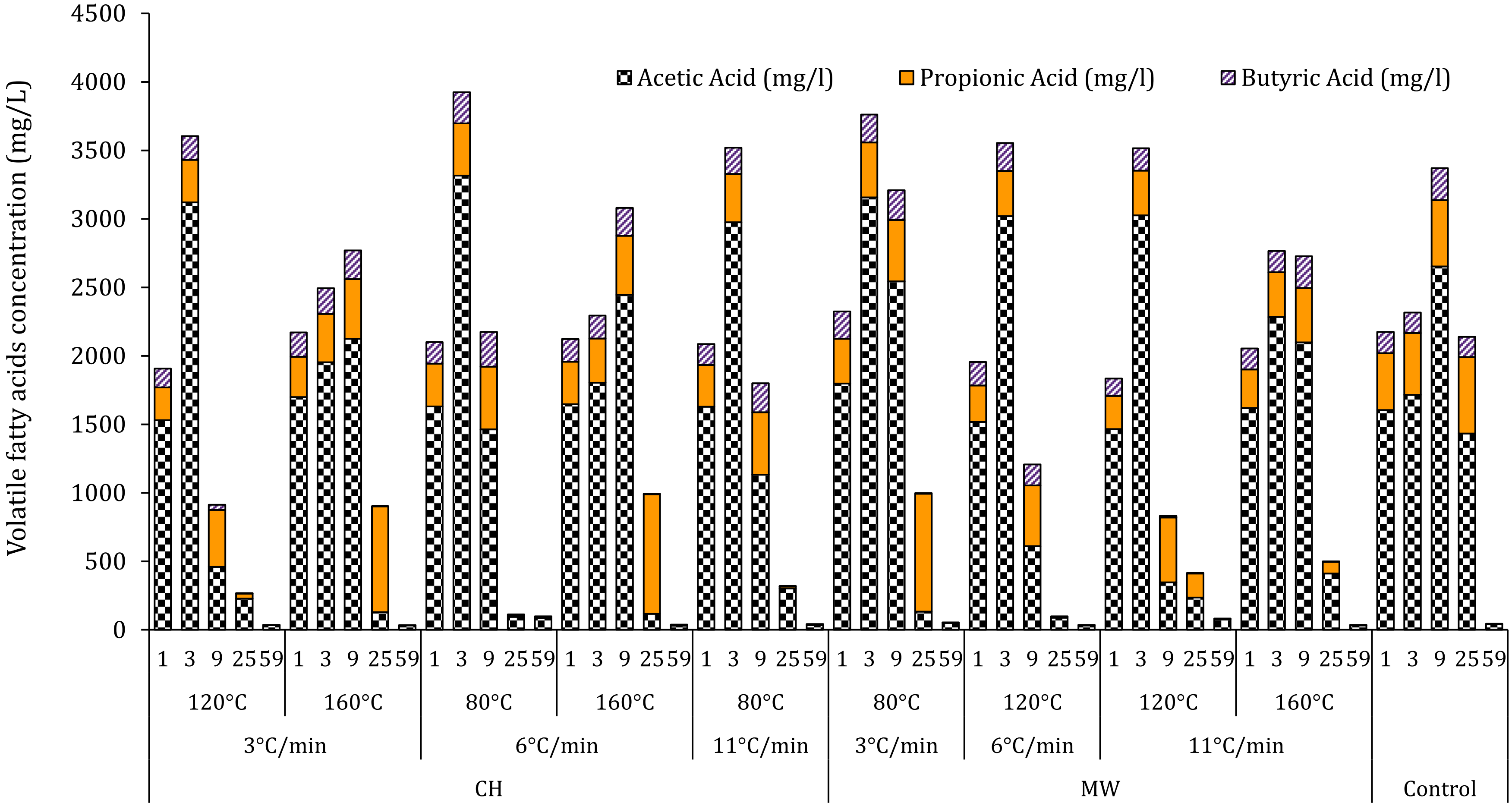


Figure 4-15. The changes in the concentration of volatile fatty acids in thermophilic digesters during the digestion period (d)

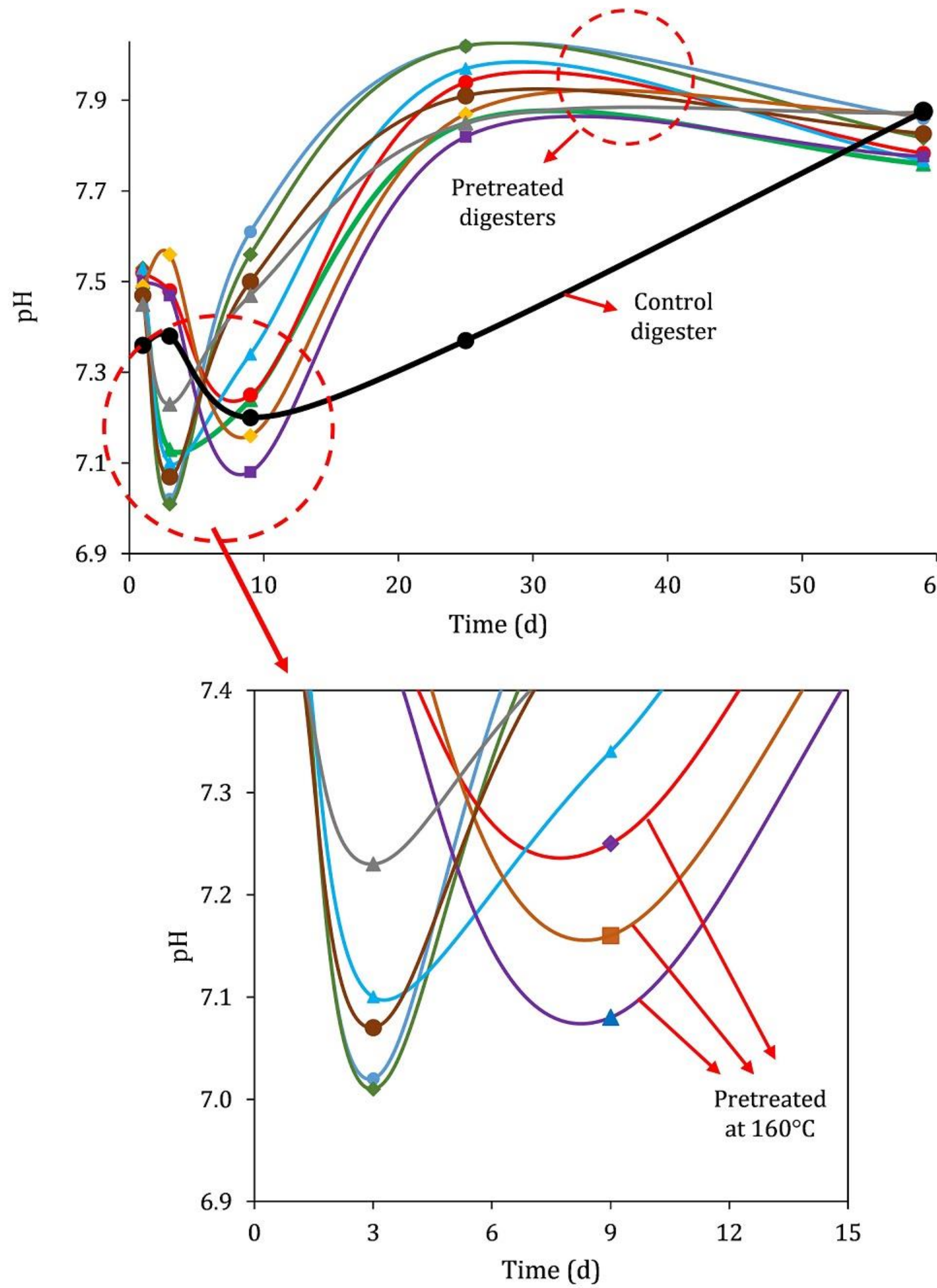


Figure 4-16. The changes in the pH of the digesters with time

The contour plots of the ultimate biogas and methane yield as a function of heating ramp rate and final temperature are presented in Figure 4-17 (a) and Figure 4-17 (b), respectively. As seen in Figure 4-17, increasing the heating ramp rate resulted in a decrease in ultimate biogas and methane yield. The ANOVA results confirmed the statistically significant effect of heating ramp rate on ultimate biogas (p-value = 0.004<0.05) and methane (p-value = 0.042<0.05) yield. The statistically significant differences in ultimate biogas (p-value = 0.000<0.05) and methane (p-value = 0.000<0.05) yield were also noticed among the digesters fed with DWSC samples pretreated at different final temperatures. The analysis of variance however proved no statistically significant effect of pretreatment method (CH vs. MW) on ultimate biogas (p-value = 0.403>0.05) and methane (p-value = 0.148>0.05) yield. These results are in agreement with the results obtained under mesophilic condition. The representative Anderson-Darling test normality plot and the representative constant variance test results for the thermophilic ultimate biogas yield data are presented in Appendix B (Figure B- 3) and Appendix C (Figure C- 3), respectively.

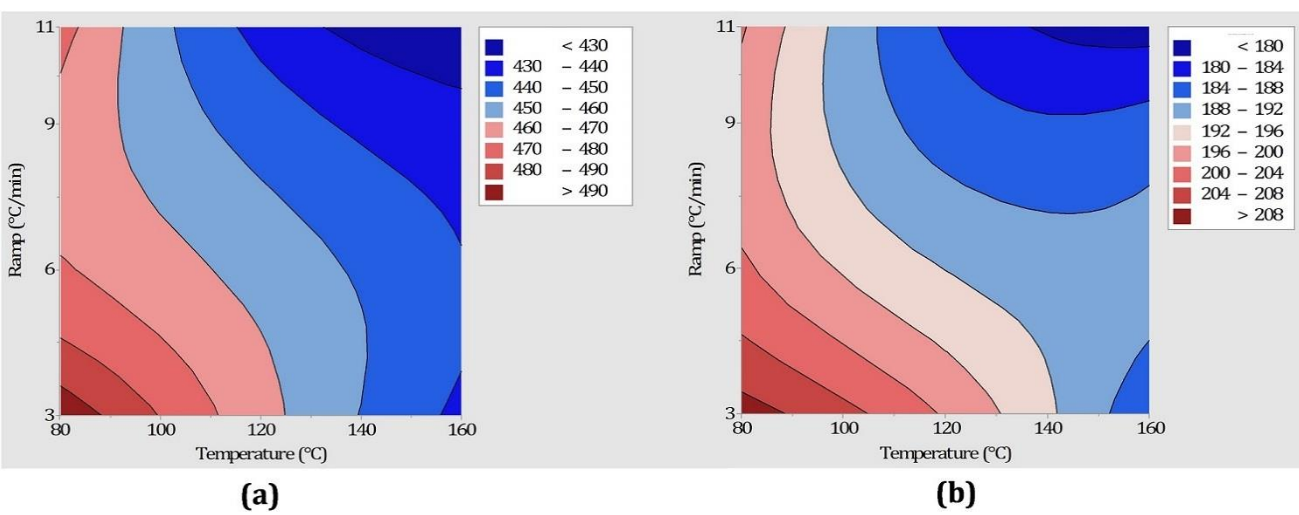


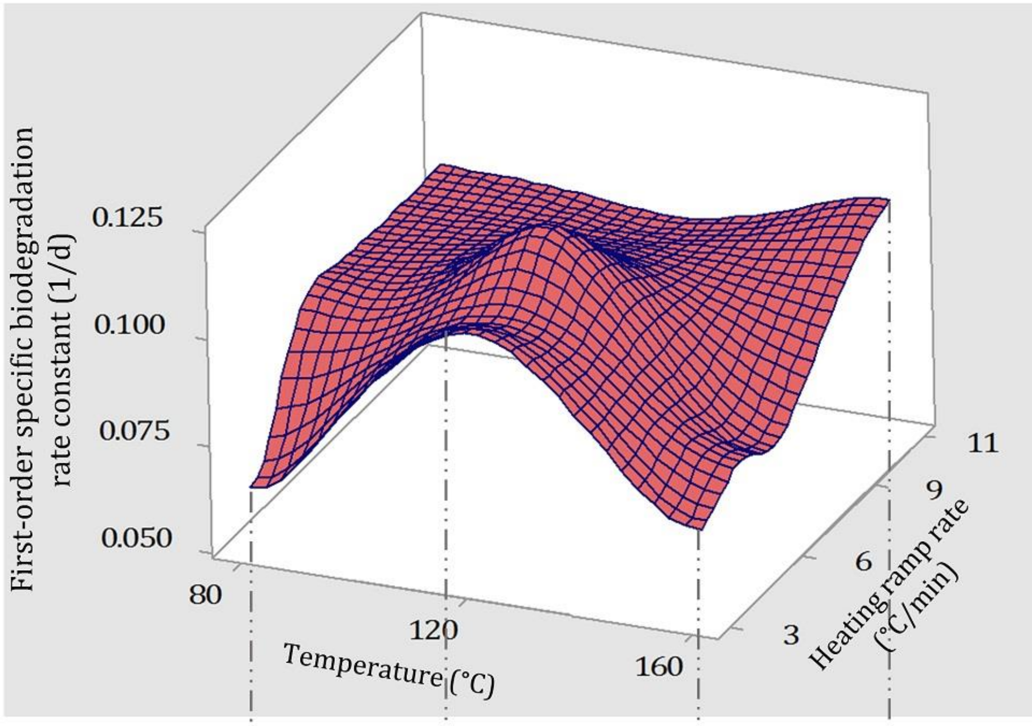
Figure 4-17. The contour plot of (a) ultimate biogas yield ($\frac{mL}{g\ VS-added}$) and (b) ultimate methane yield ($\frac{mL}{g\ TCOD-added}$) as a function of heating ramp rate and final temperature

As seen in Figure 4-17, increasing the pretreatment temperature (from 80°C to 160°C) had an adverse effect on the ultimate thermophilic biogas and methane production. According to the results, the higher the intensity of the pretreatment is, the lower the biogas and methane production will be. According to the results of main effect test, the heating ramp rate of 3, 6 and 11°C/min resulted in the average ultimate biogas yield of 466.2, 456.1 and 444.8 $\frac{mL}{g\,VS-added}$ and the average methane yield of 198.0, 192.6 and 187.5 mL/g COD-added, respectively. The pretreatment temperatures of 80°C, 120°C and 160°C also resulted in the average biogas yield of 480.9, 452.2 and 434.0 $\frac{mL}{g\,VS-added}$ and average methane yield of 203.0, 191.0 and 184.2 $\frac{mL}{g\,TCOD-added}$, respectively.

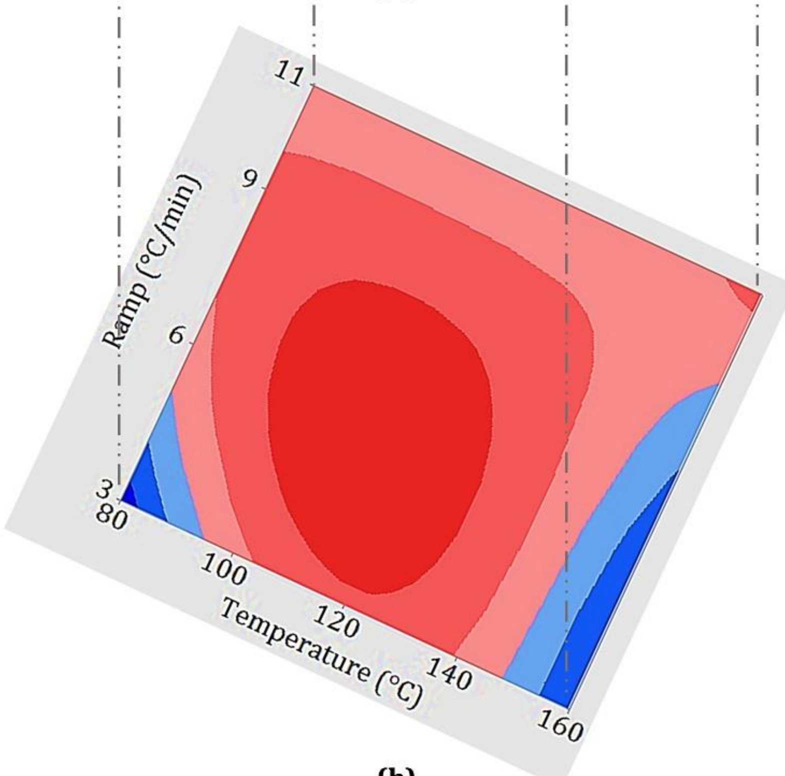
4.3.2.2. Effect of pretreatment parameters on biodegradation kinetic rate

The results of biodegradation kinetics study show higher biodegradation rate constant for all the digesters fed with pretreated DWSC compared to that of the control digester. The maximum specific biodegradation rate constant was obtained as 0.123 1/d under the heating ramp rate of 3°C/min and final temperature of 120°C. This resulted in 1.7 times higher biodegradation rate than that of the control digester. The analysis of variance proved statistically significant effect of pretreatment temperature on first-order biodegradation rate (p-value = 0.000<0.05) while there was no statistically significant effect of pretreatment method (p-value = 0.159>0.05) and heating ramp rate (p-value = 0.120>0.05).

The surface and contour plots of first-order specific biodegradation rate constant vs. heating ramp rate and pretreatment final temperature are illustrated in Figure 4-18 (a) and Figure 4-18 (b), respectively. As previously shown in Chapter 3 (Figure 3-8), the heating ramp rate of 3-8°C/min and pretreatment temperature of 120-160°C correspond to the domain of the maximum sludge solubilization. Figure 4-18 however shows that the maximum thermophilic biodegradation rate was achieved under the heating ramp rate and final temperature of 3-8°C/min and 100-130°C, respectively (the dark red region in Figure 4-18 (b)). Comparison of the thermophilic kinetics results with those of obtained under mesophilic condition reveals the interesting outcome that although the highest solubilization ratio obtained at 160°C resulted in the greatest mesophilic biodegradation rate constant due to the maximum concentration of readily biodegradable compounds, however, the refractory/inhibitory compounds which were most likely formed at 160°C had an adverse effect on biodegradation rate constant under thermophilic condition. Therefore the maximum biodegradation rate shifted from the highest temperature range (120-160°C) towards the lower temperature range of 100-130°C (Figure 4-18 (b)). Another notable finding is that the average k value of 0.08/d was achieved at both mesophilic and thermophilic conditions under a pretreatment temperature of 80°C. However, a pretreatment temperature of 160°C resulted in the average k values 0.1 and 0.08 d^{-1} under the mesophilic and thermophilic condition, respectively. These results are in agreement with the commonly known drawback of thermophilic AD which has the higher sensitivity to the operational and environmental changes along with potentially inhibitory by-products of sludge pretreatment compared to mesophilic AD.



(a)



(b)

Figure 4-18. The first-order thermophilic specific biodegradation rate constant vs. heating ramp rate and pretreatment temperature; (a) surface plot, (b) contour plot

4.4. SUMMARY

The outcomes of the batch AD study discussed in this chapter can be summarized as follows:

- Both pretreatment temperature and heating ramp rate were observed to have statistically significant effects on mesophilic and thermophilic AD performance (biogas production and solids removal), which validated the AD related part of the *Hypothesis I*.
- It was statistically proven that the two thermal pretreatment techniques (MW vs. CH) achieve similar mesophilic and thermophilic biogas production rate and extent as long as they are applied under identical thermal profiles (identical pretreatment temperatures and heating rate), which validated the AD related part of the *Hypothesis II*.
- Increasing the intensity of thermal pretreatment resulted in a decrease in anaerobic biogas production suggesting the formation of refractory compounds with less biogas/methane potential during pretreatment.
- It was observed that the greater organic solubilization ratios achieved via thermal pretreatment does not necessarily result in higher ultimate biogas production.
- In the tested temperature range of 80-160°C, the maximum mesophilic and thermophilic biodegradation rate was obtained as a result of sludge pretreatment under a final temperature of 160 and 120°C, respectively. This indicates that the highest pretreatment temperature does not necessarily correspond to the fastest thermophilic biodegradation.

- A statistically significant correlation between sludge solubilization ratio and VS biodegradation rate was proven under mesophilic condition.
- Thermophilic AD was found to be more sensitive to the inhibitory effect of thermal pretreatment at an elevated temperature of 160°C than mesophilic AD.
- Since no statistical significant effects of pretreatment method on AD performance were observed, in the case of practical (full scale) application, other criteria such as the overall input energy, ease of operation, capital cost, heat uniformity should be considered in the decision making process (to choose between MW and CH) rather than their effectiveness in terms of increasing the biodegradability of the municipal sludge.

CHAPTER 5. DESIGN AND FABRICATION OF A NOVEL ELECTROMAGNETIC SLUDGE PRETREATMENT SYSTEM AT A FREQUENCY OF 13.56 MHZ

5.1. INTRODUCTION

As previously discussed, the slow degradation of WAS which is mainly composed of microbial cells adhered together with EPS, has been known as the bottleneck of conventional AD of municipal sludge. This requires a long retention time and a large digester volume leading to high capital and operating cost (Eskicioglu et al., 2007a). Previous studies have shown that thermal pretreatment of WAS using EM heating prior to AD increases biogas production rate and extent (Eskicioglu et al., 2007b; Mehdizadeh et al., 2013; Tyagi & Lo, 2013; Vergine et al., 2014). However, the research contributions to the development and evaluation of EM sludge pretreatment techniques have almost exclusively been constrained to the commercial MW equipment operated at a frequency of 2.45 GHz (such as kitchen-type or bench-scale MW oven) (Chi et al., 2011; Hu et al., 2012; Ji Park et al., 2010; Kuglarz et al., 2013).

Although, MW heating which uses dielectric hysteresis to heat the load is most efficient at frequencies like 2.45 GHz, the implementation of sludge pretreatment at such frequencies has fundamental limitations in terms of; 1) the penetration depth of the field into the load volume, and 2) the energy efficiency of the MW power source. The limitation of the penetration depth is due to the fact that the EM field attenuates as it passes through the load resulting in non-uniform heating. This limitation would be very restrictive especially at full-scale applications at which large volumes of load need to be heated. It is also difficult to foresee how this type of heating method could be implemented in a large scale system, because the load would need to pass through structures that limit the depth of the sludge. Another drawback of MW sludge pretreatment is the low efficiency of MW power source at a frequency of 2.45 GHz. In general, power efficiency of EM sources reduces as frequency increases, and therefore designing and manufacturing a highly efficient power source at high frequencies such as 2.45 GHz can be very challenging.

Considering the limitations of the MW hydrolysis technique, new multi-disciplinary research was conducted to design, implement and evaluate a highly efficient EM heating system for pretreating TWAS that goes beyond the limitations of a single MW frequency of 2.45 GHz. This work has led to the implementation of a RF heating system at a frequency of 13.56 MHz. The frequency of 13.56 MHz was selected considering the electrical characteristics of TWAS. Among the electrical properties of the load, the conductivity and permittivity are the two important factors determining the principal mechanisms as well as the efficiency of EM heating. These characteristics were determined prior to this research by other researchers (Bobowski et al., 2012) in the School of Engineering at UBC.

In this chapter, different steps which were followed to design and fabricate a novel EM sludge pretreatment system at a frequency of 13.56 MHz is described. The main components of the custom-designed RF pretreatment system are introduced, and finally the energy-efficiency of the system is compared with that of the MW oven operated at a frequency of 2.45 GHz.

5.2. FREQUENCY SELECTION

5.2.1. Interaction of load and electromagnetic field

As stated earlier, the selection of the appropriate frequency at which the EM heating is applied is one of the key factors in designing an efficient sludge pretreatment system. In this regard, analysis of the load interaction with the EM field at different ranges of frequency is essential. It is also important to determine the principal mechanism(s) involved in heating at a given frequency. It was previously shown in Chapter 2 (Figure 2-18) that the composition of WAS is a heterogeneous mixture of different organic and inorganic compounds in which the bacterial cells are its main component. The bacterial cells are adhered together with EPS which is immersed in free (bulk) water. Most of the bacteria cells are encapsulated by a cell membrane. The bacterial cell membranes separate the extra- and intra-cellular media. Figure 5-1 illustrates a simplified equivalent circuit model that has been widely used in the previous studies for the modeling of plant and animal cells/tissues exposed to an EM field (Grimnes & Martinsen, 2014). From the structural point of view, there are similarities between the composition of the plant and animal tissues and that of WAS, since their main component is bacterial cells. Therefore, the proposed models in the literature for plant and animal cells/tissues can be used to characterize the interaction between WAS and EM field.

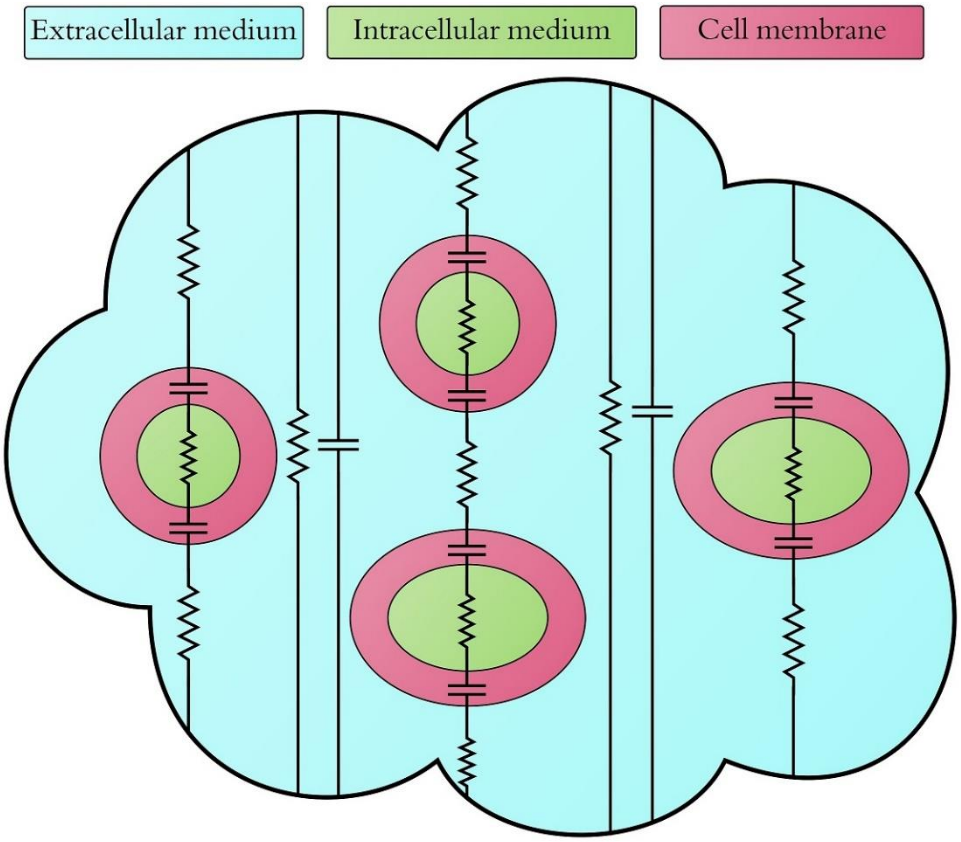


Figure 5-1. The simplified equivalent circuit model of bacterial cells exposed to an electromagnetic field

The EPS is negatively charged and surrounded by positively charged cations (Figure 2-18). Therefore, if two electrodes are inserted in, the electrical current can flow through the WAS as a result of the ionic characteristic of the extracellular medium. The current flow is frequency-dependent because of the heterogeneous structure of WAS in which each component carries different electrical characteristics. The intra- and extra-cellular media are electrolytes through which the ionic conduction current flows. The cell membrane however has much lower conductivity, functioning as a dielectric enclosed between two ionic media (intra- and extra-cellular media). Therefore, it can be modeled as a parallel plate capacitor though which the displacement current flows. Under DC or low ranges of frequency, the ionic conduction current through the extracellular medium dominates the total current flow between the electrodes. However, as the frequency increases, the displacement current passes through the cell membrane (which acts as a parallel plate capacitor) and the overall current in WAS will be composed of both the conduction and displacement currents. The intra- and extra-cellular media have different permittivity and conductivity resulting in the accumulation of charges at their interface (surface of cell membrane) which is known as Maxwell-Wagner effect.

The electrical characteristics of the municipal sludge were discussed in detail in Chapter 2 (Section 2.8). It was shown in Figure 2-19 that at low ranges of frequency, the ionic conductivity dominates the dissipation of power in WAS. On the other hand, below a frequency of about 1 GHz, the ionic conductivity term ($\frac{\sigma_{dc}}{\omega\varepsilon_0}$) increases logarithmically with reducing the frequency of the EM field, indicating that more optimum EM heating can be achieved at the lower frequencies. Considering these explanations, it may be assumed that the most optimum heating frequency is 0 Hz (or DC). However, a DC electromagnetic field changes the composition of the electrolyte (i.e., WAS) by depleting the medium ionic charges and also by generation of different gases. Electrode decomposition as a result of electrochemical reactions and accumulation of the polarization charges is another drawback of EM heating under DC current. However, it has been reported that above a frequency of 10 MHz, the electrochemical reactions and as a result the electrode polarization are negligible.

5.2.2. Other factors considered for design

In addition to the load electrical characteristics defining the load and EM interaction, there are other parameters which need to be taken into account when selecting an optimum frequency for EM heating. The frequency-dependent penetration depth of the EM wave is an example. Generally, the penetration depth of an EM wave decreases as the frequency increases, hence, more in-depth interaction of the field with the load is expected at lower ranges of frequency resulting in a more uniform heating profile across the load. As illustrated in Figure 5-2, a frequency of 13.56 MHz has the wavelength of 16.6 m in water (compared to 9.2 cm wavelength at a frequency of 2.45 GHz). This results in an almost uniform electric field over a couple of meters at 13.56 MHz. The power efficiency and the cost of a high-power EM generator are the other constrains associated to the EM heating. In addition to these, there are regulations which limit or ban the application of certain frequencies for heating purposes.

Considering the above explanations and also the fact that the selected frequency for EM heating should be one of the allocated frequency bands reserved for ISM applications, the radio frequency of 13.56 MHz was selected for the design of the EM heating apparatus used for WAS pretreatment. It is worth noting that a frequency of 13.56 MHz is very close to the region associated to β dispersion mechanism (between 100 KHz and 10 MHz) reported for biological cells/tissues (Pethig, 1984). Therefore, the EM heating at 13.56 MHz may have the potential for enhanced pathogen removal and/or biogas production due to the absorption of power by the cell membranes (β dispersion) compared to that of the MW pretreatment at a frequency of 2.45 GHz. Although, compared to the ionic conduction heating, the β dispersion effect is expected to be small and therefore may have an insignificant and/or undetectable effect on AD performance.

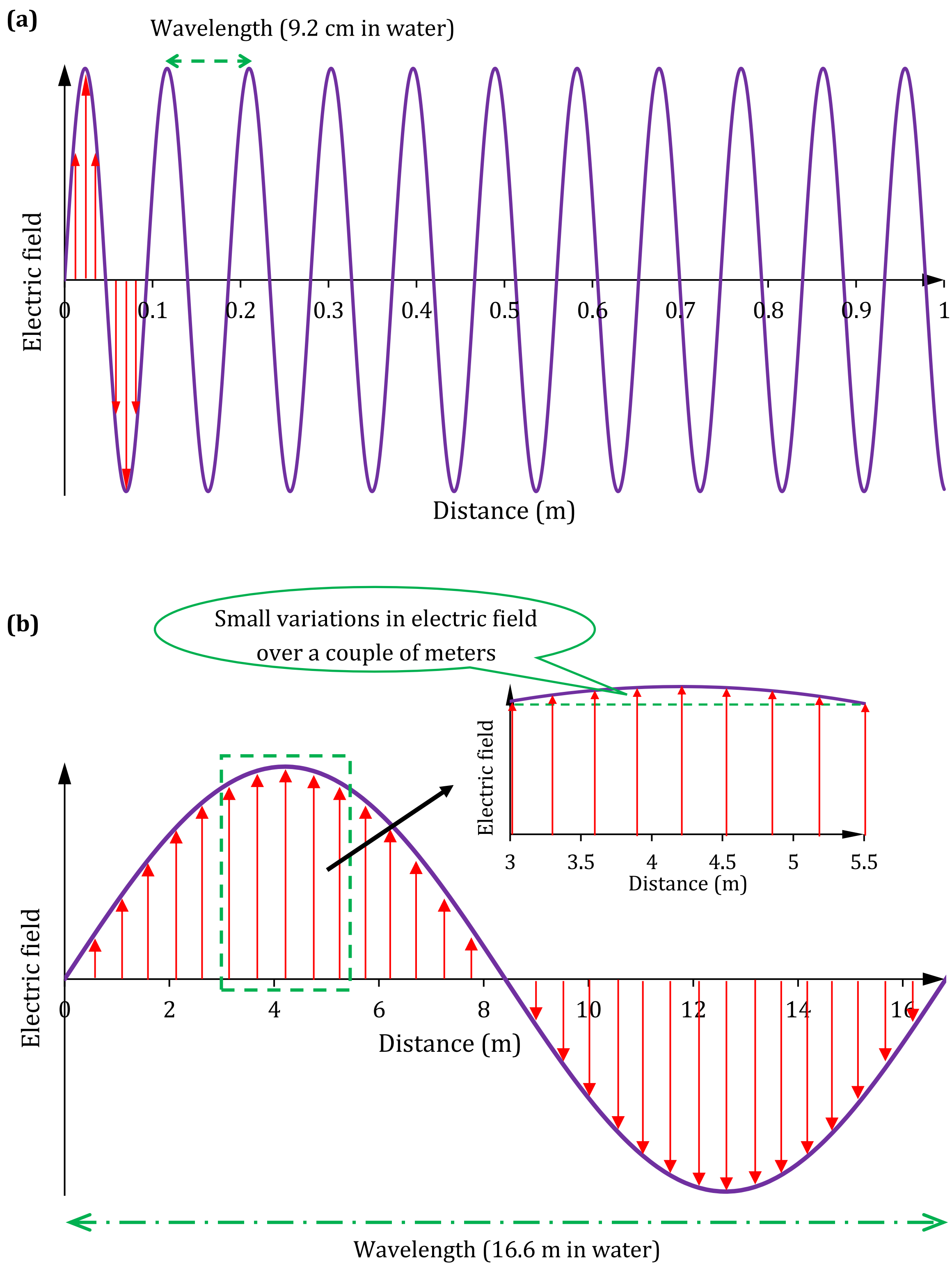


Figure 5-2. The schematic of the electromagnetic wave at a frequency of (a) 2.45 GHz and (b) 13.56 MHz

5.2.3. Power dissipation at a frequency of 13.56 MHz

The heating of municipal sludge at a frequency of 13.56 MHz is caused by the dissipation of power. The total dissipated power (P_{total}) under a time-harmonic electric field (E) can be calculated as follows (Metaxas, 1996).

$$P_{total}\ (\frac{W}{m^3}) = \frac{1}{2}(\sigma_{DC} + \omega\varepsilon_r''\varepsilon_0)|E|^2 \quad (5\text{-}1)$$

where, σ_{DC} is the sludge conductivity under the DC condition, ε_0 is the vacuum permittivity with a value of 8.8542×10^{-12} ($\frac{F}{m}$ or $\frac{s}{\Omega.m}$), ε_r'' is the imaginary part of the complex relative permittivity, and ω is the angular frequency determined as follows:

$$\omega\left(\frac{rad}{s}\right) = 2\pi f \quad (5\text{-}2)$$

in which f is the ordinary frequency measured in hertz (s^{-1}).

According to Eq. (5-1), the total dissipated power has the two components of ohmic or conduction heating loss (P_{ohmic}) and dielectric heating loss ($P_{dielectric}$) which can be determined using equations (5-3) and (5-4), respectively.

$$P_{ohmic}\ (\frac{W}{m^3}) = \frac{1}{2}\sigma_{DC}|E|^2 \quad (5\text{-}3)$$

$$P_{dielectric}\ (\frac{W}{m^3}) = \frac{1}{2}\omega\varepsilon_r''\varepsilon_0|E|^2 \quad (5\text{-}4)$$

Using the equations (5-3) and (5-4), the ratio (R) of the condition heating to the dielectric heating can be determined as follows:

$$R = \frac{\sigma_{DC}}{\omega\varepsilon_r''\varepsilon_0} \quad (5\text{-}5)$$

Eq. (5-5) allows to define the major mechanism involved in heating by determining the contribution of ohmic and dielectric heating in the total dissipated power at a given frequency. Substitution of the sludge electrical properties listed in Table 5-1 into Eq. (5-5) reveals that the conduction (ohmic) heating is the primary heating mechanism of municipal sludge at a frequency of 13.56 MHz, contributing to more than 99.3% and 96.2% of the total dissipated power in TWAS and DWSC, respectively.

Table 5-1. The electrical characteristics of KWWTP municipal sludge at a frequency of 13.56 MHz and a temperature of 25°C (adapted from Bobowski et al. (2012))

Description	**TWAS (TS = 4.5 %)**	**DWSC (TS = 18%)**
$\boldsymbol{\varepsilon_r'}$	92.5	138.8
$\boldsymbol{\varepsilon_r''}$	2.8	35.6
$\boldsymbol{\sigma_{DC}}(\boldsymbol{\Omega^{-1}m^{-1}})$	0.34	0.68

5.3. RADIO FREQUENCY SLUDGE PRETREATMENT SYSTEM DESIGN

Determination of the electrical characteristics of the WAS including its conductivity and permittivity at a frequency of 13.56 MHz was the next logical step after selecting the frequency at which the EM heating is conducted. This information was used to achieve an optimum match between the heating system and the power generator (amplifier) required for an efficient power transfer from the amplifier to the load. Table 5-1 summarizes the electrical characteristics of TWAS (at TS = 4.5%) and DWSC (at TS = 18%) generated from KWWTP which were measured by Bobowski et al. (2012). Although the sludge electrical characteristics presented in Table 5-1 were used as the reference for the design of the RF sludge pretreatment system in this project, there are some limitations to the measured data which need to be pointed out:

1- The measurements were performed at a single temperature of 25°C, however, some variation is expected at other temperatures

2- The reported values are the averages of a series of data collected from a limited number of sludge samples, however, they can vary based on the seasonal change of the sludge composition

5.3.1. Control load

Considering the fact that the ohmic heating is the dominant mechanism of sludge heating at a frequency of 13.56 MHz, the design of the RF sludge pretreatment system was initiated using equivalent salt water loads which have the same electrical characteristics as those of sludge. (Stogryn, 1971) proposed a model composed of a series of equations which defines the permittivity of sodium chloride (NaCl) solutions as a function of temperature, solution molarity, and frequency. Following the methodology proposed by Stogryn (1971), the molarity of the NaCl solutions to achieve similar electrical characteristics as those of TWAS and DWSC (listed in Table 5-1) are 0.03315 M and 0.069 M, respectively (Bobowski et al., 2012). The molarity of the equivalent salt water loads determined by Bobowski et al. (2012) were also confirmed by Ferdous (2015). Table 5-2 compares the electrical properties of TWAS and DWSC with those of equivalent salt water solutions used as controlled loads in the design of the RF pretreatment system in this project. A seen in Table 5-2, the DC conductivity of the sludge and the equivalent salt water loads are very similar. The TWAS and DWSC however demonstrate higher values for the real and imaginary part of the relative permittivity compared to those of the NaCl solutions. One explanation is that the higher ε_r' and ε_r'' measured for the municipal sludge are likely associated to the presence of the β dispersion effect which is caused by the absorption of power by the cell membranes.

Table 5-2. Comparison of the electrical characteristics of the KWWTP sludge and those of equivalent NaCl solutions (adapted from Ferdous (2015))

	Municipal sludge		**Equivalent NaCl solution**	
Description	**TWAS (TS = 4.5 %)**	**DWSC (TS = 18%)**	**0.03315 M**	**0.069 M**
$\boldsymbol{\varepsilon_r'}$	92.5	138.8	77.7	77.0
$\boldsymbol{\varepsilon_r''}$	2.8	35.6	0	0
$\boldsymbol{\sigma_{DC}}(\boldsymbol{\Omega^{-1} m^{-1}})$	0.34	0.68	0.342	0.705
$\boldsymbol{\varepsilon_r'' + \frac{\sigma_{DC}}{\omega \varepsilon_0}}$	454.0	937.0	454.1	937.6

5.3.2. Conceptual design

After the frequency of the custom-designed RF heating system was selected and the electrical characteristics of the load at the specified frequency were determined, the next step was to define the overall configuration and general geometry of the heating vessel. This step is known as "*the conceptual design phase*". Achieving uniform heating across the load was one of the key decision criteria in this step. For this purpose, several possible configurations were evaluated, and of all the available options, a parallel-plate structure was chosen. Figure 5-3 shows the distribution of the electric field in a parallel-plate structure. Ideally, the electric field (E) between the two plate with a distance of d and an electric potential difference (voltage) of V is constant and can be determined as follows:

$$E\ (\frac{volt}{meter}) = \frac{V}{d} \tag{5-6}$$

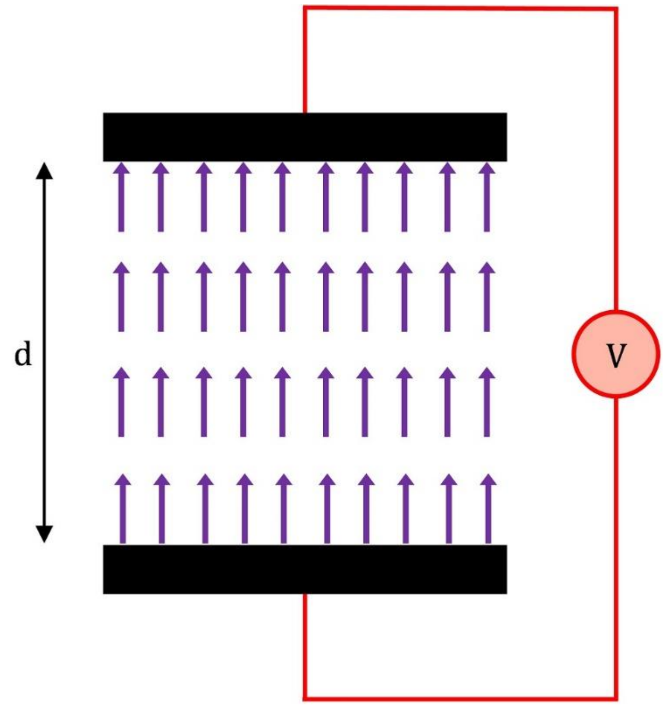


Figure 5-3. The schematic of the electric field distribution in a parallel-plate structure

The heating of the sludge in the proposed RF pretreatment is caused by a high electric field (between the two parallel plates) which has the risk of radiation to the surrounding environment. Therefore, it was decided to enclose the parallel-plate heating vessel by an external metal shield to prevent the leakage of electric field to the surroundings. Figure 5-4 illustrates the schematic configuration of the RF heating applicator as well as the distribution of electric field inside the vessel. As shown in Figure 5-4, a coaxial adapter (connector) was connected to the bottom of the metal shield. The bottom heating plate was connected to the center pin of the coaxial adapter. The top heating plate was however coupled with the external shield which was grounded and connected to the outer conductor of the adapter. The coaxial adapter was connected to the RF amplifier via a coaxial cable.

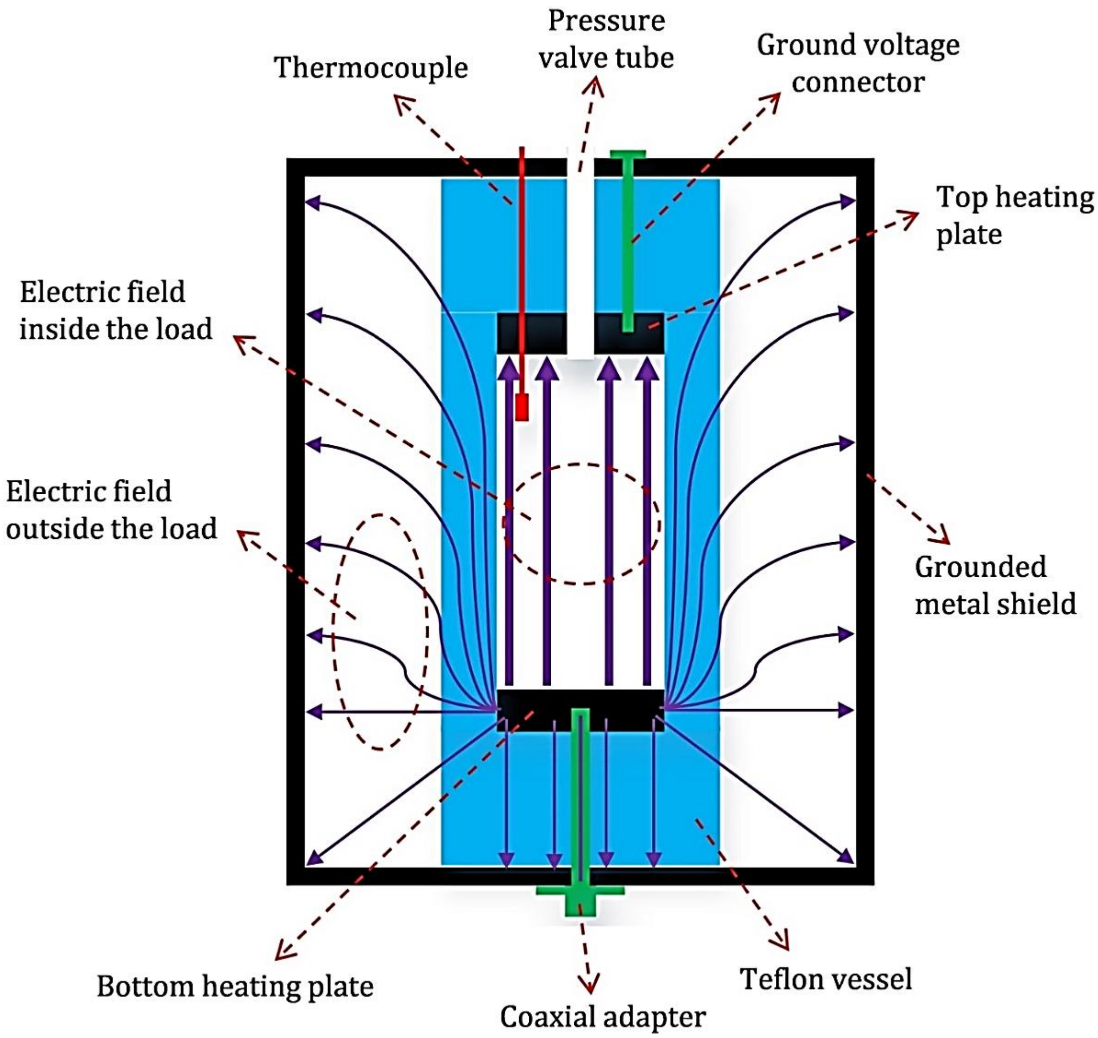


Figure 5-4. The schematic of the RF heating vessel and distribution of the electric field

5.3.3. Proof of concept

Following the conceptual design phase and before fabricating the main pretreatment system, the proposed theory of using a parallel-plate structure for sludge heating within the RF band had to be proven. For this purpose, several PVC plumbing supplies were purchased from a local store and assembled together to make a cylindrical container having the approximate dimensions of the RF heating vessel (400 mL). Figure 5-5 illustrates the configuration of the proof-of-concept prototype. As seen in Figure 5-5, two circular stainless steel electrodes were attached to the inner side of the container caps to mimic the parallel-plate structure. The preliminary experiments using the proof-of-concept prototype confirmed the feasibility of the sludge thermal pretreatment at a frequency of 13.56 MHz. The initial energy assessment results also proved that a higher energy efficiency is possible to achieve via ohmic heating at a frequency of 13.56 MHz compared to that of the dielectric heating at a frequency of 2.45 GHz.

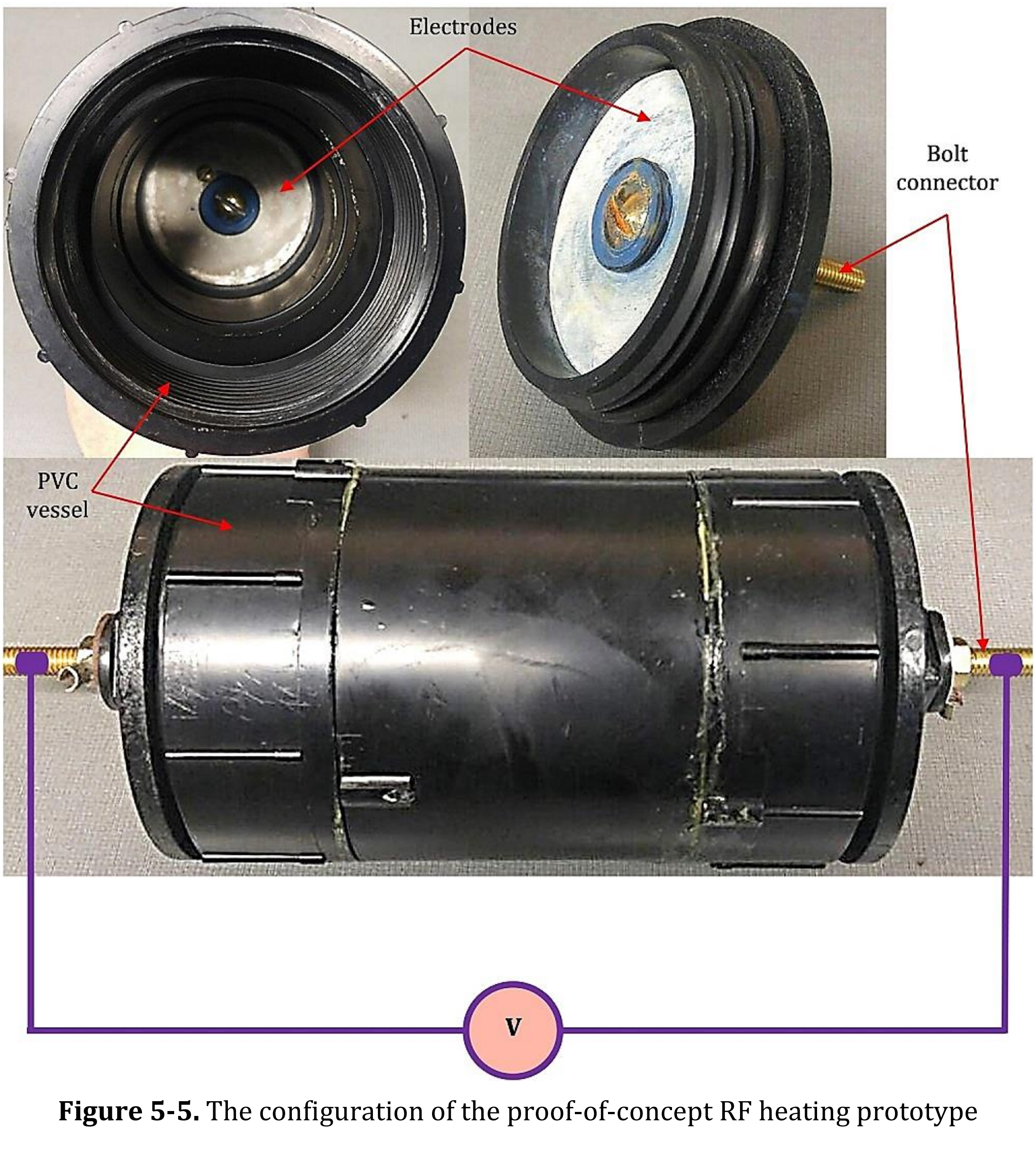


Figure 5-5. The configuration of the proof-of-concept RF heating prototype

5.3.4. Material selection

Several considerations required to be taken into account in selecting the materials for different parts of the RF heating system. Starting from the pretreatment temperature, the RF heating system is required to heat the sludge to a maximum temperature of 120°C and therefore it should be capable of handling the temperature and pressure above the boiling point. On the other hand, the body of the heating vessel which is in direct contact with the load and the heating plates needs to be dielectric in order to prevent any interference with the electric field. Hence, the heating vessel was made out of Teflon because of its low dielectric losses and inert material properties which are stable over a wide temperature range. In terms of the heating plates, they should have a high electrical conductivity. They are also required to resist the possible contact corrosion that can occur due to the direct the load. Considering these explanations, the heating plates were made out of aluminum. The outside metal shield (cylinder) was also made out of a standard aluminum pipe because of the relatively lower density of the aluminum compared to other metals.

5.3.5. Electrical modeling and simulation

The electrical modeling and simulation of the RF heating system was conducted by Md. Saimoom Ferdous, a former electrical M.Sc. student at UBC, supervised by Dr. Thomas Johnson (Ferdous, 2015). The electrical modeling was required to determine the optimum geometry of the heating vessel and to maximize the power transfer efficiency from the RF generator to the load. The electrical simulation also allowed the validation of the measured data with theoretical values.

5.3.6. Detailed mechanical design and fabrication

Detailed mechanical design as the last step before fabricating different mechanical parts of the heating applicator was performed using SolidWorks software. The exploded view of the designed RF applicator is shown in Figure 5-6. The RF heating applicator was designed to heat a 400 mL load (TWAS) at heating rates up to 15°C/min over a temperature range of 20 to 140°C. The major factors that were taken into account while designing the mechanical components of the RF heating applicator are summarized in Table 5-3.

Table 5-3. The RF sludge pretreatment system specifications

Description	**Value**
Temperature range	20-140°C
Maximum pressure	200 psi
Load volume	400 mL
Maximum heating ramp rate	15°C/min
RF amplifier maximum output power	1000 W
Maximum output power safety factor	2.4

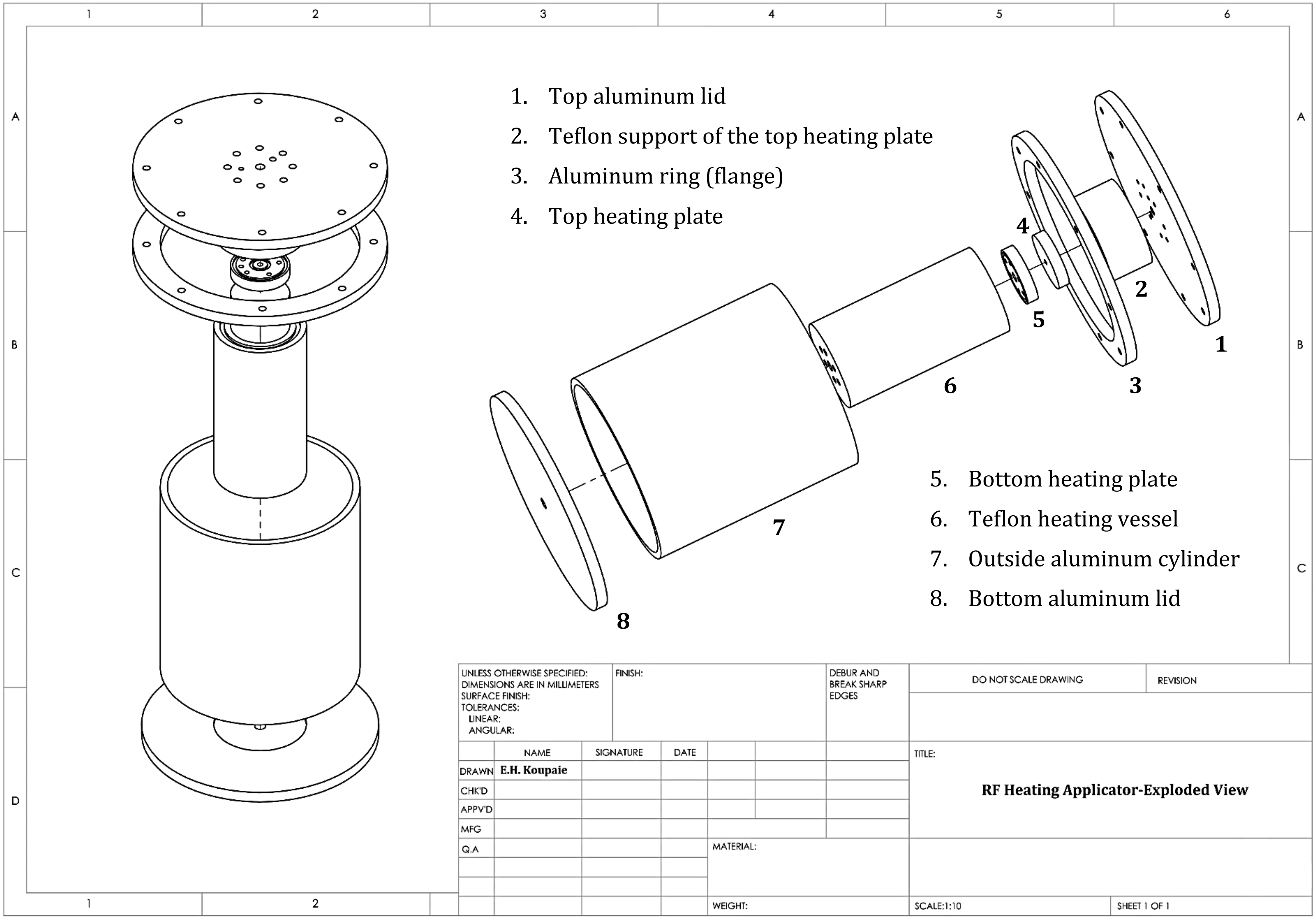


Figure 5-6. The exploded view of the RF heating applicator

5.4. RADIO FREQUENCY SLUDGE PRETREATMENT SYSTEM COMPONENTS

The operating configuration of the RF sludge pretreatment system is illustrated in Figure 5-7. In the following sections (5.4.1 to 5.4.5), the specifications of the major components of the designed RF system are described.

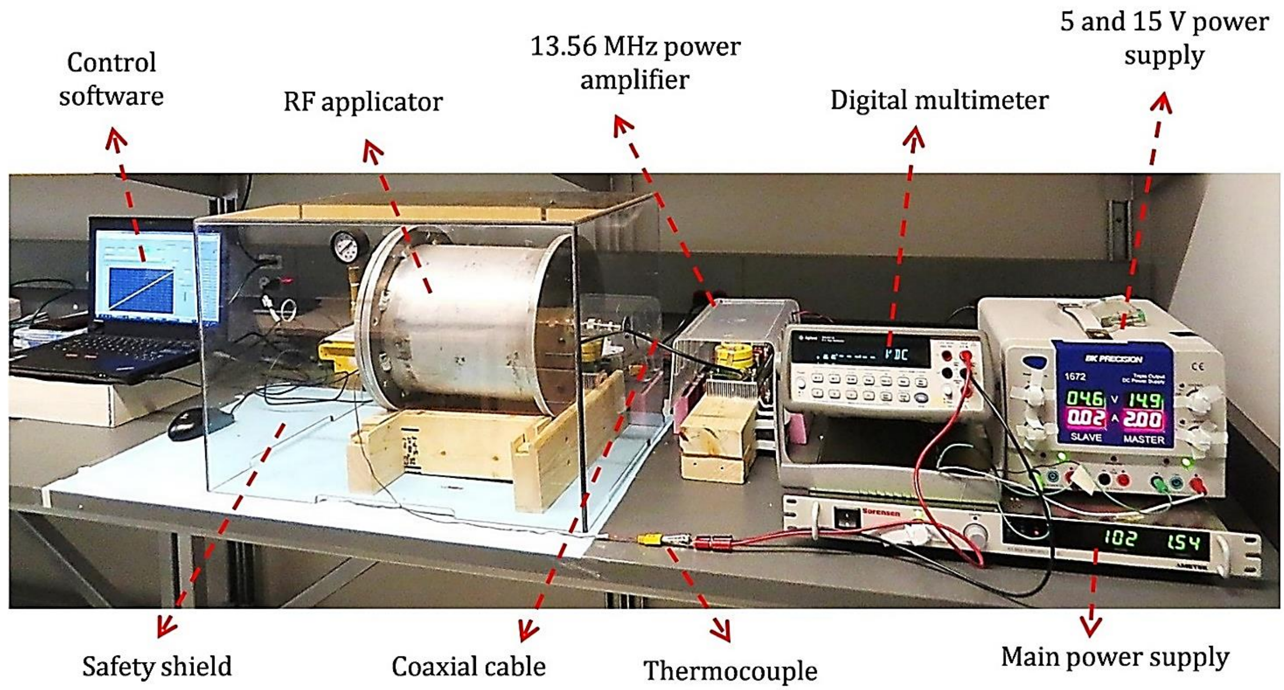
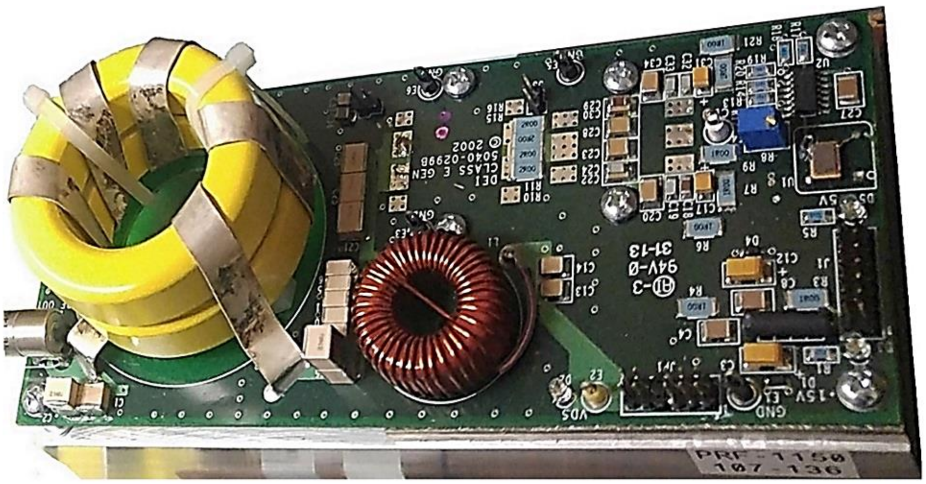


Figure 5-7. The custom-built RF sludge pretreatment system and its major components

5.4.1. Power amplifier

The required power to operate the RF heating system was generated by a 1 kW class E power amplifier (model No. PRF-1150, Directed Energy Inc., Colorado, US.). Figure 5-8 (a) illustrates the power amplifier used in this project. The cooling fans used to dissipate the heat from the amplifier during the operation is also shown in Figure 5-8 (b). For the cooling purpose, two 12 V DC fans (model No. ASB0912M) were used in parallel. The power amplifier required three external power supplies including a 5 V power supply, a 15 V power supply, and a high voltage DC power supply. The 5 and 15 V power were generated by a BK PRECISION power supply (model No. 1672). A programmable 300 V/4 Amp Ametek DC power supply (Model No. XFR 300-4) was also used as the main power supply.



(a)

(b)

Figure 5-8. The RF power generator and its cooling system; (a) the 13.56 MHz power amplifier, (b) the cooling fans used to dissipate the heat from the RF generator during the operation

5.4.2. Coaxial cable and connectors

As shown in Figure 5-9, the electrical connection between the RF power amplifier and the heating applicator was through a 30.5 cm coaxial cable (RG-58/U). The output connector of the power generator was a female BNC connector. The bottom of the heating applicator had a female N-type coaxial connector. The cable had two male BNC connectors at each end, therefore a “BNC to N” adapter was used to interconnect the cable to the applicator. All of the connectors as well as the coaxial cable had an impedance of 50 Ω.

Figure 5-9. The coaxial cable connecting the RF power amplifier to the heating applicator

5.4.3. Heating applicator

As previously shown in Figure 5-6, the heating applicator was composed of eight components. Each component was separately built at the School of Engineering machine shop and then they were assembled together. The heating plates were cut from a ½" aluminum sheet. The outside metal shield (part No. 7 in Figure 5-6) was made out of a schedule 40, 8"-diameter aluminum pipe with a wall thickness of 0.322". Figure 5-10 illustrates the assembly of the top heating plate. The heating plate had five 8 mm-diameter blind holes through which it was screwed to the Teflon support. The Teflon support was also mounted to the top aluminum plate via eight M4 screws.

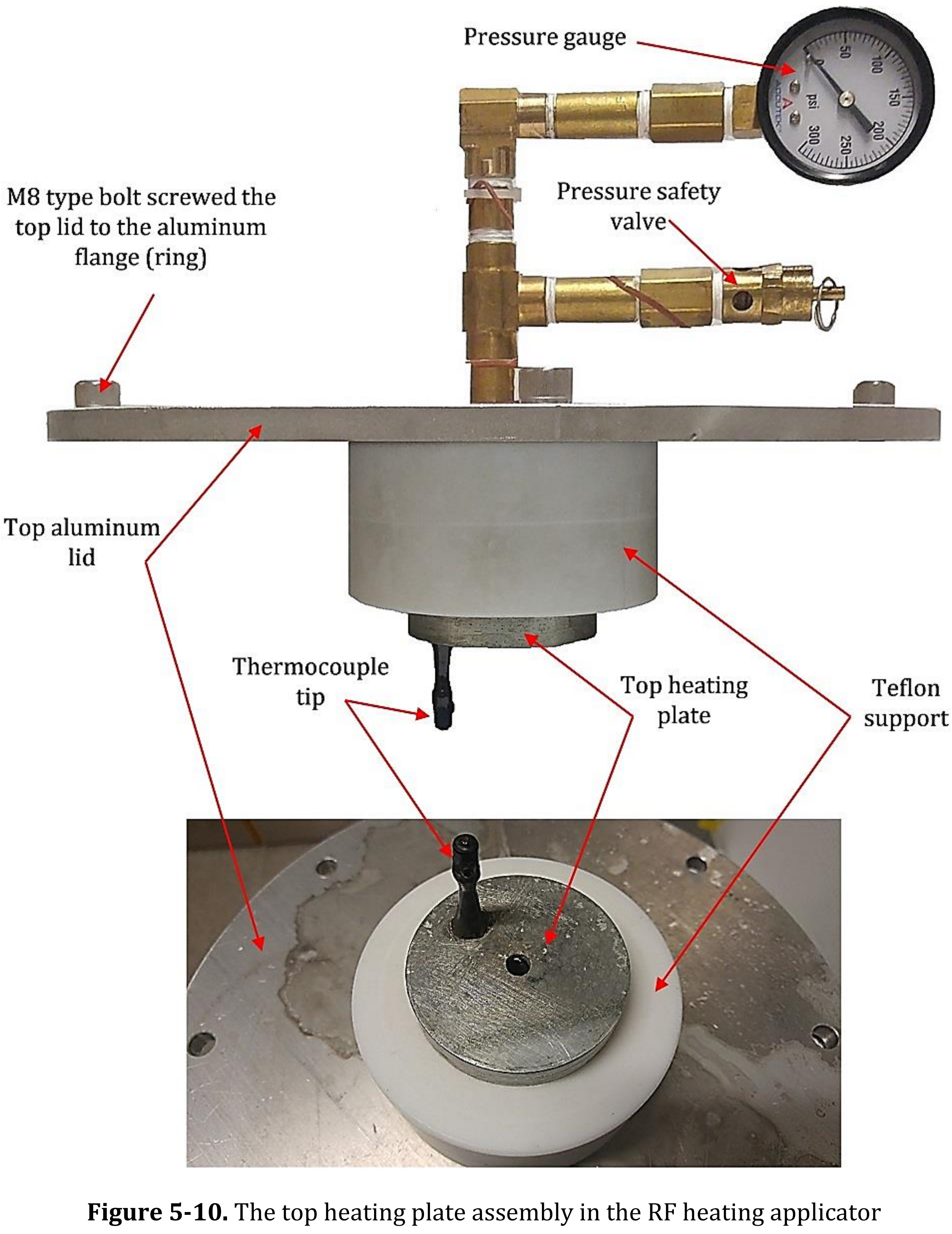


Figure 5-10. The top heating plate assembly in the RF heating applicator

Figure 5-11 shows the 400 mL Teflon heating vessel. A heating plate (the bottom heating plate) was fixed at the bottom of the Teflon vessel via six M4 screws (Figure 5-11 (b)). As seen in Figure 5-11 (c), a Viton O-ring located between the Teflon vessel and the Teflon support (shown in Figure 5-10) sealed the heating vessel during the operation.

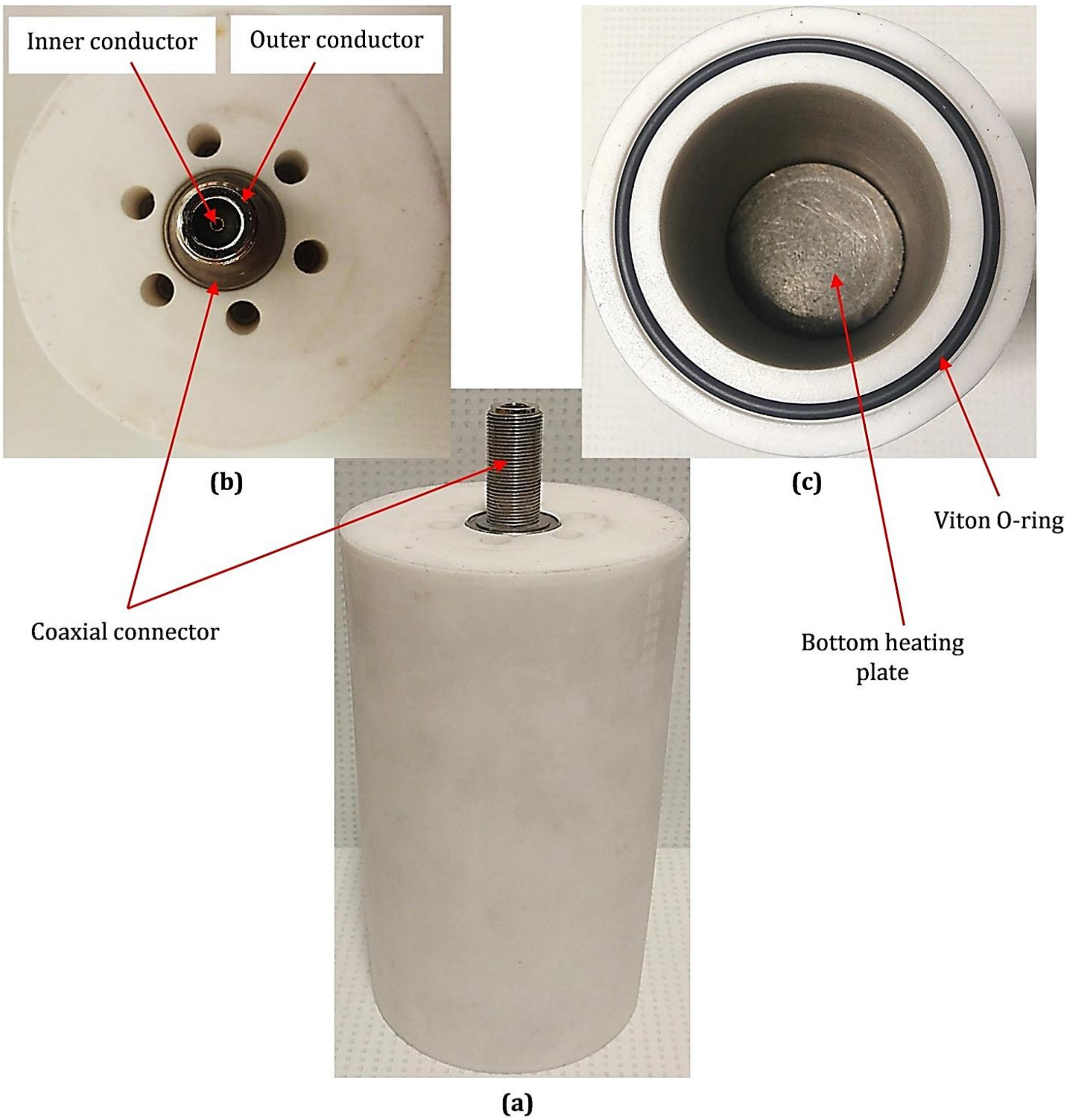


Figure 5-11. The Teflon heating vessel; (a) side view, (b) bottom view, (c) top view

The configuration of the aluminum cylinder (metal shield) and positioning of the Teflon vessel at its centre is shown in Figure 5-12.

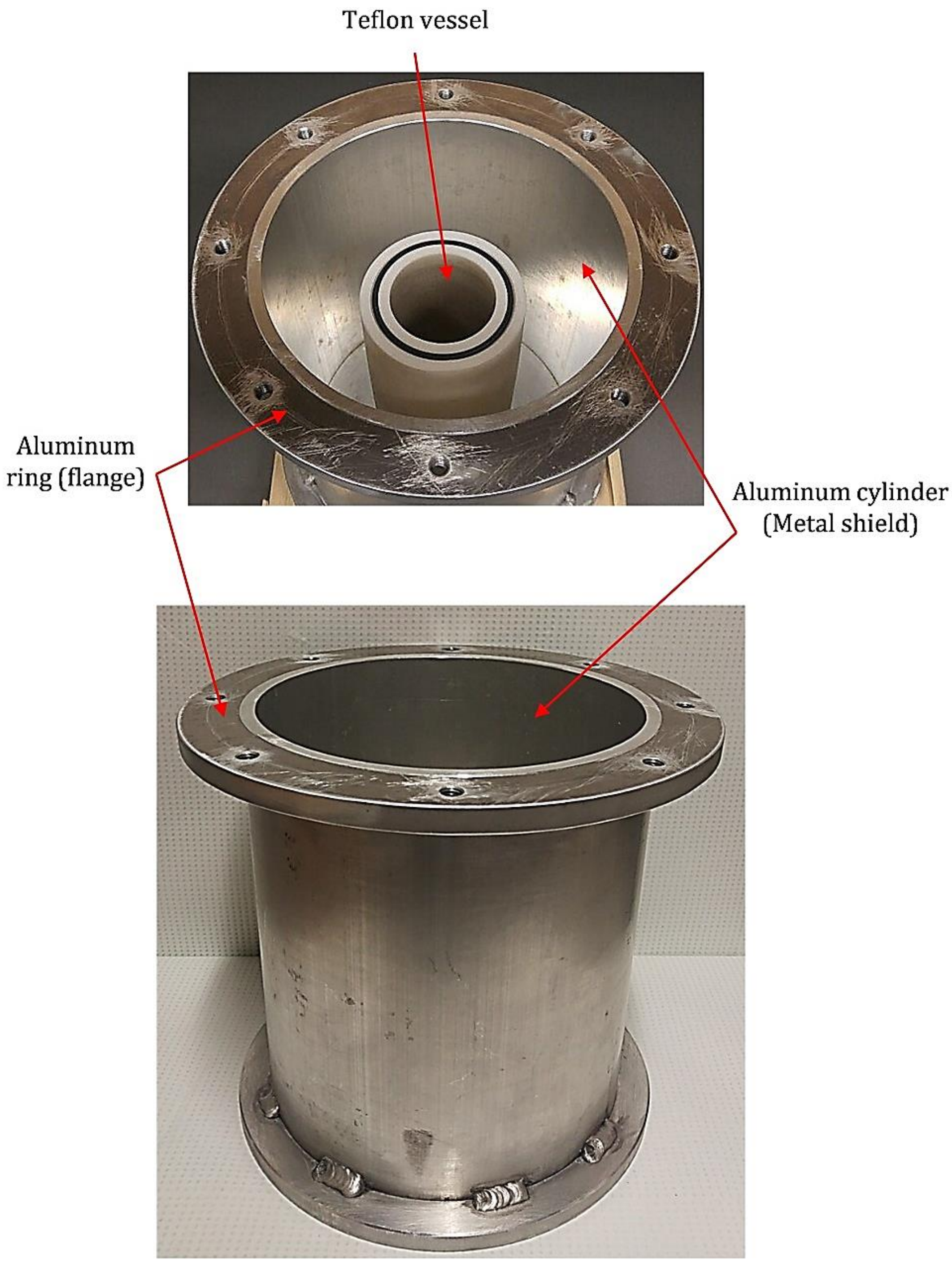


Figure 5-12. The configuration of the aluminum cylinder (metal shield) and positioning of the Teflon vessel

5.4.4. Temperature measurement system

The sludge temperature during the pretreatment process was determined by a K-type thermocouple (Model No. GK11M). The GK11M is capable of measuring the temperature at a range of -40 to 510°C, and therefore was appropriate for the sludge temperature range in this research (20 to 120°C). In order to have a direct contact between the thermocouple tip and the load, a small hole was drilled in the top heating plate all the way through the Teflon support and the top aluminum lid (shown in Figure 5-10). The thermocouple tip was then positioned 2.5 cm below the heating plate, so it will be fully submerged into the sludge. To prevent any leakage during the operation, the thermocouple hole was completely sealed using a high temperature/pressure epoxy.

The thermocouple voltage was measured by a digital multimeter (Agilent Model No. 34401A). The instant voltage measurements were recorded by a custom-developed LabVIEW program. The recorded values were then converted into temperature (°C) using the "*temperature vs. voltage*" calibration curve of the thermocouple. The relation between the thermocouple voltage and the temperature can be calculated as follows:

$$T\ (°C) = T_{amb} + \propto V \tag{5-7}$$

where, T_{amb} is the ambient or room temperature (°C), V is the thermocouple voltage (mV), and $\propto$ is the slope of the "*temperature vs. voltage*" calibration curve of the thermocouple used. For the K-type thermocouple used in this project, $\propto$ was 24.454°C/mV.

Immediately after the initial temperature tests, a significant interference was observed between the EM field through the load (sludge) and the thermocouple voltage. This interference caused large errors in the temperature measurement using the thermocouple. It was also noticed that the magnitude of the errors arbitrarily increased with increasing the EM field. Therefore, it was impossible to correct the temperature measurements by a constant correction factor. The efforts to prevent the interference by shielding the thermocouple tip was also unsuccessful. The final solution for having an accurate temperature measurement was to temporarily remove the EM field inside the load by pulsing off the applied voltage. Using this method, the DC voltage was periodically set to zero for a period of 2 seconds during which the sludge temperature was measured and then turned back on for 10 seconds. The temperature measurement with the thermocouple was finally confirmed using a mercury thermometer.

5.4.5. Input power control system

Theoretically, the amount of the power received by the load is proportional to the square of the DC voltage supplied to the RF heating system. This square relationship however does not include the change of the load impedance throughout the heating process which affect the power transfer efficiency from the amplifier to the load. It also does not consider the change in the efficiency of the power amplifier itself during the operation. Another factor which deviates the actual "voltage-power" relationship from the squared function is the thermal loss from the system which is increased with increasing the temperature gradient between the ambient and the load. Considering these explanations, an accurate open-loop calibration using a voltage input table would require trial and error. Therefore, a closed-loop LabVIEW program (as shown in Figure 5-13) was used to continuously adjust the voltage of the DC power supply based on the defined heating rate (3°C/min).

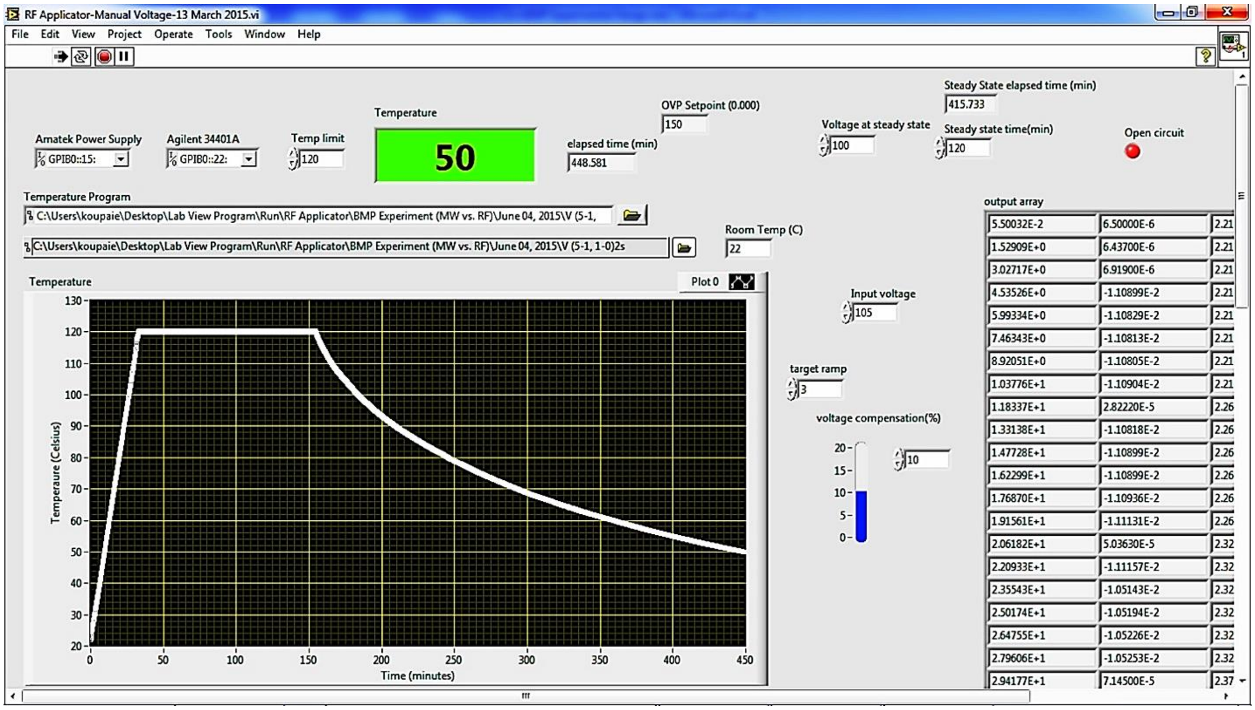


Figure 5-13. The LabVIEW power control program

5.5. ENERGY EFFICIENCY

The amount of the power received by the load (compared to the input power) depends on the efficiency of the power amplifier itself as well as the efficiency of the power transfer from the RF generator to the load. The 13.56 MHz power amplifier used in this research has an efficiency of around 85%. However, the power transfer efficiency depends on how good the impedance matching between the RF generator and the load. The maximum power is delivered to the load when the load impedance is equal to the generator impedance. In other words, the power transfer efficiency decreases as the load impedance deviates from the power generator impedance.

If P_{in} is the input power (from the wall output), then the RF generator output power (P_{out}) and the final power received by the load (P_L) can be calculated using Eq. (5-8) and Eq. (5-9), respectively.

$$P_{out} = \mu_1 . P_{in} \tag{5-8}$$

$$P_L = \mu_1 . \mu_2 . P_{in} \tag{5-9}$$

in which, μ_1 is the efficiency of the power amplifier (%) and μ_2 is the power transfer efficiency from the RF generator to the load (%) calculated using Eq. (5-10).

$$\mu_2 = (1 - |\Gamma|^2) \tag{5-10}$$

The factor, Γ in Eq. (5-10) is called reflection coefficient and is determined as follows:

$$\Gamma = \left(\frac{Z_L - Z_0}{Z_L + Z_0}\right) \tag{5-11}$$

in which, Z_L and Z_0 are the impedance of the load and the power generator, respectively.

Figure 5-14 (a) shows the change in the real and imaginary components of the load (TWAS) impedance during the RF heating for a temperature range of 20 to 140°C. The impedance measurements were carried out using a vector network analyzer (Agilent model No. 5061A). The change in the power transfer efficiency from the RF generator to the load (μ_2) as a function of temperature is also shown in Figure 5-14 (b). As seen in Figure 5-14 (a), in response to the increase of the load temperature, the real component of the impedance decreases while an increase in the imaginary part is observed. It is also demonstrated in Figure 5-14 (b) that the power transfer efficiency starts from 88% at 20°C, reaches to its maximum value (99.9%) at around 70°C and then gradually reduces to 93.9% at a temperature of 140°C. It is worth noting that the impedance of the load was measured as 52.5+3i at a temperature of 70°C. Although, these results prove the high energy efficiency of the custom-designed RF sludge pretreatment system, the energy efficiency of the RF heating system along with its effect on AD performance need to be compared with those of the MW hydrolysis at a frequency of 2.45 GHz.

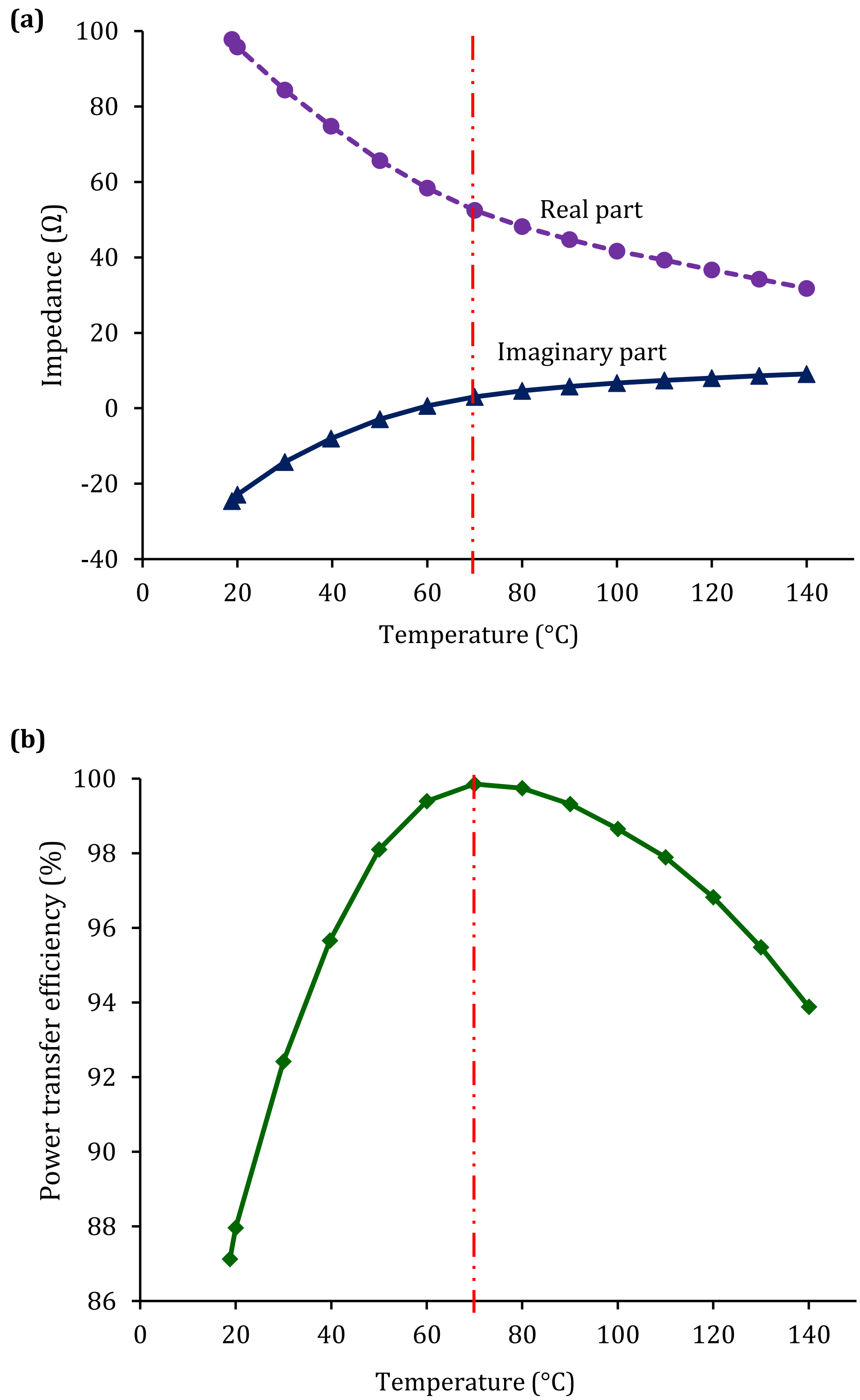


Figure 5-14. The change in the (a) impedance of TWAS during RF heating and (b) power transfer efficiency (μ_2) from the RF amplifier to the load

5.6. SUMMARY

As part of this project, a novel RF sludge pretreatment system was designed and fabricated at a frequency of 13.56 MHz for the first time. The custom-designed RF pretreatment system was aimed to eliminate the limitations of the commercially available MW heating systems (at a frequency of 2.45 GHz) used for municipal sludge hydrolysis. The heating frequency of 13.56 MHz was selected considering a variety of factors including: 1) sludge electrical characteristics such as permittivity and conductivity; 2) temperature uniformity across the load during the pretreatment process; 3) the possibility of achieving high power transfer efficiency from the EM source to the load; 4) the efficiency of the high power EM source itself; 5) the penetration depth of the EM waves; and 6) the limitations in terms of the allocated frequency bands reserved for ISM applications. The features of the custom-designed RF sludge pretreatment system at a frequency of 13.56 MHz can be summarized as follows:

- As a result of the electrolytic nature of extra- and intra-cellular materials, the ionic conduction flow dominates the heating mechanism of WAS at a frequency of 13.56 MHz, while at MW range (above 1 GHz), the dielectric relaxation of bulk water is the primary mechanism of heating.
- Considering the electrical characteristics of sludge at a frequency of 13.56 MHz, ohmic heating contributes to more than 99.3% and 96.2% of the total dissipated power in TWAS and DWSC, respectively.
- Although there is a risk of electrochemical reaction at low frequencies, at 13.56 MHz (above 10 MHz), the electrode polarization is negligible.

- The frequency of 13.56 MHz has the wavelength of about 16.6 m in water (compared to 9.2 cm wavelength at 2.45 GHz). Longer wavelength corresponds to the deeper penetration depth which is beneficial for in-depth heating of the load.
- According to the temperature measurement results, the two-parallel plate structure used in the design of the RF pretreatment system was capable of achieving uniform heating throughout the load.
- The frequency of 13.56 MHz occurs close to the band (100 KHz to 10 MHz) known for β dispersion as a result of Maxwell-Wagner effect. Therefore, WAS heating at 13.56 MHz may provide improved cell lysis due to the repeated charging and discharging of the cell membranes, which will be evaluated as part of this research.

CHAPTER 6. RADIO FREQUENCY HEATING VS. MICROWAVE PRETREATMENT FOR ENHANCED SLUDGE DISINTEGRATION AND MESOPHILIC BATCH ANAEROBIC DIGESTION

6.1. INTRODUCTION

MW sludge pretreatment systems are almost exclusively limited to a frequency of 2.45 GHz and the heating frequency is constrained by commercially available hardware. Studies using MW heating at this frequency have reported negative net energy balance (output energy as methane minus input electrical energy). This necessitates further research into more efficient thermal pretreatment technologies. In this chapter, the novel 13.56 MHz RF pretreatment system which was designed and built as part of this research is tested. The detailed information about the custom-designed RF pretreatment system is presented in Chapter 5. In the current chapter, the results of a comparison study between the RF and MW pretreatment system for enhanced solubilization of TWAS and subsequent batch AD performance are discussed.

6.2. MATERIALS AND METHODS

6.2.1. Sludge and inoculum characteristics

The comparison of the two municipal sludge hydrolysis systems (RF vs. MW) was made by applying the thermal pretreatment to the TWAS samples collected from the KWWTP. The detailed information about the wastewater treatment processes and sludge production streams of the KWWTP is presented in Chapter 3 (Section 3.2.1). Table 6-1 summarizes the characteristics of the TWAS from the KWWTP used in this study.

Table 6-1. The thickened waste activated sludge characteristics from the KWWTP

Description [a]	**Value**
pH	6.5 ± 0.1
TS (% w/w)	3.5 ± 0.2
VS (% w/w)	2.7 ± 0.2
VS/TS (%)	77.4
Ammonia (mg/L as NH_3-N)	201.4 ± 16.8
Alkalinity (mg/L as $CaCO_3$)	632 ± 128
TCOD (mg/L)	37,420 ± 574
SCOD (mg/L)	1,740 ± 350
SCOD/TCOD (%)	4.65
Total VFAs	309 ± 23

[a] TS: total solids; VS: volatile solids; TCOD: total chemical oxygen demand; SCOD: soluble chemical oxygen demand; VFA: volatile fatty acid

The mesophilic inoculum was taken from a pilot-scale AD which has been utilizing a mixture of PS and TWAS from KWWTP for more than 3 years. The pilot digester was fed two times a day at a SRT of 20 d. In order to acclimatize the inoculum to the thermal pretreatment condition, two side armed Erlenmeyer flask an effective volume of 1.3 L were set up and semi-continuously fed with thermally-pretreated TWAS for approximately 4 months under the OLR of 1.35 ± 0.10 $\frac{g\,VS}{L.d}$. To minimize the possible chronic or acute inhibition to the acid/methane formers, the most intensive pretreatment condition (temperature and stationary time of 120°C and 2 h, respectively) was selected for acclimation. In addition, one digester fed with non-pretreated TWAS was operated in order to provide acclimation to the inoculum which was later used as seed in the control (non-pretreated) digesters. The detailed information about the configuration and operation of the semi-continuous digesters is provided in Chapter 4 (Section 4.2.1). At the end of the acclimation period, the TS and VS concentration of the pretreated-acclimated inoculum were 2.04 ± 0.01% and 1.26 ± 0.01% (by weight), respectively. The TS and VS concentration of the control-acclimated inoculum were also 2.21 ± 0.01% and 1.44 ± 0.01% (by weight), respectively.

6.2.2. Thermal pretreatment systems

6.2.2.1. Custom-designed RF sludge pretreatment system

The detailed information about the electrical, mechanical and operational features of the RF sludge pretreatment system are presented in Chapter 5. In summary, the RF heating vessel consists of a parallel plate structure enclosed in an external aluminum shield. The purpose of such a configuration was to create a uniform electric field throughout the load (between the two parallel plates) and also to prevent electric field leakage to the surrounding environment. The heating vessel was made out of Teflon because of its low dielectric losses and inert material properties which are stable over a wide temperature range. The system was operated at a frequency of 13.56 MHz and was able to heat a 400 mL load at heating rates up to 15°C/min over a temperature range of 20-140°C.

6.2.2.2. Bench-scale 2.45 GHz microwave system

The detailed information about the bench-scale MW system operated at a frequency of 2.45 GHz is presented Chapter 3 (Section 3.2.2.2). In order to maximize the similarity of the operating condition between the RF and MW systems, a total volume of 400 mL of TWAS (similar to the RF heating vessel capacity) was equally divided between eight MW vessels. Therefore, the liquid volume of each MW vessel was 50 mL. To assure that the thermocouples used in the MW and RF pretreatment systems read identical temperatures, a cross calibration was performed between the two thermocouples by reading the temperature of water for a range of 20 to 100°C using the two thermocouples (Appendix D, Figure D- 1). As per Figure D- 1, the recorded temperatures with the two thermocouples deviate less than 2%.

6.2.3. Experimental design and procedure

6.2.3.1. Solubilization study

In addition to the pretreatment method (RF vs. MW), two other independent variables, pretreatment temperature (60, 90 and 120°C) and stationary (holding) time at the target temperature were included in the experimental design (Figure 6-1). The stationary times of 0, 1 and 2 h were considered in the experiments. A stationary time of 0 h means that the sludge was only being heated until it reached the target temperature. The temperature range of 60-120°C was chosen to conduct the comparison of the RF and MW systems below and above boiling point. The reason for selecting the temperature of 60°C was to see if there will be any difference observed between the two pretreatment methods due to previously described non-thermal (athermal) effects. The maximum pretreatment temperature was also limited to 120°C, since earlier research using sludge from the same WWTP showed that although higher solubilization ratio is achieved above 120°C, the net energy production is reduced due to the higher input energy consumed during the pretreatment at elevated temperatures (Mehdizadeh et al., 2013).

For the solubilization study, the experimental design included 18 combinations of the input variables (pretreatment method, temperature and stationary time) and one control (non-pretreated) scenario. Therefore, the effects of the RF and MW systems could be compared over a wide range of pretreatment conditions. The control and pretreated TWAS samples were prepared following the random order determined by Design-Expert 9 software. Based on the results of the previous research phase in which CH and MW pretreatments were compared for sludge solubilization (Chapter 3) and enhanced AD performance (Chapter 4), the thermal pretreatment was applied under the heating rate of 3°C/min in the current study. This resulted in the total heating times of 13.0, 23.0 and 33.0 min to increase the temperature of TWAS samples from the room temperature (21 ± 1°C) to a final temperature of 60, 90 and 120°C, respectively. The pretreatment of the TWAS was started immediately after it was transported from KWWTP to the UBC Bioreactor Technology Group laboratory. The pretreated samples were stored in the fridge (at the temperature of 3-4°C) and used within 48 h. Since the RF and MW pretreated samples had the same storage duration in the fridge, any possible bias to prevent solid comparison of the two pretreatment systems was minimized. Unlike the identical heating rate, the RF cooling rate was slower than that of the MW. Therefore, and in order to evaluate the possible effect of the cooling rate, two additional pretreatment scenarios were included in the experimental design. In the added scenarios, after reaching to a temperature of 90°C, the MW and RF vessels were immediately opened to eliminate the effect of cooling rate. The results obtained under these scenarios were then compared with those achieved under the scenarios of “D” and “M” as shown in Figure 6-1.

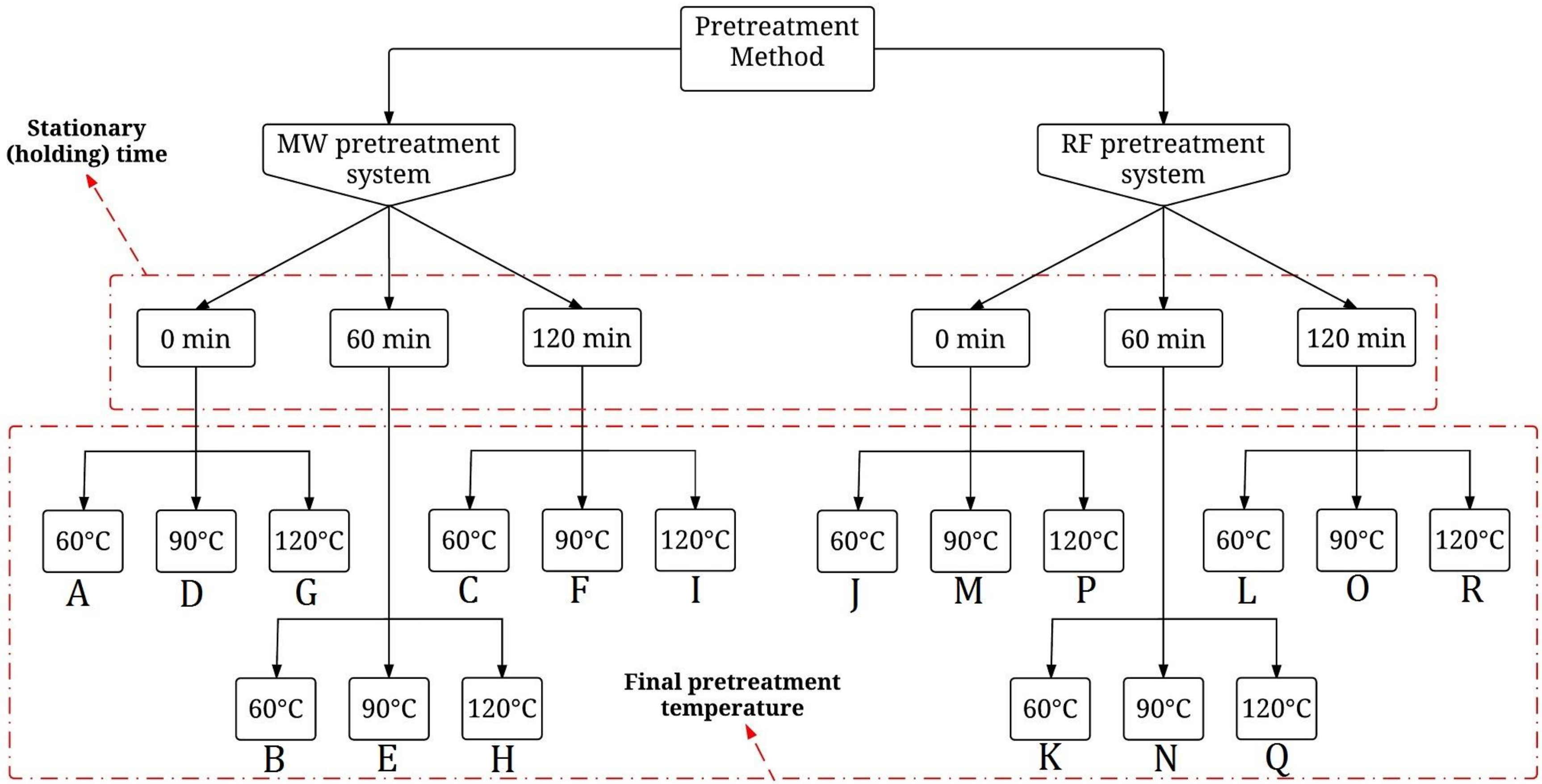


Figure 6-1. The thermal pretreatment experimental design

6.2.3.2. Batch anaerobic digestion study

In order to conduct a comprehensive comparison between RF and MW systems over a wide range of pretreatment conditions, 54 mesophilic batch anaerobic digesters (including triplicates) were set up using the pretreatment scenarios shown in Figure 6-1. In addition to the digesters fed with thermally pretreated TWAS, one set of control digesters (in triplicate) were fed with non-pretreated TWAS. In order to determine the net biogas yield corresponding to the substrate utilization only (not the inoculum), two sets of blank digesters (no substrate, only inoculum) with pretreated and non-pretreated acclimated inoculum were included in the experiment. Therefore, a total of 66 mesophilic batch anaerobic digesters (including triplicates) were operated. Each digester had a total and liquid volume of 160 and 100 mL, respectively.

The anaerobic digestion study was initiated by adding pretreated or non-pretreated TWAS samples to each bottle along with a corresponding inoculum. The volume of the substrate (TWAS) and inoculum placed to each bottle was determined considering a F/M ratio of 2.00 ± 0.1 $\frac{\text{g VS}}{\text{g VS}}$. Additional alkalinity with a concentration equivalent to 2000 mg/L of $CaCO_3$ was added to each digester in using sodium bicarbonate and potassium bicarbonate. As suggested by other researchers, in order to reduce the organic content of the inoculum and also minimize the biogas production in the blank digesters, the inoculum was starved for 3 days and degassed before mixing with the substrate (Angelidaki et al., 2009). The digesters were purged with nitrogen gas to remove the residual oxygen and sealed with rubber septa and aluminum crimp caps. The initial pressure created during the nitrogen purging was released by puncturing the septa with a needle connected to a manometer. The digesters were then placed in a shaker/incubator (Innova 44R) set at 90 rpm and a temperature of 35°C.

6.2.4. Analytical methods

The detailed information about the applied analytical procedures can be found in Chapter 3 (Section 3.2.4) and Chapter 4 (Section 4.2.4).

6.2.5. Statistical *analysis*

The statistically significant effects of the RF and MW pretreatments and two other input variables (pretreatment temperature and stationary time) were evaluated by multi-factor analysis of variance (ANOVA) at 95% confidence level (α = 0.05) using Minitab Software 17. The data were analyzed by the nonparametric statistical method of Kruskal-Wallis whenever the ANOVA assumptions (normality and constant variances) were not satisfied.

6.3. RESULTS AND DISCUSSION

6.3.1. Solubilization study

Measuring sludge solubilization ratio is one of the commonly used methods to determine the effectiveness of pretreatment methods for sludge disintegration. Thermal hydrolysis is expected to break down the sludge structure increasing the sludge solubilization. The sludge solubilization is increased due to the release of intracellular biopolymers (i.e., sugar, proteins, nucleic acids and lipids) and extracellular biopolymers (i.e., EPS) from the floc structure into the soluble phase (Tyagi & Lo, 2013). Therefore, the focus of the first part of this research was to compare the effects of the RF and MW pretreatments under identical heating profiles on TWAS disintegration. For this purpose, the solubilization of COD and three biopolymers (sugar, protein and HA) were measured and compared after pretreatment.

6.3.1.1. RF vs. MW pretreatment for enhanced COD solubilization

The SCOD concentration of the non-pretreated (control) and thermally pretreated TWAS samples are shown in Figure 6-2 (a). Figure 6-2 (b) also compares the relative (to control) improvement in COD solubilization ratio among the pretreated sludge samples. The relative (to control) improvement in solubilization ratio (SR_r) was determined using Eq. (6-1).

$$SR_r = \frac{SR\ of\ the\ pretreated\ DWSC}{SR\ of\ the\ control\ DWSC} \tag{6-1}$$

in which, SR is the sludge solubilization ratio determined via Eq. (6-2).

$$SR\ (\%) = \frac{Soluble\ fraction\ concentrion}{Total\ fraction\ concentration} \times 100 \tag{6-2}$$

As expected, the concentration of SCOD was increased after thermal hydrolysis. Among the applied pretreatment scenarios, the maximum solubilization was achieved at a temperature of 120°C and a stationary time of 2 h using the MW heating system. The application of this pretreatment condition resulted in the SCOD concentration of 11,149 ± 47 mg/L corresponding to the maximum relative (to control) improvement of 6.41 (Figure 6-2). The lowest improvement in TWAS solubilization was obtained using MW system at a temperature and stationary time of 60°C and 0 h, respectively. Under these conditions, the TWAS solubilization ratio increased by a factor of 3.14.

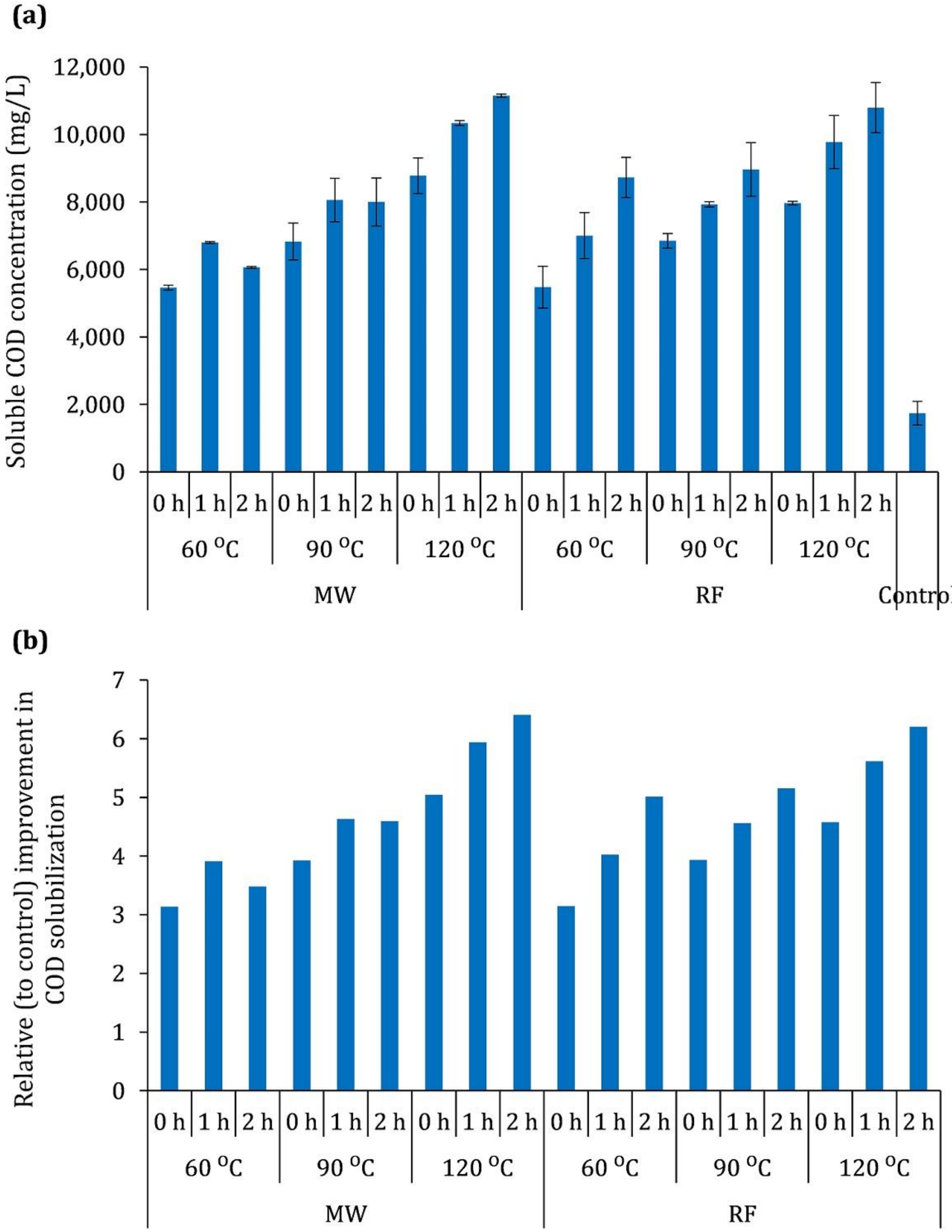


Figure 6-2. The effect of pretreatment method, temperature, and holding time on COD solubilization; (a) soluble COD concentration of the control and pretreated TWAS samples, (b) relative (to control) improvement in COD solubilization

Figure 6-3 (a) illustrates the contour plot of SCOD concentration vs. pretreatment temperature and stationary time. The main effect plot of relative (to control) improvement in COD solubilization with respect to each pretreatment parameter (pretreatment method, temperature and stationary time) is also shown in Figure 6-3 (b). As seen in Figure 6-3, the concentration of SCOD was improved by increasing pretreatment temperature and stationary time. The average SR_r ratio of 3.78, 4.47 and 5.63 were achieved at the pretreatment temperature of 60, 90 and 120°C, respectively (Figure 6-3 (b)). According to the results, the stationary times of 0, 60 and 120 min achieved an average SR_r ratio of 3.96, 4.78 and 5.14, respectively (Figure 6-3 (b)). The analysis of variance proved that the effect of pretreatment temperature on solubilization of COD (p-value = 0.000<0.05) was statistically significant. Significant differences in COD solubilization ratios were also observed among the TWAS samples exposed to the pretreatment conditions with different stationary times (p-value = 0.000<0.05).

The results described above are more remarkable when taking into account that despite the effects of both pretreatment temperature and stationary time, no statistically significant effect of pretreatment method (RF vs. MW) was observed on solubilization of COD (p-value = 0.249>0.05). These findings are in agreement with the results reported in Chapter 3 in which MW and CH pretreatments achieved similar COD solubilization of DWSC under identical heating profiles. The results provide additional proof to validate the *Hypothesis II*. The representative Anderson-Darling test normality plot for COD solubilization data is provided in Appendix B

(Figure B- 4). The representative constant variance test results for COD solubilization data are also shown in Appendix C (Figure C- 4).

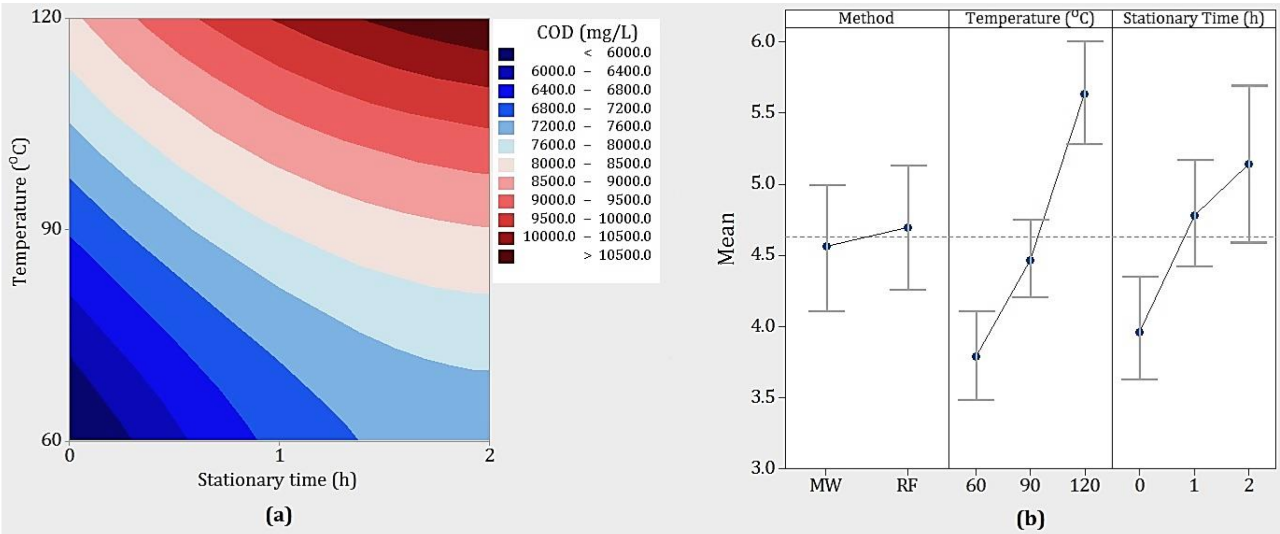
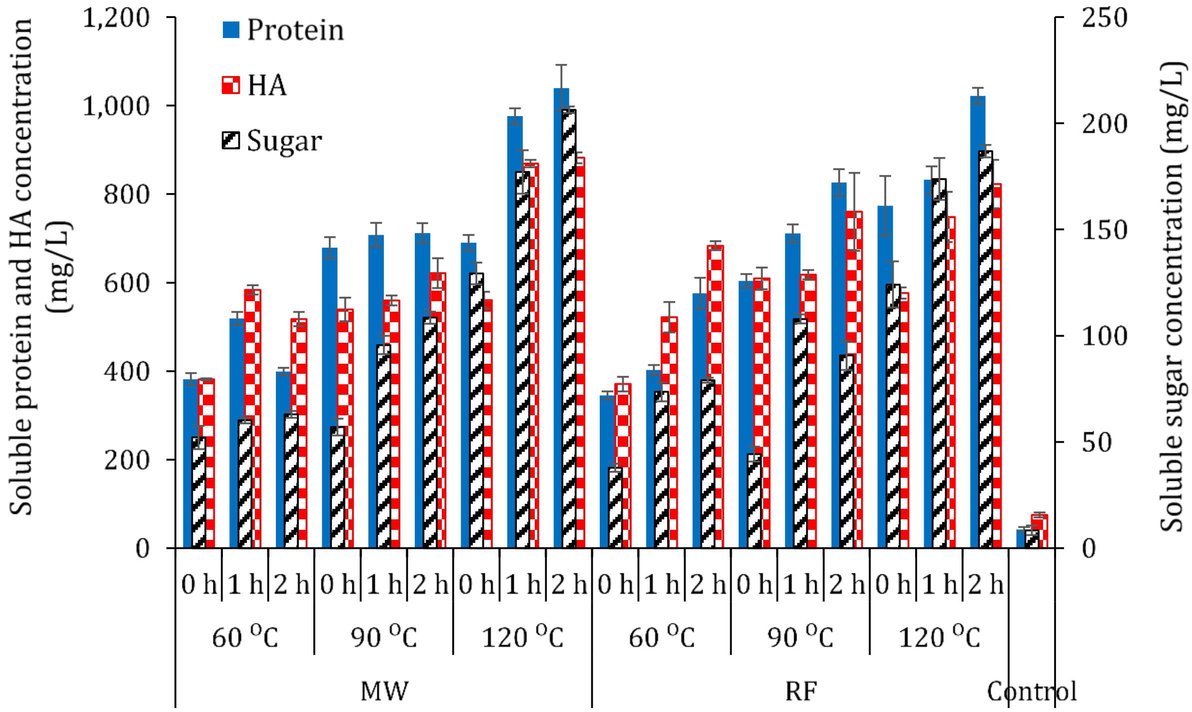


Figure 6-3. The effect of pretreatment parameters on COD solubilization; (a) the contour plot of soluble COD concentration, (b) the main effect plot of relative (to control) improvement in COD solubilization (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

6.3.1.2. RF vs. MW pretreatment for enhanced biopolymer solubilization

The concentration of soluble biopolymers (sugar, protein and HA) in the control and thermally pretreated TWAS samples are compared in Figure 6-4 (a). Figure 6-4 (b) also illustrates the relative (to control) improvement in the concentration of biopolymers after pretreatment. The highest biopolymers solubilization was obtained at the most intensive pretreatment condition (120°C for 2 h). This resulted in the relative (to control) improvement of 24.6, 24.7 and 11.8 for sugar, protein and HA, respectively (Figure 6-4). The minimum concentration of soluble sugar (37.8 mg/L), protein (345 mg/L) and HA (371 mg/L) was however obtained at the lowest pretreatment temperature (60°C) and no stationary time (0 h).

(a)



(b)

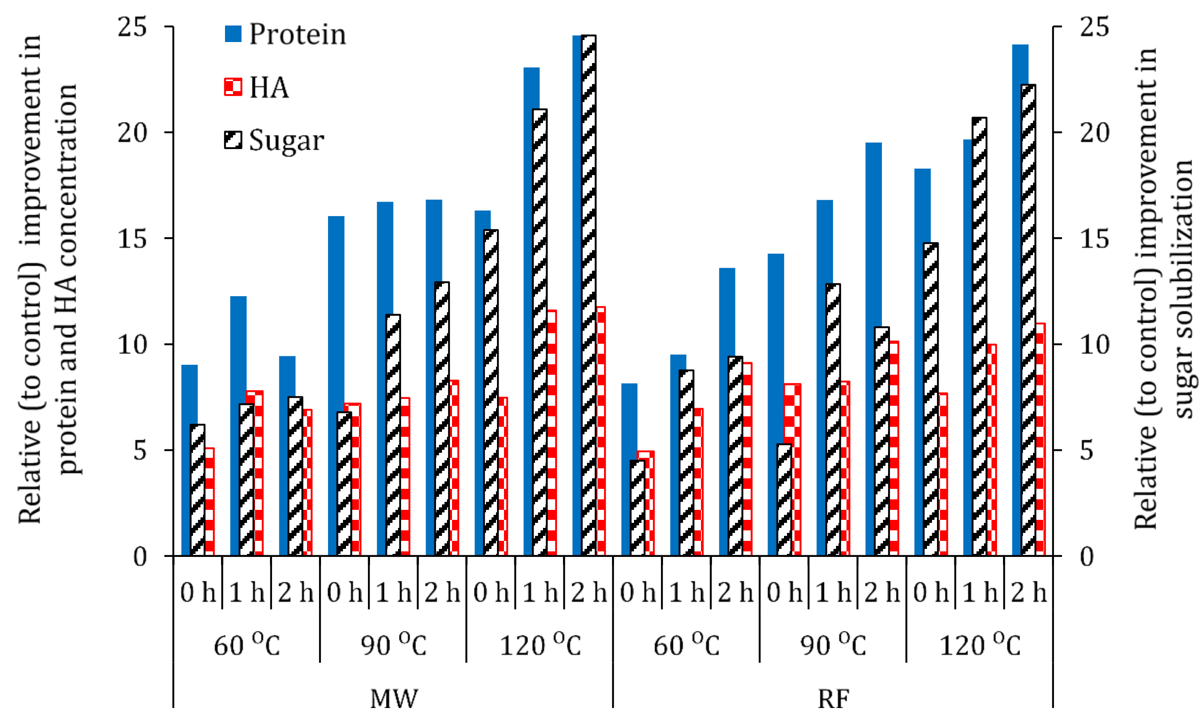


Figure 6-4. The effect of pretreatment parameters on biopolymer solubilization; (a) soluble biopolymer concentration of the control and pretreated TWAS samples, (b) relative (to control) improvement in biopolymer solubilization (adapted from Hosseini Koupaie et al. (2017))

The contour plots of soluble sugar, protein and HA concentration vs. pretreatment temperature and stationary time are shown in Figure 6-5 (a-1), (b-1) and (c-1), respectively. Figure 6-5 (a-2), (b-2) and (c-2) illustrate the main effect plot of relative (to control) improvements in sugar, protein and HA solubilization, respectively. The change in the soluble concentration of biopolymers after pretreatment demonstrated the same pattern as that of COD, suggesting that increasing the final temperature and the stationary time had positive effects on biopolymer solubilization (Figure 6-5). The statistically significant effects of pretreatment temperature and stationary time on biopolymers solubilization were also proven by the analysis of variance (p-value = 0.000<0.05). On the other hand, and consistent with the results of the COD analysis, both RF and MW pretreatments achieved similar solubilization ratios in terms of sugar (p-value = 0.227>0.05), protein (p-value = 0.923>0.05) and HA (p-value = 0.105>0.05). The results of the COD and biopolymer solubilization studies demonstrated that sludge disintegration is independent of the heating method which means that both MW and RF heating are equally effective as a thermal pretreatment process. Therefore, the selection between RF and MW pretreatment techniques is primarily determined by the energy efficiency and the scalability of the heating method to large volumes. It is worth noting that the Paired T-test analysis proved no significant effect of the cooling rate using either the MW (p-value = 0.062>0.05) or RF (p-value = 0.095>0.05) pretreatment systems.

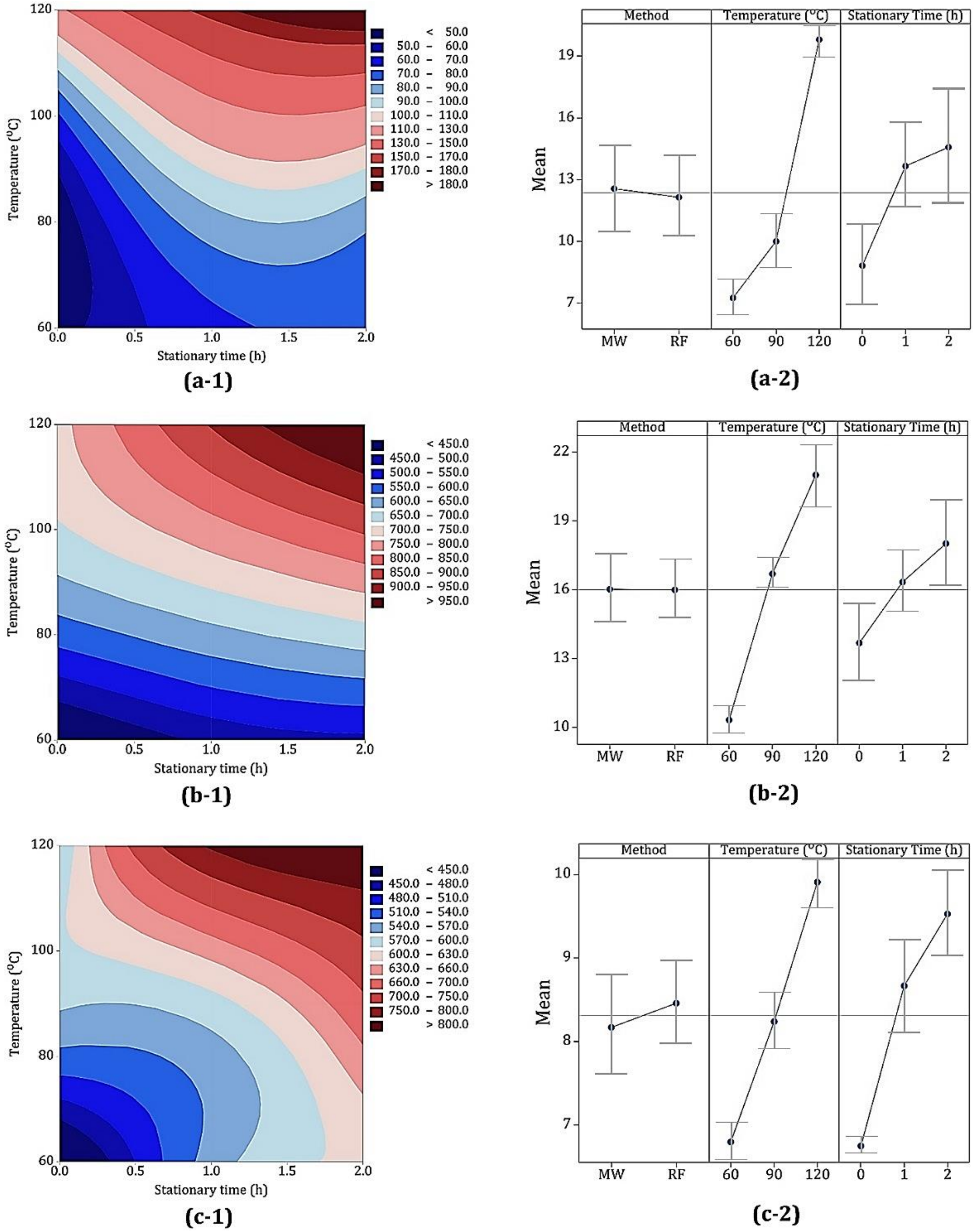


Figure 6-5. The contour plot of the soluble biopolymer concentration and the main effect plot of relative (to control) improvement in biopolymer solubilization; (a) sugar, (b) protein, (c) HA (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

6.3.2. Mesophilic batch anaerobic digestion study

6.3.2.1. RF vs. MW pretreatment for enhanced biogas production

The batch anaerobic digesters were operated for 34 days when the digesters stopped producing biogas. The ultimate biogas production in the blank digesters was less than 10% of the ultimate biogas production from the control and pretreated digesters. Figure 6-6 shows the net cumulative biogas yield (at the STP of 0°C, 1 atm) during the operating period.

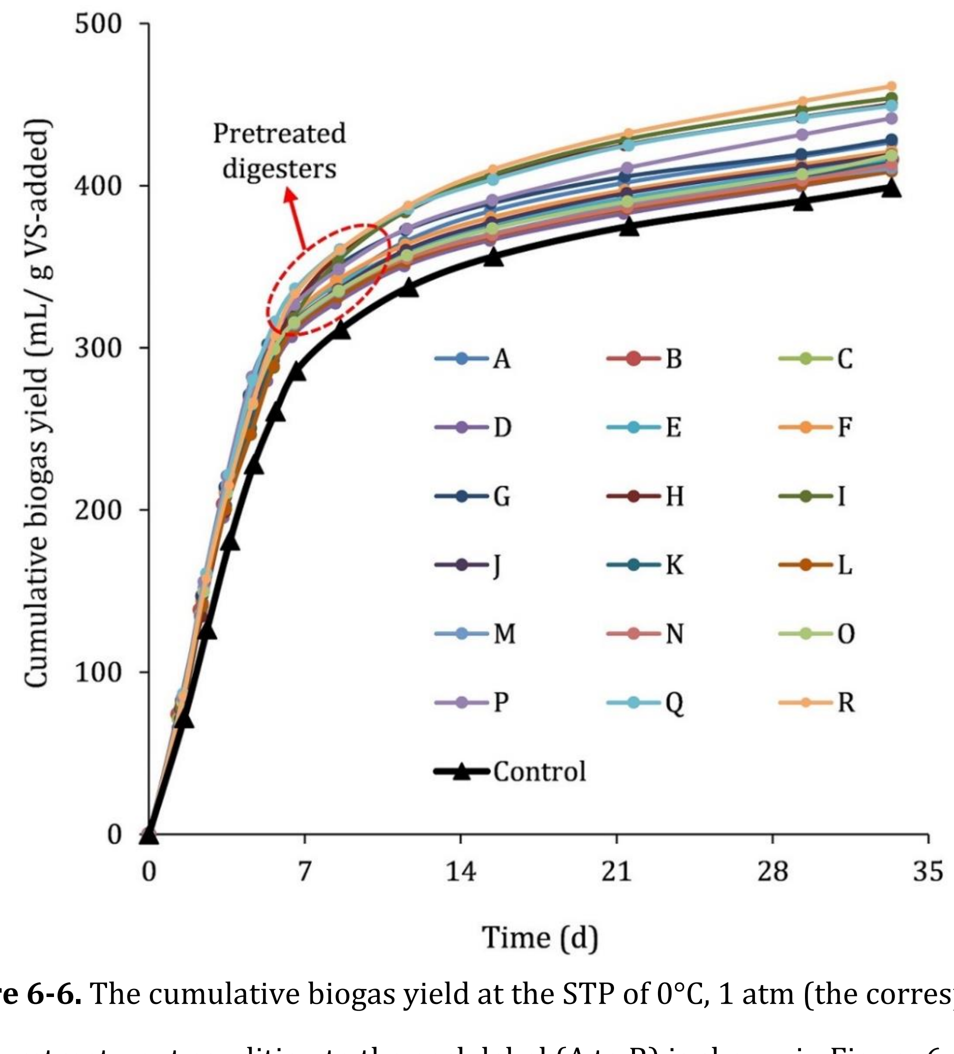


Figure 6-6. The cumulative biogas yield at the STP of 0°C, 1 atm (the corresponded pretreatment condition to the each label (A to R) is shown in Figure 6-1)

As shown in Figure 6-6, no lag-phase occurred at the beginning of the operation, confirming that inoculum acclimation prevents and/or lessens the acute inhibition. Figure 6-6 demonstrates the lower initial (during first week) and also ultimate cumulative biogas yield of the control digester compared to that of the pretreated digesters. It is worth noting that due to the addition of 2000 mg/L external alkalinity to each digester, the pH remained in the range of 7.1-7.6 throughout the entire operating period. According to the biogas composition results, the methane percentage of the produced biogas averaged 68.1 ± 1.0 % with no statistically significant difference among the digesters.

Figure 6-7 (a) compares the ultimate biogas yield (per g VS-added) in the control and pretreated digesters. The relative (to control) improvement in biogas yield is also shown in Figure 6-1 (b). The results show that the digesters fed with the pretreated TWAS at 120°C achieved the highest ultimate biogas yield. The maximum ultimate biogas yield of 461.3 ± 2.6 $\frac{mL}{g\,VS{-}added}$ was obtained using the RF heating system under the pretreatment scenario of "120°C and 2 h". This pretreatment scenario resulted in a 15.6% improvement in biogas yield relative to the control digester. Another observation is that under the temperatures of 90°C and 120°C, there is an increase in ultimate biogas yield with an increase in stationary time, similar to the TWAS solubilization results (shown in Figure 6-3 and Figure 6-5). However, under 60°C (circled area on Figure 6-7 (b)), increasing the stationary time had an adverse effect on biogas production. A possible explanation for this observation is that the low pretreatment temperature (i.e., 60°C) is not intensive enough to increase the TWAS digestibility leading to a discernible enhanced biogas production, however at the same time is able to kill or inactivate the beneficial mesophilic anaerobic bacteria in the digester feed (TWAS) that could have contributed to inocula in the digesters. These observations are in agreement with the results of another study conducted by Appels et al. (2010) in which a decrease in the biogas production was reported for the digesters fed with pretreated sludge samples at 70°C. The authors however reported positive effect of thermal pretreatment for the temperature of 90°C (Appels et al., 2010).

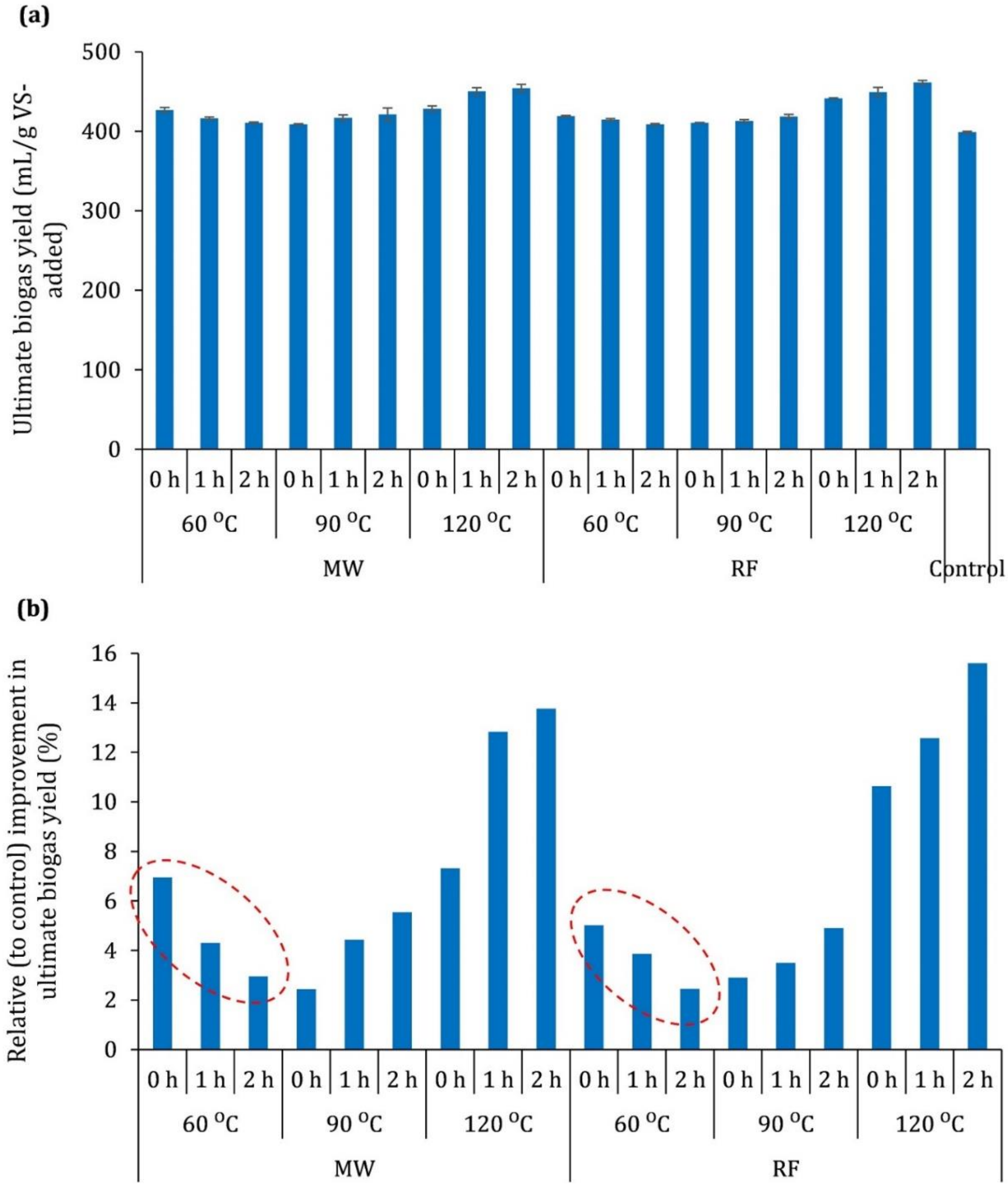


Figure 6-7. The effect of pretreatment method, temperature, and holding time on biogas production; (a) ultimate biogas yield of the control and pretreated digesters, (b) relative (to control) improvement in ultimate biogas yield after thermal pretreatment (adapted from Hosseini Koupaie et al. (2017))

As observed from the main effect plot of ultimate biogas yield (Figure 6-8) and also proven by the analysis of variance, both pretreatment temperature and stationary time had statistically significant effects on ultimate biogas yield (p-value = 0.000<0.05). There was however no statistically meaningful difference between RF and MW pretreatment techniques in terms of ultimate biogas yield (p-value = 0.690>0.05). Not only the MW and RF pretreatments achieved equal ultimate biogas yields, they also demonstrated similar biogas production patterns under all pretreatment scenarios (Figure 6-9). These observations further validate the *Hypothesis II* which was also tested in Chapter 4 that as long as the heating profile is identical, different thermal pretreatment techniques (MW, CH, and RF proposed in this study) accomplish equal improvement in anaerobic biogas production. The representative Anderson-Darling test normality plot for ultimate biogas yield data is illustrated in Appendix B (Figure B- 5). As per Figure B- 5, the Anderson-Darling test p-value is less than 0.005 which indicates that the data do not follow a normal distribution. Therefore, the nonparametric statistical method of Kruskal-Wallis was applied to the data. The nonparametric statistical analysis supported the results of the ANOVA, suggesting no statistical significant effect of the pretreatment method on biogas production (p-value = 0.749>0.05). The representative constant variance test results for ultimate biogas yield data is shown in Appendix C (Figure C- 5).

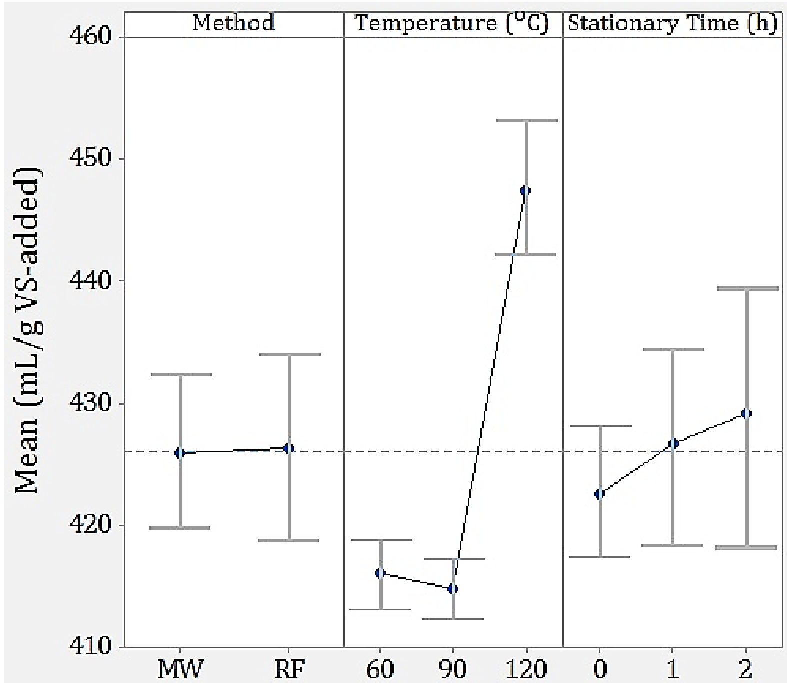


Figure 6-8. The main effect plot of ultimate biogas yield vs. pretreatment method, temperature and stationary time (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

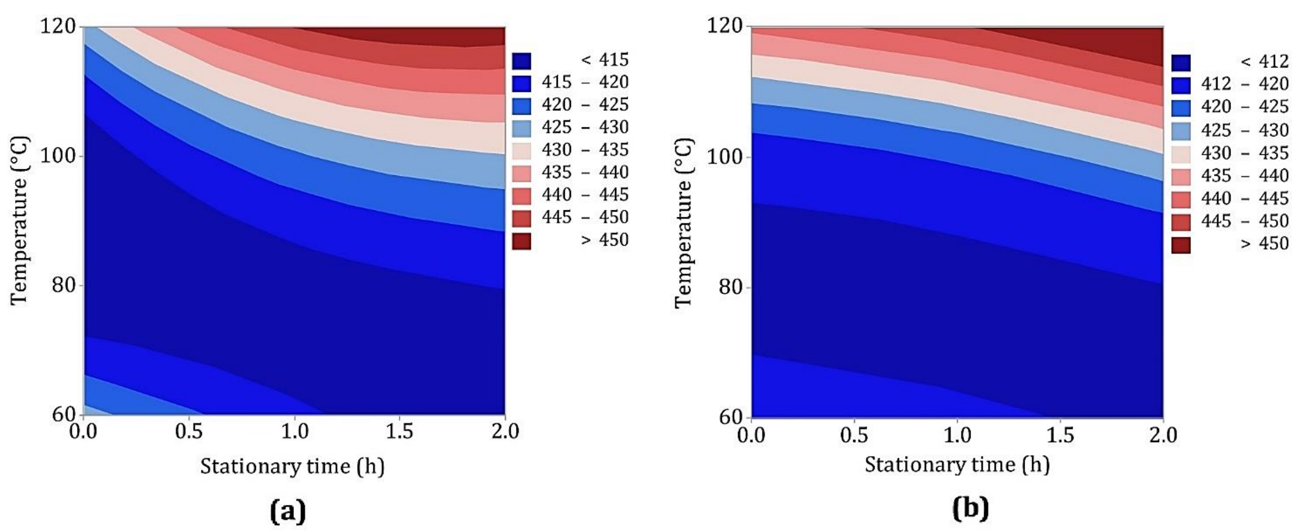


Figure 6-9. The contour plot of ultimate biogas yield (mL/g VS-added) vs. pretreatment temperature and stationary time, (a) under MW pretreatment; (b) under RF pretreatment

6.3.2.2. RF vs. MW pretreatment for enhanced solids removal

Figure 6-10 (a) compares the ultimate solids removal efficiency of the control and pretreated digesters. The relative improvement in ultimate solids removal after thermal pretreatment compared to the control digester is illustrated in Figure 6-10 (b). The maximum removal efficiency of TS (47.77 ± 0.15%) and VS (58.2 ± 0.2%) was obtained under the pretreatment temperature and stationary time of 120°C and 2 h, respectively. This observation is consistent with the biogas production data (Figure 6-7) which shows that increasing the stationary time improves the TS and VS removal efficiency under the temperatures of 90 and 120°C. The data also shows that under 60°C, the solids removal efficiency was reduced with increasing the stationary time (Figure 6-10). It should be noted that the biogas production and solids removal efficiency of WAS in digesters are functions of the SRT of the secondary treatment system where WAS is sampled from. In general, WAS generated from bioreactors with long sludge age contains higher refractory compounds with lower specific methane yields. Furthermore, duration of batch digestion also influences the organic removal efficiency. In the current study, the batch digesters were operated for 34 days, which resulted in a TS removal efficiency of 39% and VS removal efficiency of 47%. Considering the BMP assay duration of 34 days, and also the fact that no sign of inhibition (lag) was observed in the control digester, the TS and VS removal efficiencies and the biogas production obtained in this study are in the range reported elsewhere (Chi et al., 2011; Eskicioglu et al., 2007b; Lin et al., 1997).

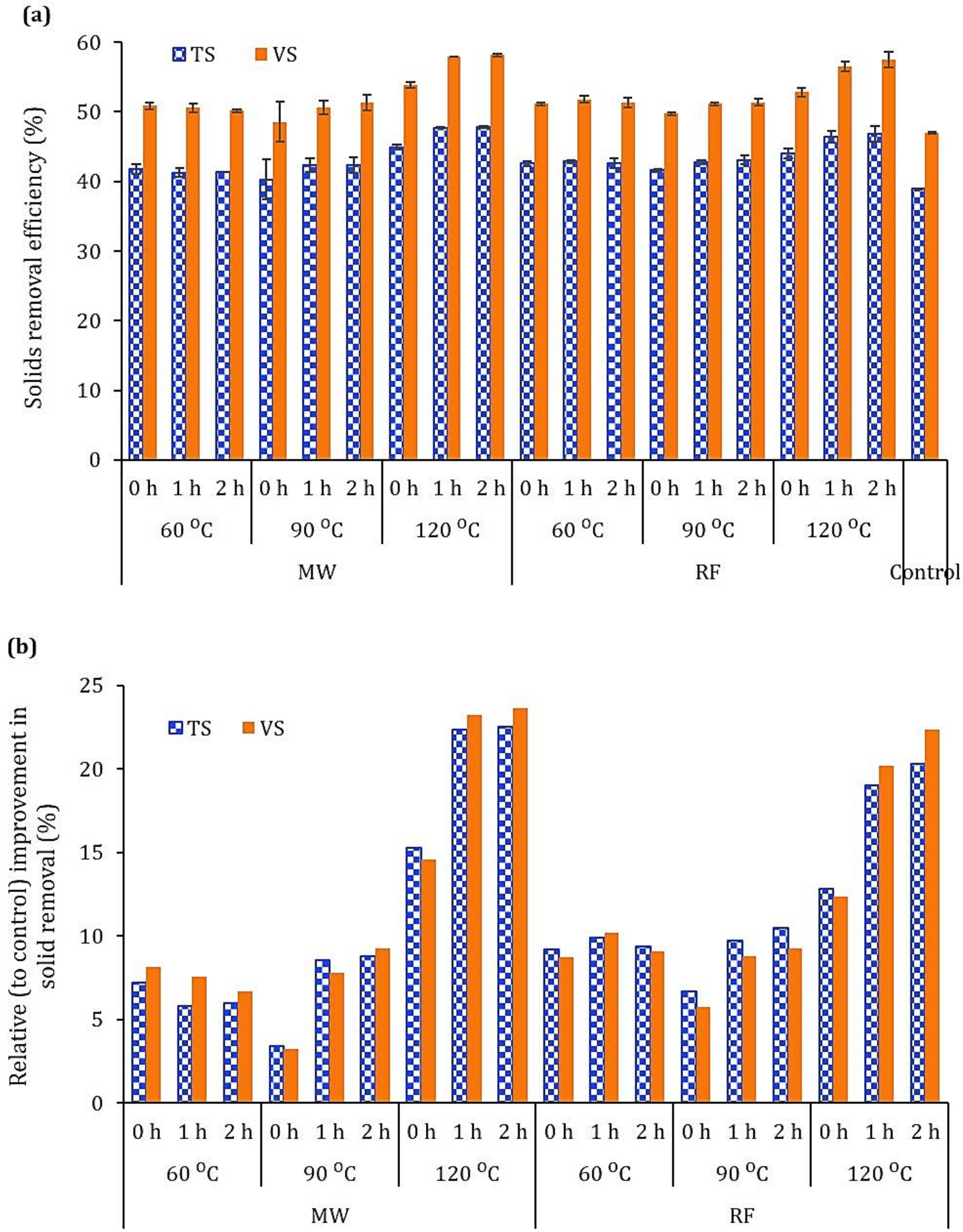


Figure 6-10. The effect of pretreatment method, temperature, and holding time on solids removal; (a) ultimate solids removal efficiency of the control and pretreated digesters, (b) relative (to control) improvement in ultimate solids removal after thermal pretreatment

Both pretreatment temperature and stationary time were observed to have statistically significant effects on the solids removal efficiency (p-value = $0.000<0.05$). According to the results of the main effect test (Figure 6-11), pretreatment temperatures of 60, 90 and 120°C resulted in an average TS removal efficiency of 42.0, 42.1 and 46.3%, and an average VS removal efficiency of 51.0, 50.5 and 56.1%, respectively. For stationary times of 0, 1 and 2 h, the average TS removal efficiency was 42.5, 43.9 and 44.0% and the average VS removal efficiency was 51.2, 53.1 and 53.3%, respectively. In contrast to the statistically significant effect of pretreatment temperature and stationary time, both RF and MW pretreatments improved solids removal efficiency by equal percentages and no statistically difference was observed for the TS (p-value = $0.293>0.05$) and VS (p-value = $0.689>0.05$) concentrations in the digesters' effluent fed with RF and MW-pretreated TWAS.

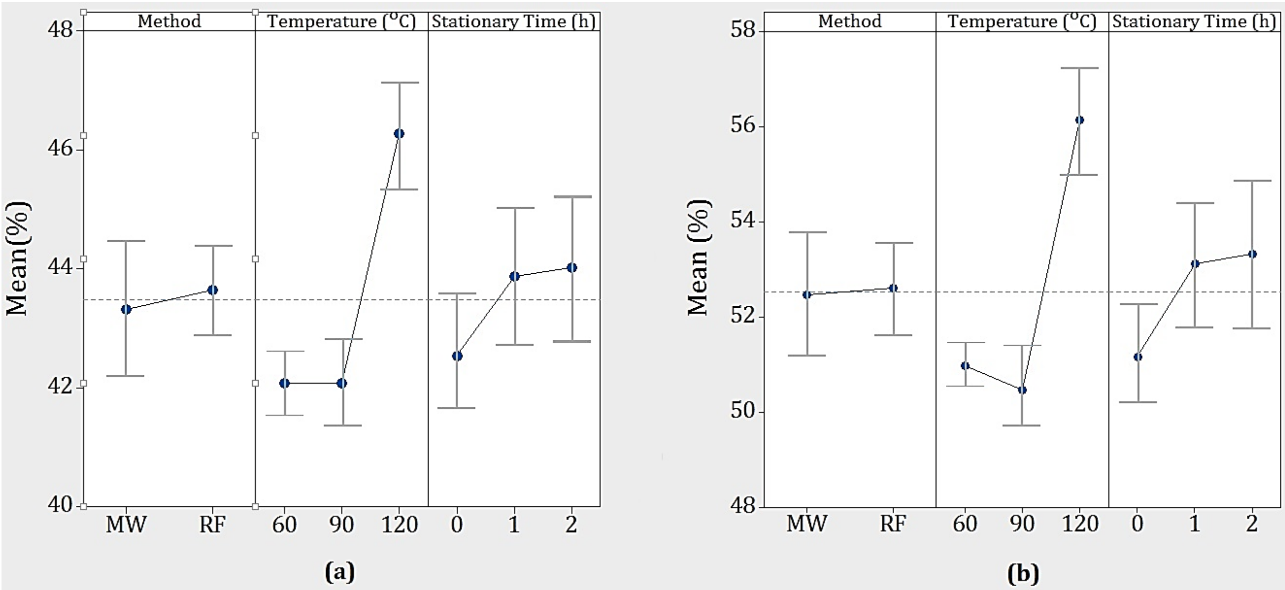


Figure 6-11. The main effect plot of ultimate solids removal efficiency vs. pretreatment method, temperature and stationary time, (a) total solids; (b) volatile solids (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

6.3.2.3. RF vs. MW pretreatment for faster biodegradation rate

The cumulative biogas production data (Figure 6-6) suggest that 65-75% of the ultimate biogas production (after 34 days of operation) was generated during the first week. Monitoring the VFA concentration also revealed that the majority of the VFAs were utilized during the first 6 days after setting up the digesters. Therefore, the first 6 days of the operating period was considered to compare the effect of RF and MW pretreatments on VS biodegradation rate. The kinetic study revealed that thermal pretreatment enhanced the rate of organics (VS) biodegradation rate in the pretreated digesters over the control digester. The maximum specific biodegradation rate constant (0.239 ± 0.003 1/d) was obtained under the pretreatment temperature of 90°C and a stationary time of 1 h which resulted in 21.7% enhancement over the control digester (0.197 ± 0.001 1/d). These results are consistent with other reported observations that although thermal pretreatment improves the overall performance of anaerobic digestion along with the biogas production rate and extent compared to the control (non-pretreated) digester, however, the most intensive pretreatment conditions do not necessarily correspond to the fastest or greatest biogas production (Dwyer et al., 2008; Toreci et al., 2009; Uma Rani et al., 2012). In agreement with solubilization and biogas data, no statistically significant differences were observed between RF and MW pretreatment methods on VS biodegradation rate constant (p-value = 0.759>0.05).

6.3.3. Comparison of RF and MW pretreatment system energy efficiency

The theoretical minimum energy (E_{th}) required to increase the temperature of a TWAS sample from an initial temperature of T_1 (°C) to a final temperature of T_2 (°C) can be determined using Eq. (5-1)

$$E_{th}(J) = \int_{T_1}^{T_2} m\, C_t dT \qquad (6\text{-}3)$$

where m (g) is the mass of the sample and C_t (J/g.°C) is the TWAS specific heat capacity. The integral form of the Eq. (5-1) allows for the inclusion a temperature dependent specific heat capacity over the pretreatment temperature (20-120°C) instead of using an average value, resulting in more accurate energy calculation. According to the literature, the solid portion of the municipal sludge has lower specific heat capacity (1.95 J/g.°C at 20°C) than that of water (4.18 J/g.°C at 20°C) (Barber, 2016; Panter, 2013; Xu & Lancaster, 2009). The solid content of the TWAS used in this study was 3.5 ± 0.2% (Table 6-1) and therefore its specific heat capacity can be assumed to be the same as that of water (water portion was 96.5%). The same assumption was made by other researchers (Mehdizadeh et al., 2013; Turovskiy & Mathai, 2006).

The total energy required to operate a pretreatment system is always higher than E_{th} which is equal to the amount of energy absorbed by the load. In order to determine the overall energy consumption of the RF system, the current and voltage supplied to the RF system was continuously recorded during the entire pretreatment period. The recorded data were then used to plot the instantaneous power as a function of time. The total RF energy consumption was calculated by integrating the P over the entire heating time. Figure 6-12 compares the actual measured energy consumption vs. the theoretical minimum energy that would be required to heat the load. It is worth noting that the energy consumption during the holding time mainly depends on how good the thermal insulation of the heating vessel is rather the efficiency of the EM generator or the power transfer efficiency. Therefore, the amount of the energy required for holding the temperature (up to 2 hours) was not included in the energy calculation presented in Figure 6-12.

In general, the overall energy consumption of an EM heating system depends on three factors: 1) the efficiency of the EM generator, 2) the power transfer efficiency from the EM generator to the load, and 3) the heat loss during the heating process. The RF pretreatment system in this research was designed to have a power transfer efficiency of more than 95% between 40°C to 120°C with the maximum efficiency of 99.9% at a temperature of 70°C. On the other hand, the efficiency of the RF generator (amplifier) was nearly constant during its operation due to the relatively low variation of the power supplied to the amplifier. Therefore, the deviation of the actual energy consumption from the theoretical minimum energy (which is equal to the energy absorbed by TWAS) was dominated by the heat loss during the pretreatment. Heat loss increases as the differential temperature between the load and ambient environment increases; therefore, lower energy efficiency is expected as load temperature rises. The measurement results in Figure 6-12 confirm this expectation and show that efficiency does decrease as temperature increases. However, RF systems with better insulation will decrease the heat loss and increase the energy efficiency at elevated temperatures as well.

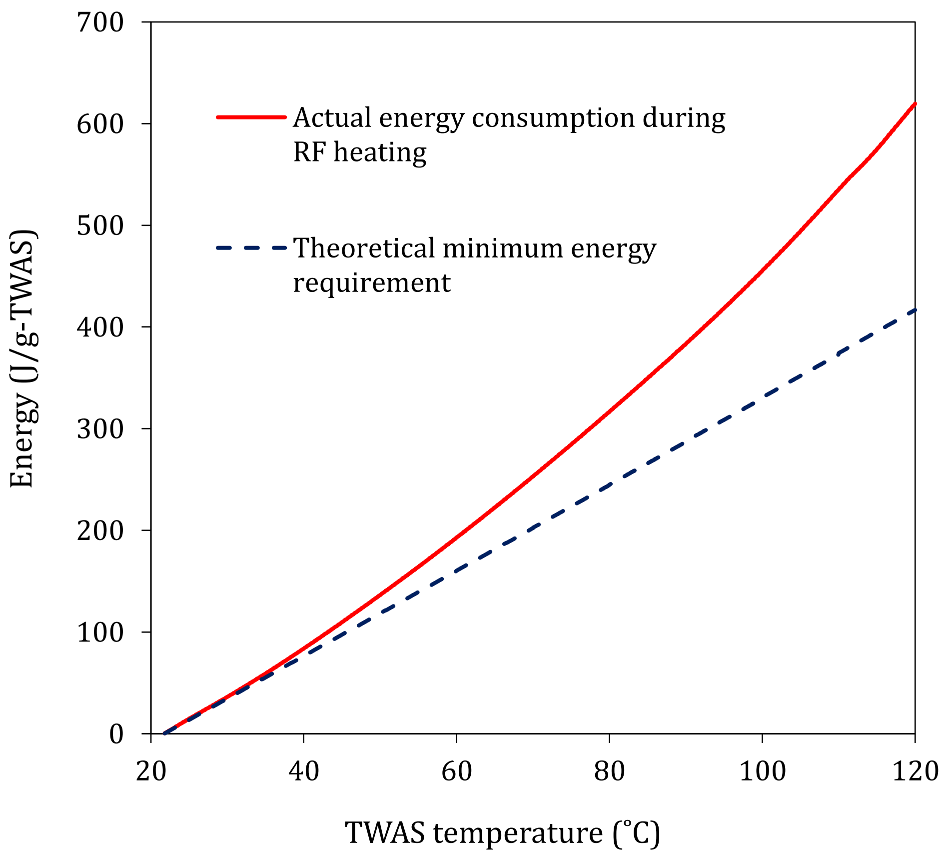


Figure 6-12. The actual RF energy consumption vs. theoretical minimum energy requirement

The overall energy efficiency (η) of a thermal pretreatment system is defined as follows:

$$\eta\ (\%) = \frac{E_{th}}{E} \times 100 \qquad (6\text{-}4)$$

in which E_{th} and E are the theoretical minimum energy requirement and the actual energy consumption of the system, respectively. For the RF pretreatment system, η was calculated using the data presented in Figure 6-12. For comparison purposes (RF vs. MW), the MW efficiency data from an earlier study led by the UBC Bioreactor Technology Group were used (Mehdizadeh et al., 2013). In the study by Mehdizadeh et al. (2013), the authors used a commercial Rogowski coil to record the instantaneous current applied to the same MW station used in this experiment (illustrated in Fig. S1). Figure 6-13 compares the overall energy efficiency of the custom-designed RF pretreatment system with that of the MW system. The overall efficiency of RF system varies between 67.3 to 95.5% over the temperature range of 25-120°C. As shown in Figure 6-13, the custom-designed RF pretreatment system was almost twice as efficient as the MW heating system which ranged from 37 to 43%. When the energy efficiency advantage of RF heating is combined with the proven fact that the type of heating method has no statistically significant difference on the effectiveness of the thermal pretreatment, it suggests that RF heating would be the preferred method compared to microwave heating. RF heating also has the advantage of uniform volumetric heating that could be scaled up to much larger systems.

Given the potential advantages of RF heating as a pretreatment method for TWAS, it is important to consider how sensitive the energy relations are to different ratios of solids content in the sludge. RF heating as designed in this system uses an ohmic heating process which is effective because of the ions in the liquid. An ionic current flows through the load which results in joule heating. As the data in the paper published by Bobowski et al. (2012) show, similar to 4% TS, ionic conductivity dominates low frequency characteristics for the sludge with a TS content of 18%. The reason is that, even at 18% TS, there is still significant liquid in the samples (more than 80%) and we expect RF heating will still be very effective. Therefore, as long as the sludge has significant liquid content, RF heating is expected to be highly efficient.

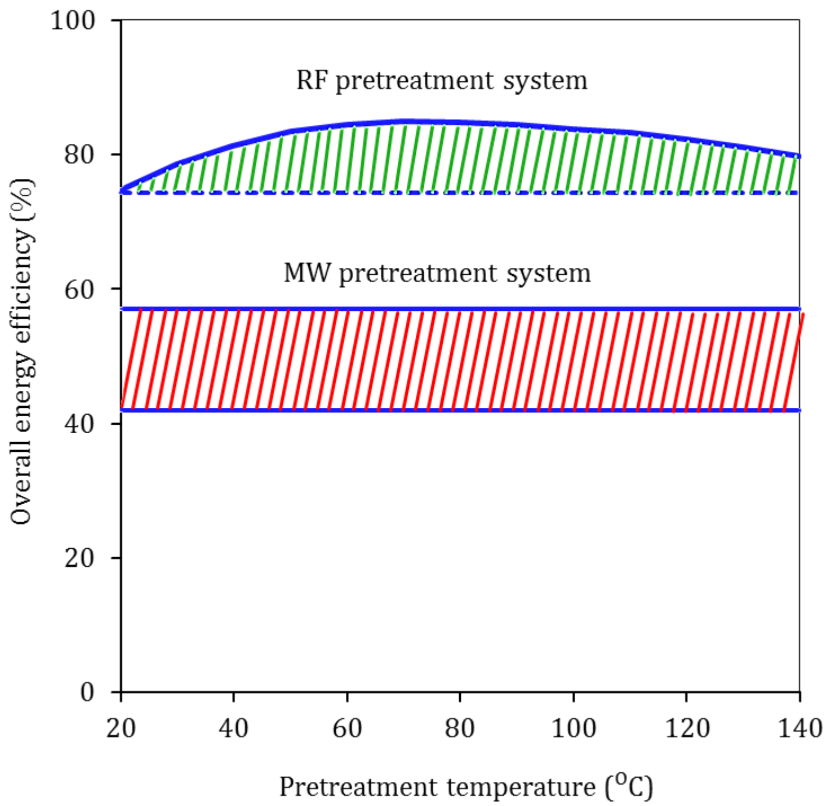


Figure 6-13. The overall energy efficiency of the RF and MW pretreatment systems

6.4. SUMMARY

In this chapter, a novel thermal pretreatment method using RF heating at a frequency of 13.56 MHz was evaluated for the first time. The effects of the system on the hydrolysis of TWAS and performance of AD were investigated. The process performance and energy efficiency of the custom-designed RF heating system was compared to that of a conventional MW system operated a frequency of 2.45 GHz. Both MW and RF pretreatment techniques improved sludge solubilization in terms of COD and biopolymers to the same extent. They also equally affected the performance of AD in terms of enhancement in biogas production and organic removals. However, for a pretreatment temperature range of 25-120°C, the overall energy efficiency of the RF heating system (67.3 to 95.5%) was two times more than that of the MW pretreatment (37-43, which validated the *Hypothesis III*. Therefore, the higher energy efficiency of the RF heating system for a target AD enhancement level makes it a proper alternative to the currently used MW pretreatment for waste sludge disintegration.

CHAPTER 7. COMPARISON OF RADIO FREQUENCY AND MICROWAVE PRETREATMENTS FOR ENHANCED SEMI-CONTINUOUS FLOW ANAEROBIC SLUDGE DIGESTION

7.1. INTRODUCTION

There are several advantages associated to the biomethane potential (BMP) assays conducted under batch flow regime, such as providing an opportunity to simultaneously evaluate the effect of a variety of experimental factors, short operation time (i.e. 1-3 months), and less sample volume required. However, the results obtained under batch mode do not represent a typical full-scale digester which is fed continuously (different flow regime) yielding different kinetics (Angelidaki et al., 2009). Furthermore, results from BMP assays need to be verified under larger-scale continuously-fed AD systems. Therefore, the objective of the last phase of this research was to evaluate the performance of semi-continuously fed mesophilic and thermophilic digesters under selected MW and RF pretreatment scenarios. For this purpose, a number of 10 digesters (pretreated and non-pretreated) with total and effective volumes of 1 and 0.5 L, were set-up, respectively. The digesters were made out of Erlenmeyer flasks and were fed once a day. The operation of the digesters was started at a SRT of 20 d and then continued at a shorter SRT of 10 d. The effects of the two thermal pretreatment methods (MW vs. RF), pretreatment temperature (80 and 120°C), and SRT on performance of the digesters were evaluated in terms of biogas (methane) production, organics removal (TS, VS, and TCOD removal), and digestate characteristics. An energy assessment was also performed to compare the net energy production achieved using the MW and RF pretreatment systems with that of the control (non-pretreated) digester.

7.2. MATERIALS AND METHODS

7.2.1. Sludge and inocula characteristics

The thermal pretreatment was applied to TWAS collected from the KWWTP. The average TS and VS concentration of the TWAS were measured as 4.02 ± 0.24% and 3.37 ± 0.20%, respectively. The mesophilic inoculum was from a pilot-scale anaerobic digester utilizing a mixture of PS and TWAS from the KWWTP for more than three years at a SRT of 20 d. The thermophilic inoculum was taken from a full-scale digester located at the Annacis Island WWTP in Vancouver (BC, Canada), which utilizes a mixture of PS and WAS. For inocula acclimation, two mesophilic and two thermophilic digesters (with an effective volume of 1.3 L each) were fed once a day with pretreated TWAS for approximately 5 months. The SRT and OLR of the digesters during the acclimation period was 20 d and 1.7 ± 0.11 $\frac{g\,VS}{L.d}$, respectively. To prevent any bias, the feed was comprised of a mixture of MW and RF pretreated TWAS (50:50% by weight) for the acclimation digesters. A pretreatment temperature of 120°C (under a heating rate of 3°C/min and holding time of 2 h) was used for the acclimation period to minimize possible inhibition of the inocula. In addition, two digesters fed with non-pretreated TWAS were operated at both mesophilic and thermophilic conditions to provide acclimation to the innocula that were later used as seed in the control (non-pretreated) digesters under similar operational condition as that of pretreated acclimation digesters.

7.2.2. Thermal pretreatment systems

7.2.2.1. Custom-designed RF sludge pretreatment system

The detailed information about the characteristics of the custom-designed RF sludge pretreatment system is presented in Chapter 5.

7.2.2.2. Bench-scale 2.45 GHz microwave system

The detailed information about the bench-scale MW system operated at a frequency of 2.45 GHz is presented in Chapter 3 (Section 3.2.2.2). To maximize the similarity of the operating condition between the RF and MW systems, a total volume of 400 mL of TWAS (similar to the RF heating vessel capacity) was equally divided between eight MW vessels. Therefore, the liquid volume of each MW vessel was 50 mL.

7.2.3. Experimental design

Figure 7-1 shows the experimental design which utilized the final temperatures of 80 and 120°C. These temperatures were included in the experimental design to compare the performance of the MW and RF pretreatment below and above the boiling point. The thermal pretreatment was applied at a heating rate of 3°C/min based on the previous research results presented in Chapter 3, 4, and 6. A rate of 3°C/min resulted in heating times of 19.7 and 33.0 min to increase the temperature of TWAS samples from room temperature (21 ± 1°C) to final temperatures of 80 and 120°C, respectively. The performance of anaerobic digesters was evaluated at both mesophilic and thermophilic conditions. The digesters were operated the SRTs of 20 and 10 d.

7.2.4. Semi-continuous anaerobic digestion

Upon acclimation, ten side-armed Erlenmeyer flasks (five in mesophilic and five in thermophilic condition) were setup to be used as lab-scale anaerobic digesters. The total and effective liquid volume of each digester was 1 L and 0.5 L, respectively. The details of the configuration and the operational features of the semi-continuous digesters are presented in Chapter 4 (Section 4.2.1). The mesophilic and thermophilic digesters were placed in the incubators set at 90 rpm and average temperatures of 35 ± 1°C and 55 ± 1°C, respectively. The operation of the digesters was started at a SRT of 20 d. A SRT of 20 d was selected for the experiment initiation to prevent any potential instability due to high organic loading rates (OLR). The average VS and TCOD loading rates under a SRT of 20 d was 1.69 ± 0.10 $\frac{g\ VS}{l.d}$ and 2.71 ± 0.2 $\frac{g\ TCOD}{l.d}$, respectively. The digesters were fed once a day (7 days/week) and operated for a minimum period of three SRTs during the steady-state period in which less than 10% variation in biogas production occurred (Ekama et al., 1986). After 85 days, the SRT was reduced to 10 d and the operation was continued for additional 40 days. The average VS and TCOD loading rates under a SRT of 10 d were increased to 3.21 ± 0.19 $\frac{g\ VS}{l.d}$ and 5.38 ± 0.8 $\frac{g\ TCOD}{l.d}$, respectively. The digester feed characteristics and loading rates at different stages of the operation are summarized in Table 7-1.

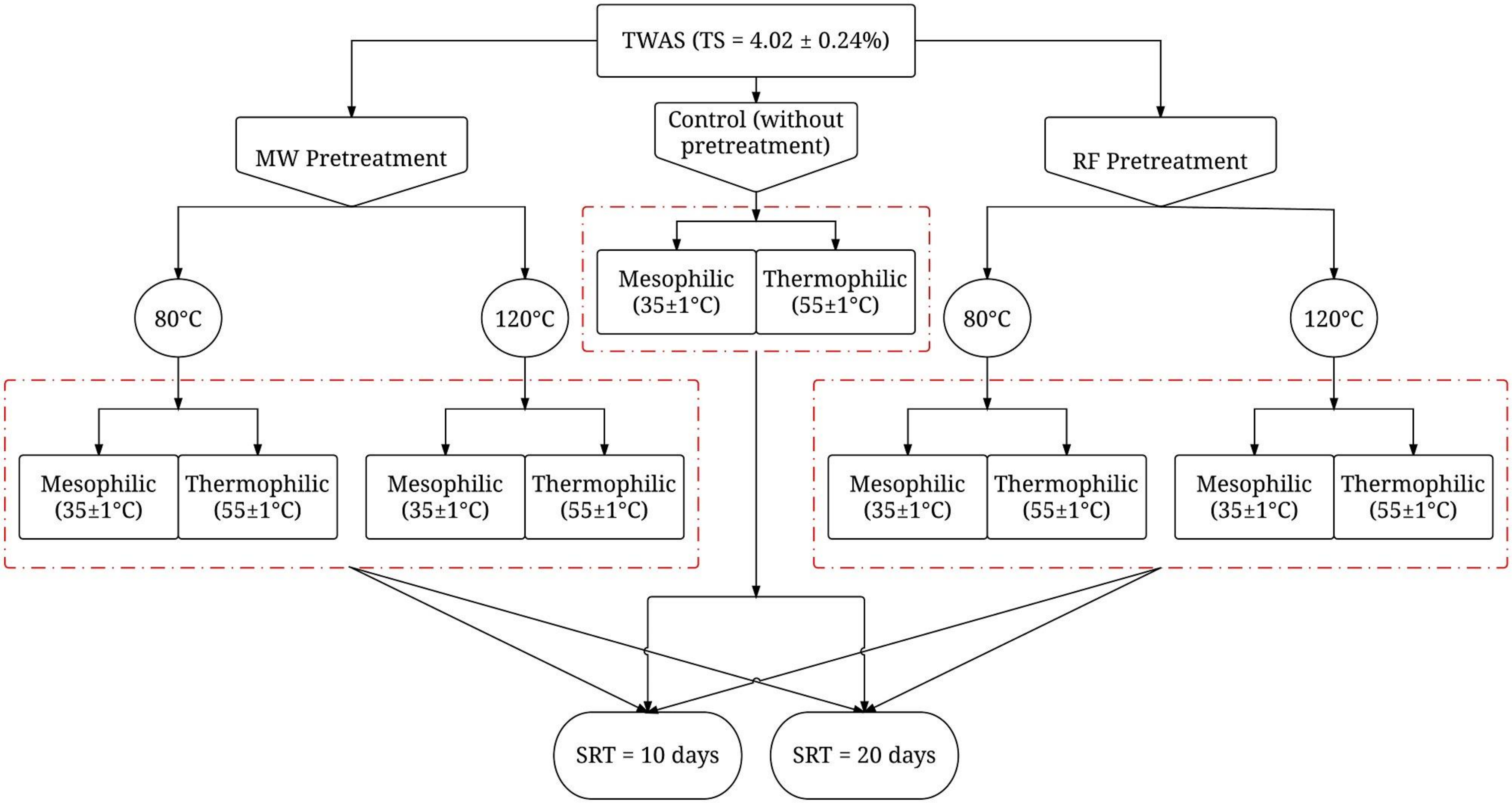


Figure 7-1. The experimental design

Table 7-1. The digester feed characteristics and loading rates at the SRTs of 20 and 10 d

Description [a]	**SRT = 20 d**	**SRT = 10 d**
Feed (TWAS) characteristics		
TS concentration (%)	4.02 ± 0.24 [b]	3.84 ± 0.16
VS concentration (%)	3.37 ± 0.20	3.21 ± 0.19
TCOD concentration (%)	54,252 ± 5,046	53,783 ± 7,974
Solids loading rate		
Daily TS added (g/d)	1.01 ± 0.06	1.92 ± 0.08
TS loading rate (g/L.d)	2.01 ± 0.12	3.84 ± 0.16
Daily VS added (g/d)	0.84 ± 0.05	1.60 ± 0.09
VS loading rate (g/L.d)	1.69 ± 0.10	3.21 ± 0.19
TCOD loading rate		
Daily TCOD added (g/d)	1.36 ± 0.13	2.69 ± 0.40
TCOD loading rate (g/L.d)	2.71 ± 0.25	5.38 ± 0.80

[a] SRT: sludge retention time; TS: total solids; VS: volatile solids; TCOD: total chemical oxygen demand; TWAS: thickened waste activated sludge

[b] Data represent arithmetic mean of measurements ± standard deviation

7.2.5. Analytical methods

The detailed information about the analytical procedures can be found in Chapter 3 (Section 3.2.4) and Chapter 4 (Section 4.2.4). Table 7-2 listed the analyzed parameters, their sampling frequency and locations during the operation of semi-continuous digesters.

7.2.6. Statistical analysis

The statistically significant effects of the input parameters (pretreatment method and temperature) were evaluated by multi-factor ANOVA at a 95% confidence level ($\alpha = 0.05$) using Minitab Software 17. The Fisher's least significant difference test was applied to compare all pairs of means.

Table 7-2. The summary of the analytical measurements

Description	Sampling frequency	Sampling location
Total and volatile solids (TS/VS)	Twice a week	Feed, digester effluent
Total and soluble chemical oxygen demand (TCOD, SCOD)	Twice a week	Feed, digester effluent
pH	Twice a week	Feed, digester effluent
Ammonia	Twice a week	Feed, digester effluent
Alkalinity (as $CaCO_3$)	Twice a week	Feed, digester effluent
Volatile fatty acids (VFA)	Weekly	Feed, digester effluent
Biogas volume	Daily	Tedlar bag
Biogas composition (in terms of CH_4, CO_2, N_2, and O_2 percentage)	Weekly	Digester gas tube

7.3. Results and Discussion

After the acclimation period, ten semi-continuously fed anaerobic digesters (control, MW-80°C, RF-80°C, MW-120°C, and RF-120°C) were setup under both mesophilic and thermophilic conditions to compare the effects of the two thermal pretreatment techniques (MW vs. RF) along with the effects the pretreatment temperature and SRT on biogas production, methane yield, organics removal, and digestate characteristics. The results obtained under the semi-continuous flow regime were then used to perform a more comprehensive energy assessment than the energy analysis conducted under the batch flow regime in Chapter 6. Table 7-3 lists the steady-state results of the mesophilic and thermophilic anaerobic digesters obtained under a SRT of 20 d. The steady-state results of the digesters under a SRT of 10 d are presented in Table 7-4.

Table 7-3. The steady-state results for the anaerobic digesters at a SRT of 20 d

	Mesophilic					Thermophilic				
Description [a]	**Control**	**MW-80**	**RF-80**	**MW-120**	**RF-120**	**Control**	**MW-80**	**RF-80**	**MW-120**	**RF-120**
Organics removal										
TS removal (%)	40.8 ± 1.4	42.1 ± 1.6	42.0 ± 2.0	44.8 ± 1.4	44.2 ± 1.9	41.5 ± 3.3	42.2± 2.4	42.0 ± 2.5	40.9 ± 2.1	42.0 ± 2.0
VS removal (%)	47.5 ± 2.1	49.9 ± 2.2	49.7 ± 2.3	51.8 ± 1.7	52.1 ± 1.7	48.5 ± 3.0	48.7 ± 1.6	48.9 ± 2.1	48.7 ± 2.0	47.8 ± 2.5
TCOD removal (%)	44.2 ± 2.6	46.7 ±3.3	47.0 ± 1.5	49.1 ± 3.2	48.8 ± 1.0	44.0 ± 1.7	45.1 ± 1.4	44.8 ± 3.0	43.8 ± 2.4	44.3 ± 3.0
Biogas and methane production										
Daily biogas production (mL/d)	262 ± 22	273 ± 19	275 ± 17	297 ± 20	295 ± 18	257 ± 21	263 ± 26	261 ± 22	260 ± 27	259 ± 22
Normalized daily biogas production ($\frac{mL}{L.d}$)	524 ± 44	546 ± 39	550 ± 34	593 ± 39	589 ± 36	514 ± 42	526 ± 53	522 ± 44	520 ± 54	517 ± 44
Methane yield ($\frac{mL}{g\ TCOD-added}$)	133 ± 11	139 ± 10	151 ± 9	140 ± 9	150 ± 9	131 ± 11	134 ± 13	132 ± 14	133 ± 11	132 ± 11
Methane yield ($\frac{mL}{g\ VS-added}$)	199 ± 17	223 ± 16	225 ± 14	243 ± 16	241 ± 15	210 ± 17	215 ± 22	214 ± 18	213 ± 22	212 ± 20
Specific methane production ($\frac{mL}{g\ TCOD-removed}$)	302 ± 25	297 ± 21	308 ± 20	298 ± 19	307 ± 19	297 ± 24	296 ± 30	302 ± 31	297 ± 25	297 ± 26
Digestate characteristics										
pH	7.32 ± 0.10	7.37 ± 0.08	7.38 ± 0.7	7.47 ± 0.05	7.48 ± 0.09	7.93 ± 0.16	7.93 ± 0.08	7.94 ± 0.12	7.95 ± 0.15	7.96 ± 0.07
SCOD (mg/L)	434 ± 32	542 ± 124	509 ± 141	769 ± 35	823 ± 61	1840 ± 176	2044 ± 122	2065 ± 135	2154 ± 110	2126 ± 186
Alkalinity (mg/L as $CaCO_3$)	2434 ± 50	2950 ± 50	2785 ± 29	3206 ± 41	3425 ± 28	3190 ± 46	3225 ± 55	3437 ± 22	3569 ± 31	3358 ± 24
Ammonia (mg/L as NH_3-N)	664 ± 100	782 ± 81	855 ± 74	956 ± 82	947 ± 81	1070 ± 87	1185 ± 98	1159 ± 106	1209 ± 96	1278 ± 126

[a] SRT: sludge retention time; MW: microwave; RF: radio frequency; TS: total solids; VS: volatile solids; TCOD: total chemical oxygen demand; SCOD: soluble chemical oxygen demand

Table 7-4. The steady-state results for the anaerobic digesters at a SRT of 10 d

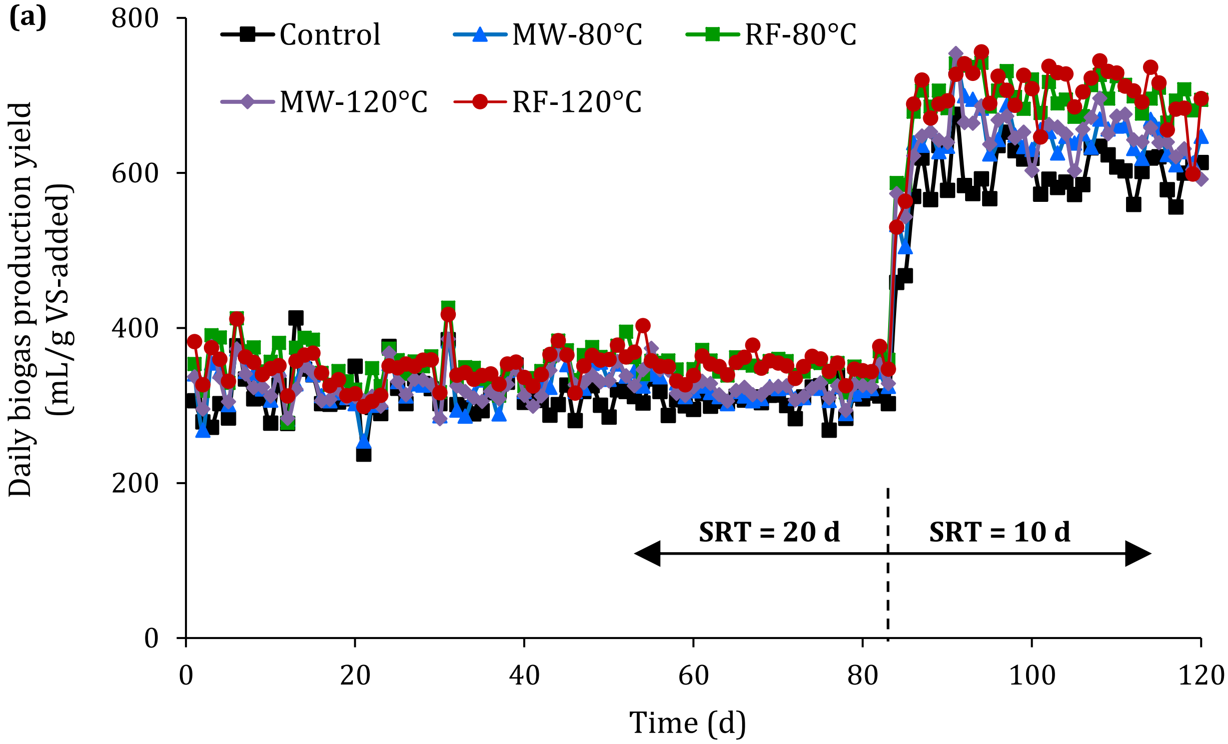
	Mesophilic					Thermophilic				
Description [a]	**Control**	**MW-80**	**RF-80**	**MW-120**	**RF-120**	**Control**	**MW-80**	**RF-80**	**MW-120**	**RF-120**
Organics removal										
TS removal (%)	32.6 ± 1.8	34.6 ± 2.2	34.6 ± 2.9	37.5 ± 1.7	37.6 ± 1.2	35.9 ± 1.4	36.4 ± 0.1	35.9 ± 0.1	37.1 ± 1.5	37.6 ± 0.9
VS removal (%)	38.3 ± 1.9	41.4 ± 2.1	40.7 ± 1.2	44.4 ± 0.4	44.5 ± 0.1	42.6 ± 1.1	42.8 ± 0.7	42.9 ± 0.1	43.9 ± 0.4	44.5 ± 0.8
TCOD removal (%)	37.1 ± 1.4	39.8 ± 2.1	39.6 ± 3.8	42.7 ± 1.5	42.7 ± 0.9	39.9 ± 1.2	40.3 ± 0.2	40.3 ± 0.9	41.4 ± 0.6	41.6 ± 2.3
Biogas and methane production										
Daily biogas production (mL/d)	501 ± 36	541 ± 34	544 ± 30	586 ± 29	587 ± 40	521 ± 40	522 ± 30	524 ± 35	537 ± 32	543 ± 40
Normalized daily biogas production ($\frac{mL}{L.d}$)	1001 ± 71	1082 ± 67	1088 ± 61	1172 ± 57	1175 ± 80	1041 ± 79	1044 ± 59	1047 ± 69	1074 ± 64	1087 ± 81
Methane yield ($\frac{mL}{g\ TCOD-added}$)	120 ± 9	130 ± 8	140 ± 7	130 ± 7	141 ± 10	125 ± 10	125 ± 7	129 ± 8	126 ± 8	130 ± 10
Methane yield ($\frac{mL}{g\ VS-added}$)	181 ± 13	209 ± 13	210 ± 12	226 ± 11	226 ± 15	201 ± 15	201 ± 11	202 ± 13	207 ± 12	209 ± 16
Specific methane production ($\frac{mL}{g\ TCOD-removed}$)	324 ± 23	326 ± 20	329 ± 16	331 ± 18	329 ± 23	313 ± 24	311 ± 18	315 ± 19	311 ± 21	315 ± 23
Digestate characteristics										
pH	7.24 ± 0.13	7.33 ± 0.08	7.32 ± 0.16	7.50 ± 0.11	7.51 ± 0.06	7.92 ± 0.10	7.95 ± 0.05	7.95 ± 0.09	7.96 ± 0.16	7.99 ± 0.14
Alkalinity (mg/L as $CaCO_3$)	2262 ± 33	2864 ± 28	2753 ± 35	3298 ± 11	3147 ± 93	2999 ± 58	3286 ± 86	3106 ± 57	3218 ± 60	3429 ± 43
Ammonia (mg/L as NH_3-N)	636 ± 24	739 ± 12	841 ± 47	920 ± 50	887 ± 41	1022 ± 43	1124 ± 31	1083 ± 12	1178 ± 66	1228 ± 32

[a] SRT: sludge retention time; MW: microwave; RF: radio frequency; TS: total solids; VS: volatile solids; TCOD: total chemical oxygen demand; SCOD: soluble chemical oxygen demand

7.3.1. Effect of MW and RF pretreatments on biogas production

The daily biogas yield of the mesophilic and thermophilic digesters (at STP) throughout the operation period are presented in Figure 7-2 (a) and (b), respectively. Neither the control nor the pretreated digesters showed any stability problems at the SRTs of 20 and 10 d. As per Figure 7-2, the difference between the daily biogas production of the control and pretreated digesters is more apparent at a SRT of 10 d compared to a SRT of 20 d. This observation is due to the fact that OLR increased at a SRT of 10 d. Therefore, the enzymes that are responsible for the hydrolysis stage are given less time to accomplish their task compared to that of the longer SRT of 20 d. Thus, the effect of thermal pretreatment which aims to accelerate the overall digestion process by breaking down the sludge structure (faster hydrolysis) will be higher under shorter SRTs.

(a)



(b)

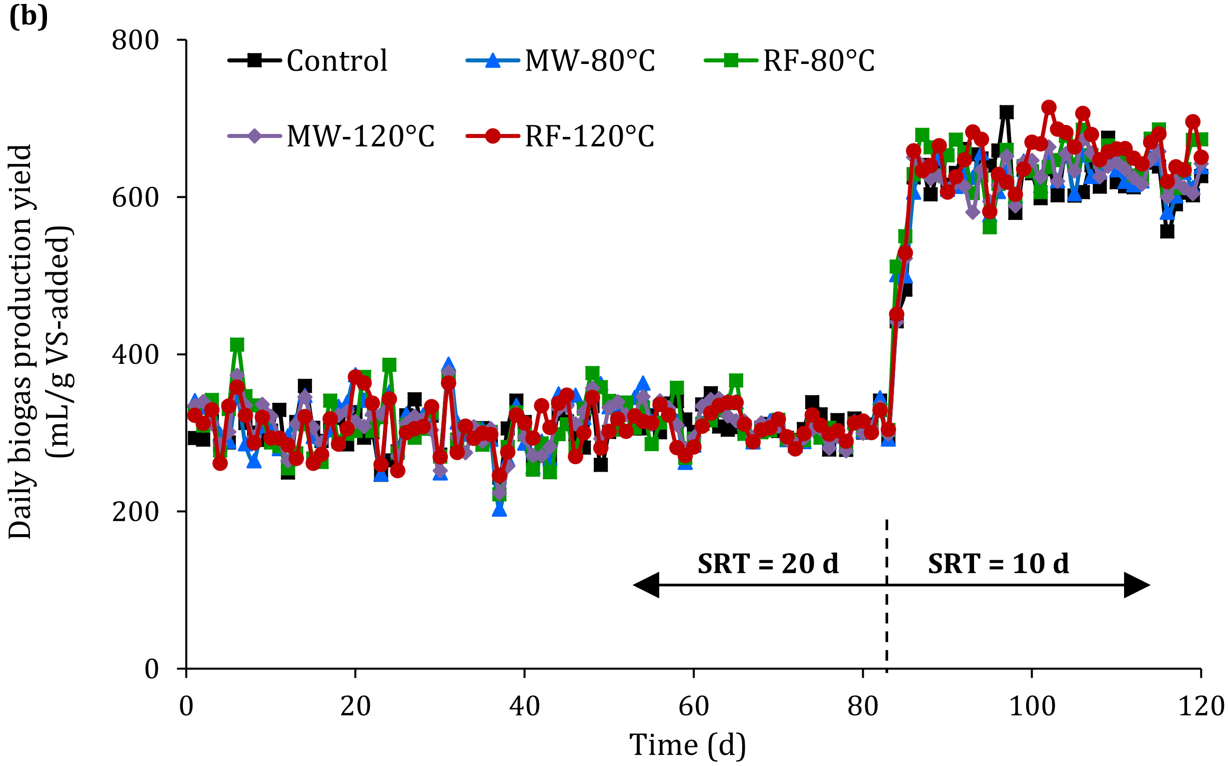


Figure 7-2. The daily biogas yield of the digesters at the STP of 0°C and 1 atm, (a) mesophilic digesters; (b) thermophilic digester

Figure 7-3 shows the relative (to control) improvement in methane yield of the mesophilic digesters fed with the pretreated sludge. Among the mesophilic digesters, the control digesters showed the lowest methane yield under both SRTs, whereas, the digesters fed with TWAS samples pretreated at 120°C achieved the highest biogas/methane yield. Under a SRT of 20 d, the maximum biogas production of 593 ± 39 $\frac{mL}{L.d}$ corresponding to the methane yield of 243 ± 16 $\frac{mL}{g\,VS-added}$ was obtained at the MW-120°C pretreatment scenario. This pretreatment scenario resulted in a 13.2% improvement for the average methane yield relative to the mesophilic control digester. Compared to these results, in another study, the methane yield of a digester fed with MW-pretreated WAS at 170°C (270 $\frac{mL}{g\,VS-added}$) was 17.4% higher than that of the a control digester fed with non-pretreated sludge (230 $\frac{mL}{g\,VS-added}$) (Chi et al., 2011). There are different factors affecting the biogas/methane production of the control and pretreated anaerobic digesters such as the pretreatment condition (i.e., temperature, rate, holding time), the digester SRT, wastewater composition, source of inoculum, and age of the sludge used as feed. With that said, the higher biogas/methane yield achieved in the control and pretreated digesters operated by Chi et al. (2011) compared to those of this research is likely due to the greater SRT (30 d) of the digesters and the higher pretreatment temperature (170°C) used in their study.

Under a SRT of 10 d, the pretreatment scenario of RF-120°C achieved the maximum biogas production of 1175 ± 80 $\frac{mL}{L.d}$ corresponding to the methane yield of 226 ± 15 $\frac{mL}{g\,VS-added}$, respectively. This corresponds to a 17.3% improvement when compared to the mesophilic control digester. As expected, the relative (to control) improvement of the mesophilic biogas/methane production was higher under a SRT of 10 d compared to that of a SRT of 20 d (Figure 7-3). According to the results of the main effect test, the pretreatment temperature of

80°C improved the mesophilic biogas production by 4.5% and 8.4% under the SRTs of 20 and 10 d, respectively. The average improvement ratios of 12.8% and 17.2% were also achieved by applying the thermal pretreatment at a temperature of 120°C under SRTs of 20 and 10 d, respectively. In a study conducted by Eskicioglu et al. (2007), the thermal pretreatment was applied on WAS using MW and CH methods up to the final temperatures of 50 and 96°C. The pretreated sludge was then used as feed in the semi-continuously fed digesters (Eskicioglu et al., 2007a). The authors reported that the relative (to control) improvement in the biogas production of the pretreated digesters was increased by reducing the SRT from 20 to 10 and 5 d which is in agreement with the results obtained in the current research. Similar findings for the effect of SRT on relative (to control) improvement of the biogas/methane production were reported by Medizadeh et al. (2013).

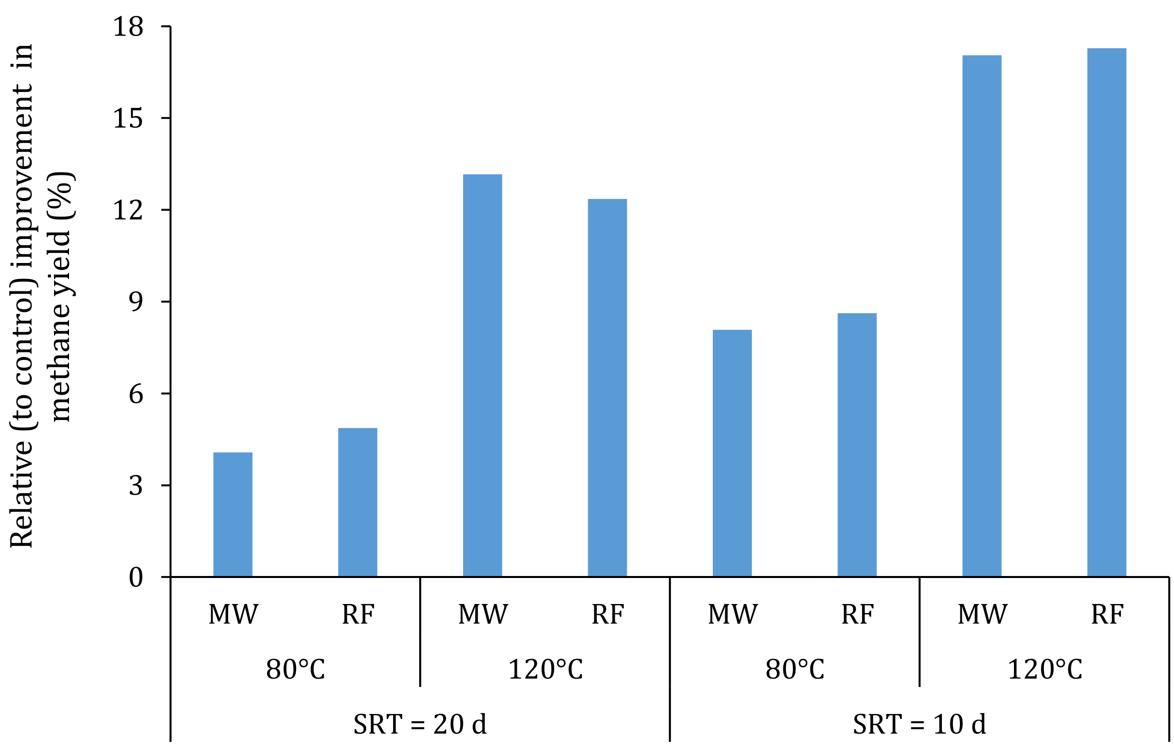


Figure 7-3. The relative (to control) improvement in mesophilic methane yield

According to the Statistical Science, when the 95% confidence interval ($CI_{95\%}$) of the means of two independent variables do not overlap, there will be a statistically significant difference between the means (at $\alpha = 0.05$) (Austin & Hux, 2002; Schenker & Gentleman, 2001). The opposite is not however necessarily true, meaning that two $CI_{95\%}$ may overlap and yet the two means can be statistically different. The $CI_{95\%}$ can be calculated using Eq. (5-1).

$$CI_{95\%} = 2SE \tag{7-1}$$

where SE is the standard error of the mean calculated using Eq. (7-2).

$$SE = \frac{SD}{\sqrt{N}} \tag{7-2}$$

where SD and N represent the standard deviation and size of the sample, respectively. The sample SD can be calculated using Eq. (7-3).

$$SD = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(x_i - \bar{x})^2} \tag{7-3}$$

where $\{x_1, x_2 \dots x_N\}$ are the observed values of the sample items and $\bar{x}$ is the mean value of these observations, while the denominator N is the sample size.

Figure 7-4 (a) and (b) illustrate the interval plot of the mesophilic biogas production at the SRTs of 20 d and 10 d, respectively. The error bars shown in the figure were calculated using the $CI_{95\%}$. As per Figure 7-4, using either the MW or RF pretreatment method, the mesophilic biogas production improves by increasing the pretreatment temperature from 80 to 120°C under both SRTs. The analysis of ANOVA also revealed the statistically significant effect of pretreatment temperature on the mesophilic biogas production at the SRTs of 20 and 10 d (p-value = 0.000<0.05). There was however no statistically significant difference between RF and MW pretreatment techniques in terms of mesophilic biogas production at a SRT of 20 d (p-value = 0.997>0.05) or the 10 d (p-value = 0.726>0.05). It is worth noting that there was no statistically significant difference among the OLR ($\frac{g\ VS-added}{L.d}$) and also the biogas methane percentage of the digesters. Therefore, the conclusions drawn from the statistical analysis performed on the normalized biogas production data (in $\frac{mL}{L.d}$) would be the same as those of the biogas and methane yield data. The representative Anderson-Darling test normality plot and the representative constant variance test results for mesophilic biogas production data are illustrated in Appendix B (Figure B- 6) and Appendix C (Figure C- 6), respectively.

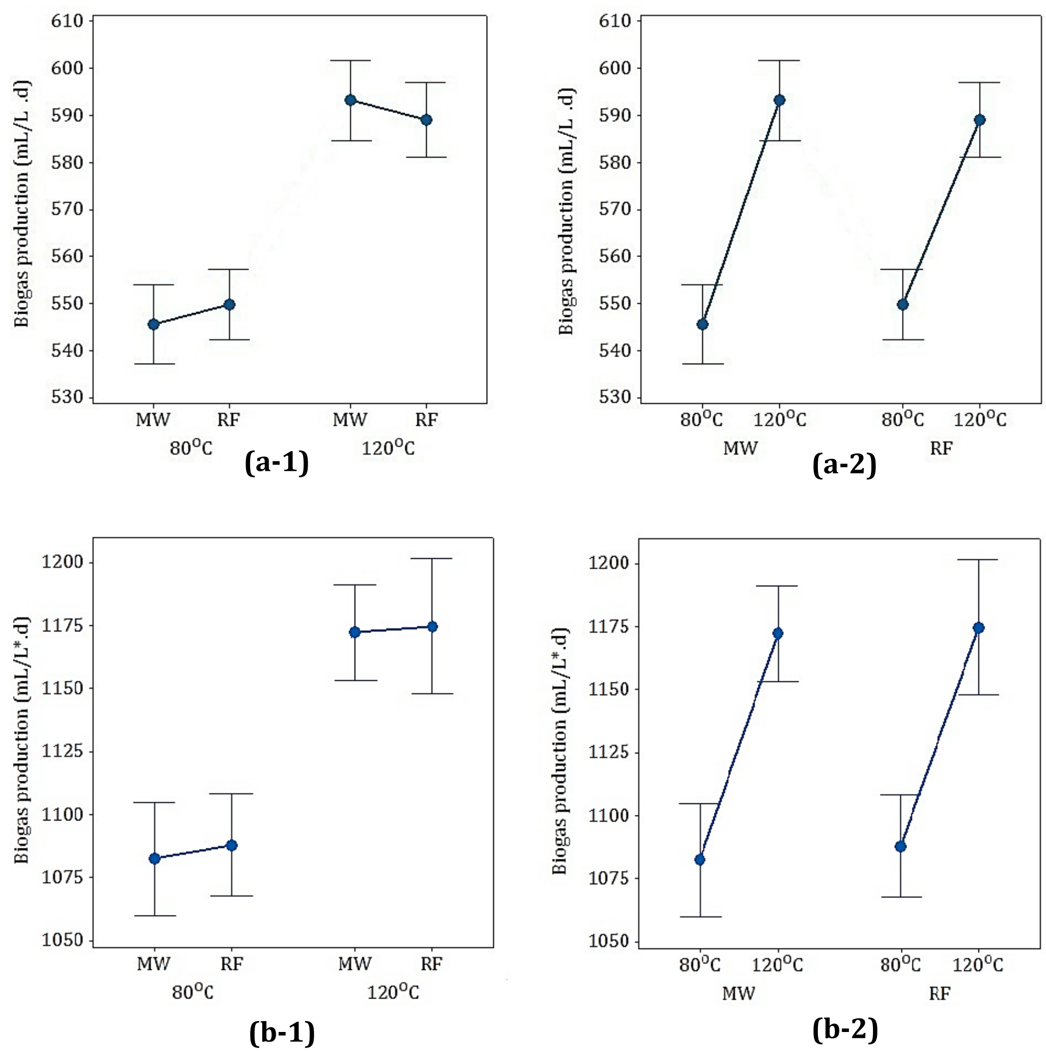


Figure 7-4. The interval plot of mesophilic biogas production at the SRTs of (a) 20 d and (b) 10 d (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

The relative (to control) improvement in methane yield of the thermophilic digesters are compared in Figure 7-5. Although, the average biogas production of the control thermophilic digester (514 ± 42 $\frac{mL}{L.d}$) was lower than that of the control mesophilic digester (524 ± 44 $\frac{mL}{L.d}$) under a SRT of 20 d, the t-test analysis proved no statistically significant difference between the mesophilic and thermophilic biogas production (p-value = 0.132>0.05). However, under a SRT of 10 d, the thermophilic control digester achieved statistically significantly higher biogas production (1041 ± 79 $\frac{mL}{L.d}$) than that of the mesophilic control digester (1002 ± 71 $\frac{mL}{L.d}$) with a corresponding p-value of 0.026.

The statistical analysis revealed no statistically significant effects of the pretreatment method (p-value = 0.588>0.05) and the pretreatment temperature (p-value = 0.327>0.05) on thermophilic biogas production under a SRT of 20 d. However, under a SRT of 10 d, the effect of the pretreatment temperature was proven to be statistically significant on thermophilic biogas production (p-value = 0.003<0.05), while there was no statistically meaningful effect of the pretreatment method (p-value = 0.486>0.05). Under this SRT (10 d), the maximum biogas production of 1087 ± 81 $\frac{mL}{L.d}$, corresponding to a methane yield of 209 ± 16 $\frac{mL}{g\ VS-added}$ was achieved at the RF-120°C pretreatment scenario. This pretreatment scenario resulted in a 4.3% improvement in the methane yield relative to the thermophilic control digester (Figure 7-5). The representative Anderson-Darling test normality plot and the representative constant variance test results for thermophilic biogas production data are illustrated in Appendix B (Figure B- 7) and Appendix C (Figure C- 7), respectively.

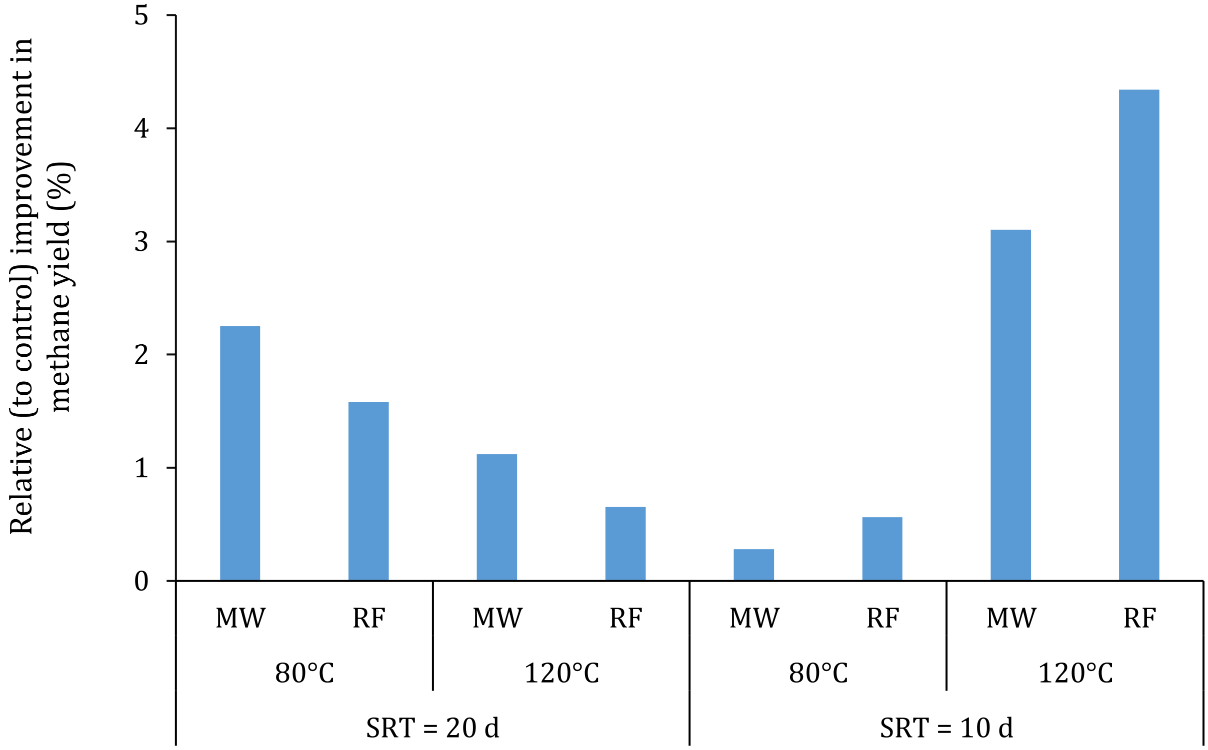


Figure 7-5. The relative (to control) improvement in thermophilic methane yield

Figure 7-6 (a) and (b) illustrate the main effect plot of the thermophilic biogas production at the SRTs of 20 d and 10 d, respectively. The error bars in Figure 7-6 were calculated using the $CI_{95\%}$ determined from Eq. (5-1). The illustrated error bars ($CI_{95\%}$ bars) in Figure 7-6 are compatible with the results of the statistical analysis. The significantly overlapping error bars in Figure 7-6 (a-1) and (b-1) agree with the statistically insignificant effect of the pretreatment method on thermophilic biogas yield at both SRTs which were proven by the ANOVA. The same conclusion can be derived for the significantly overlapping error bars in Figure 7-6 (a-2) which is consistent with the p-value > 0.05 for the effect of pretreatment temperature on thermophilic biogas production at a SRT of 20 d. However, the error bars in Figure 7-6 (b-2) do not overlap suggesting a statistically significant effect of the pretreatment temperature at a SRT of 10 d (p-value<0.05).

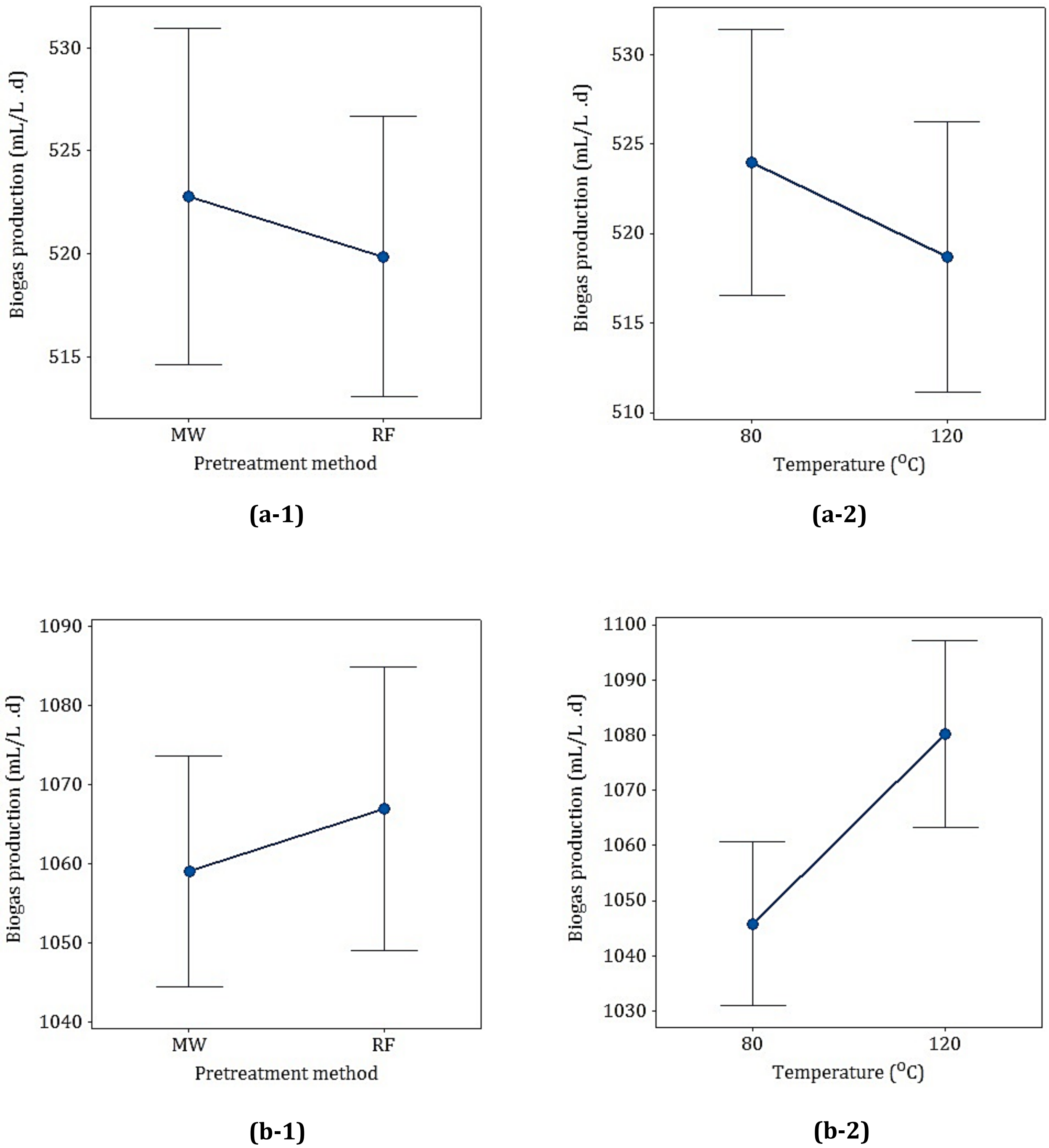


Figure 7-6. The main plot of thermophilic biogas production at the SRTs of (a) 20 d and (b) 10 d (95% confidence interval ($CI_{95\%}$) was used for error bars calculation)

Figure 7-7 compares the percentage difference (*PD*) between the mesophilic and thermophilic biogas yield at different digestion scenarios. The PD can be calculated using Eq. (7-4). The subscript i is the corresponding digestion scenario for which the PD is determined.

$$PD(\%) = \frac{MBPY_i - TBPY_i}{MBPY_i} \times 100 \quad (7\text{-}4)$$

in which $MBPY$ and $TBPY$ are the mesophilic and thermophilic biogas yield ($\frac{mL}{g\ VS\text{-}added}$), respectively.

As seen in Figure 7-7, the PD values corresponding to all the digestion scenarios under a SRT of 10 d are lower than those of a 20 d SRT. This observation agrees with the results of the statistical analysis in which the effect of thermal pretreatment on mesophilic biogas yield was proven to be statistically significant. However, under the thermophilic condition the thermal pretreatment increased the biogas yield only under a SRT of 10 d, while no statistically meaningful effect was observed at a SRT of 20 d. It should be noted that the negative value observed in Figure 7-7 means higher biogas production in the thermophilic control compared to a mesophilic control. In general, the thermophilic control digesters benefit from a faster hydrolysis rate than that of the mesophilic controls because of the higher operating temperature (i.e. 55°C vs. 35°C). This can result in higher biogas production rate and extent especially at lower ranges of SRTs when OLRs are higher (i.e. 10 d). However, when a digester is operated under a long enough SRT (i.e. 20 d), the difference between the control mesophilic and thermophilic biogas production will be minimized.

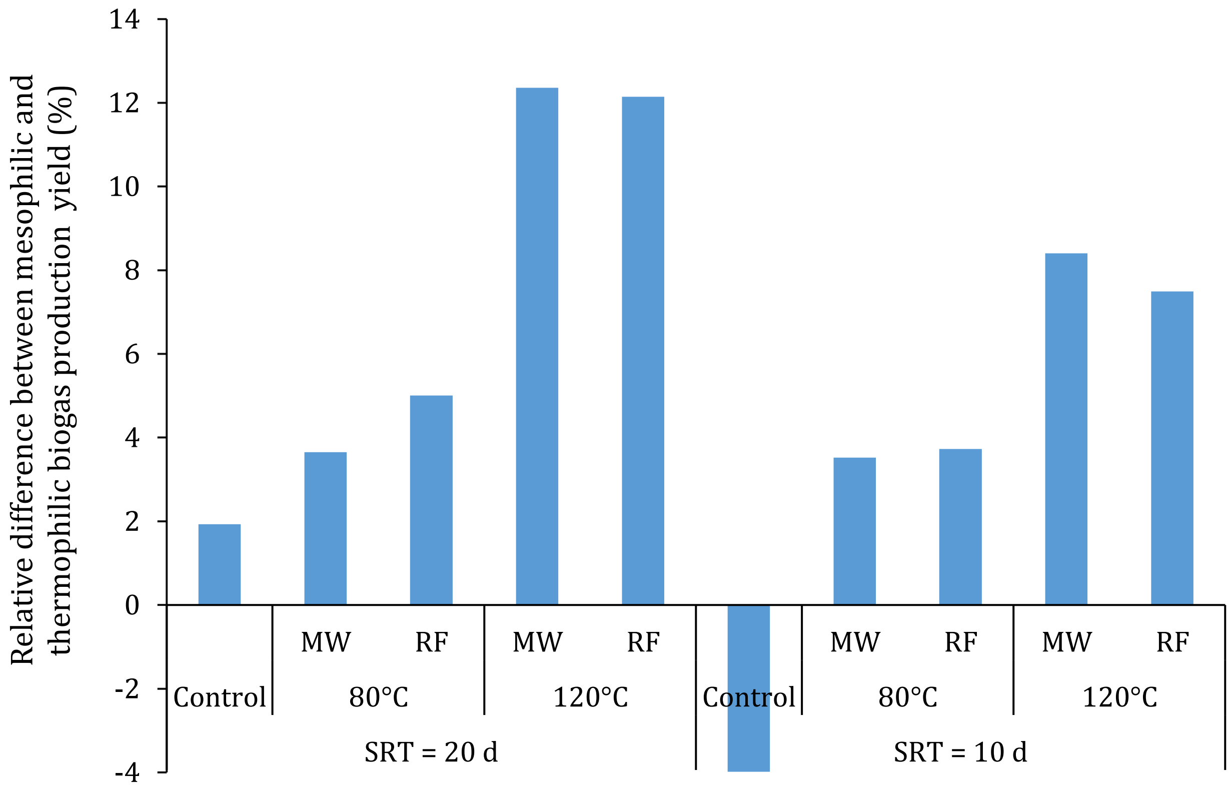


Figure 7-7. The percentage difference (PD) between the mesophilic and thermophilic biogas production at different digestion scenarios

Table 7-3 and Table 7-4 report the specific methane yield of the mesophilic and thermophilic digesters, respectively, which is comparable to the theoretical value of 350 $\frac{mL}{g\ TCOD-removed}$ (Metcalf & Eddy, 2013). In practice, the amount of methane recovered per gram of COD removed is usually less than the theoretical value because some of the COD may be used as a source of redox equivalents for microbial growth (Heidrich et al., 2011). Another factor reducing the actual measured methane yield compared to the theoretical value is the biogas leakage during the feeding and effluent discharging steps as well as during its volume measurement with the monometer.

7.3.2. Effect of MW and RF pretreatments on organics removal

The relative (to mesophilic control) improvement in solids (TS and VS) removal after thermal pretreatment is illustrated in Figure 7-8. Under a SRT of 20 d, the maximum removal efficiency of TS (44.8 ± 1.4%) and VS (52.1 ± 1.7%) was obtained at the pretreatment temperature of 120°C, corresponding to relative (to control) improvement ratios of 9.9% and 9.7%, respectively (Figure 7-8). Similar results were obtained under a SRT of 10 d at which a temperature of 120°C achieved the highest efficiency of TS (37.6 ± 1.2%) and VS (44.5 ± 0.1%). This observation is consistent with the biogas production results which shows increased mesophilic methane yield with an increase in the pretreatment temperature (Figure 7-3).

The statistical analysis proved that the pretreatment temperature had a significant effect on mesophilic TS and VS removal efficiency at both SRTs (p-value<0.05). According to the results of the main effect test, under a SRT of 20 d, the pretreatment temperatures of 80 and 120°C resulted in average TS removal efficiencies of 38.6 and 40.9%, and average VS removal efficiencies of 45.7 and 47.7%, respectively. Under a SRT of 10 d, the average TS and VS removal efficiency were increased from 31.77% and 37.7% at the pretreatment temperature of 80°C to 34.4 and 40.8% at 120°C respectively. In contrast to the statistically significant effect of the pretreatment temperature on the mesophilic solids removal efficiency, no statistically meaningful difference was observed between the TS (p-value $= 0.421>0.05$) and VS (p-value $= 0.942>0.05$) concentration of the digesters' effluent fed with RF and MW-pretreated TWAS.

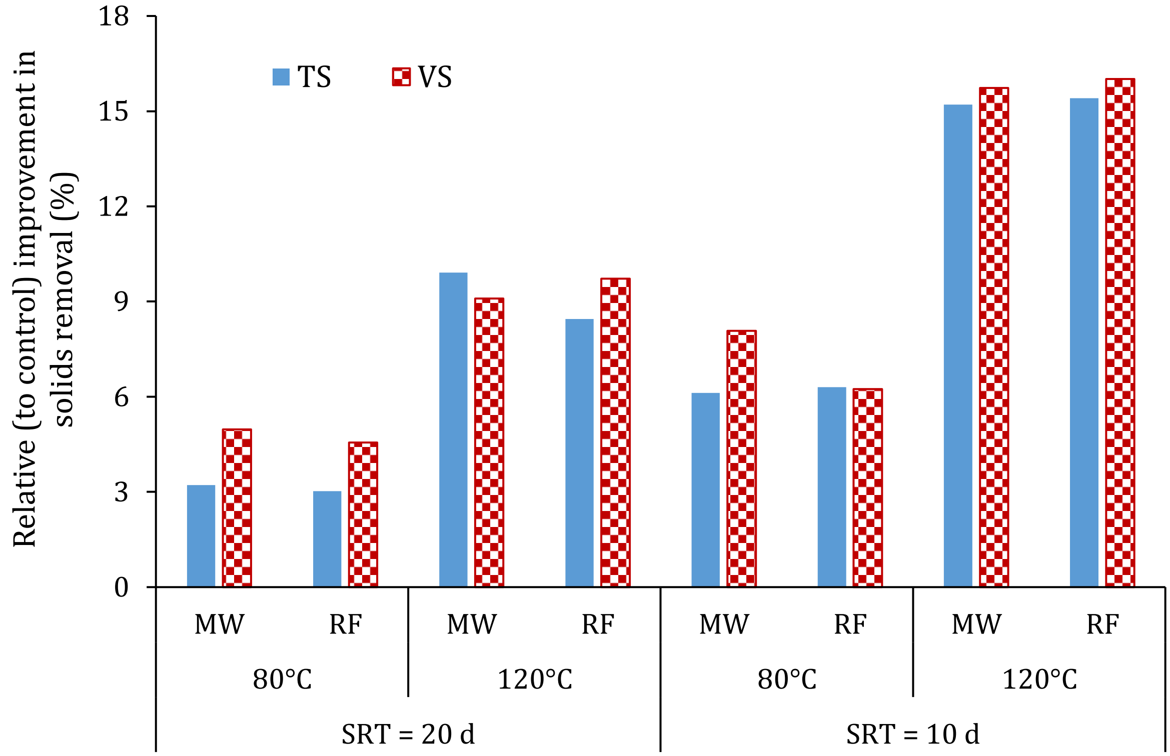


Figure 7-8. The relative (to control) improvement in solids removal after thermal pretreatment in mesophilic digesters

Although the solids removal efficiency of the mesophilic digesters decreased by reducing the SRT from 20 d to 10 d, the relative (to control) improvement of the pretreated digesters increased (Figure 7-8). This observation agrees with the analysis of the biogas data (Figure 7-3) suggesting that thermal pretreatment is more advantageous under a lower SRT. In another study (Wahidunnabi & Eskicioglu, 2014), the mechanical pretreatment was applied to WAS using a high pressure homogenizer. The authors observed higher relative (to control) improvement in the VS removal under a SRT of 14 d (24.0%) compared to that of 20 d SRT (14.2%) which is in consistent with the results obtained in this research. Similar findings were reported by other researchers (Akgul et al., 2017; Mehdizadeh et al., 2013).

The relative (to thermophilic control) improvement in the TS and VS removal efficiency after thermal pretreatment is illustrated in Figure 7-9. Despite the positive effect of thermal pretreatment on mesophilic solids removal efficiency (Figure 7-8), no statistically significant effect of thermal pretreatment on thermophilic TS (p-value = 0.306>0.05) and VS (p-value = 0.308>0.05) removal was proven under a SRT of 20 d. However, the thermal pretreatment improved the TS (p-value = 0.026<0.05) and VS (p-value = 0.001<0.05) removal in the pretreated thermophilic digesters under a SRT of 10 d, although the relative (to control) improvement ratios of the thermophilic pretreated digesters were less than those of the mesophilic ones. In agreement with this observations, the effect of sludge hydrolysis on solids removal was reported to be more discernible under mesophilic condition (compared to the thermophilic condition) by other researchers (Coelho et al., 2011; Mehdizadeh et al., 2013; Wahidunnabi & Eskicioglu, 2014). In terms of the effect of the two pretreatment methods used, no statistically significant difference was observed among the digesters fed with MW- or RF-pretreated TWAS under both SRTs. The analysis of t-test revealed no statistically significant difference between the TS (p-value = 0.410>0.05) and VS (p-value = 0.132>0.05) removal efficiency of the control mesophilic and thermophilic digesters under a SRT of 20 d. However, under a SRT of 10 d, the thermophilic control digester achieved statistically significantly higher solids removal than that of the mesophilic control digester.

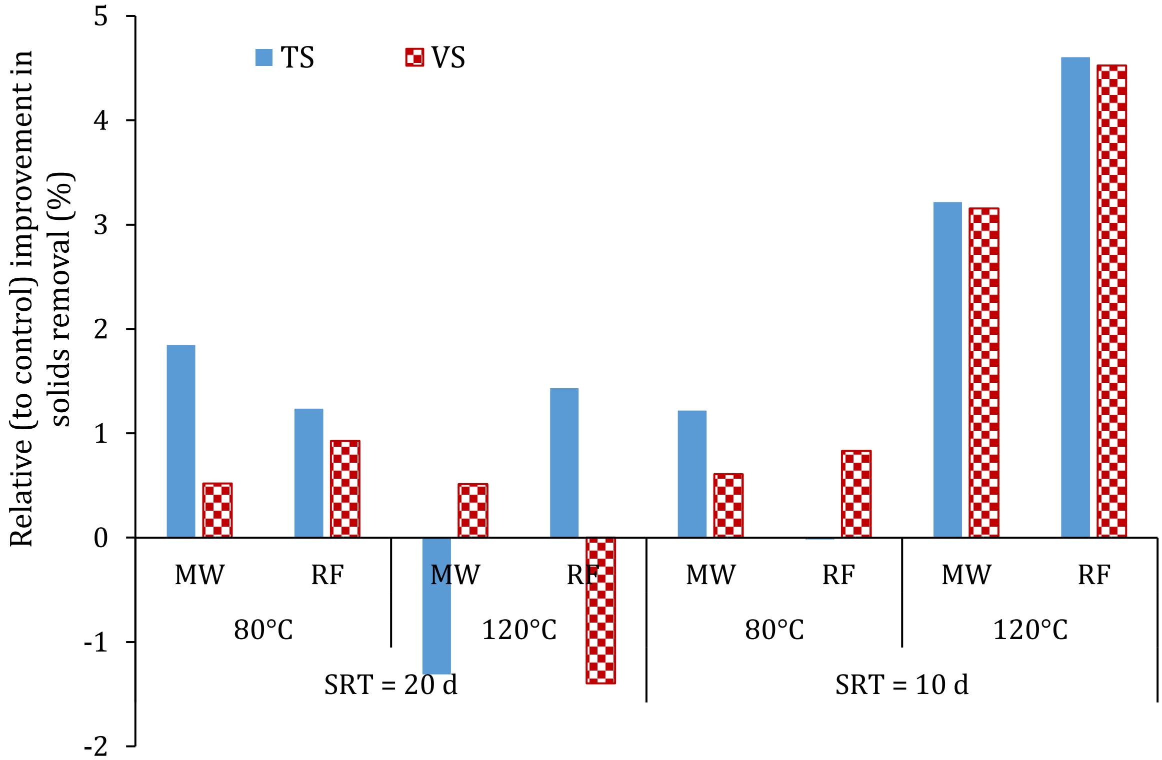


Figure 7-9. The relative (to control) improvement in solids removal after thermal pretreatment in thermophilic digesters

The results of TCOD removal of the mesophilic and thermophilic digesters demonstrated similar patterns as those of the solids removal under a SRT of 20 d and 10 d. Figure 7-10 compares the relative (to control) improvement in TCOD removal efficiency of the mesophilic digesters fed with pretreated TWAS. Under a SRT of 20 d, the pretreated digesters showed an increased TCOD removal when compared to the control digester. The pretreatment temperatures of 80 and 120°C achieved an average of 6.0% and 10.8% increase in TCOD removal, respectively. As expected, under the 10 d SRT, the effect of thermal pretreatment was more apparent resulting in an average improvement of 7.1% and 15.3% over the control digester at the temperatures of 80 and 120°C, respectively.

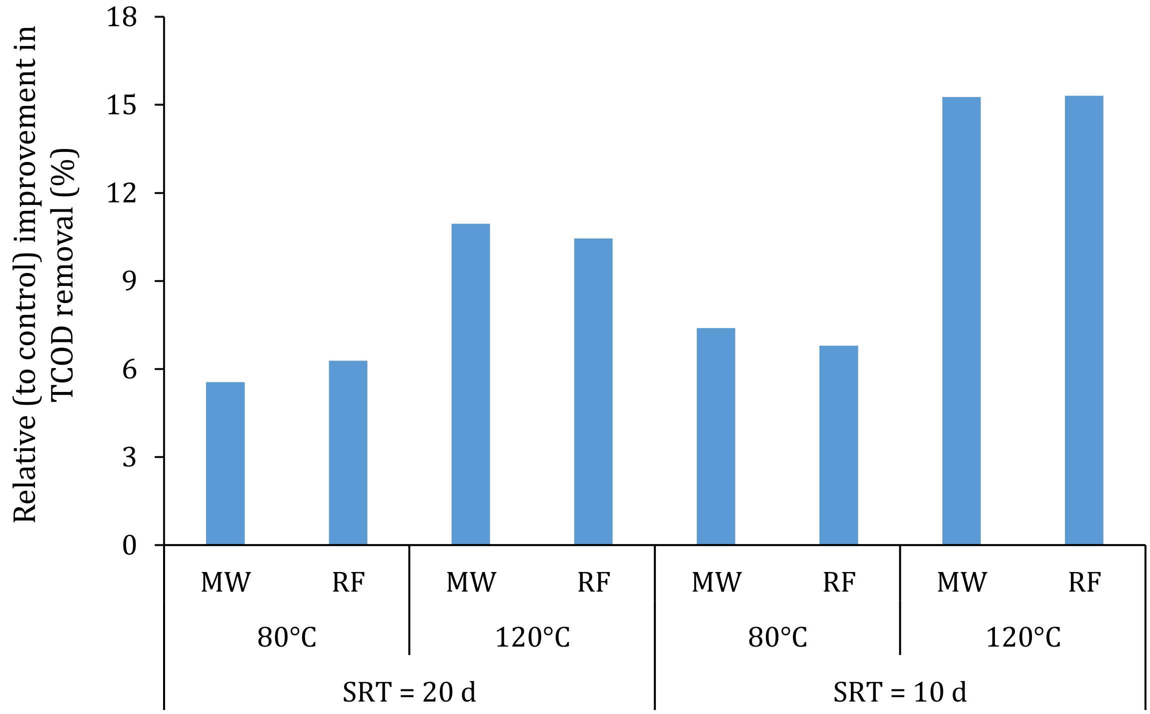


Figure 7-10. The relative (to control) improvement in TCOD removal after thermal pretreatment in mesophilic digesters

As per the ANOVA results, the pretreatment temperature had statistically significant effect on mesophilic TCOD removal at both the SRTs, with higher and lower removal efficiency at the temperature of 120°C and 80°C, respectively (p-value<0.05). No statistically significant effect of the pretreatment method (MW vs. RF) was proven for TCOD removal at both the SRTs (p-value>0.05).

The relative (to control) improvement in TCOD removal efficiency of the mesophilic digesters after thermal pretreatment are shown in Figure 7-11. Considering the results obtained under the 20 d SRT, neither the pretreatment method (p-value = 0.867>0.05) nor the temperature (p-value = 0.093>0.05) had statistically proven effects on thermophilic TCOD removal. However, under a 10 d SRT, increasing the pretreatment temperature from 80 to 120°C increased the average TCOD removal efficiency with a maximum of 4.3% improvement over the thermophilic control. This increase was achieved under the RF-120°C scenario as seen in Figure 7-11 (b). Using either MW or RF pretreatment did not affect the thermophilic TCOD removal at a SRTs of 20 and 10 d (p-value>0.05). The t-test analysis proved no statistically meaningful difference between the mesophilic and thermophilic control digesters in terms of TCOD removal under 20 d SRT (p-value = 0.751>0.05). However, the control thermophilic digester achieved higher TCOD removal (39.9 ± 1.2%) than that of the mesophilic control (37.1 ± 1.4%) at a SRT of 10 d (p-value = 0.002<0.05). This is due to the improved hydrolysis stage resulting from higher operating temperature (55°C) under the thermophilic condition compared to that of the mesophilic condition (35°C).

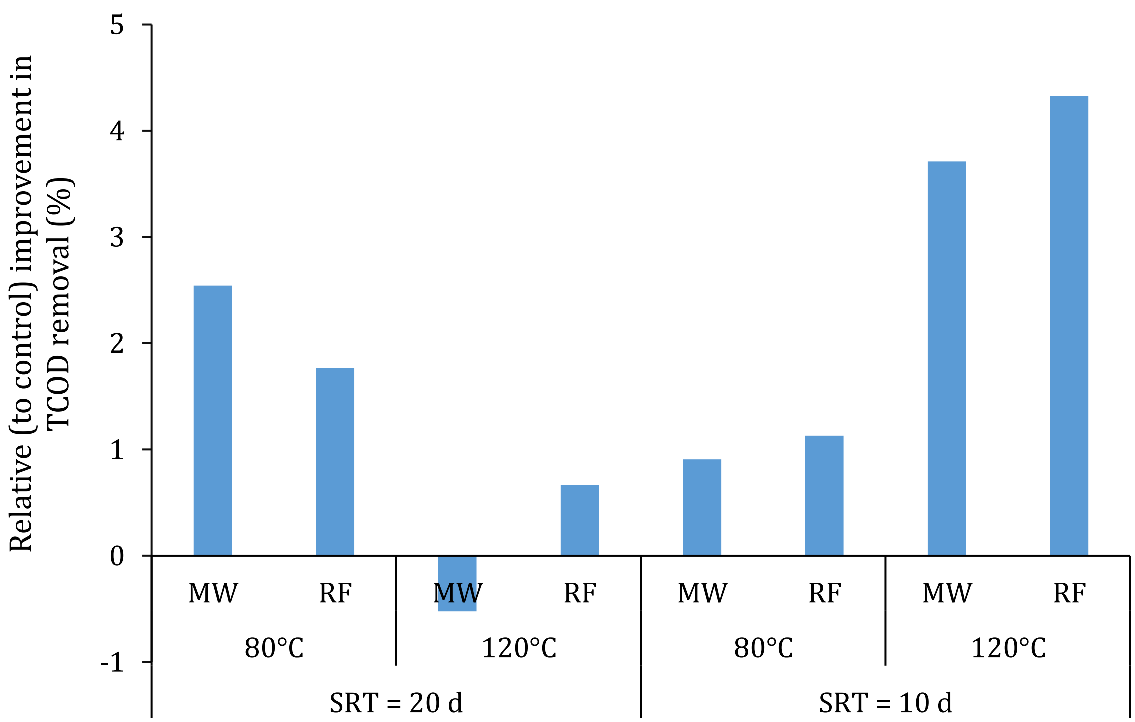


Figure 7-11. The Relative (to control) improvement in thermophilic TCOD removal after thermal pretreatment

7.3.3. Effect of MW and RF pretreatments on digestate characteristics

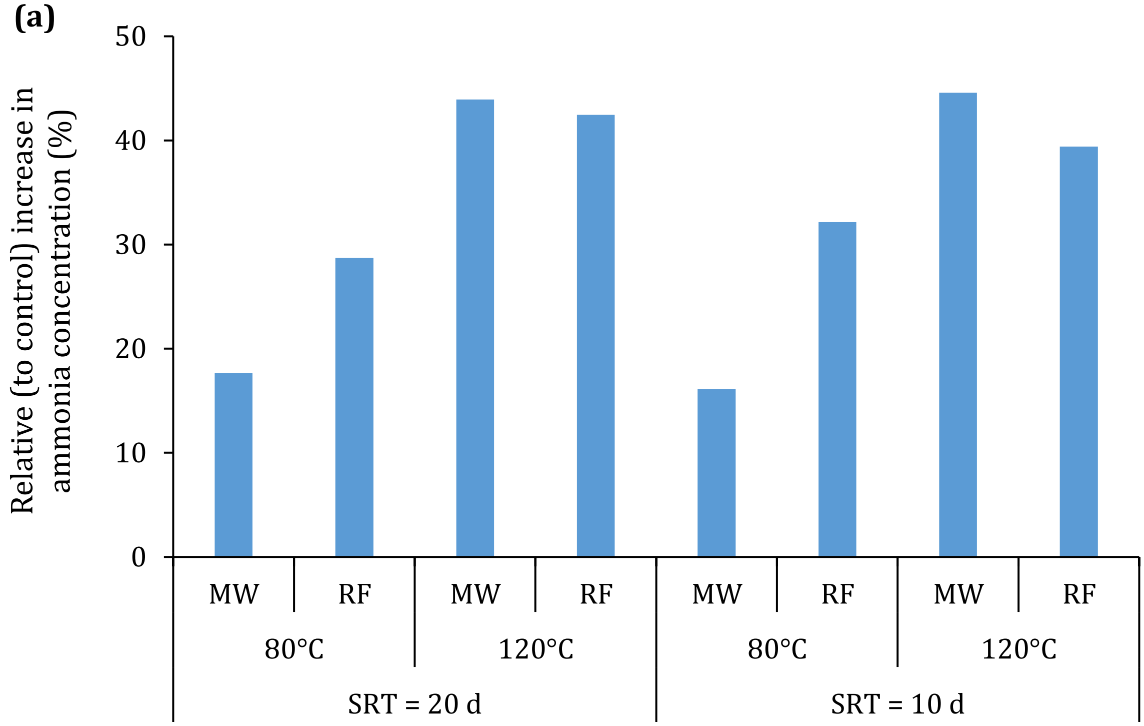
Monitoring the pH and the concentration of VFA, alkalinity, and ammonia of the digestate is common practice to assess the digester stability along with the quality of the effluent. For all of the mesophilic and thermophilic digesters operated under the SRTs of 20 and 10 d, the concentration of total VFA were measured within the safe range of < 250 mg/L (Metcalf & Eddy, 2013). Alkalinity is the most common indicator of the buffering characteristic of the digester solution required for pH control. In this study, the alkalinity concentration of the digestate representing the alkalinity content of the digester solution was monitored throughout the operation period at the SRTs of 20 and 10 d.

The average alkalinity concentration of all the digesters was more than 2000 mg/L as $CaCO_3$ (Table 7-3 and Table 7-4). The total VFA to alkalinity ratio is often used to evaluate the stability of the anaerobic digestion process (Khanal, 2009). Droste (1997) suggested a safe range of VFA/alkalinity ratio of less than 0.3-0.4. However, a range of 0.1-0.25 was reported by other researchers as an indicator of the ideal condition for the AD process (Appels et al., 2008). In this study, all the digesters showed a VFA/alkalinity ratio of less than 0.11 during the 120-day operation period.

The pH of the mesophilic digesters was in the range of 7.2 to 7.6 during the entire operation period (Table 7-3 and Table 7-4). However, the thermophilic digesters demonstrated higher pH than that of the mesophilic digesters varying from 7.8 to 8. The higher pH of the thermophilic digesters is most likely due to the greater Henry's constant for CO_2 at the temperature of 55°C compared to that of 35°C which could decrease the concentration of CO_2 at the liquid phase (Moen et al., 2003).

After the digestion process, the digestate is usually dewatered and the centrate from the dewatering process is recycled back to the wastewater mainstream which can increase the nitrogenous nutrient load to the WWPT. The reduction of nitrogen content in wastewater to a safe level is essential prior to its discharge to the environment to prevent eutrophication. Therefore, monitoring the level of nitrogenous components in the digester effluent is imperative. In this study, the concentration of ammonia as the main component of inorganic nitrogen was measured in the digestate. Determination of ammonia concentration during the digestion process is necessary due to its potential toxicity effect at high concentrations. The inhibitory effect of ammonia on mesophilic and thermophilic AD was observed for the concentration above 1,900-2,400 mg/L (Moen et al., 2003). Figure 7-12 (a) and (b) show the relative (to control) change in the ammonia concentration of the mesophilic and thermophilic digesters fed with pretreated TWAS, respectively. In this study, the ammonia concentration in all the digesters was below the toxicity threshold (1,900 mg/L). All the pretreated digesters generated more ammonia than the control digester. As per the results of the statistical analysis, the greater the pretreatment temperature, the higher the ammonia concentration will be. No statistically significant effect of pretreatment method (MW vs RF) was observed on the concentration of ammonia under the SRTs of 20 and 10 d (p-value>0.05). The higher ammonia concentration of the thermophilic digesters compared to that of the mesophilic digesters can be explained by the greater degradation of proteins at a temperature of 55°C than that of 35°C.

(a)



(b)

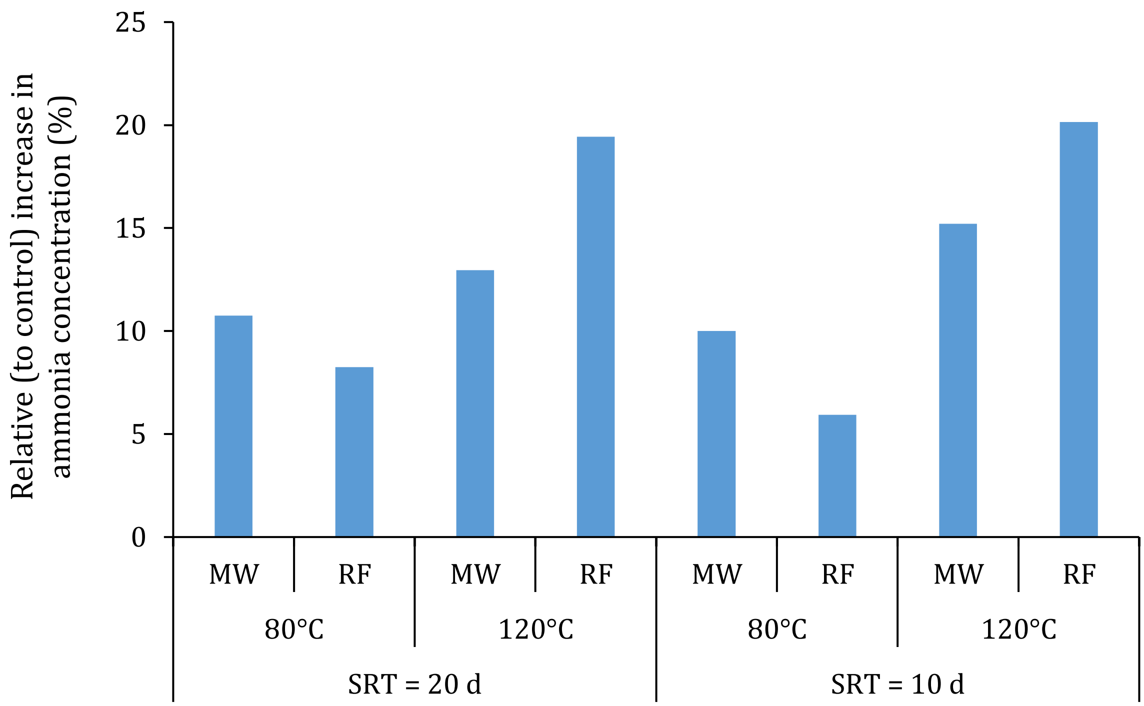


Figure 7-12. The relative (to control) increase in the concentration of ammonia in (a) mesophilic digesters and (b) thermophilic digesters

7.3.4. Energy assessment of RF and MW pretreatment systems

The output energy (E_{out}) of an AD system is defined as the amount of energy recovered during the AD process as methane (GJ/tonne TS-added). In this study, the E_{out} of the digesters was determined considering the methane energy content of 55.6 $\frac{KJ}{g\ CH_4}$ and the density of 0.715 $\frac{g}{L}$ at STP (Metcalf & Eddy, 2013). Figure 7-13 illustrates the E_{out} of the mesophilic and thermophilic anaerobic digesters. As seen in Figure 7-13 and in consistent with the biogas production data (Figure 7-3 and Figure 7-5), the thermal pretreatment had a notable effect on the output energy of the mesophilic digesters fed with pretreated TWAS. However, the effect of thermal pretreatment on enhanced thermophilic energy recovery was not discernible.

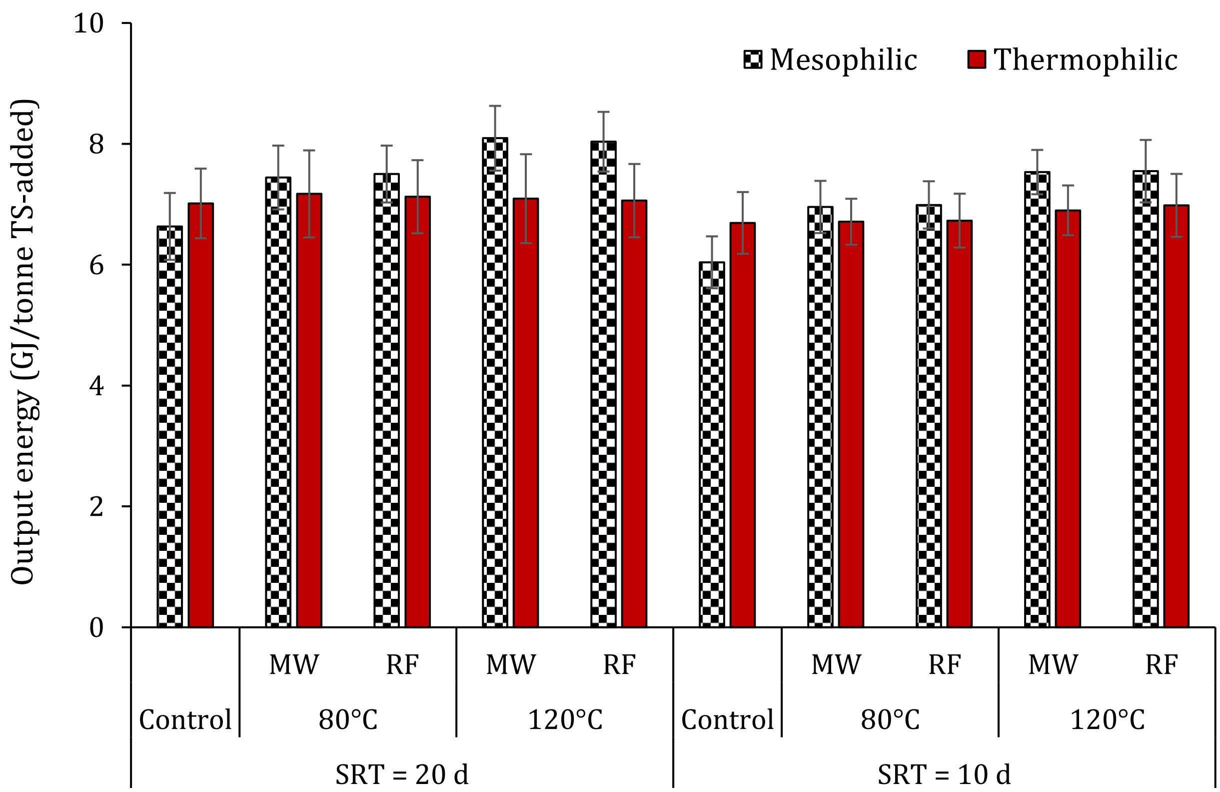


Figure 7-13. The output energy of the mesophilic and thermophilic anaerobic digesters

The overall trend of the thermal pretreatment effect on AD performance was that the higher the pretreatment temperature is, the more energy recovery (output energy) from the system will be. However, due to more energy consumption, the net energy (E_{net}) of the system may not necessarily be higher at elevated pretreatment temperatures. The net energy of an advanced AD system (pretreatment +AD) can be determined by subtracting the amount of energy consumed during the sludge pretreatment (input energy) from the amount of energy generated as methane (output energy) represented in Eq. (7-5).

$$E_{net} = E_{out} - E_{in} \tag{7-5}$$

in which, E_{net}, E_{out} and E_{in} are the system net energy, output energy and input energy (in the unit of $\frac{GJ}{tonne\ TS-added}$), respectively.

It should be noted that according to the Eq. (7-5), the net energy will be equal to the output energy for a conventional AD system without pretreatment since $E_{in} = 0$. In this study, the control digesters fed with non-pretreated TWAS are examples of a conventional AD system.

The maximum net energy that is theoretically possible to achieve (E_{net}^{max}) can be determined using Eq. (7-6).

$$E_{net}^{max} = E_{out} - E_{th} \tag{7-6}$$

where the E_{th} is the theoretical minimum required energy. The E_{th} ($\frac{GJ}{tonne\ TS-added}$) which is required to increase the sludge temperature from an initial temperature of T1 (°C) to a final temperature of T2 (°C) can be determined using Eq. (7-7)

$$E_{th}({}^{GJ}/_{tonne\ TS-added}) = \frac{1}{(TS)}\int_{T_1}^{T_2} C_T dT \tag{7-7}$$

where C_T is the TWAS specific heat capacity at a temperature of T which can be assumed to be the same as that of water ($4.18\ \frac{J}{g.°C}$ or $4.18 \times 10^{-3}\ \frac{GJ}{tonne.°C}$ at 20°C) (Mehdizadeh et al., 2013; Turovskiy & Mathai, 2006). The denominator TS is the total solids concentration of the TWAS (%, by weight).

Figure 7-14 compares the amount of the energy consumed (per tonne of TS) during the RF and MW pretreatments with the theoretical minimum required energy (E_{th}). Figure 7-14 revealed that when the MW oven is operated at a frequency of 2.45 GHz, it utilizes significantly higher energy during its operation compared to the custom-designed 13.56 MHz RF pretreatment system. The difference between the actual energy consumption during the RF pretreatment and the theoretical minimum energy requirement increased with an increase in the pretreatment temperature (Figure 7-14). As previously discussed in Chapter 5, the RF pretreatment system was designed to have a power transfer efficiency more than 95% at a temperature range of 40°C to 120°C with a maximum efficiency of approximately 100% at 70°C. On the other hand, the efficiency of the RF amplifier was almost constant during its operation. Therefore, the energy inefficiency of the RF system was primarily due to the heat loss from the body of the heating vessel during pretreatment. This explains the lower energy efficiency of the RF system at higher ranges of temperature as a result of increased heat loss caused by the greater differential temperature between the load and the surrounding environment. According to the energy analysis results, the energy efficiency of the RF system varied between 68-92% between 40-120°C which was significantly higher than that of the MW system (38-46%).

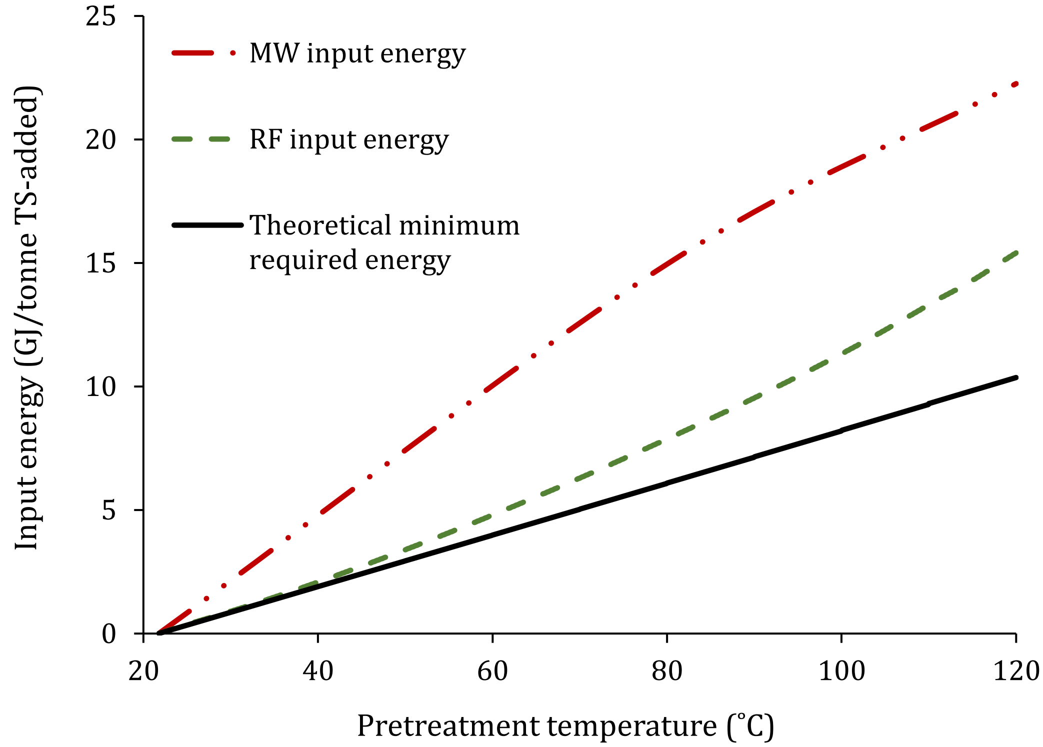


Figure 7-14. Comparison of the RF and MW input energy with the theoretical minimum required energy (E_{th})

Figure 7-15 compares the net energy of the control and pretreated digesters (per tonne of TS added to each digester) which were determined using Eq. (7-5). All the digesters fed with pretreated TWAS achieved negative net energy meaning their output energy could not compensate for their input energy. It is clear that under similar pretreatment temperatures, the net energy of the MW-pretreated digesters is significantly lower than that of the RF-pretreated digesters due to higher input energy of the MW pretreatment system. Another observation is that although the output energy of the mesophilic digesters which were fed with 120°C-pretreated TWAS were greater than that of 80°C (Figure 7-13), they demonstrated lower net energy (Figure 7-15) since they could not compensate for the additional energy required to increase the TWAS temperature from 80°C to 120°C.

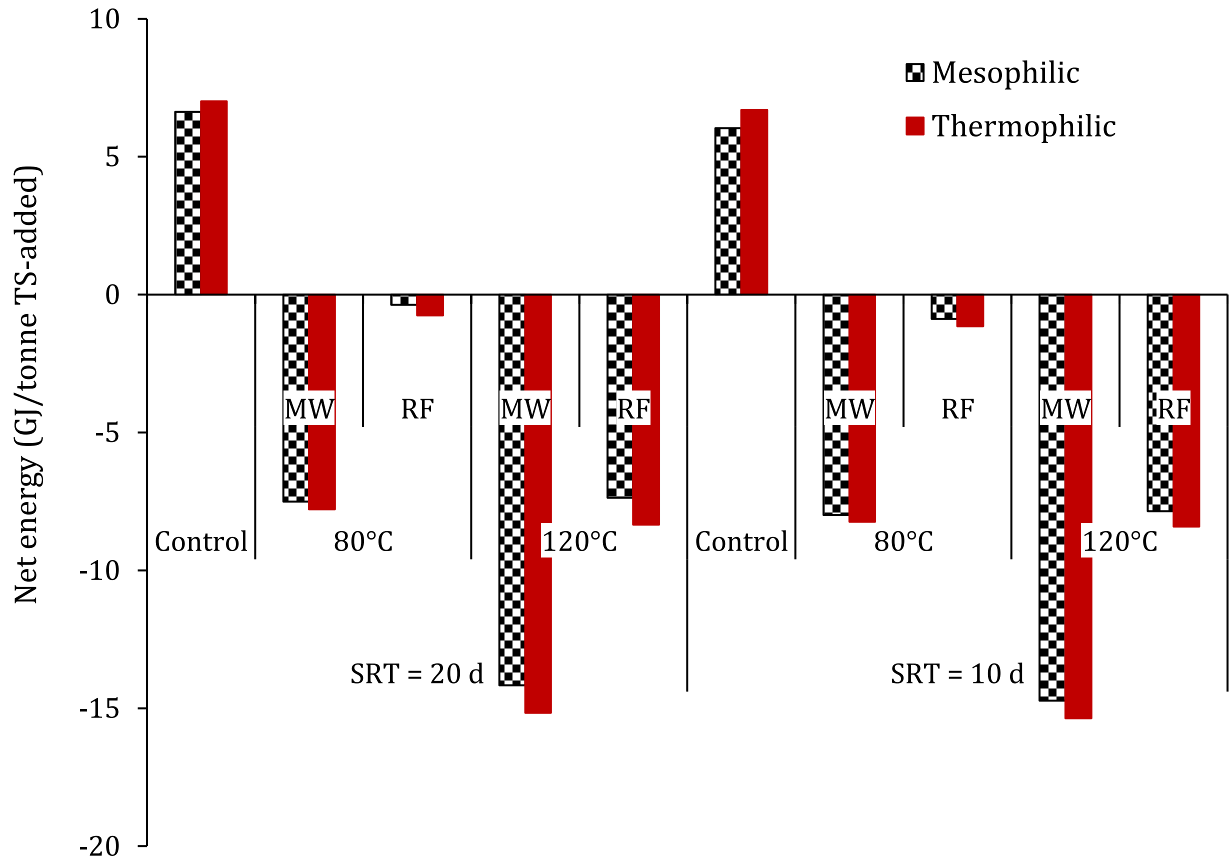


Figure 7-15. The net energy of the mesophilic and thermophilic anaerobic digesters

To increase the net energy production of an advanced AD system, the thermal pretreatment can be performed on sludge samples with higher TS content (thicker sludge) to reduce the input energy (per tonne of TS added). Additionally, it can be assumed that the output energy per tonne of TS added remains constant based on the literature that the thickening/dewatering process does not change the anaerobic digestibility of the sludge (Barber, 2016). Therefore, the net energy of the system will increase according to Eq. (7-5).

Figure 7-16 illustrates the change in the net energy production of the mesophilic digesters fed with pretreated TWAS as a function of sludge TS content and compare it with the theoretical maximum possible net energy (E_{net}^{max}). The E_{net}^{max} was determined using Eq. (7-6). The effect of the sludge TS concentration on the net energy production of the thermophilic pretreated digesters is also shown in Figure 7-17. As expected, for a given TS value, the net energy is higher for the digesters fed with RF-pretreated sludge than those fed with MW-pretreated TWAS. It is also observed that the difference between the “actual” and “theoretical maximum possible” net energy is reduced by increasing the sludge TS concentration confirming that thermal pretreatment is more beneficial at higher TS contents. From the energy point of view, among all the tested scenarios, conducting thermal pretreatment using RF heating system to a temperature of 80°C is the most feasible option as it demonstrated the minimum deviation from E_{net}^{max} at any given TS concentration.

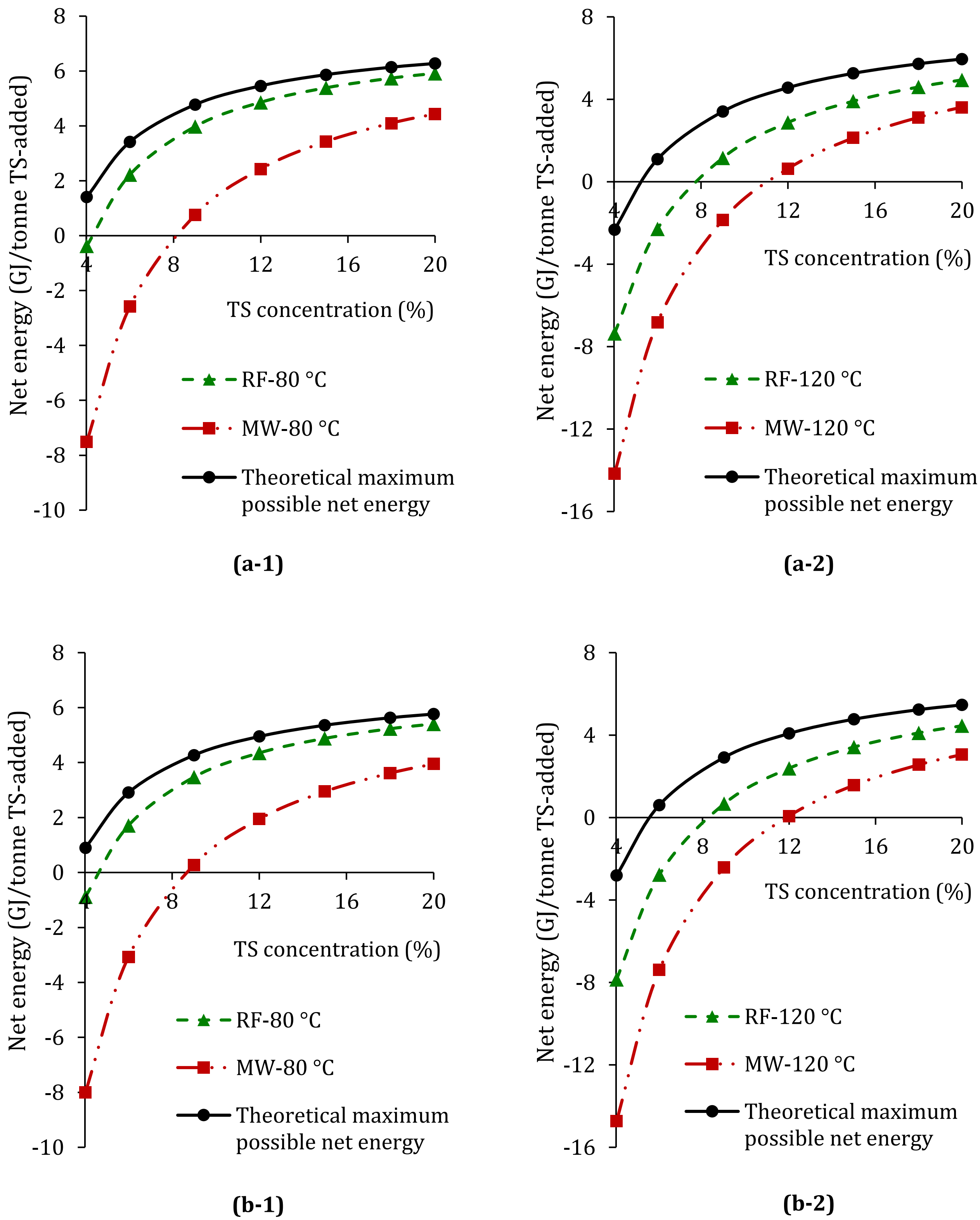


Figure 7-16. The change in the net energy production of the mesophilic pretreated digesters with TS concentration under a SRT of (a) 20 d and (b) 10 d

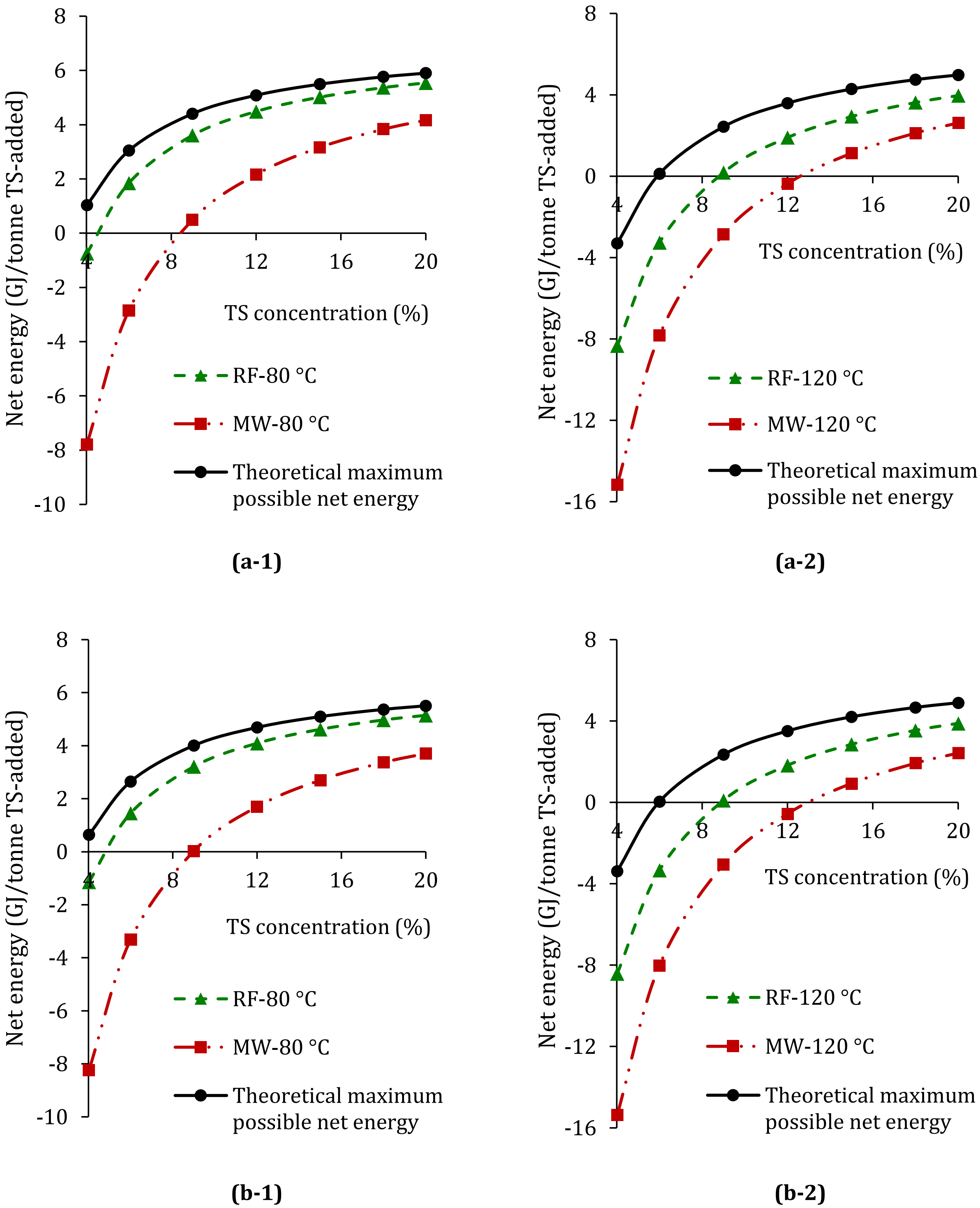
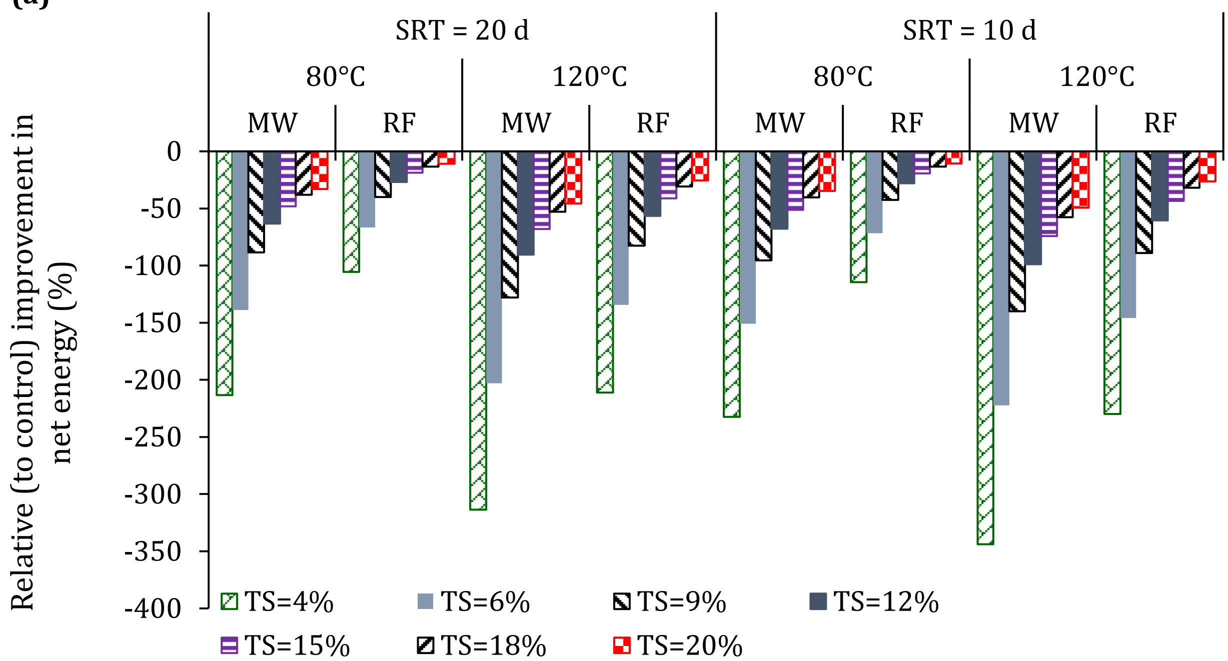


Figure 7-17. The change in the net energy production of the thermophilic pretreated digesters with TS concentration under a SRT of (a) 20 d and (b) 10 d

It was revealed by Figure 7-16 and Figure 7-17 that conducting thermal pretreatment on dewatered TWAS samples result in positive net energy production. A TS content between 12 to 20% achieves less than 8% deviation from the maximum possible net energy (E_{net}^{max}). However, taking into account the energy expenditure, the sludge pretreatment will be a feasible practice only if the improvement in the net energy (over the control digester) is greater than the energy consumption during pretreatment (input energy). In other words, it is possible to assume a situation under which the net energy production of an advanced AD system (pretreatment + AD) is positive and has small deviation from E_{net}^{max}, but still will not be practicable from the energy point of view when compared to the conventional AD scenario (control digester). The feasibility of a thermal pretreatment scenario can be assessed by comparing the net energy of the advanced AD system with that of the control (non-pretreated) AD system. In this regards, if the net energy of an advanced AD system is greater than that of the control digester, the application of thermal pretreatment prior to AD process will be beneficial from the energy standpoint.

The relative (to control) improvement in the net energy production of the mesophilic and thermophilic digesters fed with pretreated TWAS are compared in Figure 7-18 (a) and (b), respectively. Under a given pretreatment temperature, the relative (to control) improvement is higher for RF-pretreated digesters compared to that of the MW-pretreated ones (Figure 7-18). However, no matter what pretreatment scenario is used, the advanced AD system yielded lower net energy than that of the control digester even at a TS concentration of 20%.

(a)



(b)

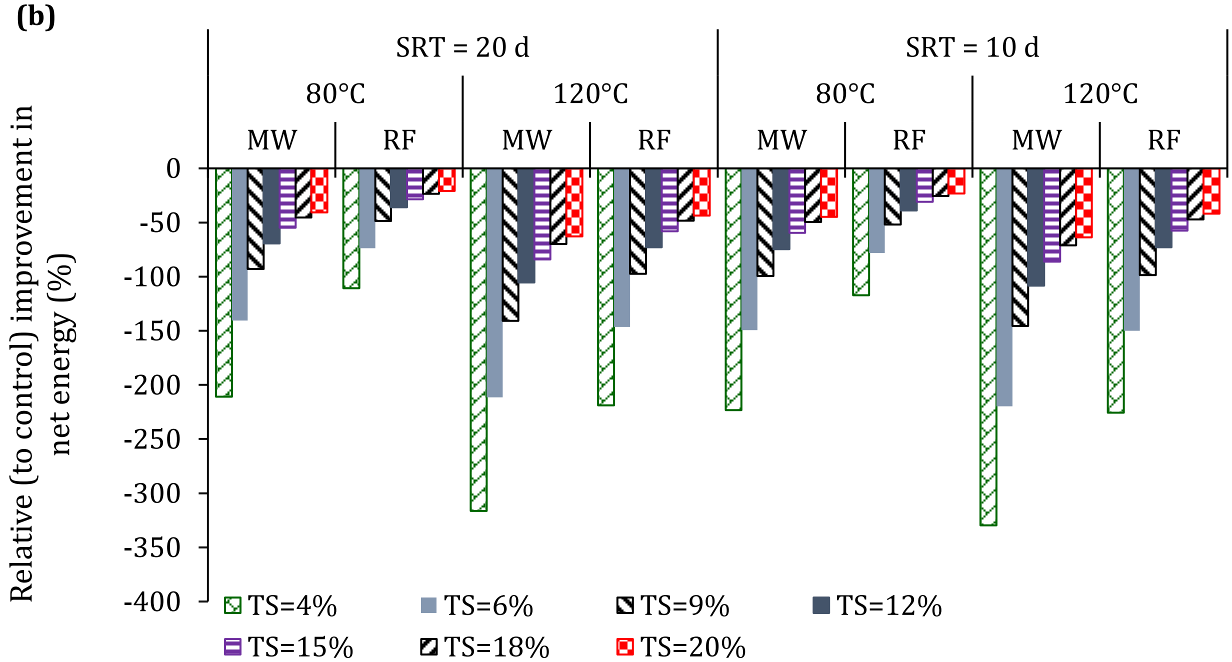
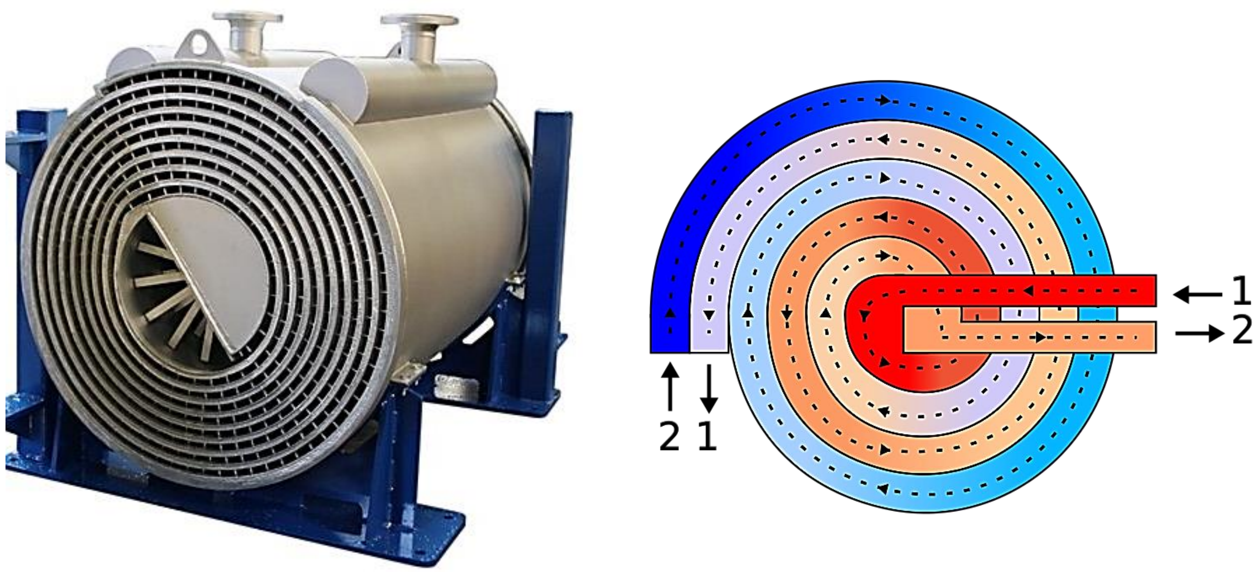


Figure 7-18. The relative (to control) improvement in the net energy production of the pretreated digesters under the (a) mesophilic and (b) thermophilic condition

The net energy of an advanced AD system calculated from Eq. (7-5) does not include the amount of the thermal energy that can be recovered from the pretreated sludge before feeding to the digester. The recovered thermal energy can be used to preheat the sludge, increase its temperature to some extent, and therefore reduce the input energy of the system. A variety of heat exchangers can be used for this purpose such as spiral, plate and tube heat exchangers. The spiral type (Figure 7-19) is however the most commonly used heat exchanger in WWTPs because of its ability to effectively handle fluids which contain solids and fibers such as municipal sludge (LINES, 2016).



(a) **(b)**

Figure 7-19. The spiral heat exchanger: (a) industrial-scale configuration and (b) schematic of heat exchange (adapted from *https://en.wikipedia.org/wiki/Heat_exchanger*)

Eq. (7-5) can be modified as follows to include the thermal energy recovery of the heat exchanger:

$$ME_{net} = E_{out} - (E_{in} - \eta E_{th}) \qquad (7\text{-}8)$$

in which ME_{net} is the modified net energy production of an advanced AD system, E_{in} is the pretreatment system input energy, E_{th} is the theoretical minimum required energy determined from Eq. (7-7), and η is the efficiency of the heat exchanger. A range of 75-90% for η is the common range suggested by the literature (Akgul et al., 2017; Gasafi et al., 2008; Lu et al., 2008; Wanner et al., 2005). In this study, $\eta = 85\%$ was considered for energy assessment. Assessing the modified net energy results revealed that under thermophilic condition, all the digesters achieved negative ME_{net} compared to the thermophilic control digester. However, under the mesophilic condition, at TS concentrations equal or greater than 15%, the RF-80°C digester showed positive improvement ratio over the mesophilic control digester (Figure 7-20). Under similar pretreatment condition, higher improvement ratios are achieved at a SRT of 10 d compared to a SRT of 20 (Figure 7-20). The overall energy assessment results suggest that for thermal pretreatment using the RF system, the TS concentration of the TWAS should be as high as 15% during pretreatment to be feasible from the energy standpoint. In agreement with these results, the TS concentration of the sludge is reached between 14.5 to 16.5% in Cambi$^{\mathrm{TM}}$ process as one of the well-established thermal hydrolysis process with more than 50 full-scale installations around the world (Mohammad & Terry, 2012). As per Figure 7-20, at a SRT of 10 d, the relative (to control) improvement in the modified net energy of 3.8%, 5.8% and 6.8% were achieved at sludge TS concentrations of 15, 18 and 20%, respectively using the RF-pretreatment. It is worth noting that the energy analysis performed in this chapter does not include the additional energy required to dewater the sludge before the pretreatment.

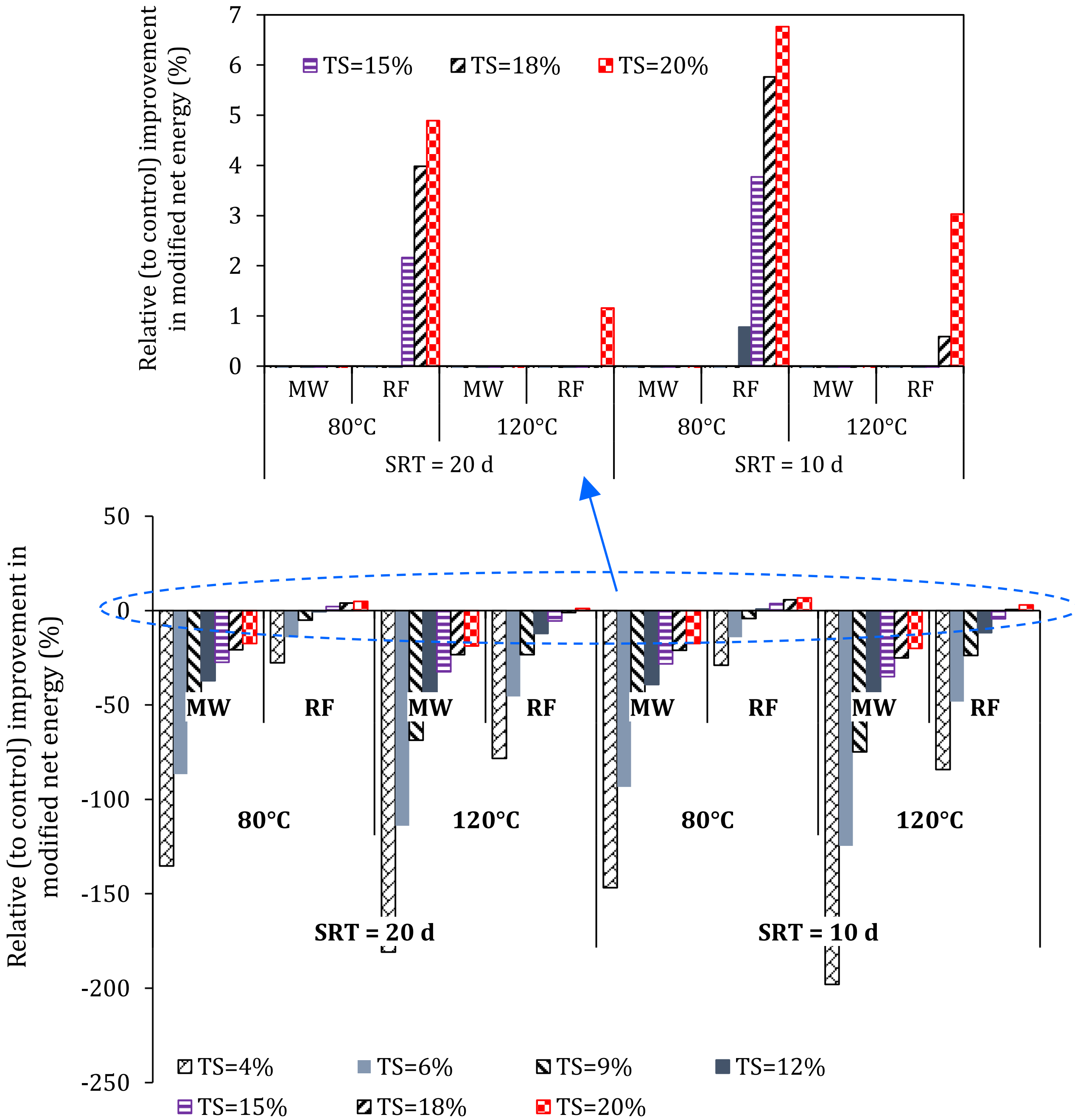


Figure 7-20. The relative (to control) improvement in the modified net energy (ME_{net}) of the mesophilic pretreated digesters

7.4. SUMMARY

The overall findings of the comparison study performed under the semi-continuous flow regime in this chapter supports the results obtained under the batch flow regime (Chapter 6). According to the results from this chapter, under both mesophilic and thermophilic conditions, the control digester demonstrated the lowest biogas yield, while the digesters fed with pretreated TWAS at a temperature of 120°C achieved the highest biogas production. The effect of the thermal pretreatment was more apparent at a SRT of 10 d compared to that of a 20 d SRT. The highest mesophilic biogas yield achieved at SRTs of 20 and 10 d was 17.3% and 13.2% higher than that of the corresponding control digester, respectively. At a SRT of 10 d, both mesophilic and thermophilic biogas production as well as the organics removal efficiency were statistically significantly enhanced with increasing the pretreatment temperature. However, at a SRT of 20 d, the effect of thermal pretreatment was statistically meaningful only under the mesophilic condition. Neither the biogas production, nor the organics removal efficiency, and the digestate characteristics were statistically significantly affected by the pretreatment methods (RF vs. MW). This proves that under identical thermal profile, the RF and MW pretreatments do not have any advantages over another in terms of improving AD performance, which validated *Hypothesis II*. As per energy assessment results, 22.26 $\frac{GJ}{tonne\ TS-added}$ energy was required to increase the TWAS temperature from room temperature (22°C) to 120°C using the MW heating system operated at a frequency of 2.45 GHz which was 44.5% higher than that of the custom-designed RF heating system at a frequency of 13.56 MHz, which validated *Hypothesis III.* It was observed that the higher the pretreatment temperature is, the more the output energy of a digester will be.

However, the digesters fed with pretreated TWAS at 120°C achieved the lowest net energy production due to the higher input energy consumption. Increasing the solids concentration of the sludge before thermal pretreatment was observed to be a significant factor increasing the net energy production of the digesters (per sludge dry tonne). Considering the energy analysis results, more than 92% of the maximum possible net energy production could be achieved by thickening the sludge to a TS content above 15%. Considering the possibility of recovering energy from the pretreated sludge, the modified net energy production of the pretreated digesters was calculated. According to the results, using the pretreatment scenario of RF-80°C and under a SRT of 10 d, the maximum improvement (to the control digester) in the modified net energy production of 3.8%, 5.8% and 6.8% could be achieved at a TS concentration of 15, 18 and 20%, respectively.

CHAPTER 8. CONCLUSIONS

8.1. SUMMARY

The research described in this dissertation fills the knowledge gap in the field of "*advanced AD of municipal sludge via thermal pretreatment*" by providing answers to the following questions; 1) Are there any advantages of choosing one of the two commonly used thermal pretreatment techniques (CH or MW at 2.45 GHz) over another for advanced AD of municipal sludge? and 2) Is there any frequency other than a frequency of 2.45 GHz (which has been exclusively used in previous studies) that can achieve more energy-efficient sludge thermal hydrolysis? Considering the experimental and analytical results discussed in the foregoing chapters, the following conclusions can be drawn from this research:

- The sludge disintegration in terms of COD, sugar, protein, and HA solubilization was increased after thermal pretreatment. Not only the pretreatment temperature, but also the heating ramp rate and holding (stationary) time had statistically significant effects on sludge solubilization ($p\text{-value}<0.05$). Increasing pretreatment temperature and holding time improved COD and biopolymer solubilization ratios, while increasing the heating ramp rate had an adverse effect on sludge solubilization.
- In agreement with the solubilization results, the pretreatment temperature, heating rate, and holding time were significant factors determining the AD performance in terms of biogas production, organics removal, and digestate characteristics. Therefore, any comparison between different thermal pretreatment methods (i.e., CH, MW, and RF)

should be conducted under identical thermal profiles, otherwise it yields non-replicable and contradictory results (as observed in the literature).

- No statistically significant effects of the thermal pretreatment methods (CH, MW at 2.45 GHz, and RF at 13.56 MHz) were observed on sludge solubilization, biogas production, organics removal, and digestate characteristics under identical thermal profiles (p-value>0.05).
- Comparison of the three thermal pretreatment methods performed in this research reject the conclusion of the previous studies in which the improved sludge solubilization or biogas production observed for the MW-pretreated samples (compared to the CH-pretreated samples) was postulated to be due to the MW non-thermal (athermal) effects. Therefore, it is inferred that any possible athermal MW effects were much smaller than the thermal effects at the applied pretreatment temperatures, so they were not discernible.
- The outcomes of this study suggest that as long as the thermal profile is identical, the thermal pretreatment method is not a significant factor determining mesophilic and thermophilic anaerobic digestibility of the municipal sludge. Therefore, selection of the most optimum pretreatment method should be based on the criteria other than the effectiveness of the pretreatment system such as the capital and operating cost of the system, the overall energy-efficiency of the system, and the uniformity of heating throughout the load during pretreatment.
- Ohmic heating as a result of the ionic conduction flow, dominates the heating mechanism in the custom-designed RF pretreatment system by contributing to more than 99% of the

total dissipated power at a frequency of 13.56 MHz. However, at a frequency of 2.45 GHz used in the commercial MW ovens, the dielectric relaxation of bulk water is the primary mechanism of heating in sludge pretreatment.

- A frequency of 13.56 MHz lies very close the band (100 KHz to 10 MHz) known for β dispersion that caused by the repeated charging and discharging of the cell membranes. However, no increased biogas production was observed as a result of further cell lysis that possibly could occur using the RF heating system (at 13.56 MHz) compared to that of MW oven (at 2.45 GHz).
- The temperature measurement results proved almost uniform heating throughout the load using the two-parallel plate structure proposed in the design of the RF sludge pretreatment system in this research. The uniform heating capability of the RF pretreatment technique will be more appreciable considering the fact that due to the thermal gradient occurred through the CH of the sludge, as well as short penetration depth of the EM waves at a frequency of 2.45 GHz, achieving temperature uniformity using either the CH or MW pretreatment is very challenging.
- The results of the semi-continuously fed digesters demonstrated that the relative (to control) improvement in biogas production as well as the TS, VS, and TCOD removal efficiency were increased as the SRT was reduced from 20 to 10 d. This observation suggests that the thermal pretreatment will be more advantageous at shorter SRTs since the control digesters are facing more challenges under higher OLRs. The effects of thermal pretreatment were more apparent under mesophilic condition since the thermophilic

digesters benefitted from an enhanced hydrolysis stage compared to the mesophilic ones at a higher operating temperature (55°C).

- As per impedance measurement results, the power transfer efficiency of the RF heating system gradually increased from 88% at 20°C to the maximum value of 99.9% at around 70°C and then reduces to around 94% a temperature of 140°C. The energy assessment results revealed that the MW heating system operated at a frequency of 2.45 GHz consumed 44.5% more energy than the custom-designed RF heating system at a frequency of 13.56 MHz to increase the TWAS temperature from 22°C (room temperature) to 120°C. The overall energy-efficiency of the RF heating system (67.3 to 95.5%) was almost two times more than that of the MW pretreatment oven (37-43%).
- Applying thermal pretreatment on 4% TS sludge samples resulted in negative net energy production under all the pretreatment scenarios at both the mesophilic and thermophilic conditions. However, the net energy production per sludge dry weight can be increased by thickening the sludge before the pretreatment. More than 92% of the maximum possible net energy production could be achieved by thickening the sludge to a TS content above 15%.
- Among all the pretreatment scenarios considered in this research, the application of RF thermal pretreatment to a temperature of 80°C is suggested as the most optimum pretreatment option from the energy point of view. As per analytical results, this pretreatment scenario results in 3.8%, 5.8% and 6.8% higher overall net energy production than that of the control mesophilic digester at a TS concentration of 15, 18, and

20%, respectively. However considering the additional complexity of applying pretreatments at the full-scale (centrifugation of the TWAS to dewater after polymer addition, potential operational issues etc.), ~7% improvement may not be a high enough incentive for the WWTP utilities to implement the RF system unless there are significant benefits in terms of other parameters (outside the scope of this research), such as dewaterability, fecal coliform and micropollutants (hormones, pharmaceuticals, and pesticides) content of the digested sludge (biosolids).

8.2. LIMITATIONS AND RECOMMENDATIONS FOR FUTURE WORK

The following studies are recommended for future work which were not pursued in the current research due to limitations in time and resources.

- Reducing the heat loss during the RF sludge pretreatment by adding an insulation layer to the air cavity in the coaxial enclosure of the system.
- Design and built a version of RF heating system that is used for thermal hydrolysis of dewatered sludge cake with a TS concentration above 12%.
- Modify the current version of the RF heating system which is operated under batch modes to a continuously operating heating system.
- Scale up the custom-designed RF heating system, so it can be used for pilot testing.
- Microscopic investigation of the thermal pretreatment effect on sludge floc structure and comparison of the results using CH, MW (at 2.45 GHz), and RF (at 13.56 MHz) heating systems.

- Explore the athermal effect of RF heating at a frequency of 13.56 MHz under high intensity pulsed electric fields.
- Comparison of the thermal pretreatment effect on pathogens and micropollutants removal, along with digestate dewaterability using the RF heating system and comparison of the results with those of MW and CH.
- Investigate the cause of the decreased biogas production by increasing the holding time under the pretreatment temperature of 60°C (observed in Chapter 6).
- Conducting a pilot-scale study utilizing a continuously-fed RF pretreatment system followed by an automated continuously-fed anaerobic digester. The results of the pilot-scale investigation can be used to perform a more realistic decision analysis to select the most optimum pretreatment method used for the full-scale application.
- Performing a comprehensive multicriteria decision making (MCDM) analysis that in addition to the energy-efficiency criterion, it includes other decision criteria such as the cost of the pretreatment system, the cost of the sludge thickening before thermal pretreatment, the reduction of the digester cost and foot print as a result of its volume reduction, the reduction of the digested sludge handling and disposal cost.

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APPENDICES

APPENDIX A – REPRESENTATIVE CALIBRATION CURVES

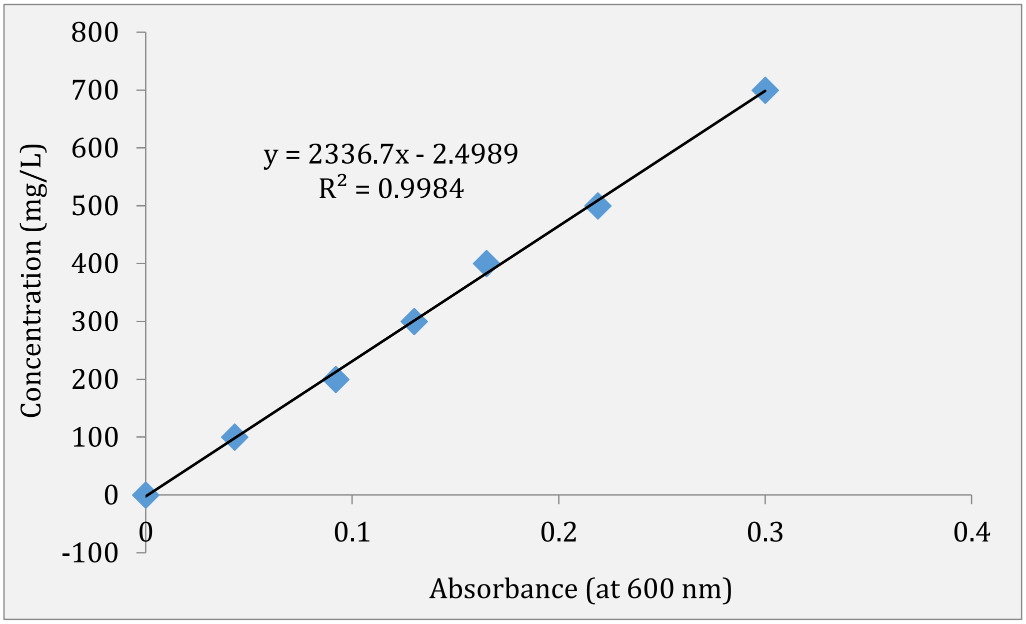

Figure A- 1. The representative calibration curve for COD determination

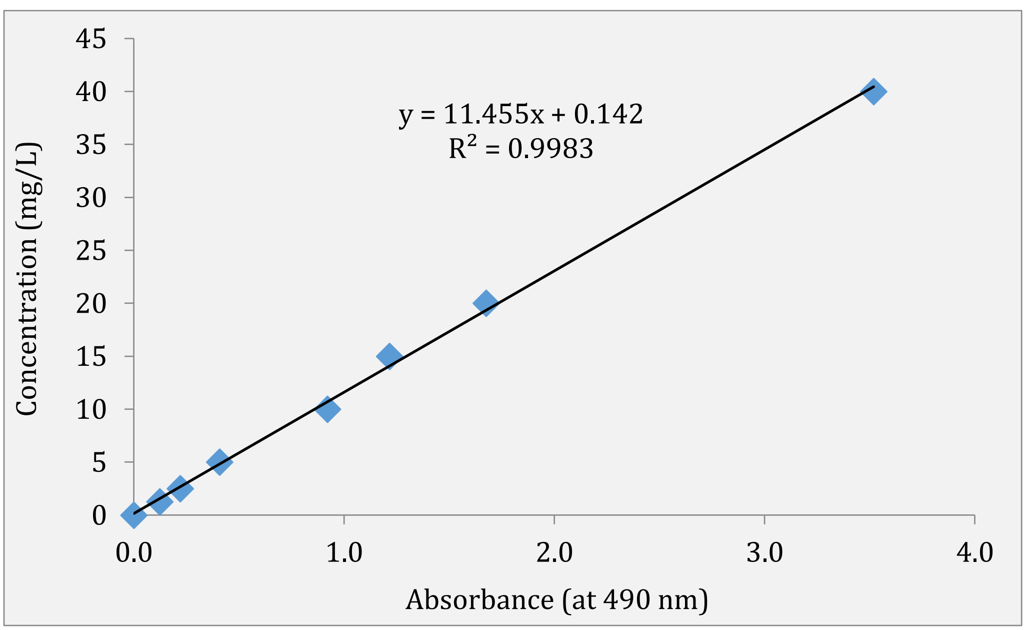

Figure A- 2. The representative calibration curve for sugar determination

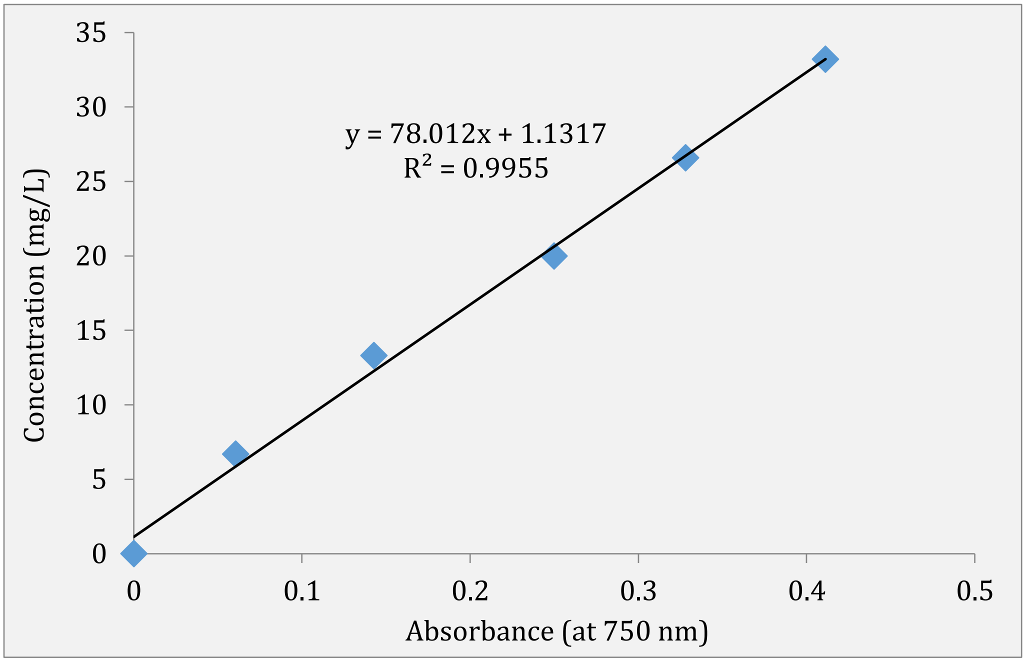

Figure A- 3. The representative calibration curve for protein determination

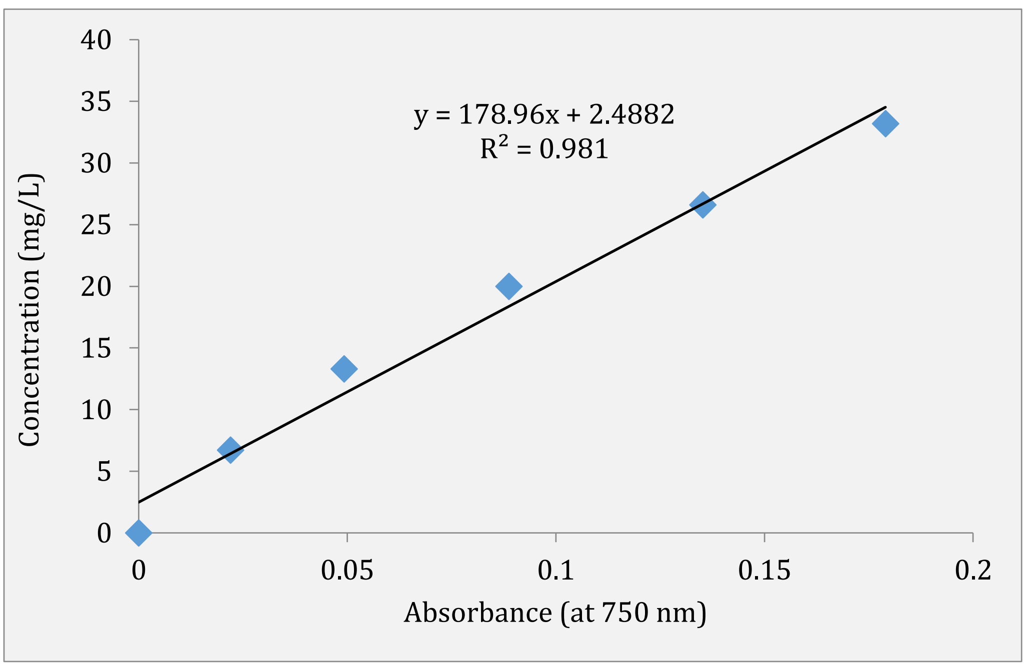

Figure A- 4. The representative calibration curve for humic acid (HA) determination

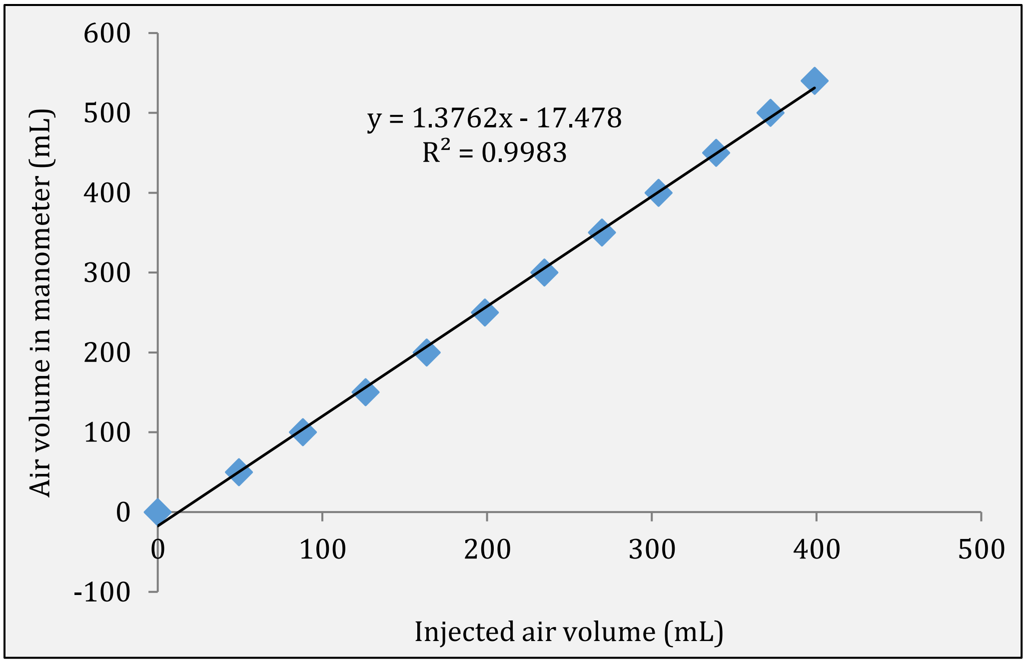

Figure A- 5. The representative calibration curve for biogas volume measurement

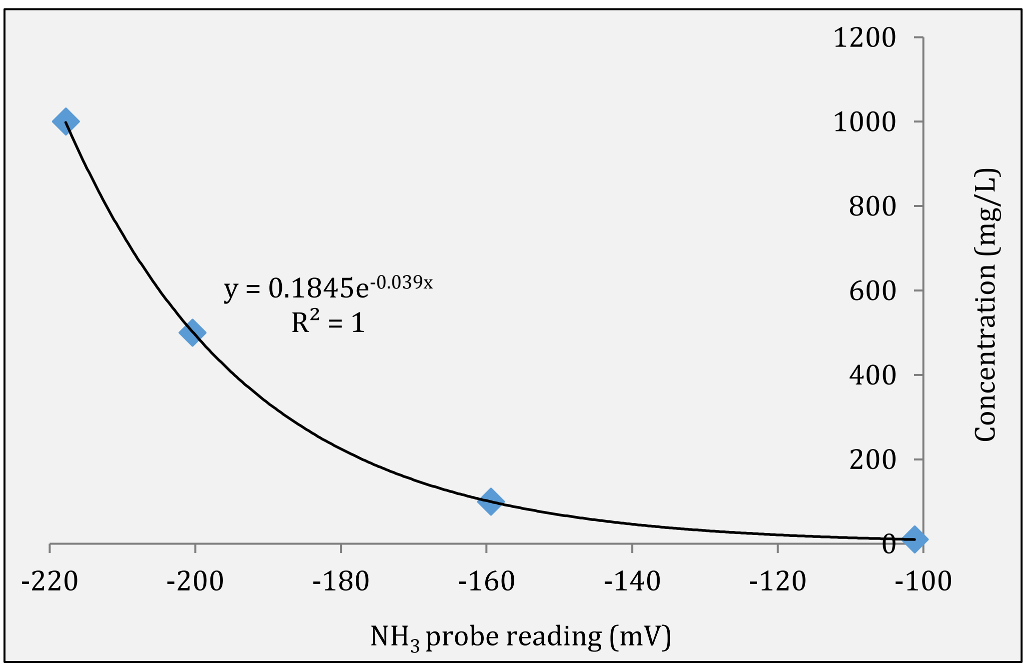

Figure A- 6. The representative calibration curve for the ammonia ($NH_3 - N$) determination

APPENDIX B – REPRESENTATIVE ANDERSON-DARLING TEST NORMALITY PLOTS

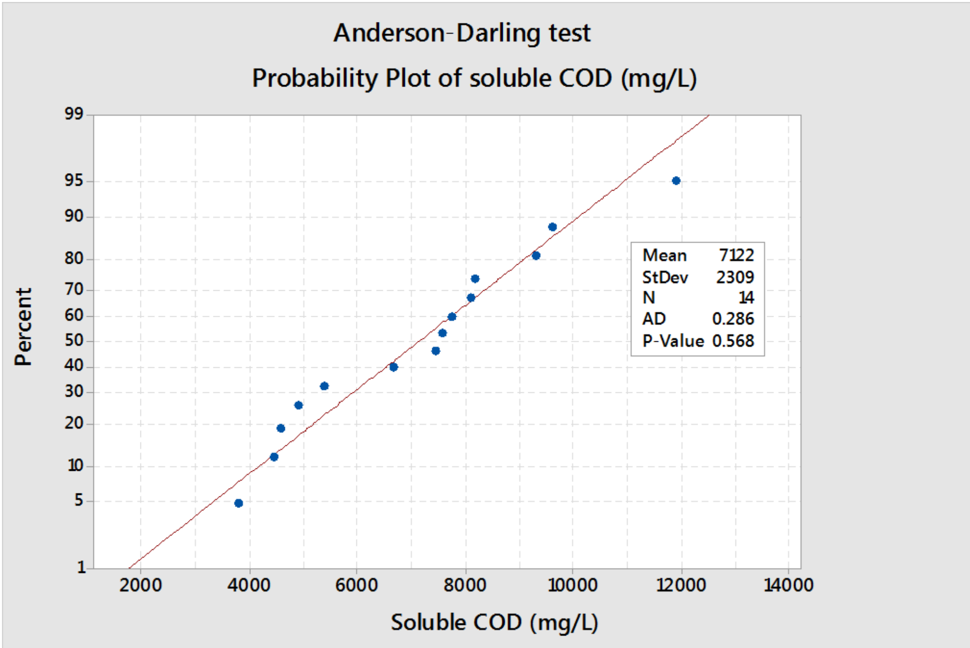

Figure B- 1. The representative Anderson-Darling test normality plot for COD solubilization data (Chapter 3)

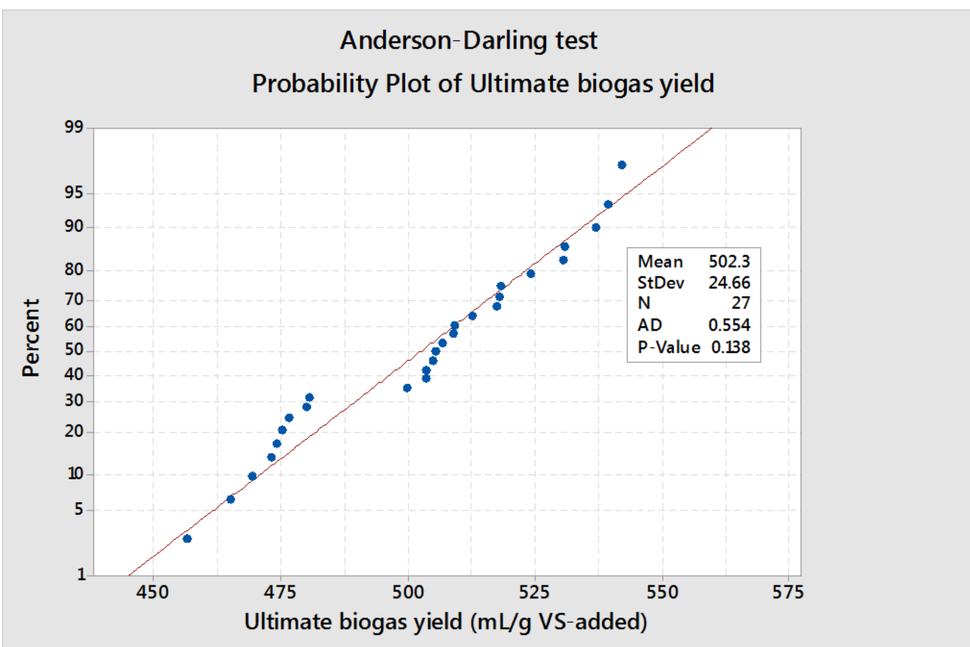

Figure B- 2. The representative Anderson-Darling test normality plot for mesophilic ultimate biogas yield data (Chapter 4)

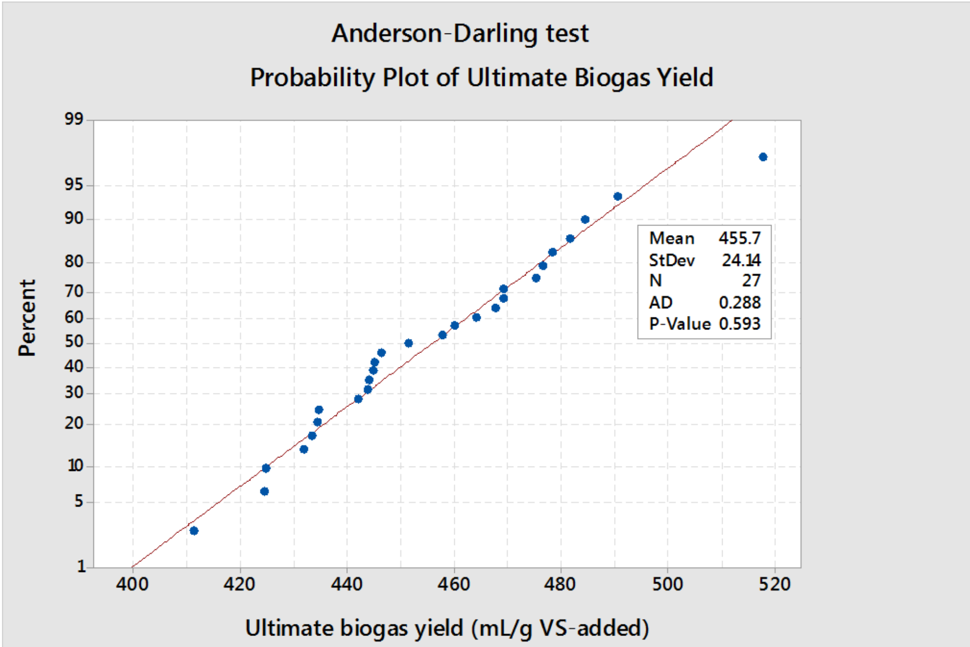

Figure B- 3. The representative Anderson-Darling test normality plot for thermophilic ultimate biogas yield data (Chapter 4)

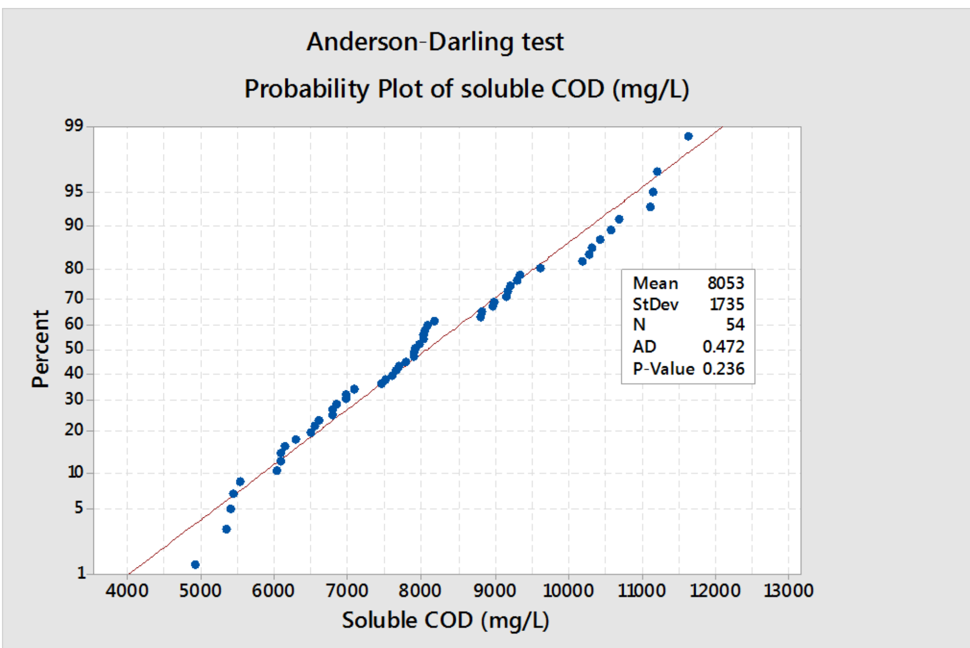

Figure B- 4. The representative Anderson-Darling test normality plot for COD solubilization data (Chapter 6)

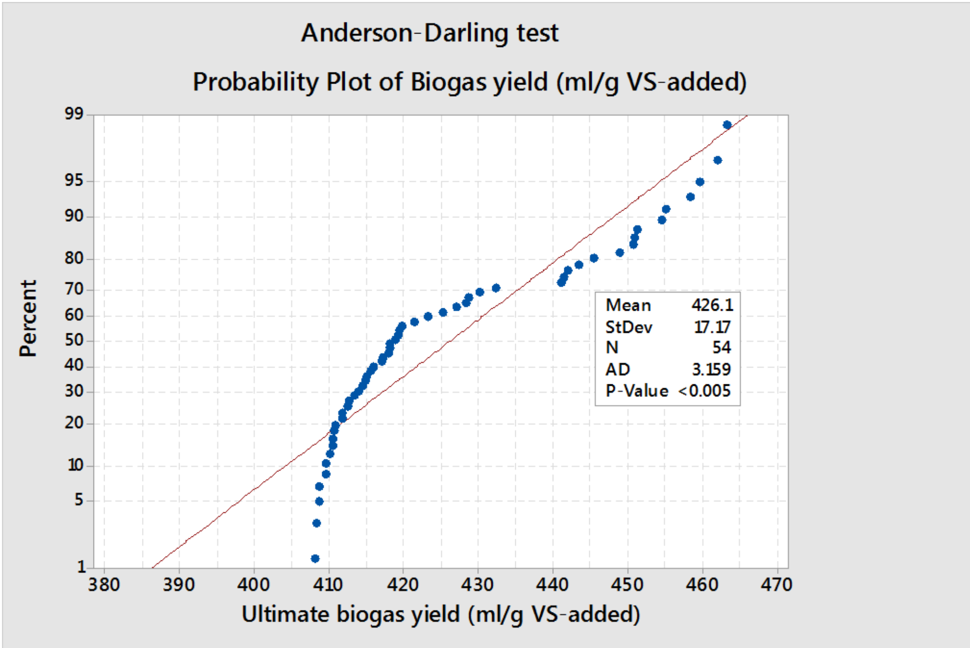

Figure B- 5. The representative Anderson-Darling test normality plot for ultimate biogas yield data (Chapter 6)

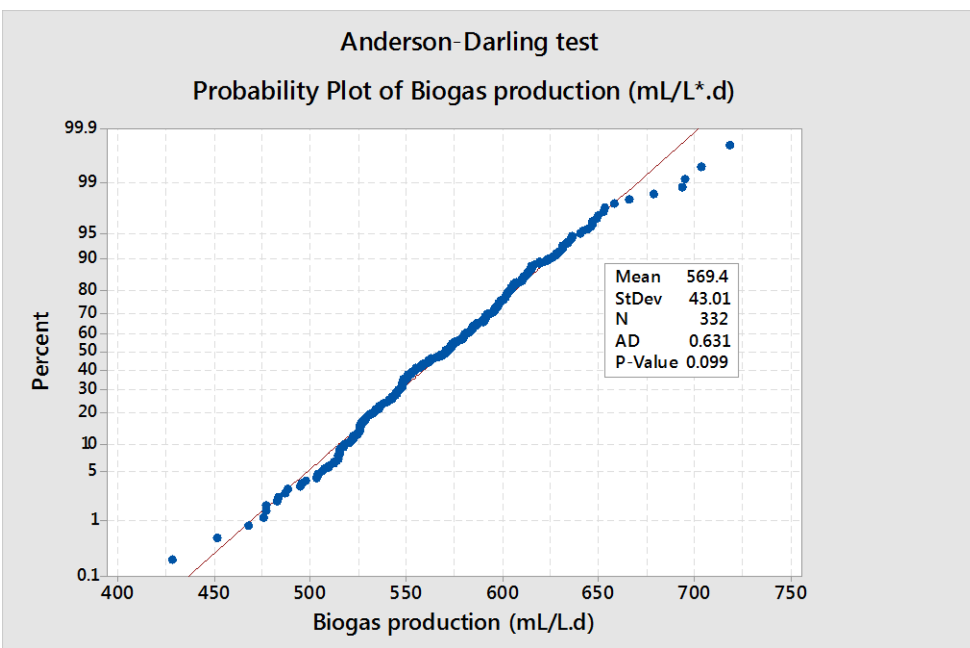

Figure B- 6. The representative Anderson-Darling test normality plot for mesophilic biogas production data (Chapter 7)

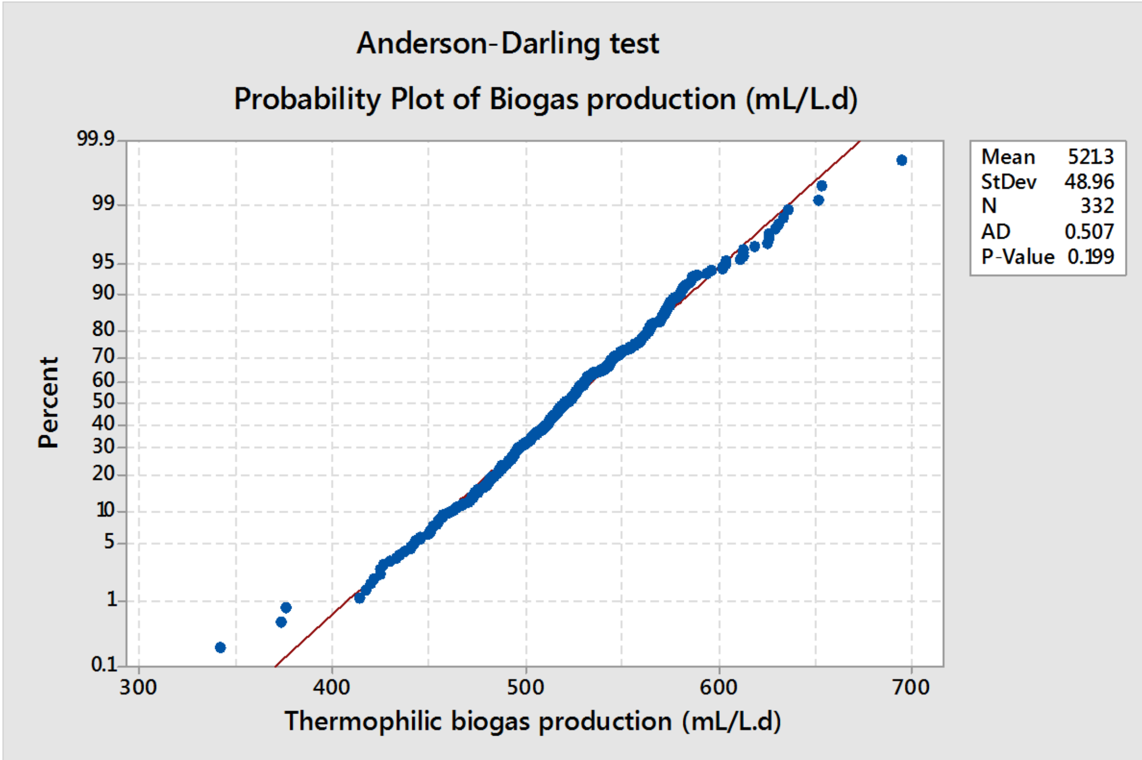

Figure B- 7. The representative Anderson-Darling test normality plot for thermophilic biogas production data (Chapter 7)

APPENDIX C – REPRESENTATIVE CONSTANT VARIANCE TEST RESULTS

```
Null hypothesis          All variances are equal
Alternative hypothesis   At least one variance is different
Significance level       α = 0.05


95% Bonferroni Confidence Intervals for Standard Deviations

           Ramp   Temp
Method  (°C/min)  (°C)  N    StDev              CI
    CH         3   120  4  125.959  ( 6.3140, 9.25284E+03)
    CH         3   160  4  436.053  (11.5567, 6.05852E+04)
    CH         6    80  3  220.158  ( 0.0000, 1.46871E+22)
    CH         6   120  2  147.581  (      *,           *)
    CH         6   160  4  112.003  ( 3.5474, 1.30216E+04)
    CH        11    80  4   90.913  ( 2.5779, 1.18062E+04)
    CH        11   120  4  246.916  ( 9.1101, 2.46432E+04)
    MW         3    80  4  100.266  ( 3.3178, 1.11579E+04)
    MW         3   120  4  244.314  ( 7.5485, 2.91174E+04)
    MW         6    80  4  152.912  ( 3.9650, 2.17150E+04)
    MW         6   120  4  186.987  ( 4.7886, 2.68865E+04)
    MW         6   160  4  105.355  ( 4.6971, 8.70165E+03)
    MW        11   120  4  447.867  (10.1275, 7.29319E+04)
    MW        11   160  4   97.326  ( 4.3779, 7.96739E+03)

Individual confidence level = 99.6429%


Method                  Statistic  P-Value
Multiple comparisons            —    0.664
Levene                       0.61    0.833
```

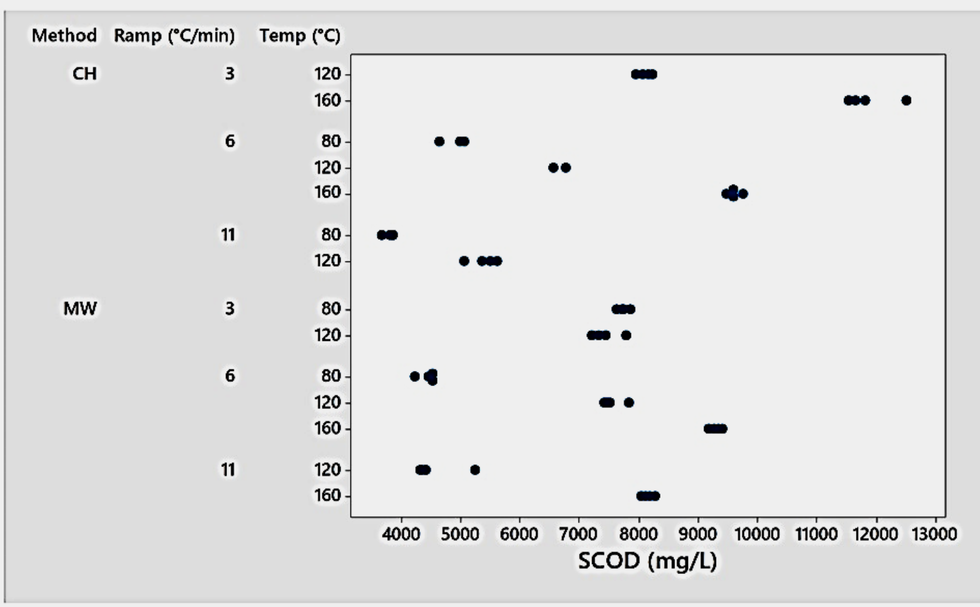

Figure C- 1. The representative constant variance test results for COD solubilization data (Chapter 3)

```
Null hypothesis          All variances are equal
Alternative hypothesis   At least one variance is different
Significance level       α = 0.05
```

95% Bonferroni Confidence Intervals for Standard Deviations

Pretreatment method	Heating ramp rate (°C/min)	Temperature (°C)	N	StDev	CI
CH	3	160	3	4.7293	(0.0000022, 133183023)
CH	6	120	3	4.9331	(0.0000023, 138920098)
CH	11	80	3	10.6135	(0.0000050, 298888590)
CH	11	120	3	6.2862	(0.0000029, 177027382)
MW	3	80	3	9.3239	(0.0000044, 262572266)
MW	3	120	3	2.9178	(0.0000014, 82167359)
MW	6	80	3	7.5870	(0.0000036, 213658522)
MW	6	160	3	8.7942	(0.0000041, 247654291)
MW	11	160	3	1.8198	(0.0000009, 51247439)

Individual confidence level = 99.4444%

Method	Statistic	P-Value
Multiple comparisons	–	0.419
Levene	0.46	0.869

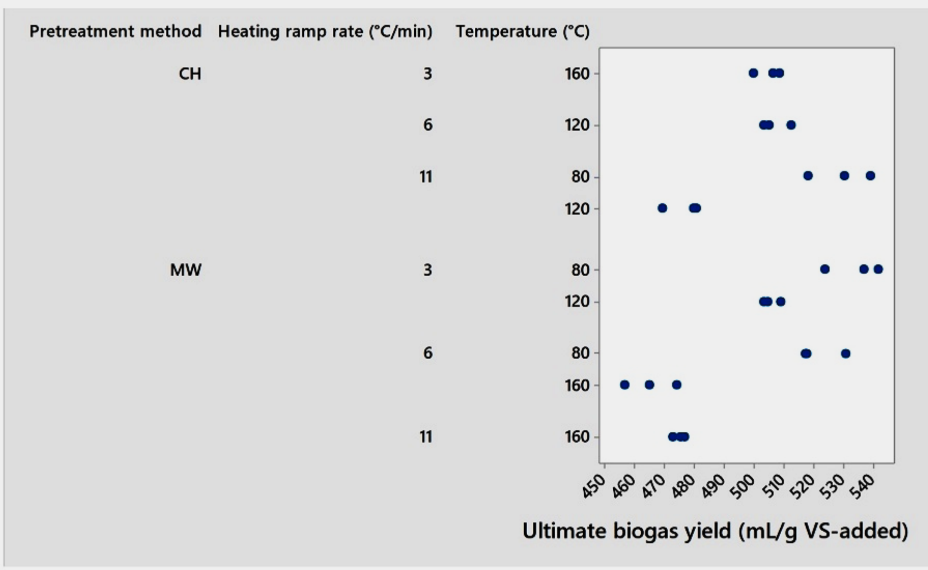

Figure C- 2. The representative constant variance test results for mesophilic ultimate biogas yield data (Chapter 4)

Null hypothesis	All variances are equal
Alternative hypothesis	At least one variance is different
Significance level	α = 0.05

95% Bonferroni Confidence Intervals for Standard Deviations

Pretreatment Method	Ramp Rate	Final Temperature	N	StDev	CI
CH	3	120	3	16.5914	(0.0000078, 467232748)
CH	3	160	3	11.4204	(0.0000054, 321612171)
CH	6	80	3	9.2018	(0.0000043, 259132762)
CH	6	160	3	5.5781	(0.0000026, 157084417)
CH	11	80	3	4.6193	(0.0000022, 130085734)
MW	3	80	3	17.7383	(0.0000083, 499529226)
MW	6	120	3	4.4323	(0.0000021, 124818757)
MW	11	120	3	7.2191	(0.0000034, 203297911)
MW	11	160	3	11.7712	(0.0000055, 331489517)

Individual confidence level = 99.4444%

Method	Statistic	P-Value
Multiple comparisons	—	0.661
Levene	0.43	0.886

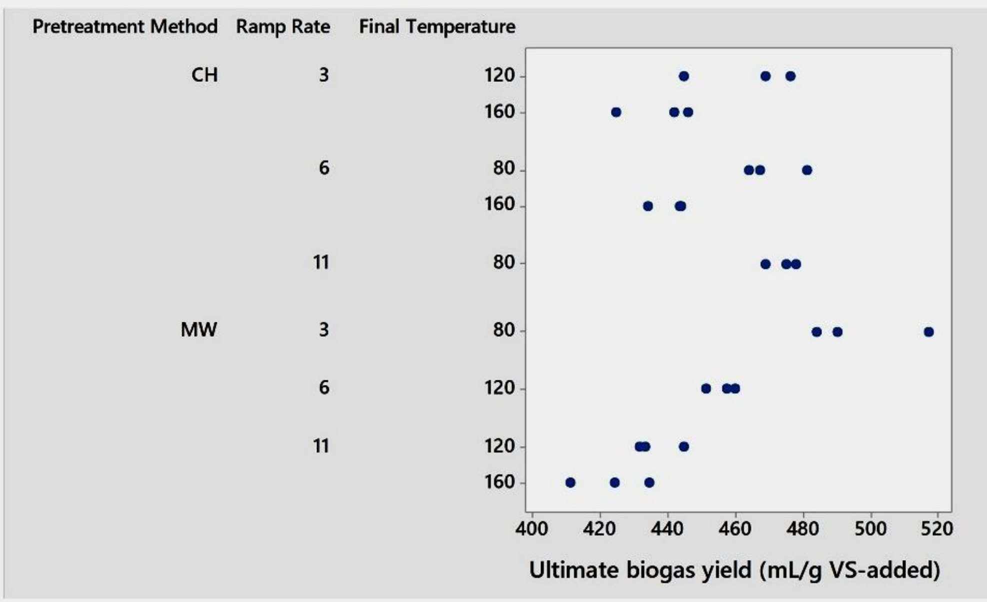

Figure C- 3. The representative constant variance test results for thermophilic ultimate biogas yield data (Chapter 4)

Null hypothesis	All variances are equal
Alternative hypothesis	At least one variance is different
Significance level	α = 0.05

95% Bonferroni Confidence Intervals for Standard Deviations

Method	Temperature (°C)	Stationary Time (h)	N	StDev	CI
MW	60	0	3	71.087	(0.0000000, 2.86766E+197)
MW	60	1	3	27.408	(0.0000000, 1.10566E+197)
MW	60	2	3	26.795	(0.0000000, 1.08092E+197)
MW	90	0	3	545.667	(0.0000000, 2.20124E+198)
MW	90	1	3	644.969	(0.0000000, 2.60183E+198)
MW	90	2	3	710.258	(0.0000000, 2.86521E+198)
MW	120	0	3	530.248	(0.0000000, 2.13904E+198)
MW	120	1	3	72.029	(0.0000000, 2.90567E+197)
MW	120	2	3	47.057	(0.0000000, 1.89831E+197)
RF	60	0	3	619.760	(0.0000000, 2.50013E+198)
RF	60	1	3	683.772	(0.0000000, 2.75836E+198)
RF	60	2	3	598.079	(0.0000000, 2.41267E+198)
RF	90	0	3	214.798	(0.0000000, 8.66501E+197)
RF	90	1	3	81.818	(0.0000000, 3.30055E+197)
RF	90	2	3	796.902	(0.0000000, 3.21473E+198)
RF	120	0	3	49.811	(0.0000000, 2.00938E+197)
RF	120	1	3	788.387	(0.0000000, 3.18038E+198)
RF	120	2	3	743.340	(0.0000000, 2.99866E+198)

Individual confidence level = 99.7222%

Method	Statistic	P-Value
Multiple comparisons	—	0.002
Levene	0.72	0.762

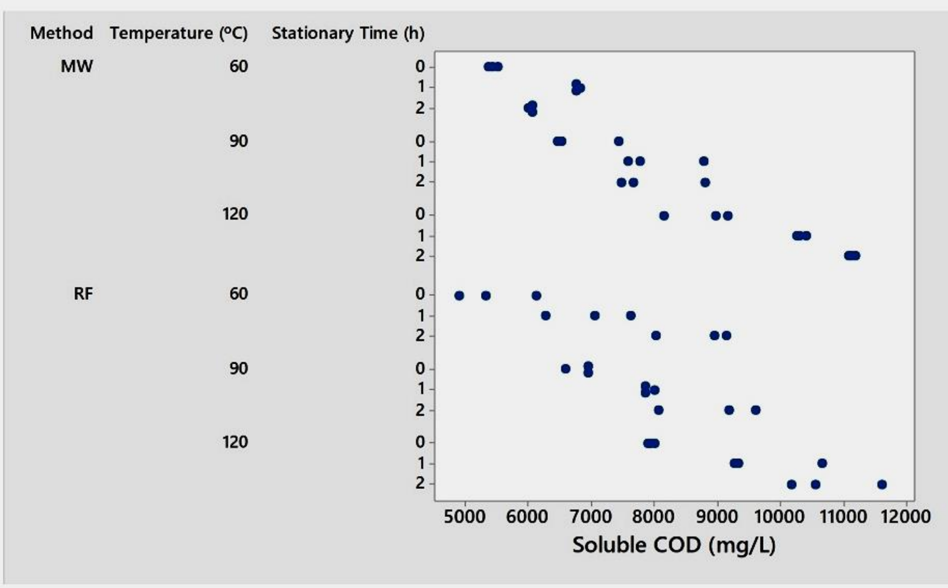

Figure C- 4. The representative constant variance test results for COD solubilization data (Chapter 6)

Null hypothesis All variances are equal
Alternative hypothesis At least one variance is different
Significance level α = 0.05

95% Bonferroni Confidence Intervals for Standard Deviations

Method	Temperature (ºC)	Stationary Time (h)	N	StDev	CI
MW	60	0	3	3.02862	(0.0000000, 1.22176E+196)
MW	60	1	3	1.53691	(0.0000000, 6.19995E+195)
MW	60	2	3	0.86654	(0.0000000, 3.49565E+195)
MW	90	0	3	0.74857	(0.0000000, 3.01976E+195)
MW	90	1	3	3.59750	(0.0000000, 1.45124E+196)
MW	90	2	3	8.10329	(0.0000000, 3.26889E+196)
MW	120	0	3	3.61461	(0.0000000, 1.45815E+196)
MW	120	1	3	4.59356	(0.0000000, 1.85306E+196)
MW	120	2	3	4.97414	(0.0000000, 2.00658E+196)
RF	60	0	3	0.86997	(0.0000000, 3.50948E+195)
RF	60	1	3	1.35887	(0.0000000, 5.48175E+195)
RF	60	2	3	0.71888	(0.0000000, 2.90000E+195)
RF	90	0	3	0.25739	(0.0000000, 1.03833E+195)
RF	90	1	3	1.61709	(0.0000000, 6.52339E+195)
RF	90	2	3	2.50751	(0.0000000, 1.01154E+196)
RF	120	0	3	0.46349	(0.0000000, 1.86975E+195)
RF	120	1	3	5.80307	(0.0000000, 2.34098E+196)
RF	120	2	3	2.60504	(0.0000000, 1.05088E+196)

Individual confidence level = 99.7222%

Method	Statistic	P-Value
Multiple comparisons	—	0.011
Levene	0.92	0.554

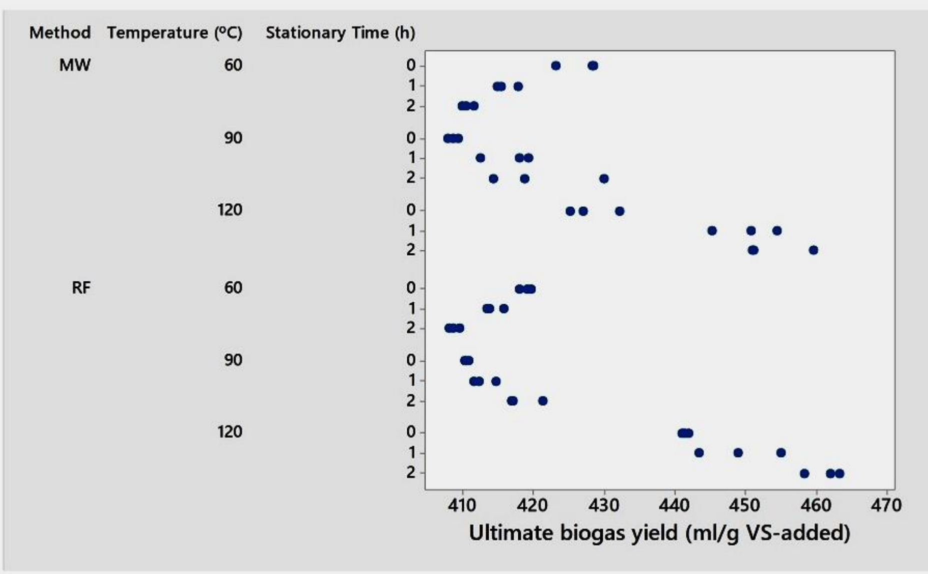

Figure C- 5. The representative constant variance test results for ultimate biogas yield data (Chapter 6)

Null hypothesis	All variances are equal
Alternative hypothesis	At least one variance is different
Significance level	$\alpha = 0.05$

95% Bonferroni Confidence Intervals for Standard Deviations

Pretreatment Method	Temperature (°C)	N	StDev	CI
MW	80	83	38.7019	(31.0324, 49.7643)
MW	120	83	39.2608	(30.6517, 51.8482)
RF	80	83	34.4923	(27.7350, 44.2268)
RF	120	83	36.2036	(28.4516, 47.4971)

Individual confidence level = 98.75%

Method	Statistic	P-Value
Multiple comparisons	–	0.789
Levene	0.42	0.739

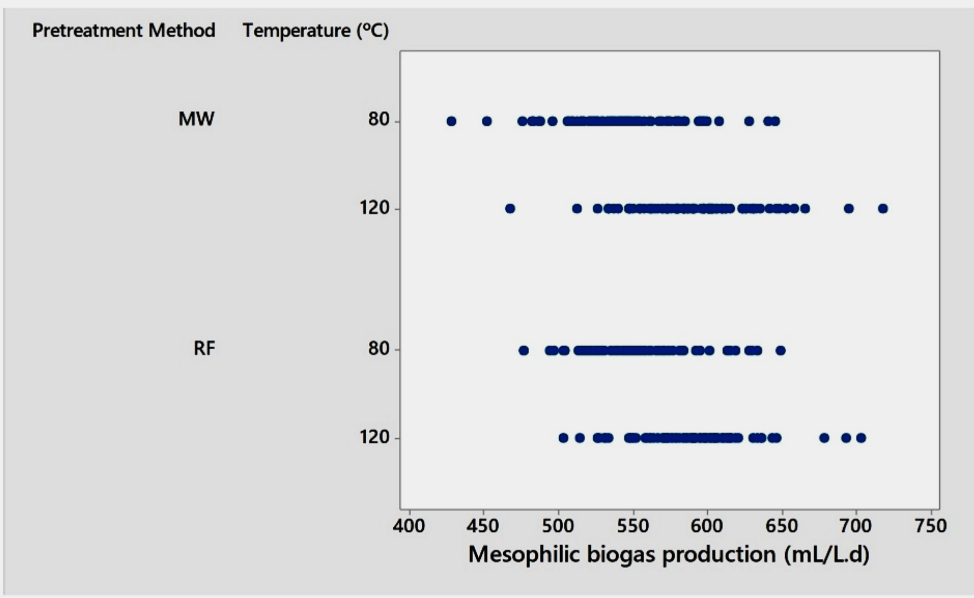

Figure C- 6. The representative constant variance test results for mesophilic biogas production data (Chapter 7)

Null hypothesis	All variances are equal
Alternative hypothesis	At least one variance is different
Significance level	α = 0.05

95% Bonferroni Confidence Intervals for Standard Deviations

Pretreatment method	Temperature (°C)	N	StDev	CI
MW	80	83	52.7898	(42.1597, 68.1510)
MW	120	83	53.9240	(42.5501, 70.4584)
RF	80	83	44.3298	(35.3971, 57.2393)
RF	120	83	44.4740	(37.2608, 54.7305)

Individual confidence level = 98.75%

Method	Statistic	P-Value
Multiple comparisons	–	0.449
Levene	1.07	0.362

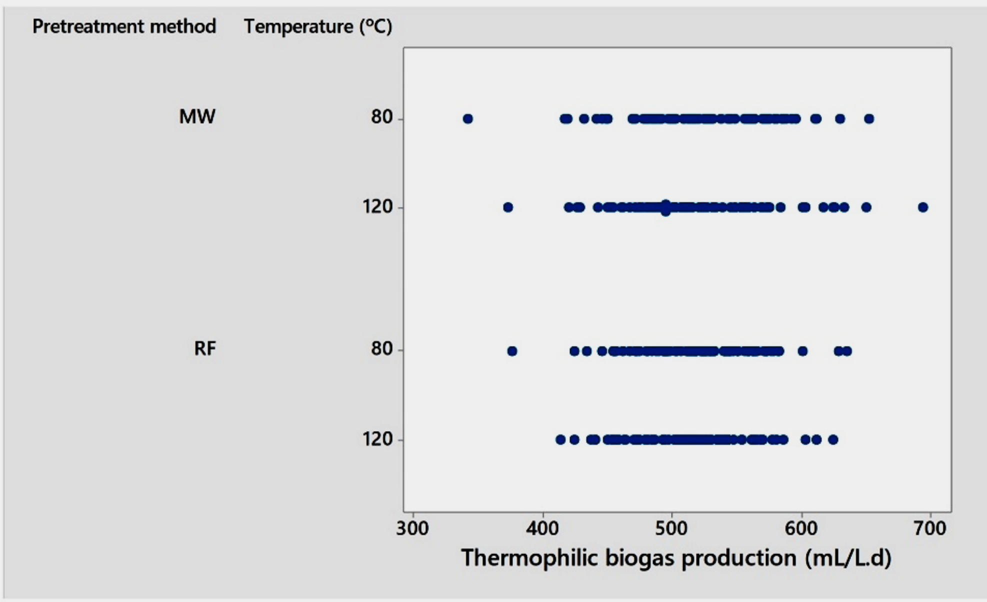

Figure C- 7. The representative constant variance test results for thermophilic biogas production data (Chapter 7)

Appendix D – Cross calibration of the MW and RF systems thermocouple

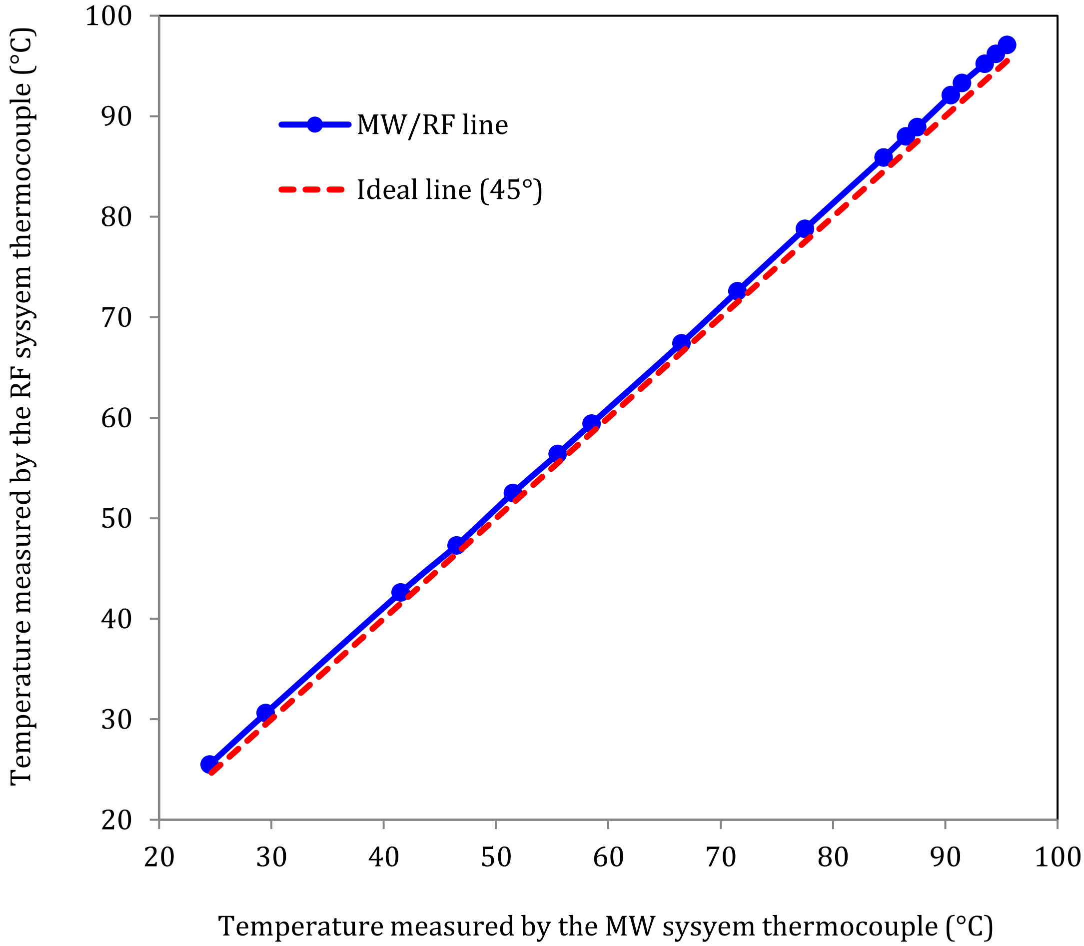

Figure D- 1. Comparison of the cross calibration line of the MW and RF systems thermocouple with the ideal line having an angle of 45°