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Study on Performance Enhancement of Anaerobic Digestion of Municipal Sewage Sludge

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DECLARATION

To the best of my knowledge and belief this thesis contains no material previously published by any other person except where due acknowledgement has been made. This thesis contains no material which has been accepted for the award of any other degree of diploma in any university.

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Conferences

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ABSTRACT

Anaerobic digestion is an energy-efficient and environmentally beneficial technology used for methane production and organic removal. One of the drawbacks of anaerobic digestion technique is the slow rate-limiting hydrolysis of organics which is the primary degradation step in the anaerobic digestion process. Different pretreatment technologies were used to enhance sludge hydrolysis and anaerobic digestion performance. Pretreatment of sludge through ultrasonic, mechanical, chemical or thermal techniques result in bacterial cell wall disruption and release of enzymes which enhance the rate of hydrolysis and biodegradation. All sludge reduction technologies are working with the principle of disintegration of cell walls, and large organic molecules. There are numerous studies on the benefits of different pretreatment techniques including ultrasonic and microwave pretreatment when the methods are applied independently and in combination with other pretreatment techniques. This research focuses on investigating effects of ultrasonic, microwave and combined microwave-ultrasonic pretreatment of municipal sewage sludge on anaerobic digester performance. The impacts were investigated in terms of biogas production, solid removal, degree of disintegration and sludge dewaterability. Combined microwave-ultrasonic pretreatment resulted in increased methane production, better COD and solid removal and improved dewaterability more than individual microwave or ultrasonic pretreatment options. Most of the work in this research is dedicated towards investigating the effects of combined microwave-ultrasonic pretreatment on the anaerobic digestibility of primary, excess activated, thickened excess activated and mixed sludge systems. Experimental setup was designed for batch and semicontinuous tests for the study of effects of microwave, ultrasonic and combined pretreatment techniques and digester operational parameters. Simultaneously operating jacketed continuously stirred digesters were fed with pretreated and untreated sludge and the digesters were continuously monitored and operation continued until steady state is achieved. Samples were collected on regular basis for analysis of total solid, volatile solids, total and soluble COD, microbial content, pH, dewaterability, ammonia, protein content, particle size and rheology. Experiments were conducted on synthetic sludge before the tests on municipal sewage sludge to understand the effects of each pretreatment technique. Samples were characterized and pretreated ultrasonically or subjected to microwave

irradiation and a combination of these techniques. The optimum pretreatment conditions were determined based on the impact of the pretreatment on sludge solubilisation, biogas production and characteristics of the digested sludge produced. The experimental results from the study on synthetic sludge showed that combined microwave-ultrasonic treatment resulted in better digester performance than ultrasonic or microwave pretreatment techniques. Mesophilic digestion of combined microwave-ultrasonic pretreated sludge produced significantly higher amount of methane after a sludge retention time (SRT) of 17 days. The combined microwave-ultrasonic pretreatment resulted in total solids reduction of 56.8% and volatile solid removal of 66.8%. The dewaterability was also improved significantly.

The experimental work on municipal sewage sludge throughout the research period was based on samples collected from Beenyup Wastewater treatment Plant. Raw primary sludge (PS), excess activated sludge (EAS), thickened excess activated sludge (TEAS), mixed sludge (MS) and digested sludge (DS) samples were characterized and subjected to different microwave and ultrasonic pretreatment conditions. Optimum pretreatment conditions for all sludge types were determined from sludge solubilisation and anaerobic digestion tests.

The effects of microwave and ultrasonic pretreatment conditions like pretreatment power, intensity, time, density and specific energy on mixed sewage sludge (MS) and thickened excess activated sludge (TEAS) characteristics and anaerobic digester performance were also investigated. The biogas production volume and kinetics, dewaterability of digested sludge, COD reduction and other sludge properties were optimized for the aforementioned ultrasonication and microwave pretreatment conditions for MS and TEAS.

The effect of Microwave pretreatment (M) was compared to Combined Microwave-Ultrasonic (CMU) pretreatment on how the two techniques enhance anaerobic biodegradability of mixed sludge. The removal of TS was 37.7 % for M pretreated sludge whereas the TS reduction for CMU pretreated sludge was 69.1%. The removal of volatile solids for CMU pretreated sludge was 21% higher than the M pretreated sample.

The effect of mixing ratio of primary sludge to excess activated sludge was also studied. Cumulative methane production of pretreated Excess Activated Sludge

(EAS) was higher (66.5 ml/g TCOD) than the methane yield from pretreated mixed sludge (44.1 ml/g TCOD). Furthermore, digested EAS showed significantly higher dewaterability. The removal of VS was improved by 50% due to the pretreatment and the release of organics and their disintegration increased the SCOD/TCOD ratio to 66% and the reduction in SCOD/TCOD ratio was 12 % higher for pre-treated TEAS resulting in increased average daily methane production rate of 782 ml/day. The average daily methane production was 592 ml/day for the untreated TEAS. Maximum percentage of methane produced was 69-71 % for pre-treated TEAS while it was 56 % for untreated TEAS. Methane: carbon dioxide ratio for pretreated TEAS was 2.51 while it was 1.93 for the untreated TEAS. Thickened excess activated sludge with greater solid concentration has resulted in a better digester performance after pretreatment.

Effects of organic loading rate and hydraulic retention time were also investigated. Combined microwave-ultrasonic pretreated sludge provided higher methane yield, volatile solid and COD removal at shorter HRT (5 days) than untreated or microwave or ultrasonic pretreated sludge.

The best digester performance was achieved for anaerobic biodegradability of Combined Microwave-Ultrasonic pretreated thickened excess activated sludge (PTEAS) mixed with untreated primary sludge (PS). The anaerobic digestion was conducted in the two continuously stirred batch anaerobic digesters for a sludge retention time of 32 days. The specific methane yield was 122 ml CH_4/g TCOD for digester 1 and 101 ml CH_4/ g TCOD for digester 2 after sludge retention time of 20 days. The amount further increased to 187 ml CH4/g TCOD for digester 1 and 116 ml CH_4/g TCOD for digester 2 after SRT of 27 days. The CH_4/CO_2 ratio reached 2.2:1 and 1.1:1 after SRT of 20days for digester 1 and digester 2 respectively.

Furthermore, Adaptive neuro-fuzzy inference application in MATLAB was used for model based optimization and prediction of digester operational parameters. Plant data collected from Beenyup wastewater treatment plant was utilized for training and validation purposes. Predictions were made on methane potential, sludge feed flow rate (organic loading rate), pH and alkalinity and the parameters that affect digester performance most were selected and optimized, the surface responses for the correlation between input and output variables were also developed. ANFIS was found to be an important tool for efficient control and optimization of operational parameters to maximize digester performance.

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ANFIS: adaptive neuro-fuzzy inference system

BWWTP: Beenyup Waste water Treatment Plant

CMU: combined microwave-ultrasonic pretreatment

CST: capillary suction time

DAFT: dissolved air floatation unit

DS: digested sludge

EAS: excess activated sludge

HRT: hydraulic retention time

MAPE: mean absolute percentage error

MS: mixed sludge

MSE: mean square error

OLR: organic loading rate

RMSE: root mean square error

RPS: raw Primary Sludge

SCOD: soluble chemical oxygen demand

SRT: sludge retention time

TCOD: total chemical oxygen demand

TDS: total dissolved solid

TEAS: thickened excess activated sludge

TS: total solids

VS: volatile solids

VSS: volatile suspended solid

WAS: waste activated sludge

CHAPTER 1

INTRODUCTION

1.1 Background

Sewage sludge handling and processing for ultimate disposal is one of the major challenges in the operation of municipal wastewater treatment plants. The production of waste activated sludge has significantly increased, as a result of increase in the number and capacity of wastewater treatment plants over time. Disposal routes are subjected to more stringent environmental policies and regulations and social constraints. Sludge treatment technologies like incineration are also quite expensive (Navaratnam, 2007)..

Biogas production through anaerobic digestion has recently captured global attention because of its substantial benefits including eco-friendly energy generation, greenhouse gases emission reduction, high organic removal, high capacity to stabilize large volume of dilute organic slurry at low cost, low biomass production, high decay rate of pathogenic microorganisms, and the capacity of producing solid residue suitable for use as soil conditioner. Anaerobic digestion reduces up to 80% of the odors in the feedstock (Ghosh et al., 1975) . It is rated as one of the most energy-efficient and environmentally beneficial technologies for bioenergy production (Weiland, 2010, Chong et al., 2012b, Müller, 2001). Generally, anaerobic digestion is a favoured stabilisation method compared to aerobic digestion, due to its lower cost, lower energy footprint, and moderate performance, especially for stabilisation (Appels et al., 2008a)

The process involves four major microbiological degradation steps comprising hydrolysis, acidogenesis, acetogenesis and methanogenesis. The hydrolysis step is a slow rate determining part of the process that lowers the gas yield and retards the kinetics. The slow degradation or hydrolysis of microorganisms also accounts for 70% of excess sludge which is the primary degradation step in the anaerobic digestion process. The microorganisms in the excess sludge contain extracellular polymeric substances (EPS) that are resistant to biodegradation which in turn limits the rate of the whole anaerobic digestion process (Tiehm, Nickel et al. 1997, Chong, Sen et al. 2012). Therefore, minimization of the amount of sludge produced coupled with the increased generation of value-added, renewable sources of energy like methane of higher quality is the best strategy for sustainable sludge management.

In an effort to improve sludge hydrolysis, biodegradability and, dewaterability, many experimental studies have been documented on pretreatment of sludge. Such technologies include ultrasonic treatment (Tiehm et al., 2001, Farooq et al., 2009, Saifuddin and Fazlili, 2009a, Apul and Sanin, 2010a), chemical treatment (Stuckey and McCarty 1978 , (Haug et al., 1978), (Penaud et al., 2000); (Tanaka and Kamiyama, 2002), microwave treatment (Eskicioglu et al., 2007c), (Park, 2011), ozone oxidation (Yeom et al., 2002), (Lin and Lee, 2002), mechanical disintegration, supercritical and subcritical water oxidation and high temperature hydrolysis (Carrère et al., 2010b). All sludge reduction technologies are working with the principle of disintegration of cell walls, and large organic molecules.

It was reported that macromolecules with a molar mass of above 40,000 are disrupted by the hydro-mechanical shear forces produced by ultrasonic cavitation (Tiehm et al., 2001). The mechanisms of ultrasonic treatment are influenced by the energy supplied, ultrasonic frequency and the nature of the sludge. Cell disintegration is proportional to energy supplied (Bougrier et al., 2005b). High frequencies promote oxidation by radicals, whereas low frequencies promote mechanical and physical phenomena like pressure waves (Portenlanger, 1999). Only Ultrasonic pretreatment method was applied on large scale Wastewater Treatment Plants (WWTP) compared to other pretreatment methods (Carrère et al., 2010b).

Microwave (MW) irradiation is another efficient sludge pre-treatment technology that enhances biodegradability, methane production and digester performance (Park 2011). It is a novel pre-treatment method for stabilization of waste activated sludge (WAS). Microwave pre-treatment of sludge increases biogas production, reduce sludge viscosity, improve dewaterability and improve pathogen decay as compared to digestion of sludge pre-treated through conventional heating and untreated sludge (Eskicioglu et al., 2007b). Microwave treatment was more cost effective as compared to conventional thermal treatment (Park, 2011). MW treatment resulted in pathogen destruction as well as thermal versus non-thermal effects (Eskicioglu et al., 2007c). MW treatment was applied to achieve higher WAS floc and cell destruction and release of extracellular polymeric substances and intracellular materials into the soluble phase compared to conventional heating, which in effect increased soluble CODs and biogas production (Saha et al., 2011a). Microwave pretreatment increased SCOD up to 4 fold, soluble protein concentration up to 1.8 fold and soluble carbohydrate concentration up to 14 fold (Zhou et al., 2010). The use of MWs in the digestion of sludge was found to increase the ratio of soluble COD to total COD (SCOD/TCOD) from 2 to 22% (Toreci et al., 2010).

There are numerous studies on the benefits of different pretreatment techniques, including ultrasonic and microwave pretreatment when the methods are applied independently and in combination with other pretreatment options (chemical and thermal pretreatment). The application of more than one treatment resulted in improved sludge biodegradation, floc destruction, cell wall disruption and release of organics due to the complementary synergy between the treatment techniques that are combined (Saifuddin and Fazlili, 2009a, Xu et al., 2010c, Saha et al., 2011a).

Microwave enhanced-oxidative pretreatment with H_2O_2 resulted in 11-34% TS, TCOD reduction and total biopolymer solubilisation (Eskicioglu et al., 2008b). Combined ultrasonic-alkali pretreatment of waste activated sludge resulted in 60% VS solubilisation. The use of NaOH weakens the cells walls increasing the disintegration effect of ultrasonication or other lysis techniques (Tyagi and Lo, 2011).

Very few researchers reported that the microwave combined with ultrasonic would be a rapid and economical method of sludge pre-treatment for enhanced biogas production. Combined microwave-ultrasonic pretreatment resulted in significant improvement in gas production, solid removal and dewaterability of municipal sludge compared to the individual ultrasonic or microwave pretreatment approaches (Saifuddin and Fazlili, 2009a, Yeneneh et al., 2013a). There is a complementary synergy between the two treatment techniques causing improved sludge disintegration, floc distruction, thermal and athermal cell wall disruption and release of organics.

Therefore, the objective of this research is to investigate the effect of ultrasonic, microwave and combined microwave-ultrasonic pretreatment when the methods are applied separately and in combination on synthetic and municipal sewage sludge.

The impacts in terms of biogas production, solid removal, COD reduction and sludge dewaterability were studied. Combined microwave-ultrasonic treatment resulted in increased methane production, better COD removal and improved dewaterability than individual microwave or ultrasonic pretreatment options. Much of the work in this research is dedicated towards investigating the effect of combined microwave-ultrasonic pretreatment on the anaerobic digestibility of primary, excess activated, thickened excess activated and mixed sludge systems. This work also aims at optimizing combined microwave-ultrasonic pretreatment conditions for enhanced digester performance and determination of optimum digester operational conditions and calculating the kinetic parameters. The last part of the research focuses on prediction of optimum operational conditions and ranges for understanding the relationship between various inputs and outputs based on historical data from Beenyup Wastewater Treatment Plant (BWWTP).

1.2 Problem statement

Most municipal mesophilic and thermophilic anaerobic digesters suffer from several limitations including low extent of solid destruction, limited gas production, process variability, process imbalance and odor problem. High energy costs associated to sludge handling and treatment are still challenges of wastewater treatment plants and the research in the area. Several technologies have been proposed as remedies to alleviate these deficiencies but still a lot of work remains undone. Hence, enhancement of anaerobic digester performance is a key point of concern in terms of making the technology more efficient and economical. This specific research focuses on searching for appropriate combined-pretreatment technology for performance enhancement of a municipal sludge anaerobic digester and process optimization for sTableand efficient operation.

1.3 Research objectives

The research has a general objective of enhancing gas generation capacity and reduction of solids and organics from the anaerobic digestion of municipal sludge, through experimental investigation of combined-pre-treatment technology and optimization of the operational parameters. The specific objectives include:

- Characterization of primary, excess activated, mixed and digested sludge from BWWTP

- Critical analysis of impact of combined pre-treatment on gas generation, solid reduction and waste stabilization dewaterability and selection of the best pre-treatment technique from tests on synthetic and actual sludge from BWWTP.
- Experimental optimization of pretreatment conditions and operational process parameters
- Model based analysis of the kinetics and operational parameters and validation with the experimental data. Predictive modelling based on adaptive neuro fuzzy logic inference system (ANFIS) application for large scale operational data.
- Comparison of experimental findings with model based prediction and historical data from BWWTP.

1.4 Scope and limitation

This research encompasses investigation of ultrasonic, microwave and combined microwave-ultrasonic pretreatment technologies to enhance gas production, ensure better solid reduction and increase kinetics of the process based on experimental anaerobic digestion research both on synthetic and real municipal sewage sludge systems. Historical data from BWWTP was also used for ANFIS based model predictions for better control and optimization of the operational parameters.

1.5 Significance of the research

This research has the following major significances

- Increased production of high quality biogas, better solid reduction and dewaterability of sludge by subjecting the feed sewage sludge to pretreatment process.
- Reduced sludge retention time and better anaerobic digestion kinetics.
- Improved dewaterability and flow characteristics of sludge
- Optimization of the operational parameters from BWWTP for better control and operation of the anaerobic digesters.
- Reduction in the operational cost of the anaerobic digestion and dewatering process.

1.6 Thesis organization

There are a total of 11 chapters in this research. The chapters are organized as follows.

Chapter one

This section of the thesis provides a general overview of the background and motivation of the research. The objectives and milestones of the research are stated, the scope and delimitations of the research are described. The organization and content of the whole research work is also presented in this part.

Chapter two

A detailed review of most published literature in the major focus areas of the research is presented in this chapter. Sludge pretreatment technologies particularly ultrasonication and microwave pretreatment technology are discussed in depth. Anaerobic digestion performance enhancement techniques such as, effect of pretreatment, optimization of operational parameters and other improvement techniques and factors affecting the performance of the anaerobic digestion process is discussed in this chapter.

Chapter three

The methodology of the experimental research is discussed in this section. The experimental work on synthetic and real municipal sewage sludge from Beenyup Wastewater Treatment Plant (BWWTP) is presented. Methods for sludge characterization and measurement of all operational parameters for the anaerobic digester are shown in this part. All analytical and instrumental techniques are discussed. A brief introduction on the modelling techniques used in the study is provided at the end of this part.

Chapter four

This chapter provides a detailed discussion on the effects of ultrasonic, microwave and combined microwave-ultrasonic pretreatment on biochemical methane potential, COD removal, solid reduction and dewaterability of synthetic sludge inoculated by real digested sludge. The three pretreatment technologies are compared and optimum pretreatment conditions for the selected technology were identified.

Chapter five

In this section, the effects of pretreatment power, time, density and intensity of ultrasonication and microwave pretreatment processes were studied. The effect of such pretreatment factors on biogas production, sludge solubilisation, dewaterability and other characteristics of the sludge is thoroughly discussed. The optimum pretreatment conditions for further research were determined.

Chapter six

The effect of microwave pretreatment was compared to that of combined microwave-ultrasonic pretreatment as an extension to the findings in chapter 5. The impact of the two pretreatment options on biogas production, sludge biodegradability, dewaterability and overall sludge characteristics was studied. Thus, the optimum pretreatment technology and conditions were selected.

Chapter seven

Impact of the selected optimum pretreatment technology on the digestibility of various sludge types is studied in this chapter. Effects of combined microwave-ultrasonic pretreatment of primary, excess activated, thickened excess activated and mixed sludge was investigated. The biogas production, solids and COD removal, dewaterability and anaerobic digester performance is discussed for each of the sludge types.

Chapter eight

The impact of pretreatment was investigated in the previous chapters for mixed sludge system by subjecting the mixed sludge to the pretreatment process after the mixing. In this chapter, the impact of pretreatment of thickened excess activated sludge (TEAS) and subsequent mixing with untreated raw primary sludge (RPS) before anaerobic digestion is presented. As the impact of pretreatment on activated sludge is more than the effect on mixed or primary sludge and because of the economic advantages associated to pretreatment of only thickened excess activated sludge portion instead of pretreating mixed sludge, different mixing ratios of pretreated TEAS to untreated RPS were compared.

Chapter nine

The effect of mixing ratio between primary and excess activated sludge, effect of organic loading rate, sludge retention time and hydraulic loading rate were studied. Anaerobic digester performance was analysed as a function of these factors. Model equations were used in this section to determine the kinetics of anaerobic digestion of the different mixing ratios. The hydrolysis rate constant, the daily methane production rate, the lag time, the cumulative methane production was predicted. Effect of organic loading rate and hydraulic retention time for pretreated and untreated thickened excess activated sludge was discussed in this section. The rheological behaviour of pretreated and untreated sludge samples is also presented in this section.

Chapter ten

Adaptive Neuro Fuzzy Logic Inference System (ANFIS) tool from MATLAB was used to model BWWTP historical data to find optimum operational conditions and predict relationship between important input and output parameters. The critical operational ranges and optimum values were also predicted using this model.

Chapter eleven

A generalized conclusion on the findings of each chapter is provided at the end. Important recommendations for future research and further investigations are presented in this part. The schematic representation on Figure 1.1 provides the overall structure of the research work and the activities undertaken at each stage to meet the stated objectives.

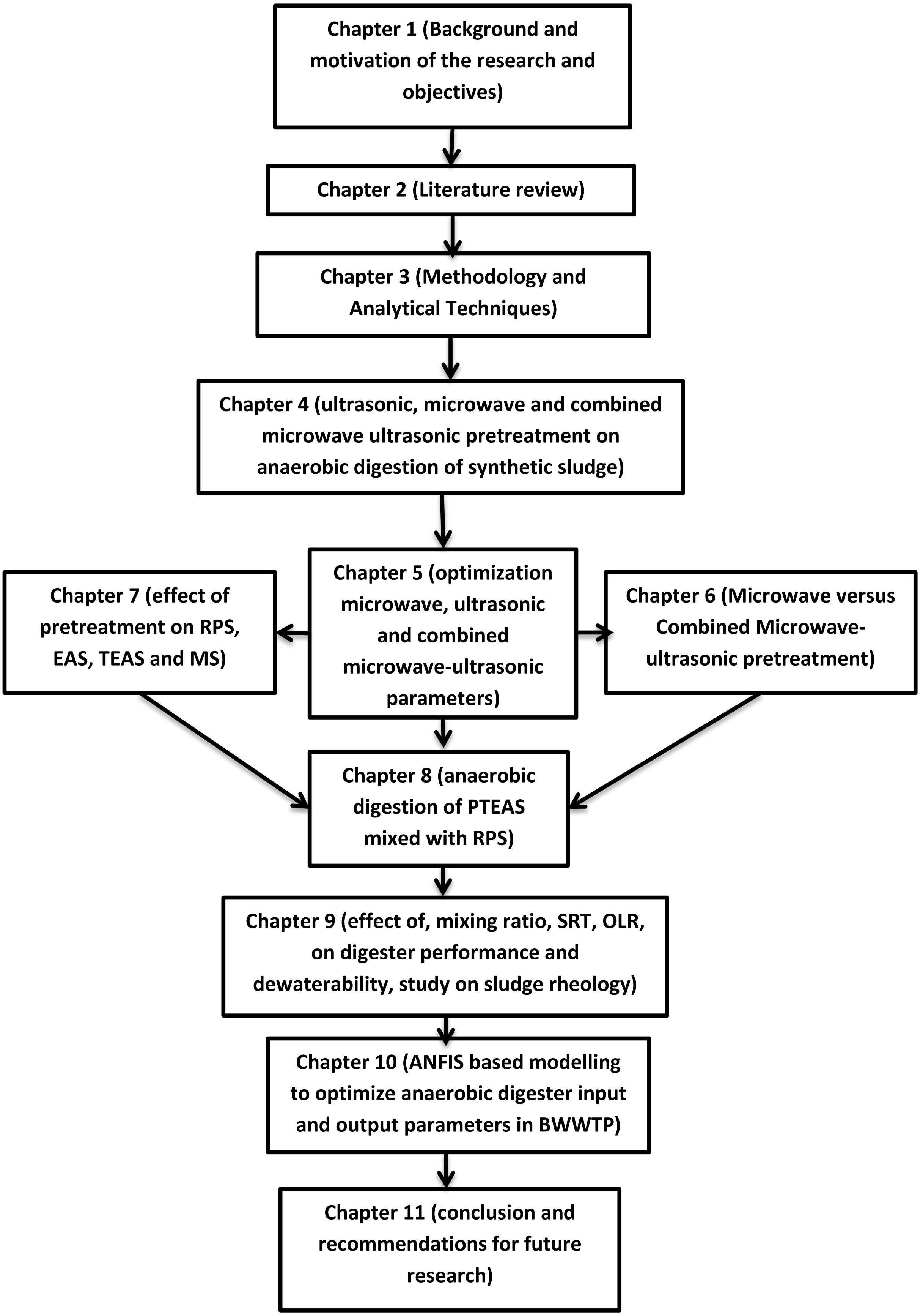


Figure 1.1 Schematic representation of the whole research work.

CHAPTER 2

LITERATURE REVIEW

2.1 Summary

This chapter provides a detailed review of relevant literature in the study area. It includes fundamental theoretical concepts of anaerobic digestion process and provides detailed analyses on all pretreatment technologies emphasizing on microwave and ultrasonic pretreatment techniques. Pretreatment mechanisms and effect of pretreatment on solid reduction, COD removal, dewaterability and anaerobic digestion kinetics enhancement are explained. Factors affecting digester performance are assessed. The research gaps and the motivations for this research work are presented in the last part of this chapter.

2.2 Types and Characteristics of Sludge from a wastewater treatment plant

Sewage sludge is a complex heterogeneous mixture of microorganisms, undigested organics such as paper, plant residues, oils, or fecal material, inorganic materials and moisture (Degremont, 1979). The undigested biomass contains very complex mixture of organic compounds comprising proteins and peptides, lipids, polysaccharides, plant macromolecules with phenolic structures (e.g. lignins or tannins) or aliphatic structures (e.g. cutins or suberins), along with organic micro-pollutants such as polycyclic aromatic hydrocarbons (PAH) or dibenzofurans (DRRSS, 2002).

As shown in Figure 2.1, sludge processed in municipal wastewater treatment plant includes raw primary sludge into the primary and preliminary treatment units, the aeration units along with the secondary sedimentation tanks convert the sludge to activated sludge which is composed of large amount of microbial biomass. The activated sludge is thickened and anaerobically digested to produce digested sludge which has lower environmental load with less organic and solid content (Zhang, 2010).

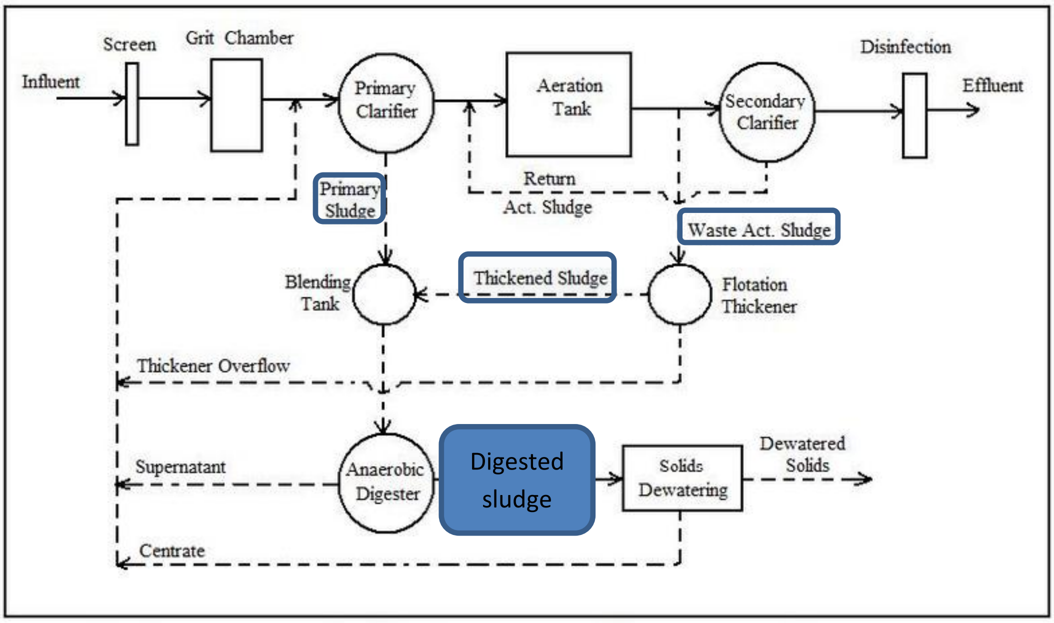


Figure 2.1 Process flow diagram for a conventional wastewater treatment plant (Tchobanoglous, 2003).

2.2.1 Primary sludge

Primary sludge which is also called raw sludge comes from the bottom of the primary clarifier as shown in Figure 2.1. It is easily digestible as it consists of highly degradable carbohydrates and fats, compared to activated sludge which consists of complex carbohydrates, proteins and long chain hydrocarbons. Hence, biogas production from primary sludge is easily digestible unless it contains less digestible complex organics like cellulose and lignin (Hanjie, 2010).

2.2.2 Excess activated sludge

Activated sludge, excess sludge or waste activated sludge is output of the secondary treatment process. Activated sludge is the result of over production of microorganisms as shown in Figure 2.1. Activated sludge is more difficult to digest than primary sludge(Hanjie, 2010).

2.2.3 Digested sludge

Primary sludge and excess activated sludge usually after a thickening process is subjected to anaerobic digestion that produces digested sludge. The digested sludge has reduced mass, it is less odorous and safer in terms of pathogen content and can be dewatered more easily than primary and activated sludge (Figure 2.1) (Houghton et al., 2002).

2.3 Anaerobic digestion technology and associated problems

Anaerobic digestion is a biological degradation of organic biomass in oxygen-deficient or free environment by a complex microbial consortium. During the process, the digestible organic biomass mainly produces methane, carbon dioxide and more biomass. The nitrogen which is not used for microbial growth will be released as ammonia (Coelho, 2012a).

Anaerobic digestion is an efficient sludge treatment technology used in a number of municipal wastewater treatment plants to stabilize organic matter. Mass reduction, methane production and improved dewatering properties of the treated sludge are the main features of this process. Biogas production through anaerobic digestion has recently captured global attention because of its substantial benefits including eco-friendly energy generation, greenhouse gases emission reduction, high organic removal from effluent and production of fertilizers. It is rated as one of the most energy-efficient and environmentally beneficial technologies for bioenergy production (Chong et al., 2012b, Müller, 2001, Weiland, 2010). Anaerobic digestion is a very effective sludge treatment technology applied in municipal and industrial wastewater treatment plants to stabilize organic matter (Park, 2011).

The process involves four major microbiological degradation steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis. One of the disadvantages of anaerobic digestion technique is the slow hydrolysis of microorganisms that accounts for 70% of excess sludge which is the primary degradation step in the anaerobic digestion process (Park, 2011). The microorganisms in the excess sludge contain Extracellular Polymeric Substances (EPS) that are resistant to biodegradation which in turn limits the rate of the whole anaerobic digestion process (Chong et al., 2012a, Tiehm et al., 1997). Anaerobic digestion is a sludge treatment used in a number of municipal wastewater treatment plants to stabilize organic matter. Mass reduction, methane production and improved dewatering properties of the treated sludge are the main features of this process (Tyagi and Lo, 2013b).

Anaerobic digesters have functional components like mechanical mixing, heating, gas collection sludge addition and withdrawal ports and supernatant outlets (Coelho, 2012a).

The advantages of anaerobic digestion process over aerobic digestion is the production of minimum excess sludge (Hanjie, 2010). Besides not needing any added chemical reagents, it can also produce a usable form of energy, as methane gas, and so reduce or eliminate (in optimal conditions) the need to supply energy to a wastewater treatment plant and the end-product methane (CH_4) results in reduced overall treatment cost (Coelho, 2012a). It is relatively cheaper to treat municipal and industrial sewage sludge compared to other sludge treatment technologies as shown in Table2.1.

Anaerobic digestion process is continuously undergoing modifications with improvement and development of new and complex technologies which are rapidly emergin (Tyagi et al., 2009). Despite the stability of the process, insufficient understanding of the biochemical and physical processes involved in the digestion process result in low CH_4 production and the accumulation of volatile fatty acids (VFAs) in the digesters. Furthermore, because of the complexity of the process, the behavior of digesters under changing organic loading rates (OLR) is unpredictable. The estimated parameters are generally case specific and difficult to adapt for system modification, since they depend on environmental conditions. Hence, thorough investigation on the effect of operational parameters is essential for all different sludge types (Noutsopoulos et al., 2013).

Table 2.1 Techniques for resource recovery (Rulkens and Bien, 2004).

Techniques	Cost of treatment	Environmental paybacks	Advancement require	Development stage	Remarks
Anaerobic digestion	Low/moderate	Energy (biogas) generation	Sludge pre-hydrolysis required to enhance biogas generation	Successfully applied at full scale	Release of phosphate and ammonia during digestion process
Incineration	High	Energy generation, minimization of biosolids quantity	Mechanical dewatering, drying, use of waste heat	Full scale	Phosphate can be recovered from ash
Co-incineration in coal fired power plant	High/moderate	Energy generation, beneficial use of inorganics	Mechanical dewatering, drying, use of waste heat	Full scale	Relative amount that can be co-incinerated is limited
Pyrolysis and gasification	High	Valuable products recovery, minimization of biosolids quantity	Mechanical dewatering, drying, use of waste heat	In development stage	Complex process, marketing of products needs attention
Wet air oxidation	Moderate	Improvement in dewatering properties of sludge	Optimization	Applied globally in practice	Process primarily focused on sludge dewatering
Supercritical water oxidation	High	Energy generation, minimization of biosolids quantity	Reactor concept, process performance	In development stage	Complex process, Corrosion and scaling problems of the reactors walls
Hydrothermal treatment	Moderate	Biogas generation, production of valuable carbon resource for denitrification, minimization of biosolids quantity	Process performance	Practical experience limited	Removal of heavy metals can be included

O’Sullivan et al. (2007) measured the concentration of intermediates like VFA and hydrogen in order to calculate the COD equivalent accounting for the component not converted to methane. cumulative SCOD increased significantly while cumulative methane is still low suggesting that intermediates formed during the hydrolysis step were possibly toxic to the methanogenic population (Vlyssides and Karlis, 2004). Anaerobic digestion has the disadvantage of very long retention time and reduced overall efficiency (40-50%). A key factor to work on for effective enhancement in anaerobic digestion is the slow hydrolysis process as it is the rate limiting step (Wei et al., 2011, Xu et al., 2010c, Yang et al., 2009). The methanogenic process is generally limited by the rate of hydrolysis of suspended matter and organic solids. This is of particular importance during the anaerobic treatment of solid wastes, slurries and manure, and wastewaters with a high concentration of suspended solids (SS), such as domestic sewage. By means of efficient pre-treatment the suspended substrate can be made better accessible for the anaerobic bacteria, optimizing the methanogenic potential of the waste to be treated (Lens and Verstraete, 2001).

2.4 Microbiology of anaerobic digestion

Organic substrates involved in anaerobic digestion can be grouped as primary substrates, which are present in the effluent or residues to be treated, intermediate substrates and final products (Batstone, 1999). The degradation process involves the microbilogial hydrolysis of complex organics to soluble products; conversion of monomers to volatile fatty acids (VFAs) by acidogenic bacteria; conversion of propionic, butyric, and alcohols to acetate, CO_2, and H_2 by acetogenic bacteria; and finally conversion of acetate and hydrogen to methane as shown in Figure 2.2 (Kaspar and Wuhrmann, 1978).

The Hydrolytic Bacteria primarily involves the breakdown of complex organic waste streams into simple sugars, fats and oils, and amino acids. This stage involves splitting of the complex organic biological molecules into simpler forms. The fermentative acidogenic bacteria convert the hydrolyzed portion into Organic acids. The fermentative acetogenic bacteria then convert the Organic acids into hydrogen,acetate and CO_2(g). Finally, the methane producing bacteria, the methanogens simultaneously produce biogas from the Acetate, or from Hydrogen and Carbon (IV) oxide as shown in Figure 2.2 (Bougrier et al., 2006).

2.4.1 Hydrolysis – it is a step mediated by extracellular enzymes, in which substrates and particles that cannot be used directly by the microorganisms are solubilized (Figure 2.2).

2.4.2 Acidogenesis – is the degradation of soluble substrates, such as amino acids and sugars that can be degraded without an external electron acceptor. The products are organic acids and alcohols (Figure 2.2).

2.4.3 Syntrophic acetogenesis and hydrogenophilic methanogenesis

Acetogenesis is the degradation of the fermentation products to acetate, using hydrogen ions or bicarbonate as external electron acceptors. This process is coupled with the methanogenesis from hydrogen, which maintains a low concentration of hydrogen which is necessary to keep the reaction thermodynamically favourable (Figure 2.2).

2.4.4 Aceticlastic methanogenesis – is degradation of acetate to carbon dioxide and methane, by highly specialized microorganisms. The anaearobic bacteria flora involved in each degradation step are different in their functions there are four digesting bacteria (Figure 2.2).

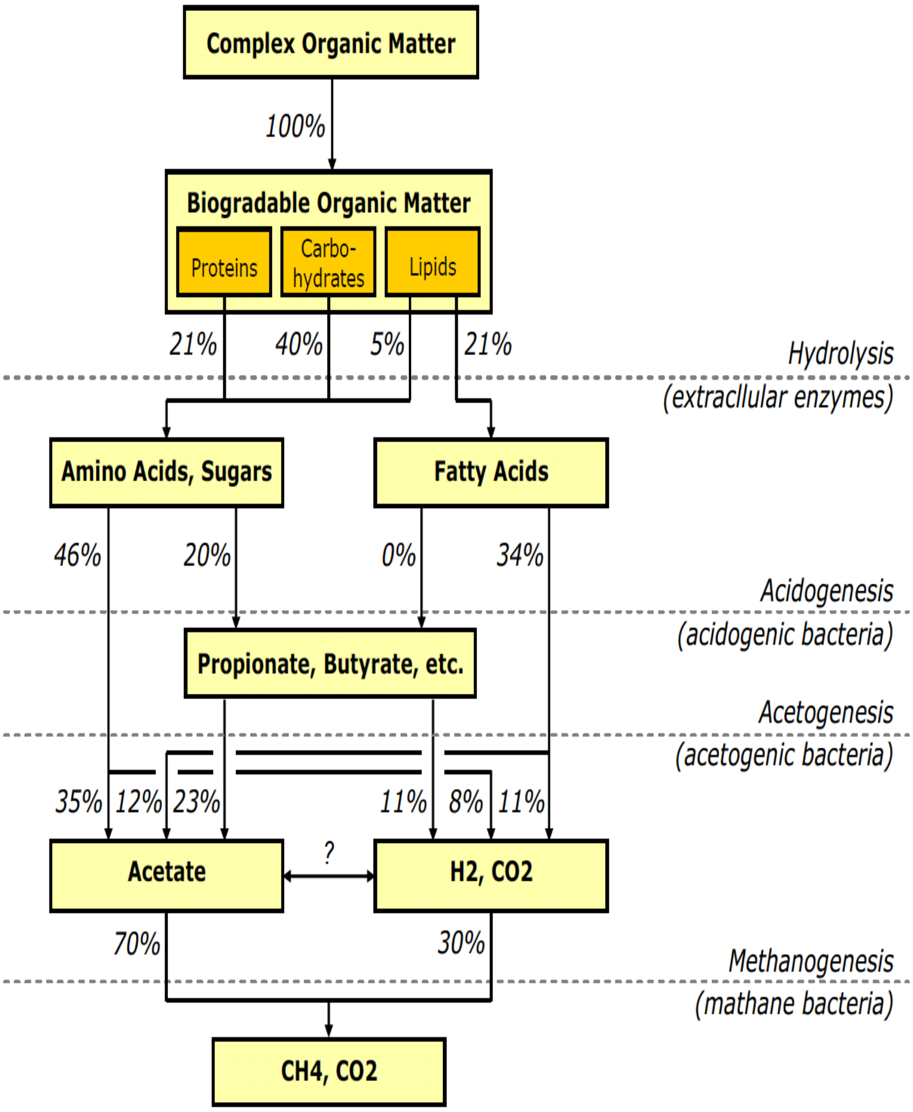


Figure 2.2 The microbiological metabolic path way and groups involved in anaerobic digestion process (Navaratnam, 2007)

2.5 Anaerobic digestion kinetics

Process kinetics plays a central role in the development and operation of anaerobic treatment systems. Based on the biochemistry and microbiology of the anaerobic process, kinetics provides a rational basis for process analysis, control, and design. In addition to the quantitative description of the rates of waste utilization, process kinetics also deal with operational and environmental factors affecting these rates. A sound knowledge of kinetics allows the optimization of performance, a more stable operation as well as better control of the process (Fountoulakis et al., 2010). Retention time relates to process kinetics, specifically the kinetics of bacterial growth, and thus is the primary factor that should be used for sizing digesters.

2.5.1 Disintegration, solubilization and enzymatic hydrolysis

Disintegration, solubilization and enzymatic hydrolysis are mostly represented in general kinetic term of hydrolysis for most practical application as hydrolysis is the slowest rate determining step in the process (Batstone et al., 2002a). Acidogenesis stage is considered to be the fastest step in the methanogenesis process. For enhanced methane production there should be a balance between the different steps of the process. For a multistep reaction the overall rate is determined by the slowest rate limiting step. The rate-limiting step in anaerobic digestion with suspended organic matter is the hydrolysis of solids (Vavilin et al., 2008a).

2.5.2 Hydrolysis rate constant

The hydrolysis rate constant can be calculated by using biochemical methane potential data from the digesters. The methane yield is a function of the reduction of organic material achieved during anaerobic digestion, which reflects on the hydrolysis rate. The hydrolysis rate constant is an indicator of the speed of hydrolysis achieved in the digesters. Enhancing the hydrolysis rate constant is an important factor. The hydrolysis rate constant can be determined using the Gompertz equation (Gadhamshetty et al., 2010). This model represents cumulative methane production as a function of the methane production potential, maximum methane production rate and duration of the lag phase. The equation is shown below.

$$M = P \times \exp\left\{-\exp\left|\frac{R_m \times e}{P}(\lambda - t) + 1\right\}\right. \qquad (2.1)$$

where: M is the cumulative methane production (mL),

P is the methane production potential (mL),

Rm the maximum methane production rate (mL/d),

λ is the duration of the lag phase (d), and

t is the duration of the assay in which cumulative methane production M is calculated (d).

According to Batstone et al. (2002), hydrolysis can be expressed using the following two conceptual models:

(a) The enzyme secreted by the organism will be adsorbed to the surface of the particle or react with soluble substrate (Jain et al., 1992).

(b) The organism attach to the surface of the particle and consume soluble products produced from the enzymatic reaction (Vavilin et al.,1996). The Michaelis–Menten kinetics may be applied for the hydrolysis of a soluble substrate, given by:

$$\frac{ds}{dt} = K.E\frac{S}{Km+S} = Vm\frac{S}{Km+S} \tag{2.2}$$

Where S, E are the substrate and enzyme concentrations, Vm = kE is the maximum hydrolysis rate, k is the maximum hydrolysis rate constant, and Km is the half-saturation rate coefficient.

2.5.3 The first-order kinetics of carbohydrate, lipid and protein degradation.

The following differential equations describe hydrolysis of protein lipid, or carbohydrate concentration as the first-order reaction in a way not linked to the bacterial growth (Vavilin et al., 2008a).

$$\frac{ds}{dt} = -kS \tag{2.3}$$

$$\frac{dP}{dt} = \alpha kS \tag{2.4}$$

Where S is the volatile solids (VS) concentration, P is the product concentration, k is the first-order rate coefficient, and α is the conversion coefficient of VS to product. After integration the product concentration is expressed as:

$$P = Po + \alpha So(1 - e^{-kt}) \tag{2.5}$$

Where Po and So are the initial product and substrate concentrations respectively. A non-linear regression may be used to estimate the values of coefficients k and α and their standard deviations. The enzyme secreted by the organism will be attached to the surface of the particle or substrate and benefit from the soluble products.

Table2.2 shows the kinetic coefficients for first order hydrolysis problem using the models discussed in equations (2.1) through (2.5).

Table 2.2 Kinetic coefficients of the first-order rate of hydrolysis (Vavilin et al., 2008b).

Substrate	***k* (day^{-1})**	**T (°C)**	**References**
Carbohydrates	0.025–0.2	55	(Christ et al., 2000)
Proteins	0.015–0.075	55	(Christ et al., 2000)
Lipids	0.005–0.010	55	(Christ et al., 2000)
Carbohydrates	0.5–2.0		Garcia-Heras (2003)
Lipids	0.1–0.7		(Garcia-Heras 2003)
Proteins	0.25–0.8		Garcia-Heras (2003)
Lipids	0.76		Shimizu et al. (1993)
Lipids	0.63	25	Masse et al. (2002)
Cellulose	0.066	35	Liebetrau et al. (2004)
Kitchen waste	0.34	35	Liebetrau et al. (2004)
Biowaste	0.12	35	Liebetrau et al. (2004)
Pig manure	0.1	28	Vavilin et al. (1997)
Proteins (gelatine)	0.65	55	Flotats et al. (2006)
Municipal solid waste	0.1	15	Bolzonella et al. (2005)
Office paper	0.036	35	Vavilin et al. (2004)
Cardboard	0.046	35	Vavilin et al. (2004)
Newsprint	0.057	35	Vavilin et al. (2004)
Food waste	0.55	37	Vavilin et al. (2004)
Forest soil	0.54	30	Lokshina and Vavilin (1999)
Forest soil	0.09–0.31	20	Lokshina and Vavilin (1999)
Slaughterhouse waste	0.35	35	Lokshina et al. (2003)
Household solid waste	0.1	37	Vavilin and Angelidaki (2005)
Primary sludge	0.99	35	(N.E. Ristow et al., 2006)
Secondary sludge	0.17–0.60	35	Ghosh (1981)
Crops and crop residues	0.009–0.094	35	Lehtomaki et al. (2005)

2.6 Enhancement of Anaerobic biodegradability by various pretreatment techniques

In anaerobic digestion of waste activated sludge (WAS), hydrolysis is considered to be the rate limiting step. Indeed, after aerobic treatment in a wastewater treatment plant, much of the organic matter in the sludge appears in the form of microbial biomass like in bacterial flocs which lessens its availability to anaerobic micro-organisms. Reduction of solids and methanization of sewage sludge can be improved by enhancing its rate limiting step, organic matter hydrolysis (Li and Noike, 1992). Therefore, WAS solubilization or disintegration, by alkaline addition,(Li and Noike, 1992, Lin et al., 1997, Navia et al., 2002, Penaud et al., 1999) thermal (Barlindhaug and Odegaard, 1996, Haug et al., 1983, Haug et al., 1978, Kepp et al., 1999)or thermo-chemical (Mustranta and Viikari, 1993, Penaud et al., 2000, Stuckey and McCarty, 1978, Tanaka and Kamiyama, 2002) pre-treatments would enhance or improve the biodegradability of the available organic mass.

Different pretreatment technologies were found to enhance sludge hydrolysis and anaerobic digestion performance (Carrère et al., 2010a). Pretreatment of sludge through ultrasonic, mechanical, chemical or thermal techniques result in bacterial cell wall disruption, disintegration of EPS and release of enzymes which enhance the rate of hydrolysis and biodegradation (Tyagi and Lo, 2011, Eskicioglu et al., 2006).

Ultrasonic, microwave, oxidative, and thermal pretreatment techniques are well documented in literature as viable methods to enhance biodegradability, hydrolysis rate and digester performance (Bougrier et al., 2006).

Pretreatment of sludge has been reported by many researchers to improve sludge hydrolysis, biodegradability and, dewaterability. Such technologies include ultrasonic treatment (Apul and Sanin, 2010a, Farooq et al., 2009, Saifuddin and Fazlili, 2009a, Tiehm et al., 2001), chemical treatment (Mustranta and Viikari, 1993, Penaud et al., 2000, Stuckey and McCarty, 1984, Tanaka and Kamiyama, 2002), microwave treatment (Eskicioglu et al., 2007c, Park, 2011) ozone oxidation (Yeom et al., 2002), mechanical disintegration, supercritical and subcritical water oxidation and high temperature hydrolysis (Carrère et al., 2010b). All sludge reduction technologies are working with the principle of disintegration of cell walls, and large organic molecules.

A significant increase in biogas production can be obtained by applying pre-treatment such as microwave irradiation, ultrasonication, ozonation, high-pressure homogenizer method, chemical pretreatment with acid or alkali etc.(Tyagi and Lo, 2013b).

Ultrasonic pretreatment is an emerging and promising mechanical pretreatment technique for the solubilisation of sludge (Pilli et al., 2011b). It was reported that macromolecules with a molar mass of above 40,000 are disrupted by the hydro-mechanical shear forces produced by ultrasonic cavitation (Tiehm et al., 2001). Only Ultrasonic pretreatment method was applied on large scale Wastewater Treatment Plants (WWTP) compared to other pretreatment methods (Carrère et al., 2010b).

Microwave (MW) irradiation is another efficient sludge pre-treatment technology that enhances biodegradability, methane production and digester performance (Park, 2011). It is a novel pre-treatment method for stabilization of waste activated sludge (WAS). Microwave pre-treatment of sludge increases biogas production, reduce sludge viscosity, improve dewaterability and improve pathogen decay as compared to digestion of sludge pre-treated through conventional heating and untreated sludge (Eskicioglu et al., 2007b).

Several studies revealed that temperature, ozone dose, ultrasonic energy density and pH have a beneficial impact on the disintegration of sludge. Different pretreatment techniques were observed to have a synergistic effect not only when they are applied independently, but also when they are systematically combined (Xu et al., 2010c). The synergy between MW irradiation and H_2O_2 based oxidative pretreatment of waste activated sludge significantly enhanced performance compared to the performance of individual pretreatment (Eskicioglu et al., 2008b).

There are numerous studies on the benefits of different pretreatment techniques including ultrasonic and microwave pretreatment when the methods are applied independently and in combination with other pretreatment options like chemical and thermal pretreatment (Valo et al., 2004, Vlyssides and Karlis, 2004).

The application of more than one treatment also resulted in improved sludge biodegradation, floc destruction, cell wall disruption and release of organics due to the complementary synergy between the treatment techniques that are combined (Saha et al., 2011b, Saifuddin and Fazlili, 2009a, Xu et al., 2010a). Microwave enhanced-oxidative pretreatment with H_2O_2 resulted in 11-34% TS, TCOD reduction and total biopolymer solubilisation (Eskicioglu et al., 2008b). Combined ultrasonic-alkali pretreatment of waste activated sludge resulted in 60% VS solubilisation. The use of NaOH weakens the cells walls increasing the disintegration effect of ultrasonication or other lysis techniques (Tyagi and Lo, 2011).

Combined microwave and ultrasonic pretreatment technique was reported in limited number of literature that the combination will be a rapid and effective method for digestion of biological materials for metal extraction (Lagha et al., 1999) starch hydrolysis (Villiere et al, 2013), enhanced heavy metal and edible oil extraction (Chemat et al., 2001) and production of ultrapure coal (Royaei et al., 2012). Combined microwave-ultrasonic pretreatment, in this study is applied for anaerobic digestion enhancement purpose and resulted in significant improvement in gas production, solid removal and dewaterability of municipal sludge compared to the individual ultrasonic or microwave pretreatment approaches (Yeneneh et al., 2013a, Yeneneh et al., 2013b).

2.6.1 Mechanical pretreatment

Mechanical pretreatment of WAS by jetting and colliding to a collision-plate at 30 bar made the sludge solubilized. The research showed that solubilization of WAS is effective to the digester performance through measuring the unit gas production, volatile fatty acids (VFA), pH, and volatile mass reduction efficiency. WAS pretreatment allowed a decrease in the digester SRT from 13 to 6 days, without major effects on process efficiency and on effluent quality. It enhanced volatile mass reduction and unit gas production (Nah et al., 2000).

2.6.2 Ultrasonic

Ultrasounds have been extensively tested in industry, particularly as pretreatment for anaerobic digestion. It has been shown that macromolecules with a molar mass above 40,000 are disrupted by the hydro-mechanical shear forces produced by ultrasonic cavitation. The mechanical forces are most effective at frequencies below 100 kHz (Portenlanger, 1999). Ultrasonic pretreatment is discussed in depth in section 2.6.7 as a major pretreatment technology of focus in this work.

2.6.3 Lysis centrifuge

Lysis-centrifuge operates directly on the thickened sludge stream in a dewatering centrifuge. It will then be re-suspended with the liquid stream.. The increase of biogas production was found to be 15–26% (Dohanyos et al., 1997).

2.6.4 Chemical treatment

Ozonation is the most extensively used chemical method. It results in partial sludge solubilisation and yield increases with ozone dose. A too high ozone dose will result in reduced apparent solubilisation due to oxidation of the solubilised component (I.T. Yeom et al., 2002). Ozonation has also been combined with anaerobic digestion as a pretreatment or post treatment and recycling back to the anaerobic digester (Goel et al., 2003).

In addition to ozone, hydrogen peroxide has also been applied as an oxidation agent to enhance the anaerobic digestion process (Valo et al., 2004, Rivero et al., 2006). The COD removal during anaerobic digestion was enhanced by means of oxidation at 90 °C with 2 g H_2O_2 g^{-1} VSS, instead of oxidation that takes place at a lower temperature 37 °C (Rivero et al., 2006).

On the other hand, alkali treatment is relatively effective in sludge solubilisation, with the order of effectiveness being (NaOH > KOH > $Mg(OH)_2$ and $Ca(OH)_2$) (Kim et al., 2003). But very high concentration of sodium and potassium ions may have synergistic inhibitory effect on the methane generation process (A.H. Mouneimne et al., 2003).

As organic matter in pretreated primary sludge was hydrolysed during pretreatment; an increase of SCOD from 1664.0 mg/L to 20472.7 mg/L was achieved, when the concentration of sodium hydroxide solution was increased to 1.2%. On the other hand, the concentration of volatile suspended solids (VSS) was reduced in the range from 6% to 19% after pretreatment which may be due to the progressive hydrolysis of the complex organic matter in the feed.

Higher alkalinity increases the buffering capacity which prevents decrease in pH or it helps to resist changes in pH caused by the addition of acids (Yunqin et al., 2010). Alkaline hydrolysis has been reported to significantly increase organic yield from acidogenesis, Tanaka et al. (1997) tested the addition of NaOH to WAS, and found a solubilization percentage of VSS of 15% for an alkaline dose of approximately 0.6 g NaOH/g VSS. The methane production was 50% higher compared to the control for a dose of 1 g NaOH/gVSS. Lin et al. (1997) tested the addition of two different concentrations of NaOH (20 and 40 meq/L) to sludge with two different solids concentrations (1 and 2%). The methane production was between 19 and 286%

higher in the sludge pretreated compared to the control sludge. The amount of soluble COD increased from a total COD/soluble COD ratio of 2 to 38% in the test with 1% TS sludge pretreated with 40 meq/L NaOH.

The chemical characterization of primary pretreated sludge after bio pretreatment shows that as organic matter was hydrolyzed during pretreatment (Yunqin et al., 2010) .

Alkalinity in anaerobic digestion system is because of the presence of hydroxides, carbonates and bicarbonates of elements such as calcium, magnesium or ammonia (Metcalf & Eddy 1991).

In addition, the increase of NH_3-N also led to an increase of alkalinity. Pretreatment helps to solubilize carbonates and phosphates, resulting in alkalinity increment (Turovskiy and Mathai, 2006).

2.6.5 Biological and thermal techniques

Biological pre-treatment improves the hydrolysis process before digestion for temperature phased anaerobic digestion (TPAD) with thermophilic (around 55 °C) or hyper-thermophilic (between 60 and 70 °C) conditions. It assists degradation of the sludge gel structure and release of bound water which enhances sludge dewaterability after treatment at 150 °C (Fisher and Swanwick, 1971) or 180 °C (Anderson et al., 2002).

WAS is the main by-product of biological wastewater treatment processes and usually consists of 70% organic matter (Wilson and Novak, 2009).

Since wastewater sludge contains significant fractions of both lipid and protein (Tanaka et al., 1997a), compounds which inhibit methanogenesis such as ammonia (Lay et al., 1999) and hydrophobic fatty acids may be products of thermal hydrolysis of proteins and lipids. Thermal biological provides a moderate performance increase over mesophilic digestion, with moderate energetic input. While increased nutrient release can be a substantial cost in enhanced sludge destruction, it also offers opportunities to recover nutrients from a concentrated water stream as mineral fertiliser (Carrère et al., 2010a).

2.6.6 Thermal pretreatment

Thermal pre-treatment can be applied for the improvement of stabilization, enhancement of dewatering of the sludge, reduction of the numbers of pathogens (Müller, 2001).

Thermal hydrolysis improves solubilisation of sludge which enhances anaerobic digestion. Several researchers have investigated thermal hydrolysis for pretreatment of anaerobic digestion (Haug et al., 1978), (Tanaka et al., 1997b). Most studies recommend an optimal temperature in the range of 160–180 °C and treatment times of 30 to 60 min.

The effect of thermal pre-treatment on the anaerobic biodegradability and toxicity of activated sludge was investigated in the study of Stuckey and McCarty (1984). It was found that WAS increased with increasing pretreatment temperature up to a maximum at 175°C, and this resulted in an increase of methane production by 27% over the control. With the compounds and cultures used, mesophilic degradation and toxicity were found to be significantly higher than the corresponding values under thermophilic conditions (Stuckey and McCarty, 1984).

Zheng et al. (1998) applied a kind of rapid thermal conditioning to sludge combined with anaerobic digestion. Sludge was heated rapidly to reaction temperature up to about 220 ^{O}C and quenched after 10–30 s. They concluded that rapid thermal conditioning would reduce the quantity of bio-solids requiring disposal, eliminate the need for polymer coagulant, improve dewaterability, increase methane production, and further reduce the concentration of pathogens.

Pinnekamp et al, (1989) studied anaerobic digestion in temperature range between 150°C and 275°C. They observed an optimum in methane production after pre-treatment at 175°C whereas at more elevated temperatures, a decrease in methane production and sludge biodegradability was observed which was attributed to the formation of toxic, refractory compounds.

Another experiment involving pre-treatment of primary and secondary sludge for 1 h at temperatures between 120°C and 220°C was described by Pinnekamp (Pinnekamp, 1989). A decrease in gas production below that of the non-pre-treated sludge was observed for temperatures higher than 180°C; however, the differences in gas yield increase at pre-treatment temperatures between 120°C and 180°C were not considerable. Digestion of the thermally pre-treated sludge resulted in an increase of 60–70% in methane production over not pre-treated sludge (Haug et al., 1978).

Li and Noike (1992) focused on the thermal pre-treatment of secondary sludge and they reported 170°C and 60 minute as the most favorable pre-treatment temperature and duration respectively, regarding COD removal and gas production during mesophilic (37°C) anaerobic digestion yielding an increase of approximately 100% compared to the untreated sludge. However, higher temperature pre-treatment has high energy requirements and is difficult to operate.

Thus, thermal pre-treatment at a lower temperature, i.e. below 100°C becomes more and more attractive. Wang et al. (1997) studied the performance of lower temperature pre-treatment (60–100°C) on mesophilic (37°C) anaerobic digestion of waste activated sludge. It was concluded that thermal pre-treatment resulted in a significant increase (30–52%) in methane yield; however, no significant differences were observed between pre-treatments at 60°C, 80°C and 100°C (Yang et al., 2010).

Methane production rate was higher after the pretreatment at 60°C compared with 80°C and 100°C. One can see that there are already numerous studies investigating the effect of the pre-treatment temperature on the anaerobic digestion of sludge. However, most studies focus on the investigation of the temperature selection and pre-treatment duration using one type of sludge while in most municipal treatment plants primary and secondary sludge streams are combined prior to anaerobic digestion (Park, 2011).

Solubilization of organic matter from samples of WAS and a mixture of primary sludge, and WAS in the order of 40–60 and 20–35%, respectively, when the treatment temperature is 170^0C. Experiments with municipal sewage sludge show that the highest yield of hydrolysis can be achieved at 165–180 ^{0}C. The pretreatment time (10–30 min) has little influence on the result. The dissolved components are readily degradable in a digestion process. In addition the dewaterability is increased (Neyens and Baeyens, 2003).

Heat treatment at lower temperature has the benefits of dewaterability with improved digestibility and at the same time avoid the problems that occurred with higher temperature heat treatments (Haug et al., 1978). Pretreatment at higher temperature resulted in decreased gas production. Thermal hydrolysis as pretreatment has hence given very good results on digester performance (Carrère et al., 2010a).

Elbing and Dünnebil, (1999) investigated the effects of thermal hydrolysis on mesophilic digestion of waste activated sludge. After pre-treatment at 135^{o}C, the volatile solids destruction in the digester increased to 135 and 235% above the reference level at an increasing 12 and 15 days retention time, respectively.

Dohanyos et al. (1997b) tried pretreatment of the sludge at 100ºC for 20 minutes. The results showed an increase of 41.8% in methane production and 27.6% in VS reduction. Tanaka et al. (1997) also tested several temperatures for a pretreatment time of 1 hour and observed that VSS solubilization was around 15% for temperatures between 115 and 150ºC and then increased further above 160ºC, reaching 30% at 180ºC.

Thermal treatment at very high temperatures greater than 170–190 °C leads to reduced sludge biodegradability despite high solubilisation efficiencies. This is usually associated to Maillard reaction (Dwyer et al., 2008) which involves carbohydrates and amino acids in the formation of melanoidins, which are difficult or impossible to degrade (Bougrier et al., 2008).

Li and Noike (1992) tested several pretreatment options varying either the temperature (between 65 and 175ºC) and the duration of the pretreatment (between 15 and 120 minutes), they found that the maximum improvement occurred for temperatures of 170ºC and 60 minutes duration. Longer times did not result in better results. The retention time in the digester could be reduced by 5 days and methane production was twice as high as the control.

Thermal hydrolysis results in a substantial performance increase with a substantial consumption of thermal energy. It is likely that low impact pretreatment method such as mechanical and thermal pretreatment improved speed of degradation, while high impact methods such as thermal hydrolysis or oxidation improve both speed and extent of degradation (Pinnekamp, 1989, Gavala, 1999).

2.6.7 Ultrasonic pretreatment

In sewage sludge treatment, ultrasound is applied as a pretreatment to improve anaerobic sludge stabilisation. The high shear forces created in the advent of cavitation can be used to improve process efficiency in sludge dewatering and to achieve sludge disintegration (Apul and Sanin, 2010a).

Due to the ultrasonic disruption of putrescible biomass in the sludge, subsequent microbial degradation occurs up to 4 times faster than in the conventional treatment. The violent collapse of cavitation bubbles in water produces shear forces that can disrupt cell membranes and kill bacteria. At lower acoustic intensities these forces weaken the membranes rendering the bacteria more susceptible to the effect of biocides. The hydroxyl radical produced during cavitation can also assist disinfection (Oh, 2006, Portenlanger, 1999).

Ultrasound is applied in water treatment and environmental applications, the destruction/transformation of organic compounds is the prime objective of fundamental and applied investigations involving ultrasound (Silva et al., 2013).

2.6.7.1 Factors affecting ultrasonication efficiency

The mechanisms of ultrasonic treatment are influenced by the energy supplied, ultrasonic frequency and the nature of the sludge. Cell disintegration is proportional to energy supplied (Bougrier et al., 2005b). High frequencies promote oxidation by radicals, whereas low frequencies promote mechanical and physical phenomena like pressure waves (Portenlanger, 1999). The full-scale installations of ultrasonication have demonstrated that there is 50% increase in the biogas generation. Besides, from energy balance calculations the average ratio of the net energy gain to electric power consumed by the ultrasound device is 2.5 (Pilli et al., 2011b).

Factors affecting ultrasonication process and impacts of sludge characteristics on sludge disintegration and biogas production and anaerobic digester performance is presented in the following sections.

2.6.7.2 Mechanism of cell disruption in ultrasonic pretreatment

The degradation of chemical pollutants is achieved by the effects of acoustic cavitation. The reaction rate is a function of the physico-chemical properties of the target compounds. Volatile and hydrophobic pollutants are degraded by thermal reactions in the “hot spot” of the cavitation bubble. Compounds which are more hydrophilic are decomposed in the bulk liquid by hydroxyl radicals produced in the cavitation bubble.

According to (Tiehm et al., 2001, Wang et al., 2005) disintegration mechanisms during ultrasonic disintegration of sludge can be:

(a) Hydro-mechanical shear forces

(b) Oxidising effect of radical OH, H, N, and O produced under the ultrasound radiation

(c) Thermal decomposition of volatile hydrophobic substances in the sludge

(d) Increase of temperature during ultrasonic activated sludge disintegration

Ultrasonic treatment involves development of cavitation (Figure 2.3), that occurs more at low frequencies, and chemical reactions due to the formation of OH^-, HO_2^-, H^+ radicals at high frequencies. In sludge treatment, low frequencies (20–40 kHz) are the most efficient(Tiehm et al., 1997)

Ultrasonic treatment involves mechanism of mechanical disruption of (Haug et al., 1978) the cell structure and floc matrix with continuous cycle of cavitation bubble growth and collapse. There are two major mechanisms of mechanical disruption as shown in Figure 2.3. Ultrasonication induces cavitation which breaks the cell walls of microbes and releases the intracellular components into the aqueous phase (Pilli et al., 2011b). Therefore, the sonication parameters affecting cavitation will affect the sludge digestion. The increased VS reduction directly translates into increased methane generation during the anaerobic digestion and less stabilized biosolids to be disposed of (Pilli et al., 2011b) . Other ultrasonic effects such as acoustic streaming, local heating, interface instabilities, agitation and cavitation may also be beneficial for solid/liquid separation (Sarabia et al.2000). The mechanisms of ultrasonic influence on sludge are not very clear, but the application of ultrasound to industrial process is relatively easy and possible (Yin et al., 2004)**.**

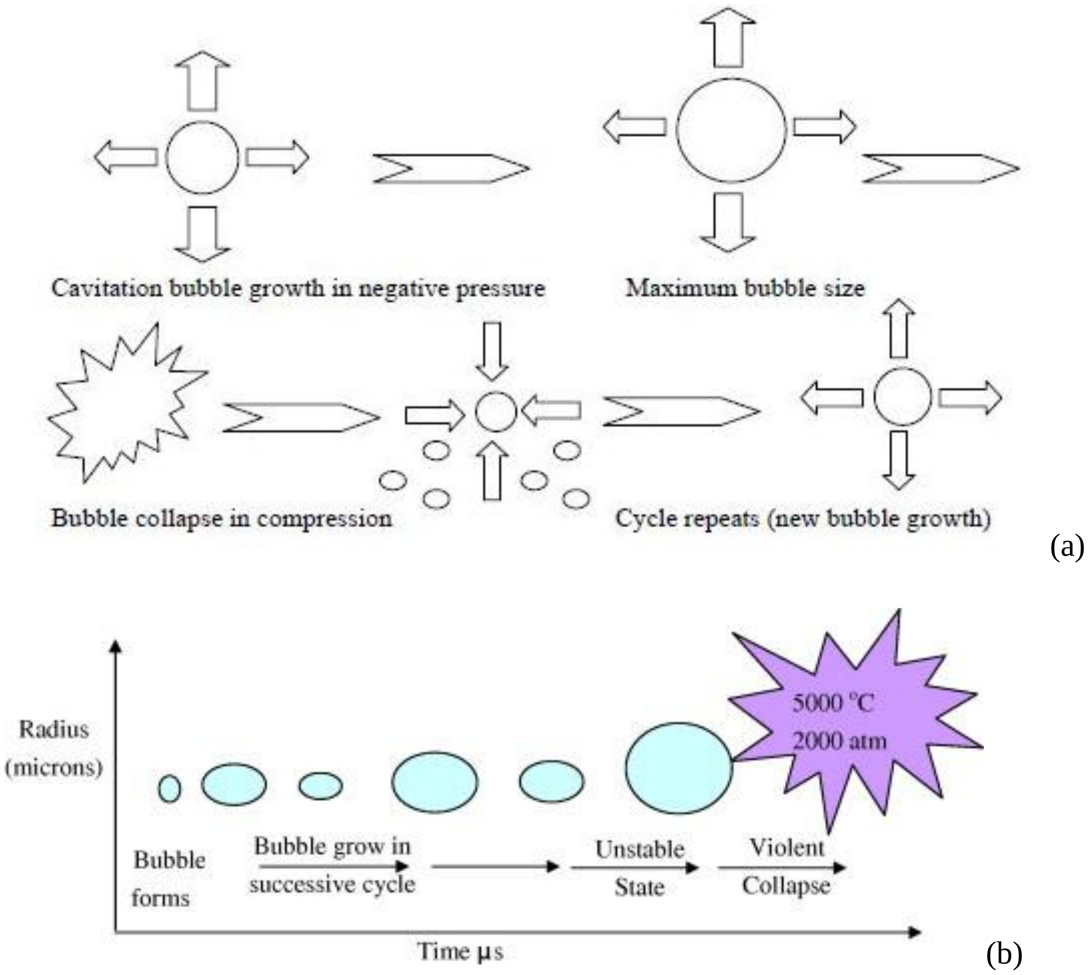


Figure 2.3 Development and collapse of the cavitation bubble (Pilli et al., 2011).

2.6.7.3 Effects of ultrasonication on sludge degradability and methane production in anaerobic digester

The primary aim of ultrasonication is to increase the sludge biodegradability to enhance the methane production at lower HRT in the anaerobic digester (Pilli et al., 2011b).

Pilli et.al (2011) reported that the pretreatment of the sludge by ultrasonication has a significant effect on the sludge biodegradability during the anaerobic digestion that increases biogas generation as well as percentage of methane in the biogas. Almost 31% reduction in sludge cake can be achieved in full-scale application and also it will increase the dewaterability of sludge.

The major factors affecting the performance of an ultrasonication unit are given in Table 2.3. The opinion of many researchers is that the effect of ultrasonic density is supposed to be more vital than the sonication time. Studies with kinetic models have shown that the effect of parameters is in the order of pH > sludge concentration > ultrasonication intensity > ultrasonic density. Mass and energy balance on full-scale studies showed that 1 kW of ultrasonic energy used generates about 7 kW of electrical energy after loss. Thus, higher amount of capital and operating cost can be overcome with significant reduction in the size of digesters operating at lower HRT, which will give a significant boost to sludge management at wastewater treatment plants (Tyagi et al., 2013).

Table 2.3 Expressions for sludge disintegration.

No.	Parameter	Expression	Unit	Reference
1	Specific energy input	$Es = \frac{P * t}{V * TS}$	kJ/kg TS or kW s/kg TS	(Feng et al., 2009)
2	Ultrasound dose	$UDo = \frac{P * t}{V}$	J/L	(Tiehm et al., 2001)
3	Ultrasound density	$UD = \frac{P}{V}$	W/L	(Tiehm et al., 2001
4	Ultrasound intensity	$UI = \frac{P}{A}$	W/cm^2	(Wang et al., 2005)

Es: specific energy in kW s/kg TS (kJ/kg TS); P: power input (kW); t: sonication time (s); V: volume of sludge (L); TS: total solids concentration (kg/L); A: surface area of the probe in cm^2.

It was reported that ultrasonic pretreatment results in disruption of cells and large sized macromolecules by the hydro-mechanical shear forces produced by ultrasonic cavitation (Appels et al., 2008b). Sonication density of 0.5W/mL and sonication intensity of 4.8W/cm^2 resulted in significant increase in soluble COD and 24.6% increase in VS reduction (Apul and Sanin, 2010b)

The rate of biogas production is directly proportional to the net rate of solubilisation. Increase in COD solubilisation results in increased methane production which will decrease the required HRT in the digester, and thereby reducing the overall size of the reactor significantly. Volatile solids reduction increases with increase in ultrasonication, which will increase the degradation efficiency of the sludge in AD.

2.6.7.4 Solubilisation of waste-activated sludge by ultrasonic treatment

Figure 2.4 shows that biogas production associated to the particulate fraction of sludge was constant for specific energy input lower than 3000 kJ/kg of total solids even if the solids concentration decreased. On the other hand, biogas production linked to the soluble part of sludge increased with ultrasonic power (Bougrier et al., 2005b). For an energy input lower than 1000 kJ/kg TS, the floc size reduction was important: d50 strongly decreased, with a reduction of about 40%. Then, particles size decreased more progressively as shown in Figure 2.4 (Bougrier et al., 2005b).

In term of biogas production, ultrasonic energy higher than 7000 kJ/kg TS is not effective. Indeed, when the supplied energy was higher than 7000 kJ/kg TS, biogas generation was constant solubilisation did not change. Moreover, biogas production linked to the particulate fraction did not depend on solid concentration for low energy input. Biogas production linked to particles was limited. But if matter was solubilised, this matter became available for bacterial action *(*Bougrier et al., 2005b*)*.

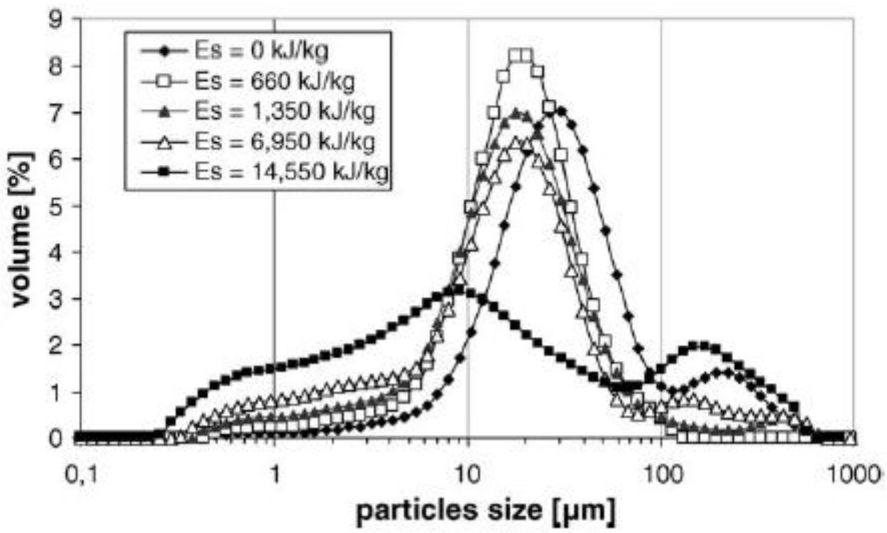


Figure 2.4 Particles size distribution for different specific energy inputs (Bougrier et al., 2005).

Sludge solubilisation is a also function of the specific energy input (treatment time and applied power) (Braguglia et al., 2008).

However, the relationship between specific energy input and sludge solubilisation is not linear, but rather follows an s-shaped curve. No significant COD solubilisation was observed at a specific energy <1000 kJ/kg TS, which shows reduced disintegration of the sludge flocs and microbial cells (Tyagi et al., 2013). Below this threshold value, all sonication energy is consumed to reduce the floc size and only the surplus energy above this threshold is used to break the cells and enable the release of organic substances into the bulk liquid (Bougrier et al., 2005). For higher specific energies, a continuous increase in COD solubilisation with the increase in specific energy input is observed. Khanal et al. (2007) suggested that an energy input of 35000 kJ/kg TS suffices for a maximum sludge solubilisation (3% TS) and that further sludge solubilisation becomes increasingly difficult at applied energies greater than 35000 kJ/kg TS.

Wang et al. (2006) reported the increase in COD solubilisation from 52 mg/L to 2581 mg/L, 7509 mg/L and 8912 mg/L for 5, 15 and 20 min of sonication, respectively, at an ultrasonic power of 0.77 W/mL. High ultrasonic power generates higher mechanical shear forces during cavitation bubble implosion (Grönroos et al., 2005), which caused higher degradation of sludge floc and higher release of soluble COD at constant treatment time (Koksoy and Sanin, 2010).

Mao et al. (2004), observed an increase in COD solubilisation by the factor of 1.2, 2.3 and 4.8 at an applied power of 2, 3 and 4 W/mL, respectively. Wang et al. (2010) suggested that below a critical level, only EPS are solubilized, while some fraction of cellular mass is also solubilized at sonication power above this critical level.

Tyagi et al. (2013) observed that the improvement in the rate of COD solubilisation was directly proportional to the increase in ultrasonic intensity.

(Neis et al., 2000) observed more than 2 fold increase in the rate of sludge solubilisation by increasing the sonication intensity from 6 W/cm^2 to 18 W/cm^2. Nevertheless, Tyagi et al. (2013) asserted that it is difficult to standardize the rate of sludge solubilisation at specific power input on the basis of available studies, due to the different treatment conditions applied in each study.

(Mao et al., 2004) studied the effect of sludge types (primary and secondary) on the sludge solubilisation by sonication. They observed 4 and 7.7 fold increase in COD solubilisation for primary and secondary sludge, respectively, after 20 min of sonication at 4 W/mL. Their study confirmed that primary sludge is more easily solubilized than secondary sludge.

(Wang et al., 1999) noticed that the soluble protein concentration increased from 50 to 1200, 3000, 5200, and 6000 mg/L, at sonication durations of 0 (control), 10, 20, 30, and 40 minutes, respectively. The protein concentration was reported to increase with increasing specific energy input (Akin et al., 2006). The release of ammonia-N concentration increased with an increase in specific energy inputs and TS concentration, as is the case for SCOD increase (Khanal et al., 2007).

2.6.7.5. Ultrasonication pretreatment and Sludge dewaterability

High energy ultrasonication treatment can disrupt flocs and increase the number of fine particles and bound water. Hence, low energy sonication is recommended. Low ultrasonication results in disruption of flocs which will refloccculate to tighter particles when flocculation agents are applied. The optimization of the ultrasonication parameter are essential for successful outcomes (Huan et al., 2009)

Water content in bio-sludge is commonly about 80–90% wt after dewatering process. The EPS and the form of water in sludge influence the structure of sludge. Adding cationic fluctuations can change the form of water in sludge and increase the flow during the dewatering process, but has little influence on the final water content (Yin et al., 2004)

2.6.7.6 Effect on sludge morphology

The mechanical shear forces generated by sonication are capable to significantly decrease the compactness of sludge by damaging the floc bridging and convert the aggregated sludge floc into micro-flocs (Jiang et al., 2011, Jiang et al., 2009).

Higher sonication time leads to the significant disruption of sludge flocs and cell membrane. Cao et al. (2006) reported that the floc binding strength became weaker after 1 min sonication time. The structural integrity was broken down after 10 min sonication time, and the flocs were completely disrupted after 30 min. Khanal et al.

(2007) reported that the floc structures were entangled within a large numbers of filaments before sonication.

The change in sludge morphology is directly dependent on the amount of ultrasonic energy input. Gradual disruption in floc structure of sludge will take place with increasing the specific energy input (26000 kJ/ kg TS) (Feng et al., 2009).

Sludge disintegration produced small flocs and dispersed cells at specific energy input of 2500 kJ/kg TS. However, only dispersed cells were observed at higher energy input of 5500 kJ/kg TS (Braguglia et al., 2008).

Chang et al. (2011) reported that the complex and non-uniform floc structure of WAS changed into more uniform smaller sizes when increasing the treatment time (from 10 to 30 min) and applied density (from 1.2 W/L to 2.4 W/L). The effect of different TS concentration on the morphology of sonicated sludge (0.86 W/L, 4 min) was studied by Akin et al. (2006). They observed almost complete disintegration of structural integrity of sludge floc for both 2 and 4% TS concentration.

2.6.8 Microwave pretreatment

Microwaves are oscillating electromagnetic energy with frequencies in the 300 MHz to 300 GHz range with the most effective range for dielectric heating between 0.915 and 2.45 GHz (Leonelli and Mason, 2010).

Most of the interactions between microwaves and materials that have chemical nature which induces electric polarization and re-orientation phenomena. The extent of change of electromagnetic energy into thermal energy is known to be dependent in practical terms on the permittivity, ε^*, which is a complex number, i.e. having real and imaginary parts, as described by the Eq. (2.5):

$$\varepsilon^* = \varepsilon^* + i\varepsilon \qquad (2.5)$$

ε', the dielectric constant, represents the ability of a material to be polarized by an external electric field and so it is a relative measure of the microwave energy density. This is often expressed relative to the permittivity of free space, ε_0, by Eq. (2.6) (Pozar, 1998):

$$\varepsilon' = \varepsilon_r \varepsilon_0 \qquad (2.6)$$

‘ε’,the loss factor, quantifies the efficiency with which the electromagnetic energy is converted to heat (Metaxas, 1996). Usually the losses due to the induction of real currents, i.e. the contribution of the electrical conductivity to heat generation is included in the effective loss factor. Sometimes a linear combination of dielectric constant and loss factor is used to account for the losses, using the loss tangent, tan δ, which is the ratio between the dissipative (including electrical conductivity losses) and capacitive behavior of the materials, according to the simplified equation below Eq.(2.7) (Ulaby, 2001):

$$\tan\delta = \frac{\varepsilon''}{\varepsilon'} \qquad (2.7)$$

The value of tan δ is then easily related to the capacity of the materials to be heated, the higher the better.

Materials are classified according to their characteristics when exposed to Microwave radiation (Coelho, 2012a). The materials can be:

Absorbers – if they absorb a great amount of the energy irradiated. An example of an absorber material is water. These materials have high dielectric constants.

Transparent – if they do not absorb energy. An example of this type of material is glass. These materials have very low dielectric constants.

Reflectors – if they reflect the waves that are applied to them. No absorption or transmission occurs in these materials. An example is metals.

Microwave ovens are generally composed of six components, the MW cavity, turntable, magnetron (the device that generates the MWs), wave guide (that directs the waves to the MW cavity), mode stirrer (that distributes the waves inside the MW cavity) and circulator (that directs the lost energy to a dummy load to protect the magnetron). Figure 2.5 represents simplified representation of the components in a microwave processing system.

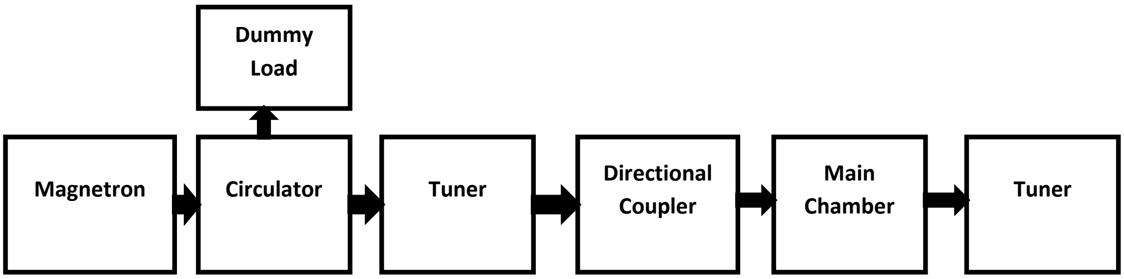


Figure 2.5 Block diagram of microwave processing unit (A. C. Metaxas, 1983).

When MWs are adequately used, heating can be accomplished in shorter time and more economically when compared with conventional heating. Some of the advantages of MW heating compared to conventional heating are ((A. C. Metaxas, 1983);

MW heating is used in many industries, besides its usual use in domestic households. It has been used in the food industry (baking, thawing, pasteurization, and drying), and in the medical industry (sterilization) among other areas (Hong, 2002).

2.6.8.1 Microwave pretreatment effects on sludge solubilization

MWs was applied to primary and WAS prior to anaerobic digestion obtaining high degrees of solubilization. For the WAS, approximately 46% of the non-soluble COD was solubilized after irradiation. For the case of primary sludge, this increase was only 12%. The pretreatment consisted in microwaving the sludge to a temperature of 60ºC (Pino-Jelcic et al., 2006).

The effect on the digestion of the sludge was measured in semicontinuous reactors with a SRT of 25 days. An increase of the biogas production of 16.4% was achieved compared to the control and of 6.3% as compared to sludge heated to the same temperature but using conventional heating. The MW heating also showed a higher inactivation of pathogenic microorganisms than sludge pretreated thermally by the conventional way (Eskicioglu et al., 2007a)

(Eskicioglu et al., 2008a) investigated the effects of MW intensity, temperature and sludge concentration on the solubilization of WAS (taken from an activated sludge unit operating at 5 d SRT). It was reported that the MW intensity had a positive effect on the solubilization of the COD but negligible effect on the biogas production of the irradiated samples. However, sludge concentration and temperature did show an influence on both parameters. The sludge irradiated at 96ºC had a greater production of biogas than the sludge irradiated at 75ºC and this sludge in turn produced more biogas than the sludge irradiated at 50ºC.

The sludge pretreated to 96ºC showed an increase of 20% in biogas production compared to the control in the essays at 3% total solids (TS). For the assays at 1.4% TS the increase in biogas production was 15%. A differentiated effect in the solubilization was reported for samples pretreated with MW and conventional heating for the same temperature, with a greater fraction of total COD being solubilized by the conventional heating, a fact that was attributed to the longer time conventional heating requires to reach the same final temperature. Microwave pretreatment can help to achieve rapid heating by orienting the water molecules in the direction of the microwave energy, whereas heat transfer by conventional heating involves transfer of energy from one molecule to another molecule as shown in Figure 2.6.

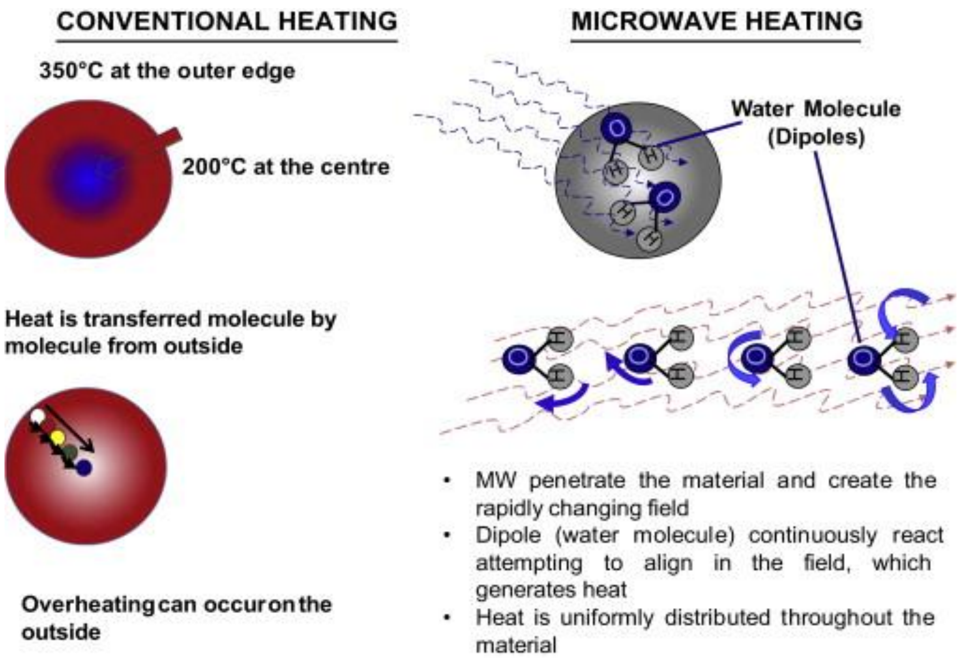


Figure 2.6 Differences between conventional heating and microwave heating (Tyagi and Lo, 2013).

Microwave treatment was more cost effective as compared to conventional thermal treatment (Park, 2011). MW treatment resulted in pathogen destruction as well as thermal versus non-thermal effects (Eskicioglu et al., 2007c). Microwave (MW) involves high frequency electromagnetic radiation which interacts with the dipolar molecules in the sludge (Eskicioglu et al., 2006). Microwave pretreatment helps to enhance rate of anaerobic digestion and dewaterability (Eskicioglu et al., 2007b). Microwave pretreatment increased SCOD up to 4 fold, soluble protein concentration up to 1.8 fold and soluble carbohydrate concentration up to 14 fold (Zhou et al., 2010).

MW treatment was applied to achieve higher WAS floc and cell destruction and release of Extracellular polymeric substances and intracellular materials into the soluble phase compared to conventional heating, which in effect increased soluble CODs and biogas production(Saha et al., 2011a). The use of MWs in the digestion of sludge was found to increase the ratio of soluble COD to total COD (sCOD/tCOD) from 2 to 22% (Toreci et al., 2010).

Thibault (2005) tested MW pretreatment of combined primary/WAS sequencing batch reactor sludge (15d SRT) and reported that applying MWs to 85ºC improved the biogas production by 16.2%. Multiple irradiation cycles to the same temperature did not improve results. The maximum sCOD/tCOD achieved in the tests using MW pretreatment was 7%.

Since water is the most abundant element in biomass, most ovens produce waves in the frequency of 2.45 GHz which is a frequency where water molecules absorb a large amount of energy, but still allow some to pass, in order to provide heating that is not limited to the surface in large samples. In this way, the heating is generated by the friction caused by rapid oscillation of water molecules, and the energy absorbed by the food is very high (A. C. Metaxas, 1983).

Microwave irradiation combined with alkali pretreatment increases biodegradability of thickened activated sludge. The degree of substrate solubilization was 18 times higher in pretreated sludge (53.2%) than in raw sludge (3.0%). Improvements in biogas production compared with the control increased as HRT was reduced to 5 days (205% higher at 5 days). Dewaterability of digested sludge deteriorated as compared to the control(Jang and Ahn, 2013a).

The use of MW irradiation in combination with other chemical methods (hybrid treatment) has been shown to synergistically enhance the efficiency of the whole process in terms of improve- ment in COD and the solubilisation of solids, which consequently enhance the digestion performance in terms of higher organics removal and biogas production. MW irradiation can be used to facilitate the recovery of valuable products from sludge, such as orthophosphate, ammonia, metals and bio-fuels. Thus, due to the synergistic effects, hybrid pretreatments can provide a more effective and economical solution compared to individual MW pretreatment methods for sludge treatment (Tyagi and Lo, 2013a).

2.6.8.2 Microwave pretreatment and Sludge dewaterability

Generally heating is known to improve sludge dewaterability. Microwave as a rapid heating method showed that significant improvement in dewaterability can be achieved after microwave pretreatment. (Wojciechowska, 2005) applied microwave irradiation for sludge conditioning and observed 73% and 84% decrease in specific resistance to filtration (SRF) of mixed sludge and anaerobically digested sludge, respectively. Microwave pretreated sludge showed better dewaterability than conventionally heated and non-pretreated sludge. Microwave pretreated sludge showed significant improvement of 17.6% and 13.8% in dewatering rates in comparison to control and conventionally heated digested sludge

Anaerobically digested sludge pretreated at 96^0C provided 40% improvement in dewaterability (by capillary suction time, CST). A significant reduction from 181 second to158 second was obtained for the CST. Furthermore, 75% improvement was reported in the dewaterability of anaerobically digested sludge that was pretreated at 175^0C (Eskicioglu et al, 2007).

2.6.8.3 Mechanism of heating and cell disruption in microwave pretreatment

Since water is the most abundant element in sludge biomass, most ovens produce waves with frequency of 2.45 GHz where water molecules absorb a large amount of energy and allow passage of part of the energy which provides heating that is not limited to the surface in large samples. Heat is produced by the friction caused by rapid oscillation of water molecules, and the energy absorbed by the biomass is very high as shown in Figure 2.7 (A. C. Metaxas, 1983).

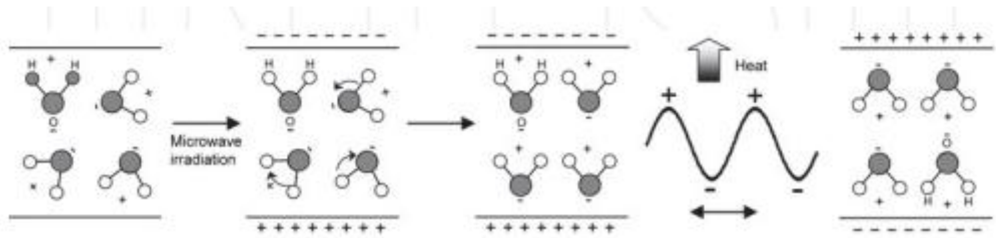


Figure 2.7 Heating mechanism of water due to microwave field (Tsuji, 2005).

The increase in the soluble chemical oxygen demand (SCOD) concentration also indicates a significant disruption of complex WAS floc structures and the release of extracellular and intracellular biopolymers (proteins and sugars) from activated sludge flocs to the soluble phase as shown in Figure 2.8 (Eskicioglu et al., 2006). Park et al. (2006) Observed 19% and 22% increases in SCOD concentration after MW pretreatment of WAS (TS-3%) at 91.2^{0}C and boiling temperature, respectively.

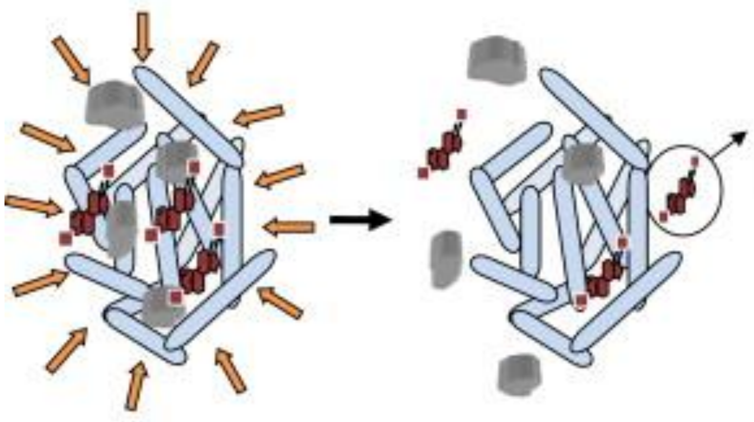


Figure 2.8 Application of microwave to sludge floc (Hamid and Eskicioglu, 2013).

2.6.9 Combined treatment techniques and their effect

Many published literature show that temperature, ozone dose, Ultrasonic energy density and pH have a beneficial impact on the disintegration of sludge not only when they are applied independently but also when they are systematically combined. The released COD levels were higher with combined pretreatment than with ultrasonic or alkaline pretreatment alone for waste activated sludge samples (Xu et al., 2010b).

Thermal treatment alone did not increase solids destruction significantly. A maximum of 15.2% increase in volatile suspended solids (VSS) destruction was observed with the oxidative treatment. A synergistic effect was observed when both treatments were combined. The increase in VSS destruction when both pretreatment techniques were applied ranged between 27.2 and 29.0%, depending on the reactors configuration (Remya, 2011) .

Thermo-oxidative treatment at low pH is important in terms of the dewaterability and color generation of digested sludge. Based on evaluation of the overall performance, thermo-oxidative treatment with acid is considered the best among the thermo-oxidative treatments examined (Takashima and Tanaka, 2008).

A microwave-enhanced advanced hydrogen peroxide oxidation process (MW/H_2O_2-AOP) was investigated to understand the synergistic effects of MW irradiation on H_2O_2 treated waste activated sludges (WAS) in terms of mineralization (perm anent stabilization), sludge disintegration/solubilization, and subsequent anaerobic biodegradation as well as dewaterability after digestion. The combined treatment enhanced organic oxidation and solubilization of particulate COD (>0.45 micron) of WAS indicating that a synergetic effect was observed when both H_2O_2 and MW treatments were combined. But the combined treatment had somehow made the kinetic of the process slower that that achieved by microwave treatment (Eskicioglu et al., 2008b).

Depending on the pollutants to be eliminated, the combination of advanced oxidation processes such as ozonation with ultrasound or an integrated ultrasonic/biological treatment can significantly improve process efficiency and economy.

Thermal pre-treatment was better than sonication or ozonation concerning sludge solubilisation. Better solubilizaiton was obtained for ultrasounds with energy of 6250 or 9350 kJ/kg TS and a thermal treatment at 170 or 190^0C. Moreover, pretreatment had effects on physicochemical characteristics of sludge samples: apparent viscosity decreased after all treatments but the reduction was more significant with thermal treatment. Median diameter of sludge flocs were reduced after sonication, increased after thermal treatment and did not change after ozonation. Finally, capillary suction time (CST) increased after ozonation, increased highly after sonication and was reduced after thermal treatment (Bougrier et al., 2006).

anaerobic digestion with thermo chemical pretreatment, resulted in total chemical oxygen demand (TCOD) reduction, volatile solid (VS) reduction, methane yield and methane biogas content of 88.9%, 77.5%, 0.52m^3/kg VS and 79.5%, respectively. These results help to determine the best hydrolysis pretreatment process for anaerobic digestion and in improving the design and operation of the large-scale treatment of WAS by anaerobic digestion with hydrolysis systems (Park et al., 2005).

the effects of various pretreatment methods (thermal, chemical, ultrasonic and thermochemical pretreatments) on the biogas production and pollutants reduction owing to solubilization enhancement, particle size reduction, increased soluble protein, and increased soluble COD. Thermo-chemical pretreatment gave the best results, *i.e.*, the production of methane increased by more than 34.3% and soluble COD (SCOD) removal also increased by more than 67.8% over the control. In this case, the biogas production, methane production and the SCOD removal efficiency were about 5037L biogas/m^3 WAS, 3367L methane/m^3 WAS and 61.4%, respectively. Therefore, it is recognized that higher digestion efficiencies of the WAS were obtained through thermochemical pretreatment of the sludge (Kim et al., 2003).

The performance of thermophilic treatment was evaluated in terms of a number of parameters that included organic removal rate (ORR) (kgVS/m^3/d and kgCOD/m^3/d), biogas and volumetric methane production rate (m^3/m^3/d), pH, total acidity (mg acetic acid/L) and acidity/alkalinity relationship. At thermophilic conditions (55°C), the OLR studied was 1.48 kgVS/m^3d (SRT: 27 days), and under these conditions the solids destruction efficiency was 53.3% VS, and the biogas produced in the digester reached 0.32 m^3/m^3(Zhao and Viraraghavan, 2004).

Enhancing, the anaerobic digestion of waste activated sludge (WAS) by the combination of temperature-phased two-stage digestion and intermediate ozonation was investigated by a continuous experiment with two processes, which consists of a thermophilic digester (55^0C), an ozone treatment reactor and a mesophilic digester (35^0C) in series. The digested sludge from thermophilic digester was ozonized in batch in an ozone treatment reactor with 5L volume. Two processes were operated at hydraulic retention times of 30 days for over 123 days. Waste activated sludge taken from wastewater treatment plant was fed as a substrate.

In the temperature phased two-stage digestion, the combination of higher rate of hydrolysis by thermophilic digestion and the decrease of soluble COD by secondary mesophilic digester resulted in higher COD reduction with improvement of the flocculation efficiency and water quality of dewatered sludge compared to the thermophilic digestion.

In spite of less amount of ozone dose in the intermediate ozonation than ozone pre-treatment, the intermediate ozonation had better effect of ozonation on performance improvement in terms of COD reduction than ozone pre-treatment.

Application of thermo-oxidative treatment resulted in the Lowering the pH of thermo-oxidative treatment is advantageous with respect to the dewaterability and color generation of digested sludge

Thermochemical treatment was found to increase volatile solids destruction from 15.2 % for individual treatment to 27.2 and 29.0%, for combined treatment depending on the reactors configuration. Unfortunately, economic balance of this application is unfavourable due to the high energy requirement for heating a low-concentrated sludge and the increased reagent dosage (Vlyssides and Karlis, 2004)

After thermochemical pretreatment, the methane production was 2.2 times higher rate than for the control sludge without any pre-treatment (Tanaka and Kamiyama, 2002).

Microwave-enhanced advanced hydrogen peroxide oxidation process (MW/H_2O_2-AOP) at Elevated MW temperatures (>80^0C) increased the decomposition of H_2O_2 into OH^- radicals and enhanced both oxidation and particulate COD disintegration of WAS samples. MW/H_2O_2-AOP generated soluble organics were slower to biodegrade or more refractory than those generated during MW irradiation. (Eskicioglu et al., 2008a). Enhanced Enzymatic Hydrolysis increased VS destruction by around 10% and biogas production by 24% (Wong, 2006).

A synergetic effect was observed when hydrodynamic cavitation was combined with alkaline treatment in which NaOH, KOH, and $Ca(OH)_2$ were used as alkaline catalysts at pH ranging from 8 to 13. As expected, the production yield of CH_4 gas increased proportionally as WAS disintegration proceeded. HC, when combined with alkaline pretreatment, was found to be a cost-effective substitute to conventional methods for WAS pretreatment (Lee and Han, 2013).

Thermo-alkaline pretreatment with prehydrolysis at 90^0C and pH 11 followed by anaerobic digestion of WAS has several advantages over conventional methods. This pretreatment resulted in a reduction of the initial VSS by about 46% and a methane production of 0.28L of methane per kg of initial VSS loading (Xu et al., 2010c).

Ultrasonic-oxidative pretreatment by a combination of Ultrasound and Ozone at optimum Temperature, O_3 dose, ultrasonic energy density and pH had a positive effect on the disintegration of sludge. The SCOD increased from 1821 to 2513 mg/l after reaction for 30 min when $NaHCO_3$ was added, which indicated that the ozone molecule played a major role in the disintegration of waste activated sludge (Cesaro and Belgiorno, 2013).

2.6.10 Combined microwave ultrasonic pretreatment

Table 2.4 illustrates that simultaneous microwave and ultrasonic irradiation overcomes the inertness of most esters and dramatically reduces reaction times (79–94% yield within 20–65 seconds). As these transformations occur under heterogeneous conditions, acceleration was interpreted in terms of enhanced heat and mass transfer. Cavitation causes liquid jets to hit the interface and the mutual injection of droplets results in fine emulsions (Cravotto and Cintas, 2007) **.**

Table2.4 Combined microwave ultrasonic irradiation for treatment of esters (Lagha et al., 1999).

Method	Time	Yield (%)
Reflux	9 hour	73
US(50W)+reflux	1.5 hour	79
MW(200W)	18 minute	80
MW+US	40 seconds	84

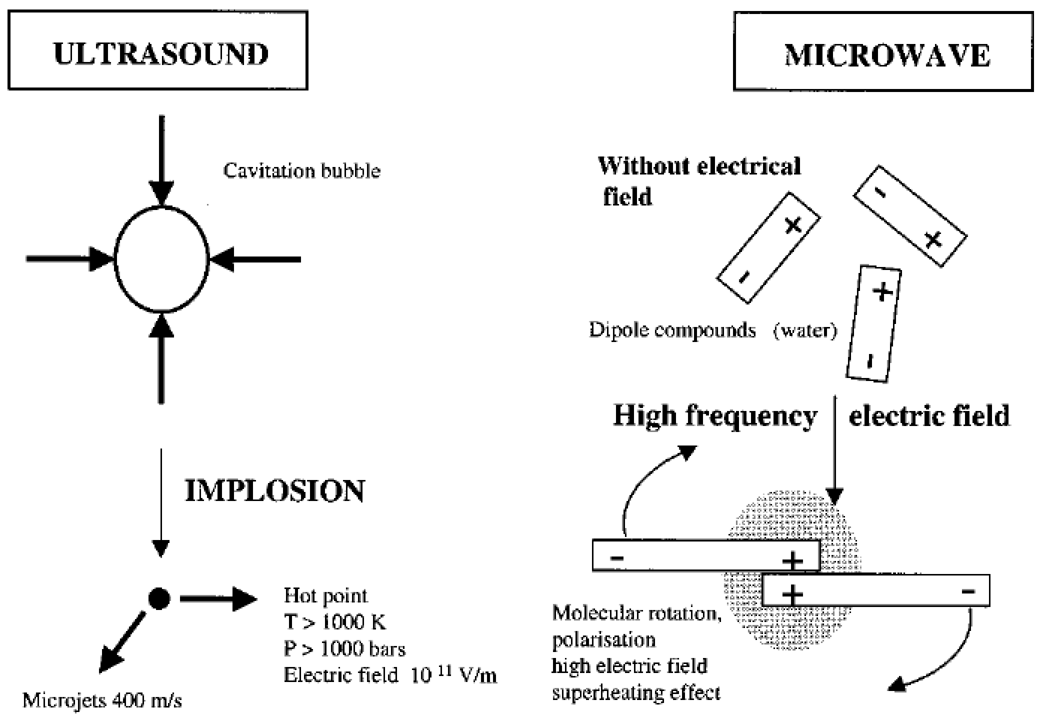


Figure 2.9 Combined microwave ultrasonic irradiation for treatment of esters (Lagha et al., 1999).

The analogy between the cavitation mode of ultrasonic pretreatment and heating mode of microwave pretreatment is given in Figure 2.9. Overall picture of flow reactors uniquely adapted for combined application of microwave and ultrasonic irradiation is given in Figure 2.10. Details of the tubing and pumping system are given in in Figure 2.11.

Figure 2.10 Overall picture of a flow reactor combining MW and US irradiation in a sequential mode (Cravotto and Cintas, 2007).

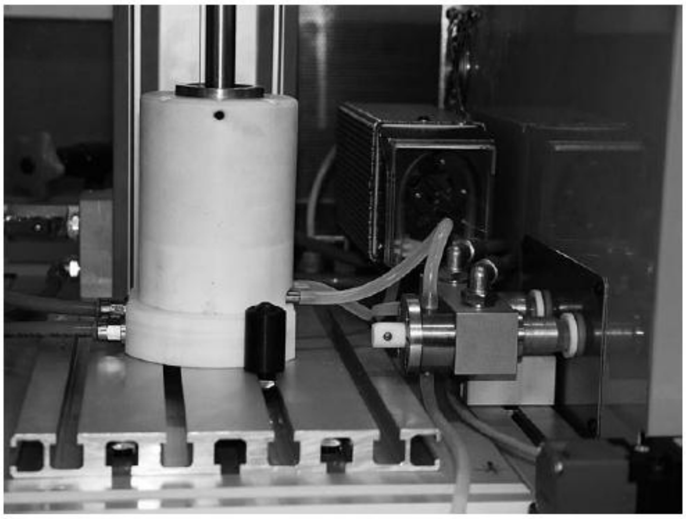


Figure 2.11 Details of the peristaltic pump and connecting tubing in a flow reactor combining MW and US irradiation (Cravotto and Cintas, 2007).

An observation was made on the conjoint application of ultrasound and microwaves that “This combination of energy sources can promote or improve a number of chemical processes such as synthesis, extraction of natural products and sample preparation in chemical analysis (Cravotto and Cintas, 2007).

(Cravotto and Cintas, 2007) discussed that combination of microwave with ultrasonic in one system is a problem as placing a metal ultrasonic horn inside a microwave zone is hazardous. There are two ways to address this problem (i) Using separate reactors one using ultrasound and another using MW with a recirculating pump to allow the liquid to be transferred from one reactor to another. (ii) or Using a single reactor with both ultrasonic and microwave units inside. The combination of microwaves with ultrasonic treatment has great potential since the two activation modes are different and provide real advantages in terms of improved heat and mass transfer. Both microwave irradiation and ultrasound definitely meet the process intensification rules through the improvement of energy transfer, the reduction of energy consumption, the reduced volumes of reactors/plants, the improved product quality, the ease of process automation as well as remote control. It can be can conclude that both methods of activation will have significant applications in many areas.

2.7 Effect of other process parameters on anaerobic digestion performance

Operation of the anaerobic digestion process requires stringent control and optimization of loading rate, (organic and hydraulic), mixing, carbon:nitrogen ratio, volatile and total solid content, pH, temperature, concentration of volatile acids, hydrogen and ammonia, for safety and better enhancement of the gas generation and solid reduction capacity of the system (Mata-Alvarez et al., 2000).

2.7.1 Effect of Temperature

Temperature inside the digester has a major effect on the biogas production process. There are different temperature ranges during which anaerobic digestion can be carried out, psychrophilic (<30^0C), mesophilic (30–40^0C) and thermophilic (50–60^0C) are common temperature conditions. However, anaerobes are most active in the mesophilic and thermophilic temperature range (Saha et al., 2011a). Angelidaki and Ahring (1994) observed that when the NH_3 load was high, reducing temperature below 55^0C resulted in an increase of biogas yield and better process stability with reduced VFA concentration. The production of ammonia could be marginally reduced by operating a thermal hydrolytic pretreatment process at 150^oC. The effect of temperature on the hydrolysis of proteins, lipids, and polysaccharides, both in pure form and as part of the macromolecular makeup of primary and secondary wastewater sludge, has been studied. (Wilson and Novak, 2009).

2.7.2 Ammonia

High ammonia content can be very toxic to the microrganisms and contributes to the reduction in methane production. Ammonia concentration tends to be high in protein rich sludge (cattle dung) and excessive ammonia inhibits anaerobic digestion process (Khalid et al, 2011). A study on high-paper municipal and industrial waste showed that the lowest final ammonia nitrogen concentration relate to the highest production of methane (Poggi-Varaldo et al., 1997). Ammonification of protein by thermal hydrolysis may be an important consideration in the event that methanogenic inhibition becomes apparent. Additional ammonia produced from WAS correlates well to its higher total protein content relative to PS. Solids loading rate may provide a more meaningful control against ammonia induced methanogenic inhibition (Wilson and Novak, 2009).

2.7.3 Effect of pH

pH is an important parameter affecting the growth of microorganisms during anaerobic digestion. pH of the digester should be kept within the desired range of 6.8–7.2 by controlling loading rate. The amount of carbon dioxide and volatile fatty acids produced during the anaerobic digestion process affects the pH in the digester.

Jain and Mattiasson (1998) found that above pH 5.0, the efficiency of CH_4 production was more than 75%. It is possible to increase gas yield and reduce retention time by addition of inoculum (Sreekrishnan et al., 2004). Neutralization of pH enhances the development of microbial activity and the humification process seems greatly influenced by means of neutralization. In the case of neutralization by lime, the intense oxidation of organic compounds occurred and humification involved polyphenol condensation. Neutralization of pH by phosphate could be considered the best treatment that allows good stabilisation of organic matter and high preservation of nitrogen in humic form (Hafidi et al., 2005)

The conversion in methanogenic aggregates, information on the pH gradients and the pH dependency of the growth is indispensable. Batch experiments showed that acetate uptake by aggregates was not coupled directly to methanogenesis. Consumed acetate was not converted instantaneously to methane, suggesting the conversion to proceed via a pool of acetate or reserve material (Beer et al., 1991).

2.7.4 Volatile fatty acid (VFA)

Anaerobic fermentation can proceed normally when concentration of volatile fatty acids, acetic acid in particular is below 2000 mg/l. VFA are needed in small amount as part of the intermediary step in the methanogenic cycle, and accumulation of VFA can lead to drop in pH which results in digester inhibition. A continual drop in pH leads to failure of the digester (Carucci et al., 2005).

2.7.5 Organic loading rate (OLR)

Organic loading rate significantly affects the biogas yield and the performance of anaerobic digesters (De la Rubia et al., 2006). When OLR was varied from 346 kg VS/day to 1030 kg VS/day, gas yield increased from 67 to 202 m^3/day. There is an optimum sludge feed rate for a particular size of plant that produces highest amount of gas beyond which further increase in the amount of feed sludge will not result in proportional increase in gas production. Anaerobic digestion of primary sludge was found to be feasible with organic loading rates (OLR) of 1–1.4 kgVS/m^3d and hydraulic retention times (HRT) of 16–32 d resulting in methane yields of 190–240 m^3CH4/t VS fed. Also the highest tested OLR of 2 kgVS/m^3d and the shortest HRT of 14–16 d could be feasible, if pH stability is confirmed (Sreekrishnan et al., 2004).

2.7.6 Hydraulic retention time (HRT)

It is possible to carry out methanogenic fermentation at low HRT’s without stressing the fermentation process at mesophilic and thermophilic temperature ranges (Zennaki et al., 1996).

At 20 day HRT, reduction of volatile solids and COD was higher for microwave pretreatment and thermal pretreatment than the untreated sample. The VS and COD removal improved with decreasing HRT. The greatest improvement occurred for HRT of 5 days with 29% improvement for microwave pretreated and 14% improvement of the thermally pretreated sample as compared to the control . The COD removal was 53% and greater for microwave pretreated sludge and 38% better for the thermally treated one. This finding shows how increased OLR affects the performance of the untreated sludge (Park and Ahn, 2011b).

2.7.7 Solid concentration

The sludge disintegration efficiency of WAS by the ultrasonic sound is increased with decreasing TS content. However the anaerobic digestibility efficiency is increased with increasing TS content. Therefore the TS contents should be optimized by considering both the disintegration efficiency and the anaerobic digestibility efficiency.

The amount of fermentable volume of slurry is defined as solid concentration. Ordinarily 7–9% solids concentration is best-suited (Zennaki et al., 1996). The biogas yield increased, reaching 0.46 $m^3/(m^3$ day) at 37^0C and 0.68 $m^3/(m^3$ day) at 55^0C respectively.

Baserj (1984) reported that anaerobic digestion became unstable below a total solids level of 7% (of manure) while a level of 10% caused an overloading of the fermenter.

2.8 Anaerobic digester modelling studies

Anaerobic digestion modelling is an established method for assessing anaerobic wastewater treatment for design, systems analysis, operational analysis, and control. Anaerobic treatment of domestic wastewater is a relatively new, but rapidly maturing technology, for the advantage of low cost, and moderate-good performance it provides (Husain, 1998).

To model the whole biogas digestion process based on biological and physico-chemical background, the kinetics of bacterial growth, substrate degradation and product formation have to be considered (Gerber and Span, 2008).

2.8.1 Steady state models

It is desirable to operate digesters at steady state conditions. Unstable conditions arising from stress of the biological population lead to a reduction in methane production. Stress of the biological population may occur as a result of short residence time leading to bacterial wash-out, inhibition by high VFA levels or toxicity due to high ammonia concentrations.

The kinetics of bacterial growth, substrate degradation and product formation have to be considered, to model the whole biogas digestion process based on biological and physico-chemical background, (Gerber and Span, 2008).

The primary indexes used to evaluate digester performance include pH, TS, VS, TCOD, SCOD, ammonia-nitrogen, methane production, and methane content of the produced biogas (Komatsu et al., 2007).

Much simpler model structures may be required for design and hydraulic. Other applications such as model-based process control require a minimalist, model, with defined structural elements (Bernard et al., 2001).

2.8.2 Models for complex wastewater

Complex model structures are ideal for complex process analysis, complex models can either be minimalist or inclusive.

Minimalist refers to a model the minimum number of steps required for a specific purpose. These mainly encompass control models such as (Bernard et al. 2001) or instrument development.

Inclusive are those that include all processes and components found in a specific or even complex wastewater. This category can also include simplified inclusive models, in which several steps have been lumped. This is separate from minimalist, since the structure of minimalist models is often based on numerical considerations.

Decay rate and uptake rate are usually comparable. When the values for both are low the robustness of the kinetic models won't be efficient There are now indications that high decay rates are more valid, based on observations from continuous mixed systems (Batstone et al., 2002b).

To model the whole biogas digestion process based on biological and physico-chemical background, the kinetics of bacterial growth, substrate degradation and product formation have to be considered (Gerber and Span, 2008).

The sludge disintegration efficiency of waste activated sludge by the ultrasonic sound is increased with decreasing TS content. However the anaerobic digestibility efficiency is increased with increasing TS content. Therefore the TS contents should be optimized by considering both the disintegration efficiency and the anaerobic digestibility efficiency. The primary parameters used to evaluate digester performance include pH, TS, VS, TCOD, SCOD, ammonia-nitrogen, methane production, and methane content of the produced biogas (Komatsu et al., 2007).

Much simpler model structures may be required for design and hydraulic. Other applications such as model-based process control require a minimalist, model, with defined structural elements (Bernard et al., 2001).

2.9 Anaerobic digestion modelling using ANFIS

Fundamentals of neural fuzzy modelling for anaerobic wastewater treatment systems have been presented comprehensively by (Tay and Zhang, 2000). Thus, the introduction focuses on the architecture and implementation of the conceptual model for brevity. As the model development is based on the conceptual neural fuzzy model developed by (Tay and Zhang, 1998), the conceptual neural fuzzy model is briefly introduced to provide a better understanding of the development of the model.

Basic concepts and methodology of fuzzy logic theory, neural network technology and their integration can be found elsewhere, such as (Lin and Lee, 1996).

Tay and Zhang developed a conceptual neural fuzzy model for three different high-rate anaerobic treatment configurations. Their model predicted the volumetric methane production (VMP), TOC and VFA with high accuracy using OLR, hydraulic loading rate, alkalinity loading rate, and VMP, TOC and VFA prediction of the previous day as the input parameters.

ANFIS models were used to predict effluent VS concentration and methane yield in the anaerobic digester fed with pre-thickened primary sedimentation sludge. They found out that the model results showed good agreement with real WWTP data between observed and predicted values. The applicability of the model is simple and does not require complex reactions and due to highly nonlinear structure of the ANFIS model, it was shown that a complex system such as anaerobic digestion could be easily modelled. (Cakmakci, 2007)

As ANFIS demonstrated its ability to construct any nonlinear function with multiple inputs and outputs in many applications, its estimating performance was investigated for a complex wastewater treatment process at increasing organic loading rates from 1.1 to 5.5 g COD/L d. Approximation of the ANFIS models was validated using correlation coefficient, MAPE and RMSE. ANFIS was successful to model unsteady data for pH and COD within anaerobic digestion limits with multiple input structure (Erdirencelebi and Yalpir, 2011). The details of the structure of the ANFIS model is provided in chapter 10.

As the adaptive capability of the neural network and reasoning ability of the fuzzy logic are combined in ANFIS modeling, It was implemented in need of a fast responsive and flexible model to a highly complex anaerobic treatment process. The ANFIS models developed were successful in predicting the effluent parameters of pH and COD within anaerobic digestion limits at an OLR range of 1.1–5.5 g COD/L d. (Perendeci et al., 2008)

It was proved that steady conditions at a large OLR range can be modeled with its structure and used in the controlling of an anaerobic reactor’s influent pH and COD in high-strength dairy wastewaters where input parameters usually occur at a highly fluctuating level due to dense acidification reactions in the influent. Enlarging of the database and/or frequency of monitoring will serve to reduce the error level and improve the predicting capability of the model.

On-line and off-line monitoring of the influent pH and COD, respectively, will enable the regulation of the COD concentration in the influent using the proposed model. As ANFIS can be trained with new data or seasonal changes, the control system based on the model can be adapted or updated continuously by the user, providing a great potential for application in the controlling of anaerobic digesters.

Anaerobic process must be monitored and controlled to avoid instability. Extensive modelling of anaerobic process is complex as many biochemical reactions occur inside the anaerobic digester.

Therefore, models are required to analyse detailed descriptions of anaerobic process, such as modelling of methane production volatile solid and COD reduction, alkalinity and VFA changes. Anaerobic digestion being a biological process, modelling of this process is complex. Hence, a simplified modelling tool is essential to understand the interaction between input and output parameters.

2.10 Conclusion

- Anaerobic digestion performance enhancement can be achieved by improving slow rate-limiting hydrolysis step. Biogas production, solid reduction, dewaterability, pathogen removal and process kinetics can be enhanced using different pretreatment technologies. Ultrasonic, microwave, chemical, thermal, mechanical and biological pretreatment techniques can be applied to increase digester performance as individually or in combination. The combination of two pretreatment techniques (thermo-oxidative, thermal-alkaline, ultrasonic-oxidative, microwave-oxidative, microwave-ultrasonic) have superiority over individual pretreatment options as the combination improves the digester performance highly compared to the individual techniques.

- Rapid heating and heat penetration effects, energy efficiency, non-contact heating, Athermal effects, selectivity, space saving, flexibility and many more other benefits make microwave pretreatment an interesting candidate for the selection in the combination study.

- Ultrasonic pretreatment has significant effects on physical chemical and biological properties of sludge. It results in improvement of solid removal, biogas production and process kinetics. It has been applied at industrial scale. But the process requires further enhancement through optimization of factors like ultrasonic density, duration of treatment, intensity and specific energy input against the effects on process. Ultrasonication coupled with other pretreatment techniques enhances its effect on anaerobic digestion process.

- Combined microwave-ultrasonic pretreatment is the noble pretreatment combination applied for the enhancement of anaerobic digestion of municipal sewage sludge in this study. The enhanced performance obtained in combined microwave-ultrasonic pretreatment in organic synthesis and extraction and the benefit of other combined pretreatment techniques over individual pretreatment options for enhanced sludge solubilisation and biogas production are the bases for this research work. The combination of microwaves with ultrasonic treatment has great potential since the two activation modes are different and provide real advantages in terms of improved heat and mass transfer.

- Primary sludge, excess activated sludge and mixed sludge have distinctively different biochemical composition, rheological property, response to pretreatment, biodegradability and methane potential, floc size and dewaterability. Studying effect of pretreatment technologies and biodegradability of each of the sludge types is beneficial for the selection of appropriate pretreatment technology and pretreatment condition, better design and operation of digesters (Zhang, 2010).

- All the required pretreatment and digester operational parameters have been experimentally analysed in this study. The synergistic improvement effect of the combined microwave-ultrasonic pretreatment has been compared to microwave alone and ultrasonic alone pretreatment options. The pretreatment options require optimization for efficient performance. The impacts on biogas production, solid removal, pathogen removal, dewaterability, process kinetics (SRT, HRT) should be assessed. Digester operational parameters should also be optimized accounting for the pretreatment. This research addresses these gaps using experimental and modelling techniques. Kinetics models were used to analyse the kinetics parameters and adaptive neural network inference model was used to analyse plant operational data.

CHAPTER 3

METHODOLOGY AND ANALYTICAL TECHNIQUES

3.1 Introduction

This research on the enhancement of municipal sludge anaerobic digestion performance by pretreatment technology combines both experimental and modelling techniques. The experimental work was conducted both on synthetic sludge prepared in the laboratory from sources that simulate municipal sewage sludge and real sewage sludge from Beenyup Waste Water Treatment Plant (BWWTP). The experimental work on synthetic sludge was conducted before the tests on real sludge to understand the effect of various pretreatment technologies on anaerobic digestion performance, to select the best pretreatment technology and identify the optimum pretreatment and digestion conditions.

The municipal sewage sludge samples used throughout the research were collected from BWWTP. Raw primary sludge, excess activated sludge, thickened excess activated sludge, mixed sludge and digested sludge samples from BWWTP were characterized and subjected to different microwave and ultrasonic pretreatment conditions. Optimum pretreatment conditions for all sludge types were determined from sludge solubilisation and anaerobic digestion tests.

This chapter comprises all the experimental work performed to determine the best pretreatment technology and anaerobic digestion conditions. The detailed experimental procedures and methods are discussed in the respective specific chapters. Brief methodology section at the end discusses the modelling techniques involved in this research work. The overall structure of the experimental work in the research is schematically represented in Figure 3.1. Characterization of all sludge samples was initially conducted as shown in stage1; the characterized samples were pretreated ultrasonically or subjected to microwave irradiation and a combination of these techniques and the optimum pretreatment conditions were determined based on the impact of the pretreatment on sludge solublization biogas production and characteristics of the digested sludge produced (stage 2). Sludge samples subjected to optimum pretreatment conditions were anaerobically digested and the digester performance was investigated under different operational conditions (stage 3). The kinetics of anaerobic digestion for the experimental studies was investigated and optimum operational conditions were predicted using ANFIS for a historical data from BWWTP (stage 4).

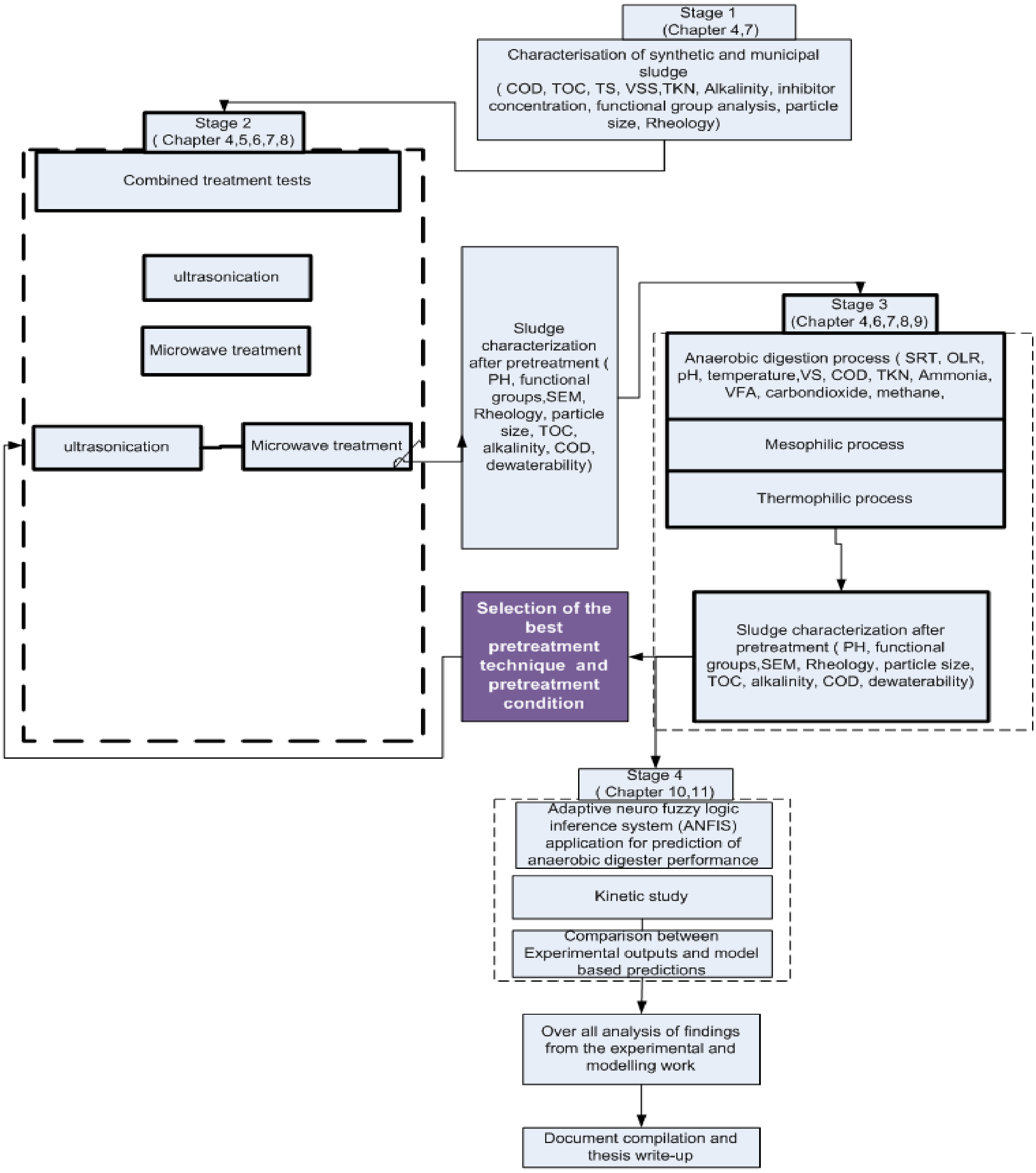


Figure 3.1: Overall schematics of the research work

3.2 Process description of Beenyup Wastewater Treatment Plant (BWWTP)

Beenyup Waste Water Treatment Plant (BWWTP) is one of the four major waste water treatment facilities in Perth, Western Australia. This wastewater treatment plant has a treatment capacity of 135 million litres of sewage sludge per day. Currently, the treatment plant serves 660,000 inhabitants in northern suburbs from Quinn's Beach to Scarborough and inland to Dianella and Bayswater to the foothills east of Midland. The wastewater in the plant originates mainly from household kitchens, bathrooms, toilets and laundries. Wastewater flowing into Beenyup is more than 99 per cent water. Most of the treated wastewater is discharged to the ocean. (Corporation, 2009)

3.2.1 History of Beenyup waste water treatment plant

Historically, small local treatment plants used to serve early sub-divisions in the northern suburbs. The establishment of BWWTP in 1970 enabled centralized treatment at a larger facility. The first stage of the permanent plant started operation with a capacity of 3.6 million litres a day. This plant utilised the extended aeration process. The plant was upgraded to treat 27 million litres a day using the conventional activated sludge process in 1978. Also at this time a gravity outfall system was commissioned which enabled the treated effluent to be discharged into the Indian Ocean off Ocean Reef. Incineration technology was employed for sludge disposal.

Further upgrades were commissioned in 1984 to enable the plant to treat 54 million litres a day. The sludge digestion facilities were commissioned in 1990 replacing the sludge incineration process. New secondary treatment technology became operational in 1996 with a capacity of 112.5 million litres a day. State-of-the-art odour control and further facility enhancements were completed in 2005 to increase the plant's capacity to 120 million litres a day. In 2007 the treatment capacity of BWWTP increased to 135 million litres a day. The plant will soon be upgraded to a capacity of 200 million litres/day to serve 1.1 million people (Corporation, 2009).

3.2.2 Preliminary treatment

Municipal wastewater flows into the BWWTP from three main sewers sources that combine at the inlet channel. First, screenings process with five step screens (with

6mm openings) removes large material such as rags and plastics from the inflow. The removed material named as screenings, is later washed and compacted ready for disposal to an engineered landfill site. After screening, inorganic material (grit) settle in grit removal tanks while the organic material stays suspended in the wastewater. Water is drained from the settled grit by a screw conveyor, then the grit is washed and sent to an approved landfill site, together with the screenings (Corporation, 2009).

3.2.3 Primary treatment

The waste water then enters the primary treatment process after preliminary treatment. The primary treatment process consists of six rectangular tanks and a raw sludge pumping system. The wastewater remains in the tank sufficiently long enough until 90 per cent of the solids settle down to the floor of the tanks while the oil and grease floats to the top of the tanks. Mechanical scrapers push the settled solids to a hopper at the inlet end of the tank and the oil and grease is collected at the opposite end of the tank. The settled solids are pumped to the sludge treatment area while the oil and grease is sent back to the head of the plant where it is absorbed onto the rags during the screening process and slowly removed (Corporation, 2009).

3.2.4 Sludge treatment process

The thickened excess activated sludge is mixed with raw sludge from the primary sedimentation tanks with raw sludge: thickened excess activated sludge ratio of 3:1 and transferred to a two-stage heated anaerobic digestion process. The digesters involve biological treatment through bacterial action followed by dewatering in centrifuges. The sludge is converted into a residue (biosolids) that is an excellent soil conditioner for agricultural use. Methane gas produced in the digestion process is used to provide the fuel for the digester’s heating and mixing requirements. Any excess methane is burnt off through a waste gas burner at $750^{o}C$ to destroy any odorous gases. A portion of the biosolids produced in this process is transported by trucks to agricultural areas where it is applied, under strict guidelines, to paddocks for use as soil improver. The remainder of the biosolids produced is used as an ingredient in commercial compost for landscaping (Corporation, 2009).

3.2.5 Odor removal

Odorous gases collected from the covered parts of the plant are discharged to chemical scrubbing towers for treatment. These chemical scrubbing towers remove the hydrogen sulphide and other odorous gases from the extracted air and release the treated air to the atmosphere through a 50-metre high stack. The height of the stack ensures good dispersal and dilution of any residual odours. Bio filters and activated carbon adsorption processes remove any remaining malodours gases which were not removed in the chemical scrubbing towers (Corporation, 2009).

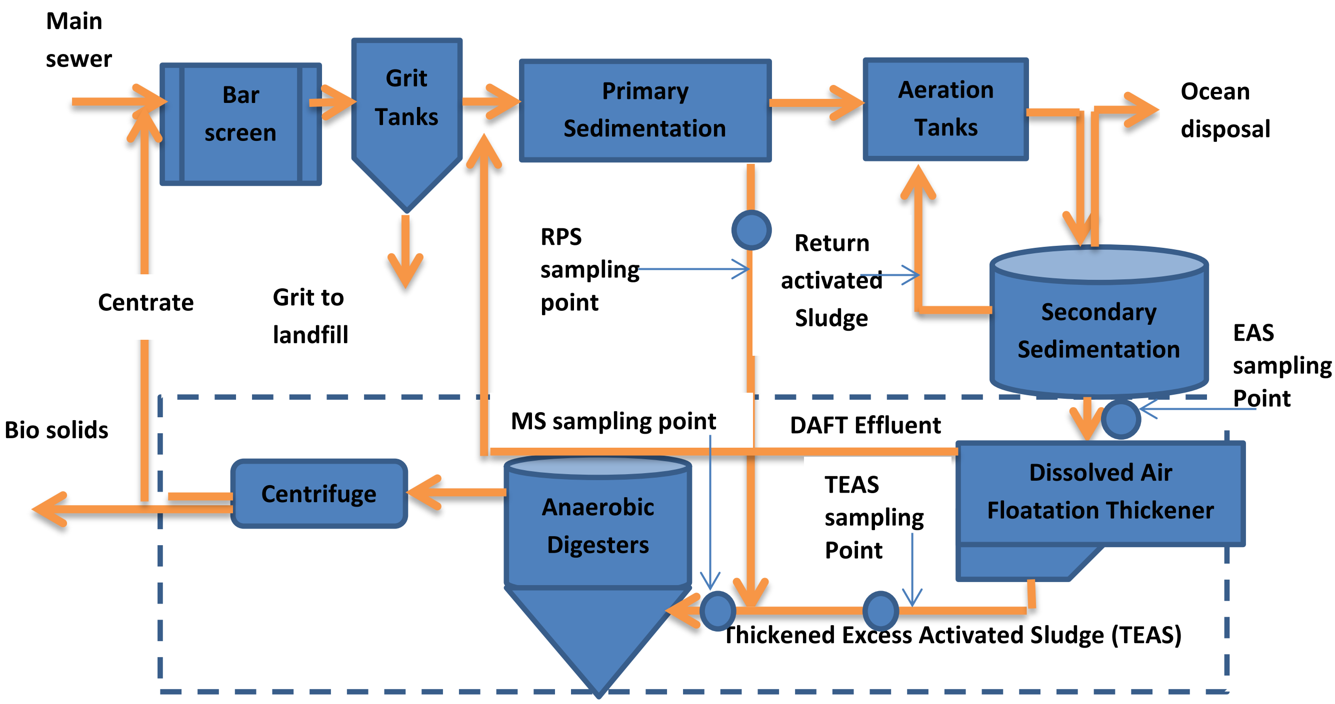


Figure 3.2 Process flow for municipal sewer treatment in BWWTP (this research focuses in the section enclosed in broken lines, the small circles shows the sludge sampling points.)

3.3 Sample collection and characterization

Primary, excess activated sludge, thickened excess activated sludge, and digested sludge samples were collected from BWWTP at the sampling points indicated in Figures 3.2-3.5 in all the experimental work carried out in this research. All sludge samples were characterized based on the physico-chemical and biological parameters including pH, COD, TS, VSS, functional group analysis, particle size, rheology etc. Such characteristic parameters were measured using the equipment and the methods described in Table 3.1. The pre-treatment tests were conducted, after determination of such characteristic parameters.

Table 3.1: parameters for sludge characterization and anaerobic digester performance tests.

Test parameter	**Instrument (equipment) or method of measurement**
COD	Oxidation with COD reagent and colorimetric analysis on ORION UV/Vis spectrometer
NH_4-N	Ion-selective probe
VFA (acetic, butyric or propionic acid)	GC with Flame ionization detector
Hydrogen sulphide	Gas meter
Methane	ThermoFisher SCIENTIFIC GA 2000 plus gas analyzer
Oxygen	GA 2000 plus gas analyzer
Carbondioxide	GA 2000 plus gas analyzer
Dewaterability (capillary suction time)	Type 304 CST equipment
Gas volume	Gas displacement technique and Wetgas meter.
pH	pH meter
VS,TS	Standard method (APHA et al., 2005)
VSS	Standard method (APHA et al., 2005)
Rheology	Rheometer
Particle size distribution	Mastersizer 2000
Functional group analysis	Fourier transform infrared spectroscopic technique (FTIR)
Sludge morphology and microbial structure	Scanning Electro-microscope (SEM)
Protein,	Bradford method

3.3.1 Primary sludge sampling

There are 6 Primary sludge sedimentations tanks. The raw primary sludge was collected from Primary Sedimentation Tank (PST) no. 4 based on the recommendation of the engineers and operators at BWWTP to get a representative raw primary sludge. The sludge sample was well sealed and stored in the refrigerator at a temperature less than 4^0C. Fresh samples were always collected and utilized throughout the course of the research.

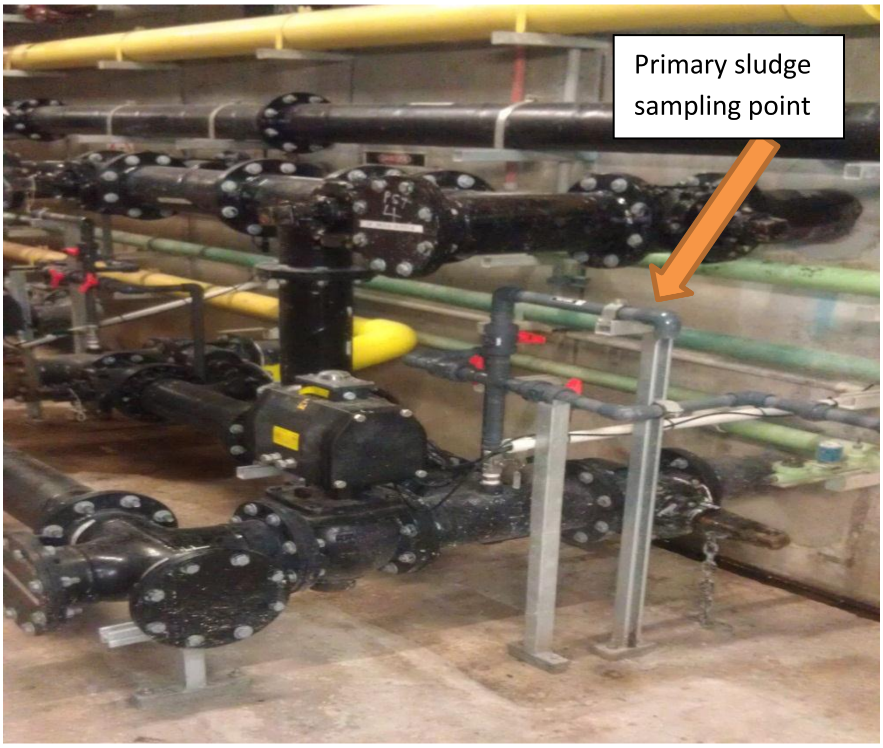


Figure 3.3: Primary sludge sampling point in the primary Gallery (BWWTP)

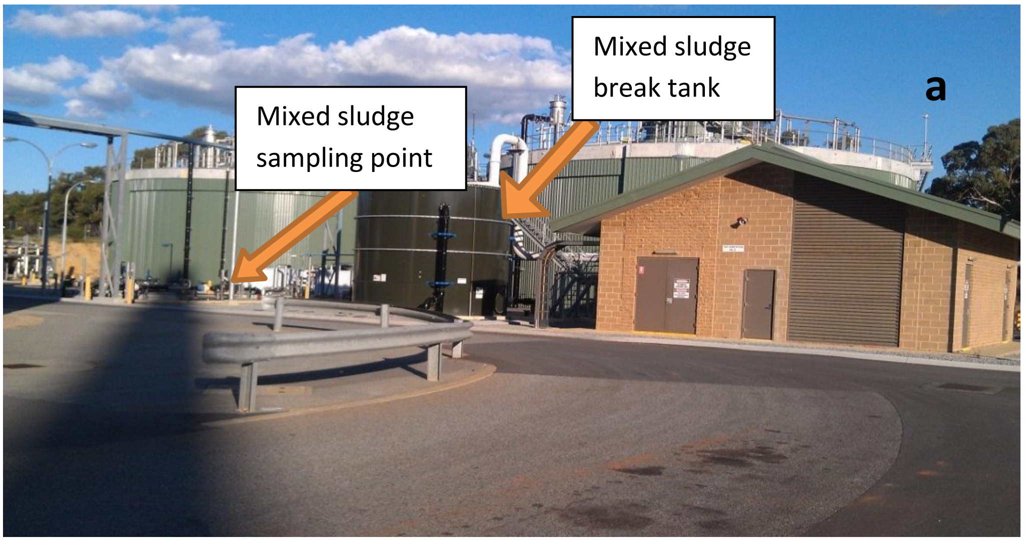
3.3.2 Excess activated and thickened excess activated sludge sampling

Excess activated sludge was collected from module 4 before thickening in the Dissolved Air Floatation Unit (DAFT). Thickened excess activated sludge was collected from the discharge of the DAFT before the mixing of thickened excess activated sludge with primary sludge for the anaerobic digestion process.

Figure 3.4: Thickened activated sludge sampling point at the DAFT units

3.3.3 Mixed sludge sampling

Mixed sludge sample was collected from the mixed sludge sampling point after the sludge break tank where the mixing of the thickened excess activated sludge and Raw Primary Sludge (RPS) takes place. The mixed sludge is composed of RPS and TEAS with a ratio of 3:1 or 2:1.



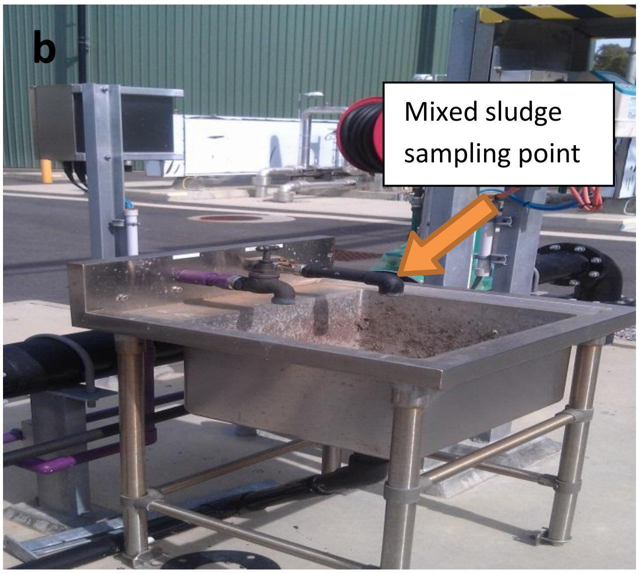


Figure 3.5: (a) Mixed sludge sampling area (b) close-up picture of mixed sludge sampling point

3.3.4 Digested sludge sampling

Digested sludge sample was collected from centrifuge number 2 in the dewatering section. The digested sludge sample collected from this spot was mostly used for inoculation of digesters and characterization tests. The digested sludge was stored in the refrigerator under 4^0C like the other sludge samples.

Figure 3.6: Digested sludge sampling point (centrifuge Number 2) in the dewatering section of BWWTP.

3.4 Sludge characterisation, analytical and instrumental methods

3.4.1 pH

pH was measured with WP-90 and WP-81 conductivity/TDS-pH/temperature meter equipped with a glass electrode according to Standard Methods (Federation, 2000). pH was measured before and after pretreatment and during the anaerobic digestion process on regular basis. pH measurement was performed immediately to minimize contact of the sample with air. During the biochemical methane potential (BMP) assay, pH of fresh samples was measured immediately after the sample was taken. The electrode was rinsed with distilled water before each measurement. The temperature was maintained constant during the measurement of pH and the equipment was calibrated periodically using buffer solutions at pH 4 and 9 (Eskicioglu et al., 2006).

3.4.2 Total and soluble chemical oxygen demand

Total and soluble chemical oxygen demand were determined by using oxidation method with HACH COD reagent and colorimetric analysis on ORION UV/Vis spectrometer from Cole Parmer. Total chemical oxygen demand was measured by taking 1 ml of representative sample measured in a micro pipette and diluting it in 50 ml of distilled water. 2 ml of each sample was transferred to each HACH-COD vial and the mixture was thoroughly homogenized and all the vials were heated in the COD- reactor (digester) for 2 hours at 150^0C. The COD vials were cooled and COD was determined using ORION UV/Vis spectrophotometer designed for this specific purpose. Soluble chemical oxygen demand was determined by centrifuging the sample at 5000 rpm for 10 min to separate the supernatant from the solid sludge and filtering the supernant in Whatman (45 μm) filter paper. The COD measurement was then conducted on the filtrate exactly in the same way as total COD (Park et al., 2004).

3.4.3 Ammonia-Nitrogen

Standard Method 4500 NH_3-F APHA 2000 was employed to measure the dissolved ammonia (NH_3(aq) and N-H) concentration. Equipment used for the measurement was an ammonia electrode model 95-12 and WP-90 and WP-81 conductivity/TDS-pH/temperature meter. 25 or 40 mL samples (depending on availability) were placed in an 80mL beaker with the ammonia electrode. One ml of 10N sodium hydroxide was added into the sample to raise the pH value to above 11 and release the ionic ammonia into free (gas) ammonia prior to measurement. The electrode was inserted into the sample to confirm that the pH has reached 11. Calibration curve was prepared prior to ammonia concentration reading of samples to verify proper electrode operation. The standard ammonia concentration points chosen were 10, 100, and 1000 mg/L based on the range of ammonia concentration in the samples examined (Saha et al., 2011a).

3.4.4 Total and volatile solids

The total and volatile solids content were determined according to Standard Methods for the Examination of Water and Wastewater. For the determination of the total and volatile solids, sludge samples were first dried at 105°C and then the residue was ignited at 550°C until the weight of the samples becomes constant. Evaporating dish of 100 ml, muffle furnace, oven, desiccator, analytical balance (capable of weighing to 0.1 mg), magnetic stirrer, glass-fiber disks without organic binder (Whatman grade 934AJ etc), filtration apparatus and drying oven were used for the tests on solid content (Federation, 2000).

Procedure:

I. Total Solids (TS):

- Ignite clean evaporating dish at 550 ± 50°C for 1 h in a muffle furnace.
- Cool in desiccator, weigh (B).
- Store in desiccator until ready for use.
- Choose a sample volume that will yield a residue between 2.5 mg and 200 mg, put in a beaker and stir using a magnetic stirrer.
- Pour to the prepared evaporating dish, and weigh (C).
- Evaporate to dryness in an oven at 98°C. If necessary, add successive sample portions to the same dish after evaporation.
- Continue drying at 103 to 105°C for 1 h, cool to balance temperature in an individual desiccator containing fresh desiccant, and weigh (A).
- Repeat cycling of drying, cooling, desiccating, and weighing until a constant weight is obtained or until weight loss is < 4% of previous weight or 0.5 mg, whichever is less. This residue is known as Residue A.

II. Total Volatile Solids (VS):

- Have furnace up to temperature before inserting sample (usually 15 to 20 min ignition are required).
- Transfer Residue A above to the furnace at 550 ± 50oC, and ignite for 1 h.
- Let dish or filter disk cool partially in air until most of the heat has been dissipated.
- Cool in desiccator to balance temperature and weigh (D).
- Repeat cycle of igniting, cooling, desiccating, and weighing until weight loss < 4% or previous weight.

Calculation:

$$\%\ \text{Total Solids} = \frac{(A-B)\times 100}{C-B} \qquad \text{(Equation 3.1)}$$

$$\%\ \text{Volatile Solids} = \frac{(A-D)\times 100}{A-B} \qquad \text{(Equation 3.2)}$$

Where:

A = weight of dried residue + dish, mg,

B = weight of dish,

C = weight of wet sample + dish, mg, and

D = weight of residue + dish after ignition, mg.

III. Total Dissolved Solids (TDS):

- Insert disk with wrinkled side up into filtration apparatus. Apply vacuum and wash disk with three successive 20-ml volumes of distilled water. Continue suction to remove all traces of water. Discard washings.
- Ignite cleaned evaporating dish at 550 ± 50oC for 1 h in a muffle furnace. Store in desiccators until needed. Weigh immediately before use (B).
- Choose sample volume to yield between 2.5 and 200 mg dried residue.
- Filter measured volume of well-mixed sample through glass-fiber filter, wash with three successive 10-ml volumes of distilled water, allowing complete drainage between washings, and continue suction for about 3 min after filtration is complete. If more than 10 min are required to complete filtration, increase filter size or decrease sample volume but do not produce less than 2.5 mg residue.

- Transfer filtrate to a weighed evaporating dish and evaporate to dryness in an oven at 98^{0}C. Add successive portions to the same dish after evaporation if necessary.
- Continue drying for at least 1 h in an oven at 180 ± 2^{o}C, cool in desiccators to balance temperature and weigh.
- Repeat drying cycle of drying, cooling, desiccating, and weighing until a constant weight is obtained or until weight loss is less than 4% of previous weight or 0.5 mg, whichever is less (A).

Calculation:

$$\text{mg Total Dissolved Solids/L} = \frac{(A-B)\times 1000}{\text{Sample volume,ml}} \qquad \text{(Equation 3.3)}$$

Where:

A = weight of dried residue + dish, mg,

B = weight of dish, mg.

3.4.5 Alkalinity

Bicarbonate alkalinity was measured as alkalinity according to Standard Method (Federation, 2000). pH value change was measured with WP-90 and WP-81 conductivity/TDS-pH/temperature meter with the electrode of the pH meter inserted in the sample during the titration. The quantity of acid needed to reach pH of 4.5 was recorded. The titration endpoint of pH 8.3 was not tested as all samples presented a pH value below 8.3. The electrode of the pH meter was stored in a large volume of distilled water and thoroughly rinsed with distilled water before each use (Ahn et al., 2009).

3.4.6 Elemental analysis

The elemental composition (carbon, hydrogen, nitrogen and sulphur content) of primary sludge, excess activated sludge, mixed and digested sludge was analysed using micro elemental analyser. Sludge samples were dried at 105^{0}C for 2hrs and desiccated overnight before the analysis. The sludge sample mass used for the measurement was less than 2 mg for all the sludge samples. The percentage composition of total carbon, total hydrogen, total Nitrogen and sulphur were determined for all the sludge types collected from BWWTP.

3.4.7 Measurement of biogas composition (CH_4/CO_2/O_2/NH_3/H_2S)

The biogas composition was measured using ThermoFisher SCIENTIFIC GA 2000 plus. The Gas meter is designed to measure, volume percentage of methane, carbon dioxide, oxygen and other gases. The Gas analysis was conducted by pumping the biogas online from the digester into the gas analyser at a rate of 8 ml/s. The analyser is equipped with internal suction pump making it useful for online measurement. The concentration of ammonia and hydrogen sulphide was monitored by the meter in addition to the other components of the biogas. Gas Chromatographic technique was used to confirm the accuracy and consistency of the biogas composition measured using GA 2000 plus biogas analyser.

3.4.8 Temperature

Temperature measurement was conducted using WP-90 and WP-81 conductivity/TDS-pH/temperature meter during all analytical techniques to ensure consistency of the results. Temperature during the digestion process was maintained constant using the water bath heater which pumps the water flowing in the jacket of the digesters. Mesophilic (36-37 ^{0}C) and thermophilic (55^{0}C) anaerobic digestion conditions were tested in this research (Yeneneh et al., 2013b).

3.4.9 Particle size analysis

The particle size distribution of feed, intermediate and digested sludge samples was determined using Malvern Mastersizer 2000 ® Laser Diffraction Particle Size Analyser. The instrument uses lazer diffraction technique to quantify particle diameter as d(0.1), d(0.5) and d(0.9) values which indicate that 10%, 50% and 90% of the particles measured were less than or equal to the sizes stated respectively. It utilizes dual-wavelength detection system. A short wavelength blue light source is used in conjunction with forward and backscatter detection. The sludge samples were exposed to He–Ne laser and a refractive index of 1.58 was used for the sludge test. Surface weighted and volume weighted mean diameters were also determined. The average surface area (m^2/g) was also measured using this technique. The particle size data relates well to the mass transfer rate and sludge dewaterability (Yu et al., 2009).

3.4.10 Rheological measurement

In this study, the rheology of raw primary, thickened excess activated and mixed sludge were studied for different microwave-ultrasonic pretreatment conditions. Homogenised samples of feed, intermediate and digested sludge were subjected to rheological measurement on HAAKE MARS Rheometer from Thermo SCIENTIFIC for the rheological tests during anaerobic digestion (Eshtiaghi et al., 2013). The shear stress versus shear rate and viscosity versus shear rate curves were plotted for raw untreated and microwave-ultrasonic pretreated sludge samples at various pretreatment conditions. The effect of solid concentration and temperature on the rheological properties of different types of sludge was also investigated (Civelekoglu and Kalkan, 2010).

3.4.11 Microbial content

Microbial content was measured using Coliscan Kit. Coliscan kit incorporates two special chromogenic substrates which interact with the enzymes galactosidase and glucuronidase to produce pigments of contrasting colours. The presence and number of coliforms and E. coli can be determined by counting. General coliforms produce the enzyme galactosidase and the colonies that grow in the medium have a pink colour. E. coli produces both galactosidase and glucuronidase and therefore grows as dark blue to purple colonies in the medium. It is simple to count the blue/purple colonies (E. coli) which indicate the number of E. coli per sample. The pink colonies indicate the number of general coliforms per sample. The combined general coliform and E. coli number equals the total coliform number. Any non-coloured colonies which grow in the medium are not coliforms, but may be members of the family Enterobacteriaceae. Since the Coliscan contains inhibitors, most other bacterial types will not grow. The bacterial count method applied in this research involves dilution of 1ml of sludge sample from digesters in 50 ml of distilled water. 1 ml of each diluted sample was then mixed with the coliscan easygel solution and the easygel-sludge mixtures were allowed to set on petridish. The cultures were kept at room temperature (25-30^{o}C) for 48 hours before the coliform and Ecoli colonies were counted. The microbial content in 100ml sample was predicted from this count (Tune and Elmore, 2009).

3.4.12 Total protein

Bio-Rad Assay was used for the determination of total protein concentration in the hydrolysis and methanogenesis stages. One part of the Bio-Rad reagent was mixed with 4 parts of ultra-pure water. Standard solution of gamma bovine serum (IgG) was prepared in the range of 0.2 mg/ml to 1.5 mg/ml. Sludge samples withdrawn at different SRT from digesters were diluted 50 times. 200 microliter of four standard protein samples and the unknown diluted sludge samples were mixed with 5 ml of the Bio-Rad reagent. After thorough mixing the samples were analysed on UV/Vis spectrophotometer. Calibration curve was first developed using the known standards of IgG (Pervaiz and Sain, 2012).

3.4.13 Dewaterability

The dewaterability of the different sludge samples was measured using capillary suction timer (Type 304 CST equipment). Samples were placed at room temperature for 1-2 hours before the test to ensure sample temperatures were 20-25°C for all testing. The CST paper was placed between contacting sensors and a stainless steel funnel (hollow cylinder) was placed on top of the sensors. 3-5 mL of sample was slowly introduced to the funnel. The time required for sludge water to flow from the first sensor to the second sensor determines the dewaterability as CST in seconds (Yuan et al., 2011b).

3.4.14 Optical Microscope and Scanning Electro-Microscope imaging

Optical Microscope (Olympus LG-PS2) and Scanning Electron Microscope (Philips XL30) with magnification of 20,000- 30,000 times were used for the sludge of sludge floc size shape and morphology as a function of microwave and ultrasonic Pretreatment and anaerobic digestion process(Yuan et al., 2011a).

3.4.15 Fourier Transform Infrared Spectroscopy (FTIR)

The essential characteristic functional groups and the chemical alteration that happened in the course of the digestion process were qualitatively analysed using

Fourier Transform Infrared Spectroscopy (FTIR) (Perkin Elmer Spectrometer 100) (Imam et al., 1995).

3.5 Pretreatment of Sludge Samples (equipment and techniques)

3.5.1 Microwave pretreatment

Microwaves are electromagnetic energy. In the electromagnetic spectrum, microwave radiation occurs in an area of transition between infrared radiation and radio frequency waves. A frequency of 2450 MHz is mostly used for the microwave unit to avoid interference with other equipment and appliances. This frequency of 2450 MHz is the cause of alignment of molecules followed by returning to disorder ultimately resulting in very fast heating (Mehdizadeh et al., 2013). The typical MW instrument used for heating has six major parts: MW generator (magnetron), waves guide, MW cavity, mode stirrer, a circulator and a turntable. MW energy is produced by the magnetron, propagated by the wave guide and injected into the MW cavity where the mode stirrer distributes the incoming energy in different directions. MWs are effectively reflected by the metallic walls and form standing waves (Eskicioglu et al., 2008a, Park, 2011, Saha et al., 2011a, Toreci et al., 2011). Samples were subjected to microwave pretreatment at different pretreatment time, power and density.

3.5.2 Ultrasonication

The ultrasonication unit utilized was SONICs digital ultrasonication unit with titanium tip. This equipment can deliver a maximum power of 500 Watts at a frequency of 20 kHz. The stack equipment of ultrasonic processor used had a 3-16 μmp-p converter, a 3:1 gain booster and a 2:1 gain probe of 2.54 cm (1 in) diameter. The amplitude could thus be modulated from 6 to 90 %. The ultrasonication chamber used for batch operation was a common borosilicate 250 ml and 500ml glass beaker. The lowest 3 cm of the probe was immersed in the solution. The pulse during all ultrasonication tests was 55/5. This depth was enough to avoid air introduction and scum formation in the media (according to sounds produced and visual observations) which would reduce the acoustic transmission and enhances ultrasonication efficiency. Besides, this depth was shallow enough to allow the entire sample to be mixed by acoustic streaming and cavitation. The diameter of the beakers (approximately 7 cm for 250 ml beaker and 13 cm for 500 ml beaker) allowed the half wave length (around 12.7 cm at 20 kHz) to be fully created in this containers. The diameter of the beaker was selected to avoid the introduction of unwanted wall effects. The effects of change in ultrasonic density, ultrasonic intensity and ultrasonic duration (pretreatment time) were investigated (Apul and Sanin, 2010a, Bougrier et al., 2005b, Muller et al., 2009, Park et al.).

3.5.3 Combined Microwave-Ultrasonic pretreatment

The innovative combined pre-treatments were performed in experimental conditions described in Table 3.2. The sequence of pretreatment was in the order of first microwave followed by ultrasonic pretreatment. The optimum pretreatment condition for the two techniques was selected after thoroughly investigating effect of pretreatment power, time, density and intensity for both pretreatment techniques (Lagha et al., 1999, Yeneneh et al., 2013b).

Table 3.2: Pre-treatment types and experimental conditions

Type of pre-treatment	**Test conditions and ranges**	**Anaerobic reactor type**
Ultrasonic	0.3 - 1W/ml, 20 KHZ, 50-150W, 4-12 min	Batch CSTR, HRT<= 20days
Microwave treatment	80W- 800w , 2450 MHz, 1-5 min	Batch CSTR, HRT < =20days
Combined Microwave-ultrasonic treatment	Microwave followed by ultrasonic pretreatment in the test ranges shown for each of the pretreatment techniques.	Batch CSTR, HRT<= 20days

The samples were characterized after conducting all the pre-treatment tests and the best pre-treatment conditions which resulted in better improvement of sludge characteristics and with better gas generation, solubilisation, sludge reduction were chosen and used for further anaerobic digestion tests. The digesters used for anaerobic digestion experiment were designed to operate simultaneously. All jacketed digesters were supplied with hot water flowing through the jackets to maintain mesophilic working temperature (36-37 ^{0}C) and magnetic stirrers for agitation. All the digesters have sludge feeding and withdrawal ports and gas-line extends to the inverted cylinders where the biogas is collected for volume measurement. Buffer bottles between the inverted cylinders and the reactor prevent water and condensable matter from entering the cylinder. Nitrogen gas was used to purge air out of the reactors before inoculation. All reactors were inoculated 2 to 5 days before the sludge substrate is introduced. Tube outlets were designed near the buffer bottles for gas composition analysis purposes. The overall reactor setup is shown in Figure 3.6 and 3.7.

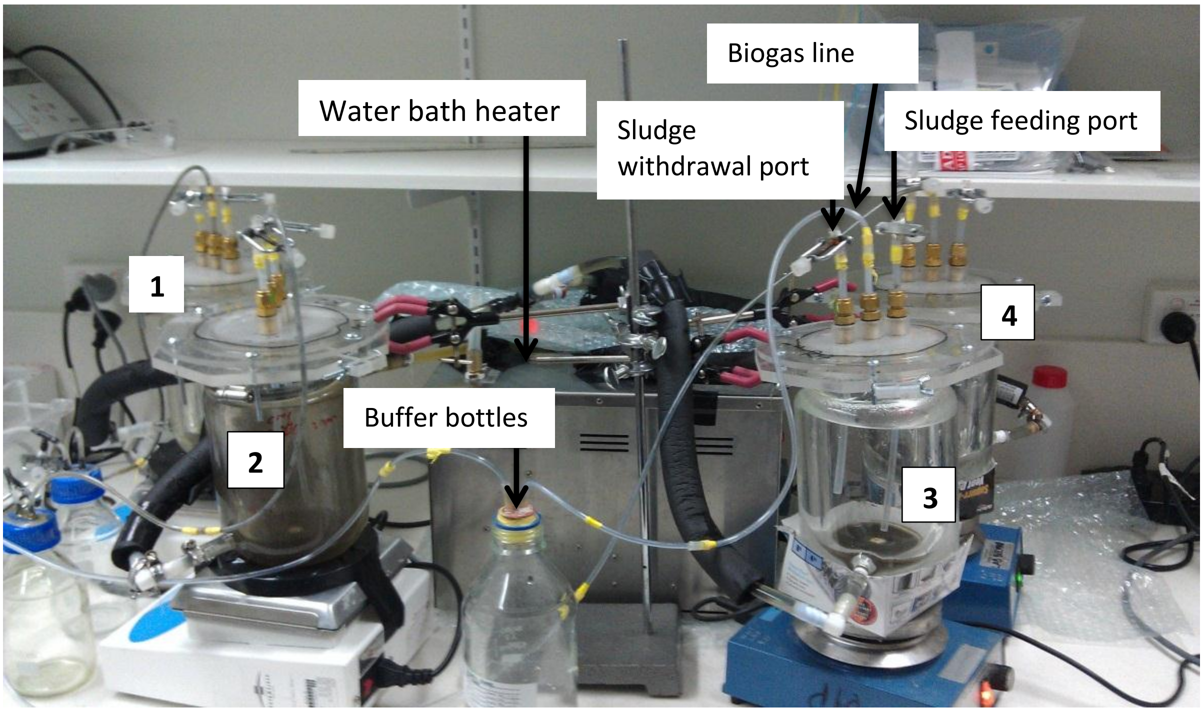


Figure 3.7: Simultaneously operating CSTR set-up for anaerobic digestion tests during inoculation

Figure 3.8: CSTR set-up for tests on effect of pretreatment and other operational parameters on anaerobic digestion

3.6 Biochemical Methane Potential tests and digester performance analysis

3.6.1 Batch methane potential tests on continuously stirred tank reactors

Sludge samples were introduced to the continuously stirred batch reactors for the study of effect of pretreatment, sludge retention time, organic loading rate, pH and temperature on biochemical methane potential, sludge biodegradability, solid reduction capacity, process kinetics and dewaterability. Analysis of both liquid and gas samples were performed periodically as indicated in Table3.4. When the equilibrium sludge retention time anticipated is reached the characteristics of the digested sludge were analysed and compared with that of the reactor feed sludge. The biogas produced was continuously measured by liquid displacement technique.

3.6.2 Continuous methane potential tests on continuously stirred small scale jacketed reactors

In the semi-batch operations, continuous charging of the feed sludge took place on a daily basis for each of the reactors according to the predetermined doses shown in Table3.4. The performance of the reactors for varying SRT and OLR, pretreatment of feed sludge and dose of inhibitors will be investigated. Besides, several parameters including sludge rheology, dewaterability, particle size, microbial content and reduction in TS, VS & COD and several other parameters shown in Table3.3 shall be measured regularly.

Table 3.3: Analysis of effect of SRT and OLR for digestion of pretreated samples

Parameter to be tested for the pretreated mixed sludge feed	**Ranges and conditions for the experiment**
Effect of organic loading rate	3.96 -15.6 gTCOD/l/Day
Effect of sludge retention time	5, 10, 15, 20 days of SRT

3.6.3 Model equations and modelling tools used for analysis of anaerobic digester

In this research work, the kinetics of the anaerobic digestion process was determined using Gombertz model and hydrolysis rate equations. Historical data from BWWTP was used to develop a predictive model that determines the optimum operational condition and optimize the major control parameters. The relationship between important input and output parameters in the anaerobic digestion process was also identified using Adaptive Neuro Fuzzy Logic Inference System (ANFIS) in MATLAB. The details of this study and the modelling tools are discussed in chapter 10.

CHAPTER 4

EFFECT OF ULTRASONIC, MICROWAVE AND COMBINED MICROWAVE-ULTRASONIC PRETREATMENT ON ANAEROBIC DIGESTION OF SYNTHETIC SLUDGE

Abstract

This chapter discusses the effect of ultrasonic, microwave and combined microwave-ultrasonic pretreatment on biogas production, solids removal, and dewaterability of anaerobically digested synthetic sludge. A comparison was made between the three pre-treatment techniques conducting the digestion tests under similar conditions on the same synthetic sludge sample inoculated by digested sewage sludge. The experimental results depict that the combined microwave-ultrasonic treatment (2450 MHz, 800 W and 3 min microwave treatment followed by 0.4 W/ml and 10 min ultrasonication) resulted in better digester performance than ultrasonic or microwave treatment. Mesophilic digestion of combined microwave-ultrasonic pretreated sludge produced significantly higher amount of methane (147 ml) after a Sludge Retention Time (SRT) of 17 days. Whereas, the ultrasonic and microwave treated sludge samples produced only 30 ml and 16 ml of methane respectively. The combined microwave-ultrasonic treatment resulted in total solids reduction of 56.8% and volatile solid removal of 66.8%. Furthermore, this combined treatment improved dewaterability of the digested sludge by reducing the capillary suction time (CST) down to 92 seconds, as compared to CST of 331 seconds for microwave treated and 285 seconds for ultrasonically treated digested sludge samples. Optimisation tests were also carried out to determine the best combination.

4.1 Introduction

There are numerous studies on the benefits of different pretreatment techniques including ultrasonic and microwave pretreatment when the methods are applied independently and in combination with other pretreatment options (chemical and thermal pretreatment) (Valo et al., 2004, Vlyssides and Karlis, 2004). There are only very limited literature on the application of combined microwave-ultrasonic pretreatment for improved anaerobic digestion (Yeneneh et al., 2013b). Therefore, the objective of this chapter is to investigate the effect of three promising pretreatment methods (ultrasonic, microwave and combined microwave-ultrasonic pretreatment) when the methods are applied separately and in combination on synthetic sludge inoculated by real digested mixed sludge. The impacts in terms of biogas production, solid removal, COD reduction and sludge dewaterability were studied. Combined microwave-ultrasonic treatment resulted in increased methane production, better COD removal and improved dewaterability than individual microwave or ultrasonic pretreatment options. The findings of the study on synthetic sludge formed the basis for further investigations conducted on real sludge system discussed in chapters 5 to 10.

4.2 Materials and methods

4.2.1 Sampling and characterization of digested seed sludge

Digested sludge was collected from the dewatering unit (Centrifuge No. 2) at BWWTP. Digested sewage sludge is abundantly available and consists of a broad spectrum of microorganisms and thus usable as seed for reactor acclimation purpose (Qamaruz-Zaman and Milke, 2008). The characteristics of the collected digested sludge are shown in Table 4.1.

Table 4.1: Characteristics of digested sludge seed used for inoculation in the experiment

Parameter	Digested Seed sludge
TS (%)	1.3
VS (% TS)	78.7
COD (g/l)	15.9
pH	6.98
CST (seconds)	110
Surface-average mean particle size (μm)	49.6
Conductivity (mS)	5.74

4.2.2 Preparation of synthetic sludge and its characterization

The synthetic sludge used in the study was prepared by mixing 800 mg/l of peptone, 2720 mg/l of glucose, 560mg/l of meat extract, 1500 mg/l of sodium bicarbonate, 38 mg/l of calcium chloride, 42 mg/l of magnesium sulphate heptahydrate, 320 mg/l iron sulphate heptahydrate and 60 mg/l potassium dihydrogen phosphate. The targeted COD of the synthetic sludge was 40000 mg/l. The characteristics of the synthetic sludge are given in Table4.2.

Table 4.2: Characteristics of the synthetic feed sludge used in the experiment

Parameter	Synthetic sludge
TS (%)	4.7
VS (% TS)	75.5
COD (g/l)	40
pH	7.0
CST (seconds)	35.4
Surface-average mean particle size (μm)	0.284
Conductivity (mS)	13.42

4.2.3 Characterization of sludge fed into the reactors

The synthetic sludge characterized according to the data provided in Table 4.2 was pretreated under the conditions described in Table 4.4. The pretreated synthetic sludge samples were charged into the four reactors and homogenized through mechanical mixing with the digested sludge seed. Samples were withdrawn from each anaerobic digester for characterization purpose. The characteristics of sludge fed to the four reactors are presented in Table 4.3.

Table 4.3: Characteristics of sludge fed into each of the four reactors used in the experiment

Parameter	Reactor 1 (M)	Reactor 2 (U)	Reactor 3 (MU)	Reactor 4 (R)
TS (%)	4.3	4.7	4.4	4.6
VS (% TS)	71.7	80	71.2	70
COD (g/l)	44.1	40	37.8	34.7
pH	7.2	7	7.1	7

4.3 Various sludge pre-treatment methods

Digested sludge (DS) collected from BWWTP (see section 4.3.1) was introduced as seed to inoculate the anaerobic digesters. The reactor acclimation took place for duration of 2 days using 50 ml (20% of total reactor volume) of this digested sludge sample. The four reactors were fed with 200 ml (80% of total volume) of pretreated synthetic sludge samples. The different conditions of pre-treatment used in this study are shown in Table 4.4.

Table 4.4: Various sludge pre-treatment techniques and test conditions

Sample	Pre-treatment method	Conditions
1	Microwave treatment (M)	2450 MHz, 800 W, 3 min
2	Ultrasonic treatment (U)	0.33 W/mL, 178,000 Joules, 90% amplitude, 55/5 pulse, 20 min
3	Combined microwave-ultrasonic treatment (MU)	Microwave: 2450 MHz, 800 W, 3 min Ultrasonic: 0.4 W/mL, 48,000 Joules, 90% amplitude, 55/5 pulse, 10 min
4	Microwave-ultrasonic treatment (MU) for optimization tests	Microwave: 2450 MHz, 800 W, 1 min, 2 min, and 2 min Ultrasonic: 0.4 W/mL, 48,000 Joules, 90% amplitude, 55/5 pulse, 10 min, 10 min and 6 min

Combined microwave-ultrasonic treatment (MU) was conducted in two steps. The microwave treatment was carried out at 2450 MHz and 800 W for a period of 3 min in the first step. The ultrasonic treatment was performed in the second step at 48,000 Joules of ultrasonic energy, 55/5 pulse, 90% amplitude, 0.4 W/mL ultrasonic energy density for a period of 10 minutes. Optimisation tests were carried out after this pretreatment study to obtain the best pre-treatment combination as shown in section 4.6.6.

4.4 Experimental setup for methane potential tests

The biochemical methane potential tests were conducted in 500ml continuously-stirred batch anaerobic reactors where the volume of the reaction mixture was 250 ml. These simultaneously operating four single-stage reactors were kept at a mesophilic temperature of 37.5^0C and were first fed with 50 ml digested sludge as seed. The reactors were acclimated with the digested sludge for 2 days and were separately fed with 200 ml of synthetic sludge pretreated by the various methods described in Table 4.4. The pH was maintained at 7.0 using sodium hydroxide and hydrochloric acid. The methane generated was allowed to pass through buffer tanks to remove any condensate before the gas volume was measured in inverted cylinders by water displacement technique (Federation, 2000). The biogas composition and other parameters were continuously monitored using the methods described in section 3.4.7 until biogas generation ceased at SRT of 17 days.

4.5 Analytical methods

The sludge samples were analysed for total solids, volatile solids, chemical oxygen demand, pH, ammonia, conductivity, dewaterability and particle size using the methods and instruments discussed in section 3.3.1 through 3.3.11.

4.6 Results and discussion

4.6.1 Effect of various sludge pre-treatment methods on methane production

The anaerobic degradation took place during a sludge retention time of 17 days. As shown in Figure 4.1, the cumulative biogas produced after 17 days of SRT for microwave-ultrasonic, ultrasonic, microwave treated and raw untreated sludge samples was 16.7,5.6, 3.8, 2.7 ml biogas /g feed sludge respectively. The specific methane yield was 15.6, 4.02, 1.5, 1.4 ml CH_4/g $TCOD_{added}$ for microwave-ultrasonic, microwave, ultrasonic and raw untreated sludge respectively (Figure 4.2). These results clearly show the significant improvement on biogas production and specific methane yield achieved by the application of combined microwave-ultrasonic treatment. Ultrasonic and microwave pre-treatment techniques also resulted in increased biogas production and methane yield by initiating fast internal heating and disintegration of flocs and cells. Ultrasonic pretreatment was reported to release extra cellular polymeric substances (EPS) which consist of short-chain organic matter by disintegrating the flocs. Ultrasound waves attack the bacterial cell walls and facilitate the release of exo-enzymes that assist the breakdown of organic materials into readily biodegradable fractions (Tiehm et al., 2001). This results in a significant enhancement of the bacterial kinetics which in turn contributes for volatile solids degradation and increased biogas production (Bougrier et al., 2005a). The enhancement of the kinetics of biogas production for ultrasonically treated sludge at the initial stage of the biodegradation process is due to the disintegration of cell walls and release of soluble organics after the 20 min ultrasonic treatment. The increase in digestion efficiency is proportional to the degree of sludge disintegration (Tiehm et al., 2001).

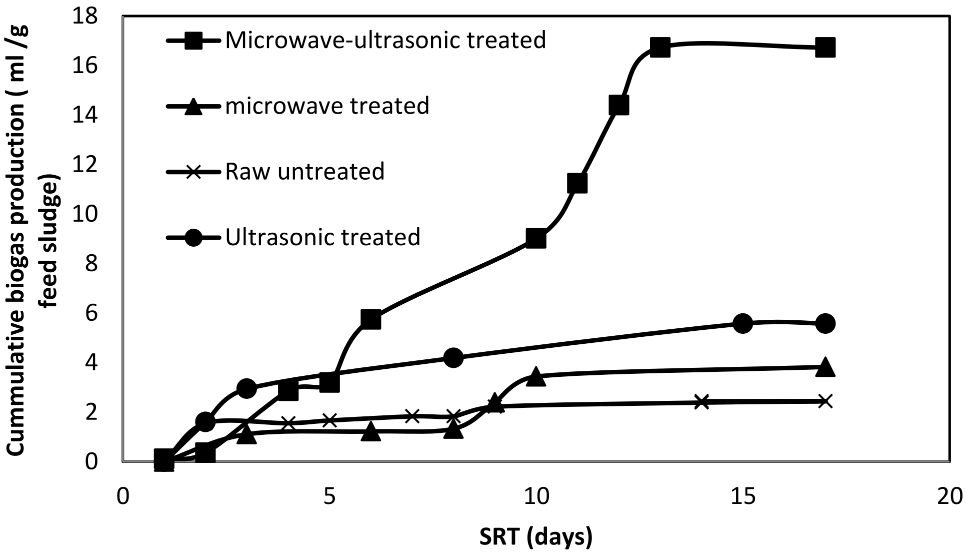


Figure 4.1: Effect of various pretreatment techniques on cumulative biogas production

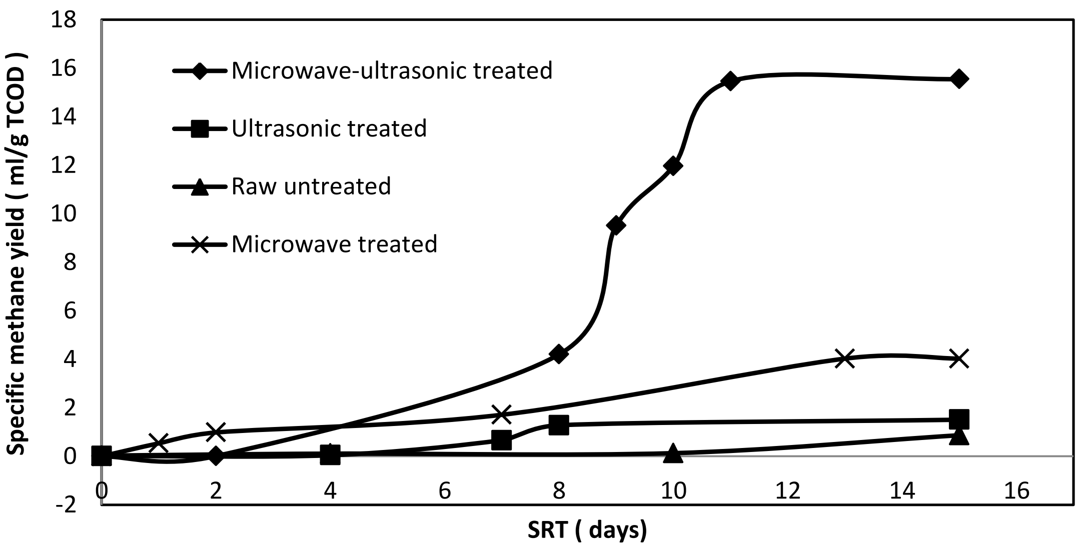


Figure 4.2: Effect of different pretreatment methods on specific methane yield.

In case of microwave treatment, there is higher total volatile acid formation because of the rapid internal heating that destroys the cell walls of microorganisms and disintegrates organics (Saifuddin and Fazlili, 2009b). The combination of the two treatment techniques does not result in additive effect; rather complementary synergy between the two treatment techniques causes enhanced sludge disintegration, floc destruction, thermal and athermal cell wall disruption and release of organics (Eskicioglu et al., 2008b). Efficient and faster disintegration is achieved by the microwave treatment as it helps to achieve internal heating rapidly; whereas the ultrasonicaton assists cavitation, floc size reduction and promotes formation of highly reactive radicals that facilitate destruction of organics.

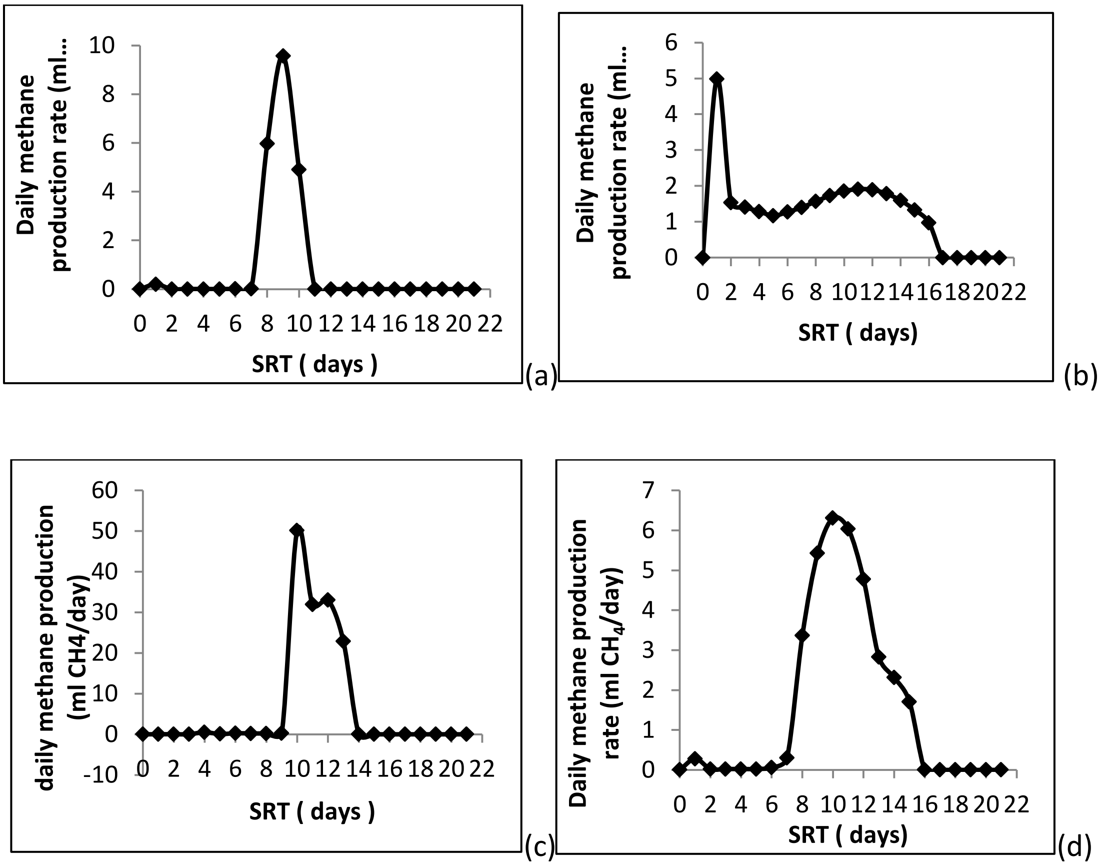


Figure 4.3: Daily methane production rate (ml CH_4/day) for (a) microwave pretreated sludge; (b) ultrasonic pretreated sludge; (c) microwave-ultrasonic pretreated sludge; (d) raw untreated sludge.

According to the daily methane production trend given in Figure 4.3, the process took about 7 days to complete the hydrolysis stage and significant methane production was achieved after the completion of the hydrolysis step. This trend appears to be consistent in all the reactors except the reactor with untreated sludge which had much slower hydrolysis rate (Figure 4.2). The methane production reached its peak after almost 10 days of operation and the process can be considered to have reached to a point of no methane production at 17 days of SRT. The methane production on daily basis also shows that microwave-ultrasonic-treated sludge produced the highest amount of methane, followed by microwave-treated sludge.

4.6.2 Effect of various pretreatment methods on solid removal

Pre-treatment resulted in a significant reduction of the solid content of the feed sludge. The percentage reduction in TS was 51.6 %, 63.5% and 56.8% for microwave, ultrasonic and microwave-ultrasonic treated sludge respectively as shown in Figure 4.4 Combined microwave-ultrasonic treatment resulted in an intermediate TS reduction which was better than that of microwave-treated but less than that of ultrasonic-treated sludge. On the other hand, the volatile solid reduction was 64%, 79.3% and 66.8 % for microwave, ultrasonic and microwave-ultrasonic-treated samples respectively. Increased solid removal for ultrasonically treated sludge is due to greater disintegration of cells and solubilisation of organics which ultimately reduced to methane and other gases. It can also be observed that increased solubilisation does not guarantee increased methane production, as the later depends on methanogens performance and other factors (Saha et al., 2011b).

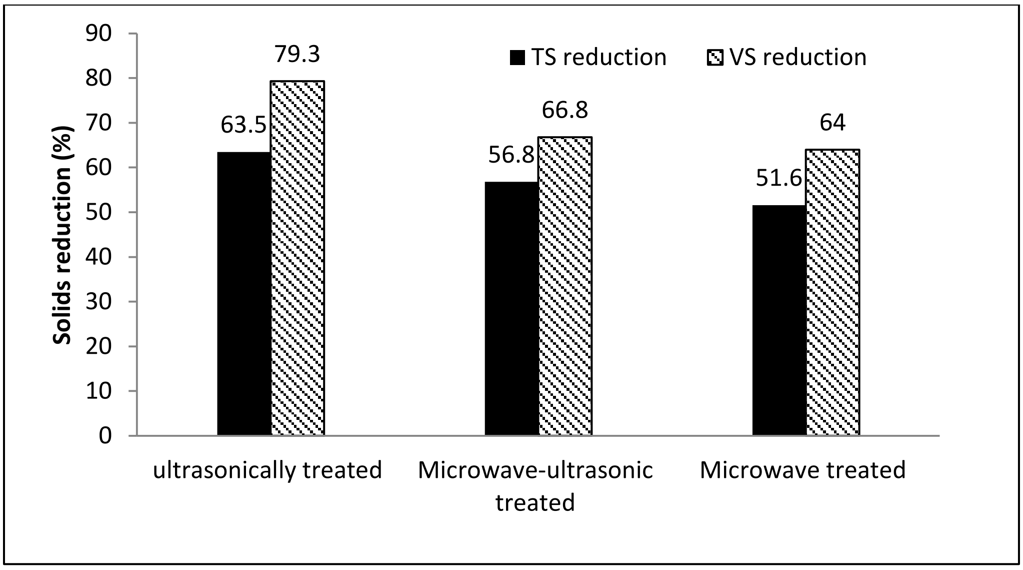


Figure 4.4: Solids reduction (TS and VS) for different pre-treated sludges

4.6.3 Effect of various pre-treatment methods on sludge dewaterability

Dewaterability of the four sludge samples was measured using the capillary Suction Timer (CST). Figure 4.5 shows dewaterability of different digested pretreated sludge samples. Microwave-ultrasonic treated sludge had significantly higher dewaterability or shorter CST as compared to the other pretreatment options. The CST for ultrasonicated and microwave treated samples was relatively longer, this is attributed to high degree of floc disintegration and higher specific energy of ultrasonication, resulting in the reduction of some filamentous materials and biopolymers which may have caused bulking (Bougrier et al., 2006). The dewaterability of raw untreated sludge was the shortest as the average particle size for this sludge type is relatively bigger as shown in Table 4.5 resulting in better dewaterability and lesser availability of hydrophilic biopolymers that hold water molecules as compared to pretreated sludge samples.

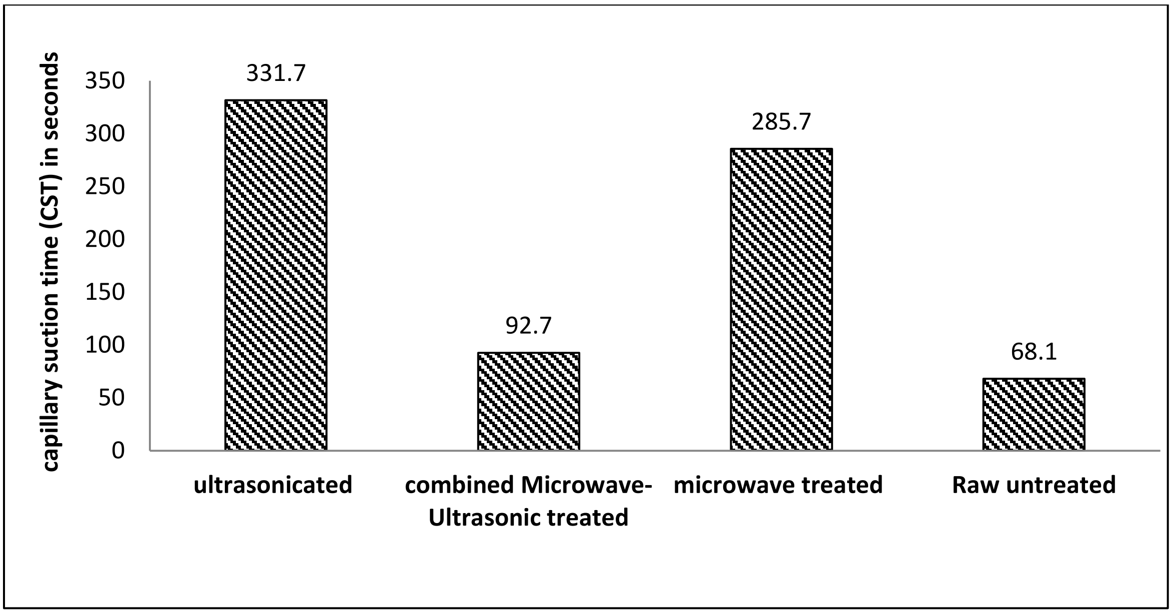


Figure 4.5: Dewaterability based on capillary suction time after anaerobic digestion

4.6.4 Functional group analysis based on FTIR-ATR spectra

FTIR bands around 1100-1000 cm-1 particularly around 1070 cm-1 representing the occurrence of polysaccharides were of lower intensities for the pretreated digested sludge samples confirming the disintegration of polysaccharides. The intensity changes of bands around 2925-2950 cm-1 show the decomposition of fatty acids and lipid components. The disappearance of bands of amide I around 1630-1650 cm-1 particularly with combined microwave- ultrasonic treated digested sludge testifies the enhancement in the decomposition of proteins for the combined pretreatment. Microwave treated sludge showed a similar trend. Figure 4.6 presents the FTIR spectra of digested sludge pretreated by the various methods.

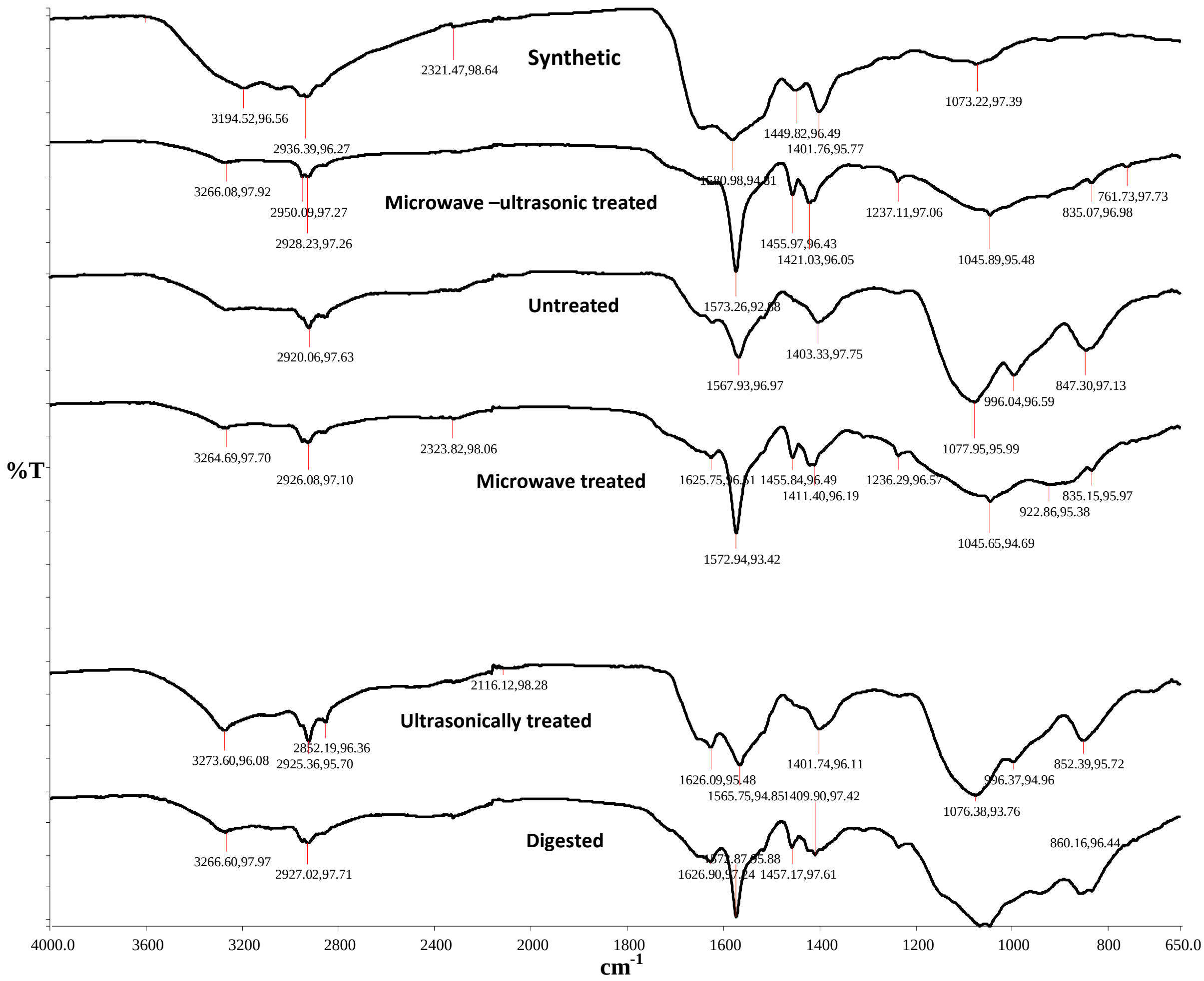


Figure 4.6: FTIR spectra of synthetic and pretreated digested sludge

4.6.5 Particle size distribution of various pretreated sludge samples

The particle size distribution of each of the digesters is shown in Table 4.5. Different size distributions were obtained for each of the treatment types. This is likely to be due to the differences in biodegradability of the different types of feed sludge. The d(0.1), d(0.5) and d(0.9) values indicate that 10%, 50% and 90% of the particles measured were less than or equal to the size stated. According to the distributions shown in Table4.5, ultrasonically treated sludge sample appeared to have smaller particles as compared to the distribution of particles from the other pre-treatment techniques. This is expected to be linked to the cavitation and increased disintegration of the sludge particles that has happened during ultrasonication of the feed. The average particle size increased from ultrasonically treated sludge to the untreated sludge sample. The sludge specific surface area was derived from the particle size distribution. The specific surface area data quoted in Table4.5 clearly illustrates that the smaller particles contributed more in terms of specific surface area than the larger size fractions. The smaller particle sizes are indicative to the disintegration that happened because of pre-treatment which has ultimately assisted the release of organic matter, the increase in digestion and the biogas production.

Table 4.5: Particle size distribution of different digested sludge samples

Type of sludge	**d(0.1)**	**d(0.5)**	**d(0.9)**	**Specific surface area (m^2/g)**	**Surface-weighted mean (µm)**	**Volume-weighted mean (µm)**
Ultrasonically treated	9.918	31.676	76.987	0.314	19.082	41.84
Microwave-ultrasonic treated	10.435	32.35	74.82	0.293	20.48	40.88
Microwave treated	10.986	35.403	84.529	0.283	21.185	45.3
Raw untreated	12.313	38.297	97.661	0.249	24.14	48.8
Digested feed sludge	24.946	85.248	219.065	0.121	49.55	113.7

4.6.6 Optimisation of process parameters for combined microwave-ultrasonic pre-treatment

The optimum operating conditions for the microwave-ultrasonic pre-treatment method were determined by comparing four combinations 1 min, 2 min and 3 min of microwave followed by 10 min of ultrasonication and 2min of microwave followed by 6 min of ultrasonication. These different pre-treatment combinations showed that shorter ultrasonication time and energy with more microwave time (2 minute microwave treatment and 6 minute ultrasonication) resulted in improved efficiencies in terms of methane production (Figure 4.7) and dewaterability (Figure 4.8) The percentage reductions in total and volatile solids are more or less similar for all the pre-treatment conditions (57% and 68% respectively).

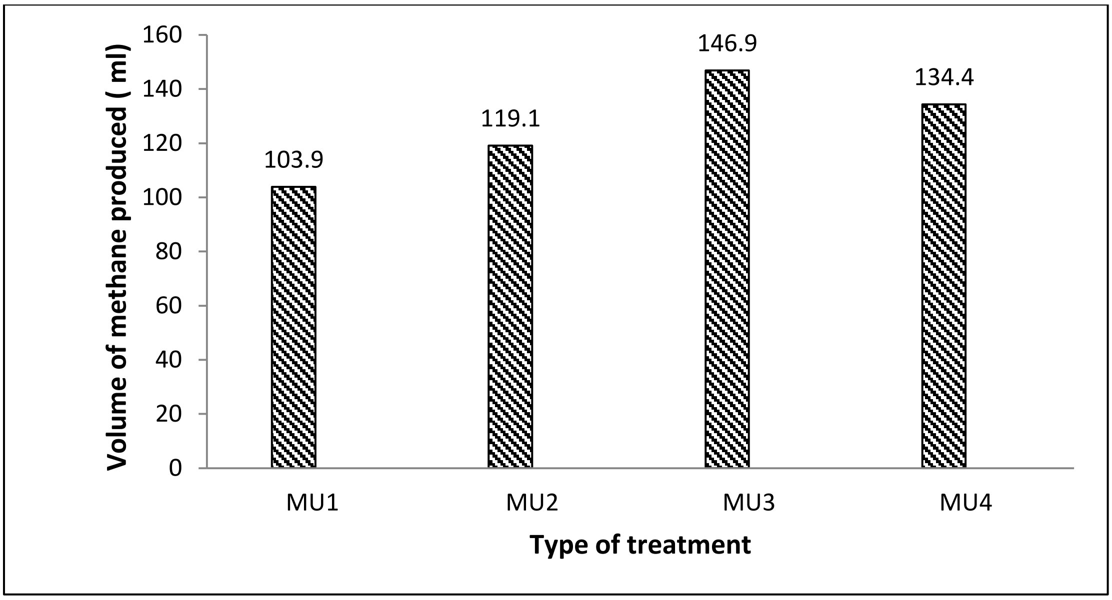


Figure 4.7: Cumulative methane production from the anaerobic digestion of different microwave-ultrasonic pretreated samples. (MU1= 1 minute microwave pretreatment followed by 10 minute ultrasonic treatment, MU2= 2 minute Microwave pretreatment followed by 10 minute ultrasonic pretreatment, MU3= 2 minute microwave pretreatment followed by 6 minute ultrasonic pretreatment, MU4= 3 minute microwave pretreatment followed by10 minute ultrasonic pretreatment)

The amount of methane produced from this pre-treated sludge (2 min of microwave followed by 6 min of ultrasonication) was higher than what was achieved in the other combinations. As the results for different combinations show, a mild ultrasonication is sufficient for disintegration of flocs and organics, whereas relatively higher microwaving assists in faster thermal and athermal cell wall disruption and organic degradation. The dewaterability measured in capillary suction time was found to be as low as 43 seconds for this pre-treatment. Although more experiments are still required to expand the range of the treatment times, the implication of this in terms of energy consumption of the pre-treatment process is promising. This is because of the fact that the shorter the ultrasonicaton time and power, the smaller will be the energy consumed in the process. Besides, the energy requirement for microwave treatment is far less than that of ultrasonic treatment favouring the optimum outcome observed in this work.

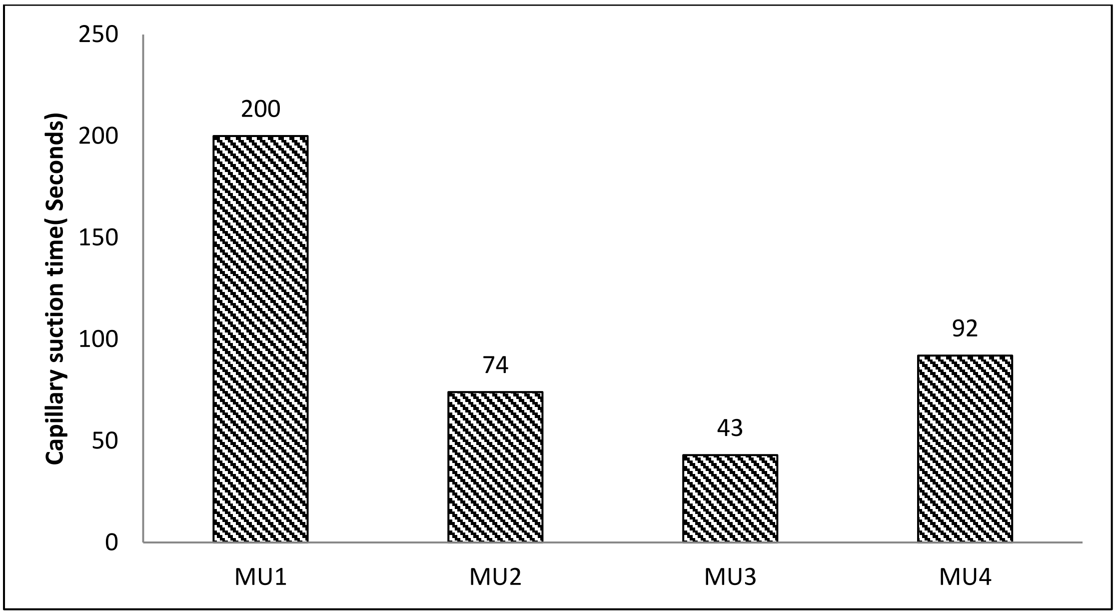


Figure 4.8: Dewaterability of sludge after microwave-ultrasonic treatment at different conditions

4.7 Conclusions

Combined microwave-ultrasonic pretreatment significantly improved methane production, solid removal and dewaterability under the pretreatment and operating conditions specified. The optimum combination for the two pretreatment techniques was found to be 2 min of microwave treatment followed by 6 min of ultrasonication. Particle size distribution for ultrasonically treated sludge was found to be smaller than the size distributions for combined microwave-ultrasonic-treated, microwave-treated or untreated sludge. FTIR bands for combined microwave-ultrasonic-treated digested sludge confirmed the increased polysaccharide, protein and fatty acid decomposition as compared to the other techniques. Microwave-treated sludge also showed a similar trend. The combination of the two treatment techniques did not result in direct additive effect. There is rather a complementary synergy between the two treatment techniques causing enhanced sludge disintegration, floc destruction, cell wall disruption and release of soluble organics. Combined microwave-ultrasonic pretreatment effects on anaerobic digestion of real municipal sewage sludge will be addressed in the following chapters. In chapter 5 the optimization of the pretreatment conditions is investigated in depth based on the findings of the pretreatment and anaerobic digestion study on synthetic sludge. Chapters 6,7,8,9 address anaerobic digestion and biodegradability studies on various sludge types and digestion conditions for selected pretreatment condition.

CHAPTER 5

OPTIMIZATION OF MICROWAVE, ULTRASONIC AND COMBINED MICROAVE-ULTRASONIC PRETREATMENT CONDITIONS FOR ENHANCED ANAEROBIC DIGESTION

Abstract

This chapter describes the effect of microwave and low frequency ultrasonic pretreatment power intensity, time, and density on mixed sewage sludge (MS) and thickened excess activated sludge (TEAS) characteristics and anaerobic digester performance. Key parameters affecting the efficiency of ultrasonic and microwave treatment were optimized and the effect of change in ultrasonication and microwave pretreatment conditions on sludge solubilisation and other sludge characteristics were analysed. Ultrasonication power, density and time have individual significance on the sludge solubilisation process. Three mixed sludge samples pretreated under three different ultrasonication powers (80W, 100W and 150W) and microwave conditions of (2450MHz, 3min, 800W) were digested in batch anaerobic continuously stirred tank digesters for a sludge retention time of 25 days. Moreover, other three mixed sludge samples were pretreated at three different ultrasonication durations of (4min, 6 min and 10 min) and microwave treatment condition of (2450Hz, 6 min, 800W). The samples were later subjected to continuously mixed anaerobic batch digesters. Effects of microwave density and pretreatment time on solubilisation of TEAS were investigated for treatment densities of 3.2 W/mL, 4.6 W/mL and 6.4 W/mL and treatment duration of 1-7 minutes. TEAS was pretreated at the optimum microwave pretreatment conditions followed by ultrasonic pretreatment at ultrasonic densities of 0.5W/mL, 0.66 W/mL and 1 W/mL and ultrasonication times of 1-12 minutes. Higher sludge degradability, higher volatile solid removal and better digester performance was achieved for the anaerobic digester with lower ultrasonication power of 80W, ultrasonication time of 6 min and ultrasonic density of 0.32W/ml for mixed sludge. The biogas production volume and kinetics, dewaterability of digested sludge, COD reduction and other sludge properties were optimized for the aforementioned ultrasonication and microwave pretreatment conditions for MS and TEAS as well.

5.2 Introduction

Ultrasonic and microwave pretreatment are a function of pretreatment power, time, density and pH during the pretreatment process. The pretreatment conditions directly affect the degree of sludge disintegration and solubilisation which in turn influence the gas production, solid removal, dewaterability and flow characteristics of the sludge and overall operation cost of the wastewater treatment plant (Fernández-Cegrí et al., 2012, Saha et al., 2011a). Microwave irradiation and ultrasonication are energy intensive processes that the cost effectiveness of these techniques has to be addressed. In this chapter, effect of change in ultrasonic and microwave pretreatment power, time, density and intensity on biodegradability of sludge is discussed. The optimum combined treatment power and time that maximizes gas generation, improves dewaterability and solid removal is also presented**.** Optimisation of pretreatment duration, intensity and density contributes significantly by reducing operational cost, duration of pretreatment and results in improved digester performance, better sludge quality and dewaterability (Wang et al., 2005). The best pretreatment technology and the optimum pretreatment conditions employed in this study were selected based on the findings presented in this chapter.

5.3 Materials and methods

The effects of microwave and ultrasonic power, time and density on sludge solubilisation and degree of disintegration of organics were assessed when the pretreatment methods are applied alone and in combination. Mixed and thickened excess activated sludge samples used in the study were obtained from Beenyup waste water treatment plant. The mixed sludge was composed of 75% primary sludge and 25% thickened excess activated sludge. The characteristics of mixed sludge and thickened excess activated sludge samples used for optimization study are shown in Table 5.1.

Table 5.1 Characteristics of the feed mixed and thickened excess activated sludge samples used in the study.

Characteristic parameter	Mixed Sludge (MS)	Thickened excess activated sludge (TEAS)
Total solid	27g/L	45 g/L
Volatile solids	23.5g/L	40.5g/L
TCOD	37,950mg/L	35600 mg/L
SCOD	4600 mg/L	3200 mg/L
pH	6.9-7.1	7.1

The characterized samples were subjected to microwave, ultrasonic and combined microwave-ultrasonic pretreatment. Table 5.2 presented detailed experimental conditions for pretreatment including time, power and density. Sludge solubilisation, dewaterability, pH and other parameters were measured after each pretreatment and the effect of each pretreatment factor on these parameters was investigated. The degree of disintegration of sludge was measured after microwave, ultrasonic and combined microwave-ultrasonic pretreatment at different pretreatment conditions. Pretreatment density and duration of pretreatment was varied. The SCOD/TCOD ratio was measured by using standard COD measurement colorimetric technique for each untreated and pretreated sludge sample (Park et al., 2004). Each sample was first diluted 50 times and filtered on Whatman filter paper type 1PS-110mm and then SCOD was measured. Three continuously stirred batch digesters each with working volume of 250ml and five other digesters were inoculated by digested sludge (DS) from BWWTP. The two sets of digesters were charged with Mixed and thickened excess activated sludge samples pretreated as shown in Table 5.2 to undergo mesophilic (36.5^{o}C) digestion for a sludge retention time of 28 days and 25 days respectively. Biochemical methane potential (BMP), dewaterability, solid and COD removal test were performed on sludge samples from each of the digesters. The performance of the anaerobic digesters and sludge characteristics were analysed using different analytical and instrumental techniques presented in Section 3.3.1 through 3.3.11.

Table 5.2 Types and conditions of pretreatment.

test	Pre-treatment method	Test conditions
1	Microwave treatment: Effect of Pretreatment power (Mixed sludge)	Frequency= 2450 MHz, time= 3min Power=(800 W, 640W, 400W, 240W, 80W)
2	Microwave treatment: Effect of pretreatment time (Mixed sludge)	Frequency= 2450 MHz, Power= 800W Time= (1 min, 2min, 3 min, 5min)
3	Ultrasonic treatment : Effect of pretreatment power (Mixed sludge)	0.4 W/mL: ultrasonic density, 55/5:Pulse, 6 min: time, Power=(150W@90%Amplitude,100W@65% Amplitude, 80W @45% Amplitude)
4	Ultrasonic treatment : Effect of pretreatment time (Mixed sludge)	0.4 W/mL: ultrasonic density, 90%: Amplitude, 55/5:Pulse, Power: 150W, Time= (4 min, 6 min, 8 min, 12 min)
5	Combined Microwave-Ultrasonic treatment (MU) with varying ultrasonic time. (mixed sludge)	Microwave: 2450 MHz, 800 W, 3 min and ultrasonic treatment: 0.4 W/mL density, 90% amplitude (140W), 55/5 pulse, time: 4min, 6min, 10 min
6	Combined Microwave-Ultrasonic treatment (MU) with varying ultrasonic power. (mixed sludge)	Microwave: 2450 MHz, 800 W, 3 min and ultrasonic treatment: 0.4 W/mL density, 90% amplitude (140W) 75% amplitude (100W) 70% amplitude (80W), 55/5 pulse, time: 6min
7	Effect of ultrasonic density and pretreatment time (TEAS)	Ultrasonication conditions (amplitude = 81%, power= 100 W, pulse 55/5, probe depth = 2cm, Time = 1min, 3 min, 6min, 8 min, 12min Ultrasonic density: 0.5W/ml, 0.66 W/ml and 1 W/ml)
8	Effect of microwave density and pretreatment time on TEAS	Microwave irradiation conditions (power= 640W, Frequency= 2450 MHz, time 1 min, 3 min, 5 min, 7 min) Microwave density (3.2 W/ml , 4.2 W/ml and 6.4 W/ml)
9	Combined Microwave Ultrasonic pretreatment (Effect of pretreatment density and time For TEAS)	Microwave: 2450 MHz, 3.2W/mL and 4.2 W/mL ,3 min and Ultrasonic treatment density: 0.66 W/mL, 0.55 w/ml and 1 W/mL. 90% amplitude, 55/5 pulse, pretreatment time: 1 min, 3 min, 6 min, 8 min, 12min,

5.4 Result and discussion

5.4.1 Effect of Microwave pretreatment temperature, density and time on sludge solubilisation

The average temperature of the sludge after microwave heating and the degree of sludge solubilisation is shown in Figure 5.1 and Figure 5.2 (a). The microwave energy is transformed into heat derived from the internal resistance to rotation. Temperature rise during sludge heating is related to heat generated as a result of the absorption of the microwave energy by water, or by organic components which undergo constant or induced polarization (Jang and Ahn, 2013b). Thus, the thermal activation and sludge solubilisation in the sludge samples is due to the absorption of microwave energy by water and organic complexes available in the sludge sample. Microwave heating is due to absorption of microwaves radiation by water (ZHAO Xiang, 2009). Microwaves pretreatment has benefits like rapid heating, pathogen destruction, and ease of control system. The factors influencing microwave irradiation of the dielectric materials include temperature, radiation time and penetration. Optimum pretreatment conditions obtained in this study confirm the benefits. A maximum temperature of 80^0C was chosen to avoid vaporization of liquid (Coelho, 2012b).

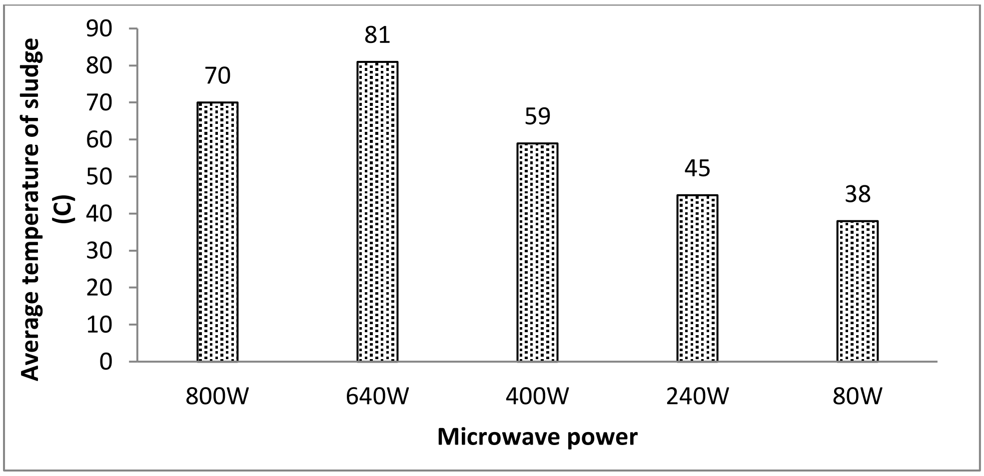
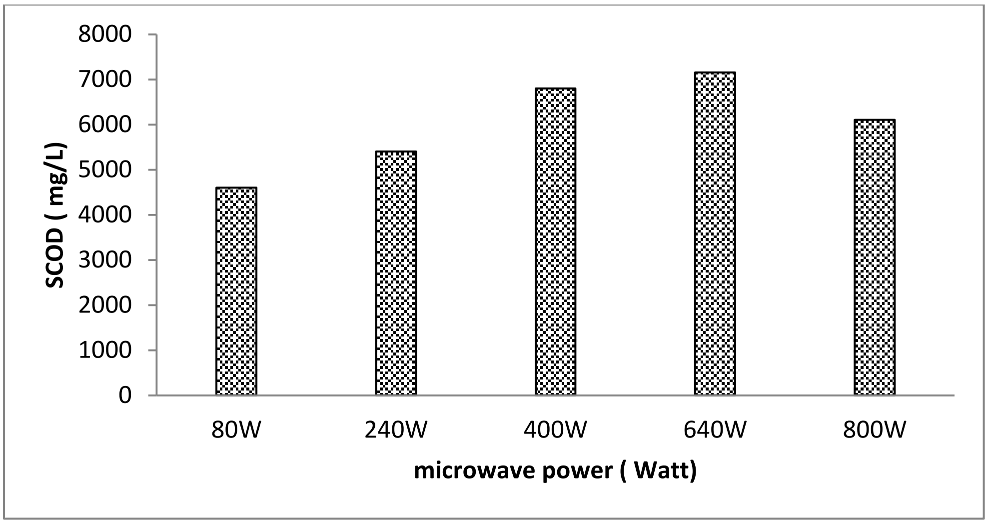
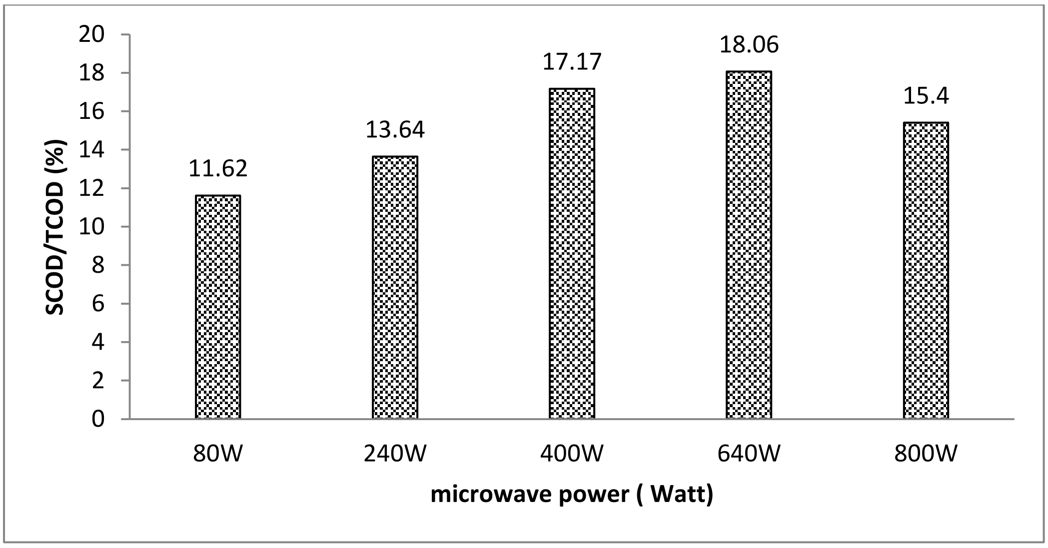


Figure 5.1 Effect of microwave power on temperature.

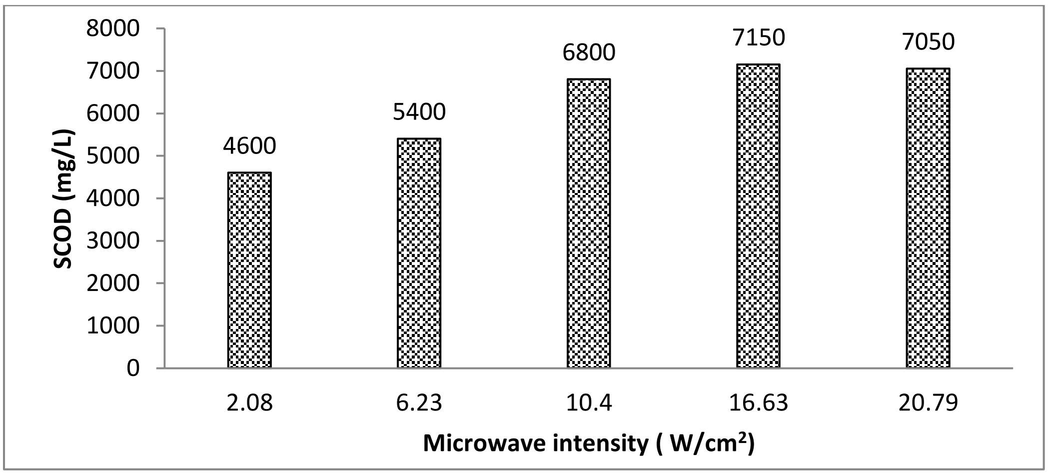
Increased solubilisation in SCOD occurred due to the microwave pretreatment as shown in Figure 5.2(a) and (b). Sludge solubilisation increases with increasing temperature for the microwave pretreatment at different power intensities. This is because of the fact that the heat generated in the process is the main physical factor causing the solubilisation of sludge flocs. Flocs in activated sludge are composed of a polymeric matrix made up of variable quantities of extracellular polymeric substances (EPS) such as proteins, carbohydrates, humic substances, glycoproteins, lipids, and nucleic acids with the bacterial cells embedded in the mesh (Urbain, 1993). However, the most prevalent substances are proteins and carbohydrates. The increase in SCOD is due to the release of such components (Miron et al., 2000). Park et al (2010) investigated the effects of microwave pretreatment temperature, output power and solid concentration in the sludge. Each of these factors affected the pretreatment process. Correspondingly, the highest mixed sludge solubilisation occurred at microwave pretreatment power of 640W as shown in Figure 5.2 (a) and (b) and pretreatment duration of 3 min for the treatment power range considered in this study. On the other hand, higher SCOD release was achieved for microwave pretreatment duration of 5 minutes for thickened excess activated sludge. Despite high solid concentration of thickened excess activated sludge, sludge disintegration and SCOD release was higher for mixed sludge. This is because of the primary sludge portion of the mixed sludge which consists of greater concentration of biodegradable organics. The general trend in SCOD concentration (mg/l) indicates that with increasing microwave pretreatment power, intensity and pretreatment time, the degree of sludge disintegration and solubilisation increases. The hydrolysis of large organic molecules, cell wall lysis and disintegration of sludge is intensified by the microwave pretreatment.



(a)



(b)



(c)

Figure 5.2 change in SCOD (a) and SCOD/TCOD ratio (b) versus microwave power, change in SCOD with respect to microwave intensity (c).

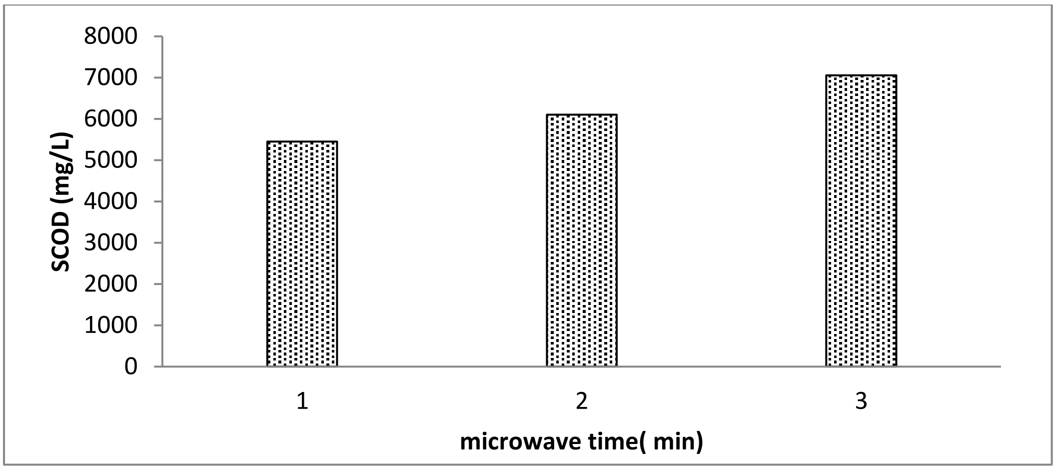


Figure 5.3 Effect of microwave pretreatment time on sludge disintegration of mixed sludge.

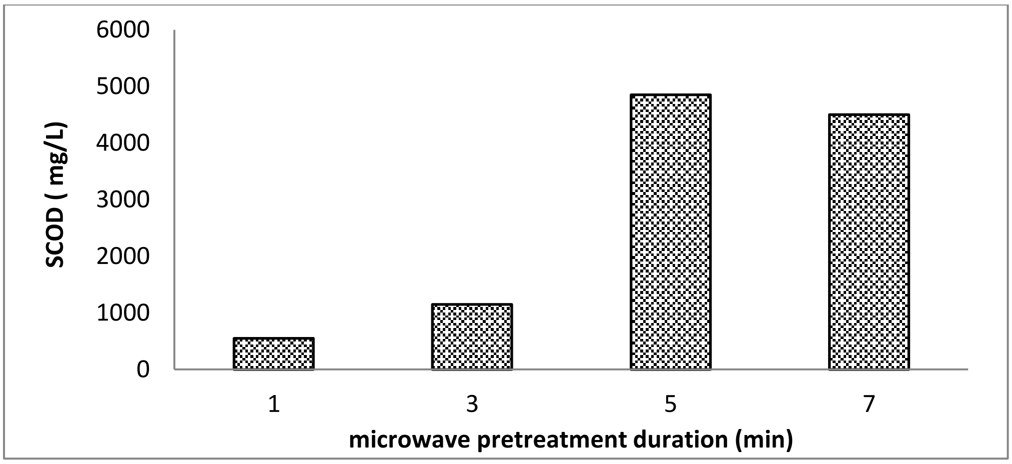


Figure 5.4 Effect of microwave pretreatment time on sludge solubilisation for Thickened excess activated sludge.

Specific energy of sludge solubilisation calculated according to equation 5.1 (Kuglarz et al., 2013) shows that for the combined microwave-ultrasonic pretreatment, microwave pretreatment duration of 5 minutes resulted in the highest degree of sludge solubilisation with the least energy consumed. Hence, the optimum microwave pretreatment duration is 5 minutes for TEAS during the combined pretreatment as shown in Figure 5.5.

$$E_{SCOD} = \frac{P * tD}{V * SCOD} \qquad (5.1)$$

Where, E_{SCOD} = Specific energy consumption (KJ/g soluble COD released)

t = exposure time (s)

P= power of the microwave heater (KW)

V= volume of sludge treated (L) and

SCOD = soluble organic matter released into the liquid phase. (mg/l)

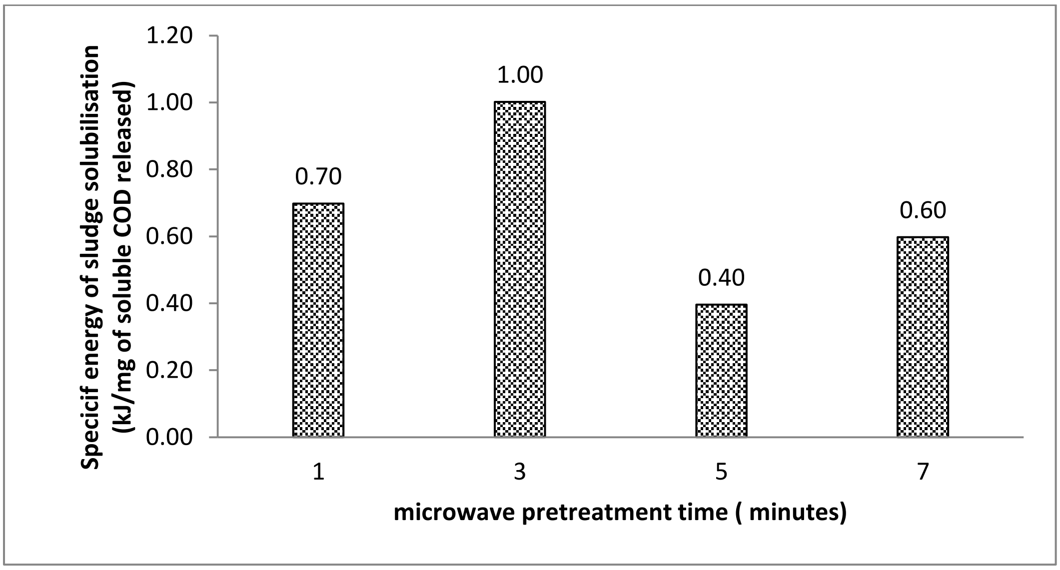


Figure 5.5 Specific energy of sludge solubilisation as a measure of energy consumption (kJ/mg of SCOD released) for thickened excess activated sludge.

5.4.2 Effect of ultrasonic power (intensity), density and pretreatment time on sludge solubilisation

Ultrasonic pretreatment significantly increased the degree of sludge solubilisation and anaerobic digestion performance. Ultrasonication pretreatment for a short duration of time has resulted in breakdown of macro flocs and micro biodegradable organics to a reasonable degree (Oh, 2006). Shorter sonication duration is preferred in this study as the sonication is coupled with microwave pretreatment to make use of the advantage synergistic combined pretreatment provides over individual pretreatment. Besides, the economic benefits in terms of reducing pretreatment cost by reducing ultrasonication time are significant. Degree of sludge solubilisation for mixed sludge sample was highest for 12 minutes of pretreatment time as shown in Figure 5.6.

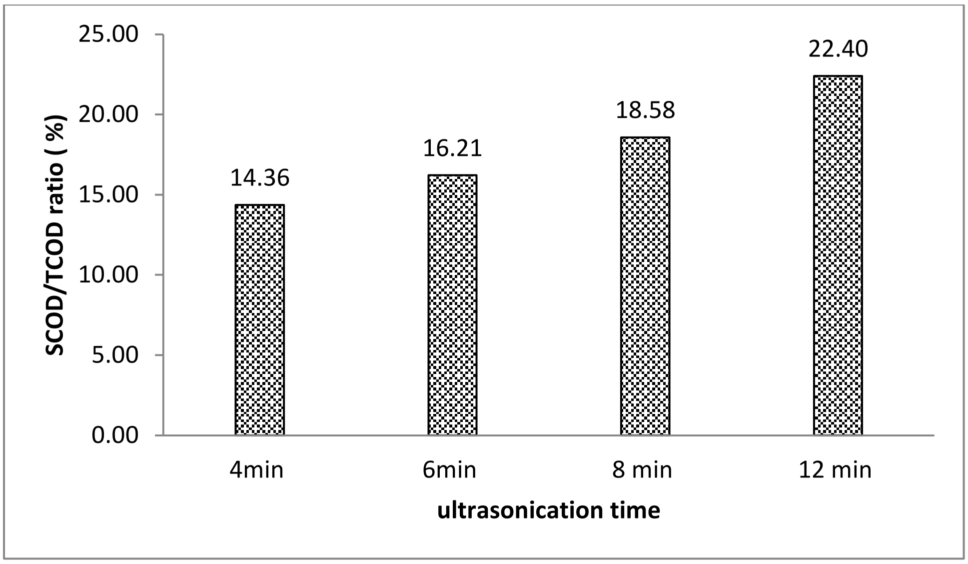


Figure 5.6 Effect of ultrasonication time on mixed sludge solubilisation.

Sludge solubilisation for combined pretreatment was far better than individual treatment techniques where the SCOD was almost doubled after the combined treatment proving the potential for higher methane production (Figure 5.7 and 5.8).

Sonication density plays a significant role in cavitation bubble formation (Urbain, 1993). Particle disruption can be optimized by sonication of sludge sample at high sonication density and shorter sonication time. The particle disruption study against duration of sonication revealed that macro flocs are affected than micro flocs. Larger surface area of exposure favours higher particle disruption. Combined pretreatment with ultrasonication density of 0.52 W/ml resulted in higher degree of sludge solubilisation of 41 % for sonication time of 8 minutes (Figure 5.7 and 5.8). This pretreatment condition has the advantage of less specific energy consumption compared to other pretreatment densities with similar sonication time. Generally, the degree of sludge solubilisation is the highest at this condition with the least amount of energy consumed as shown in Figure 5.9.

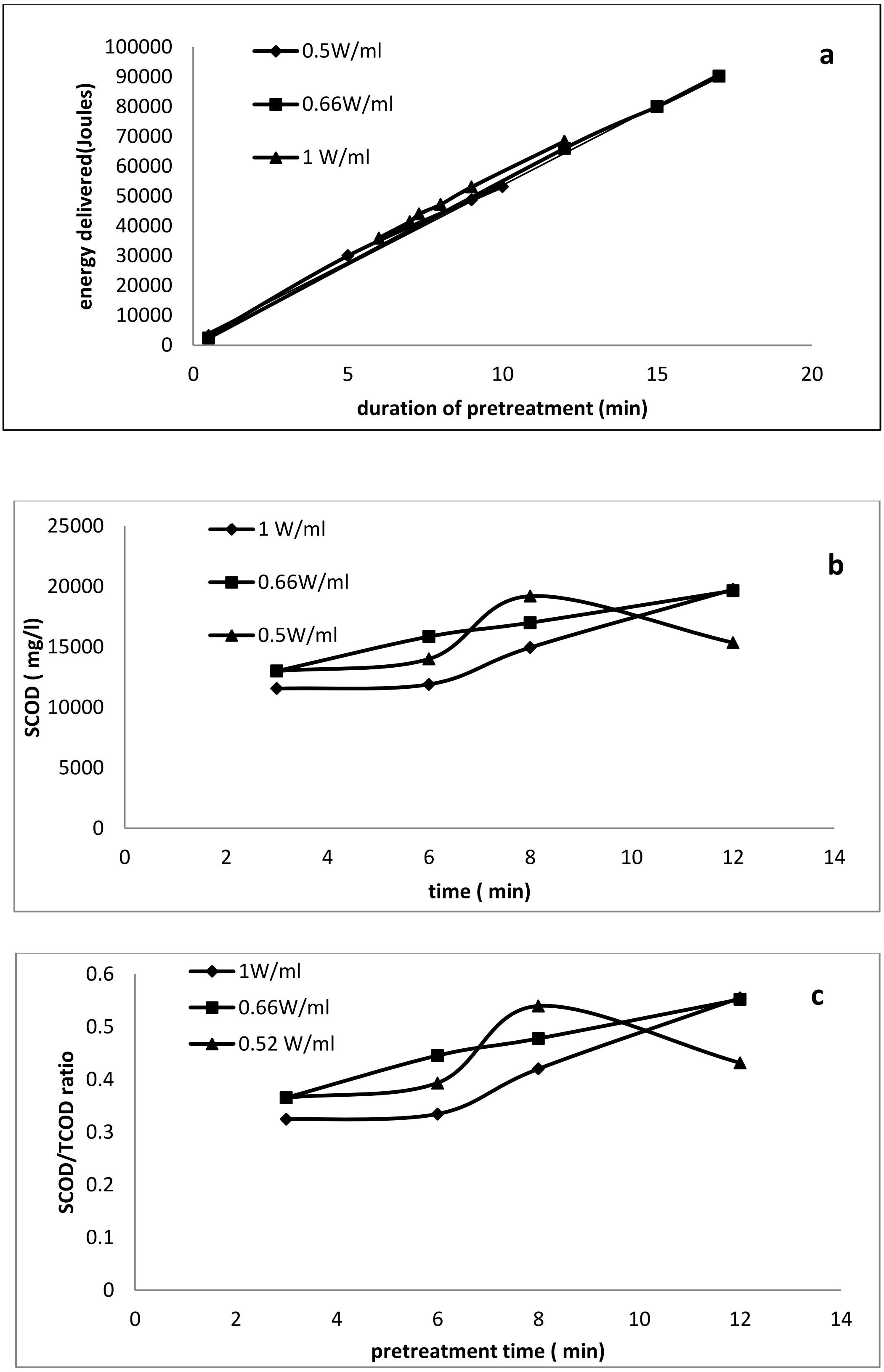


Figure 5.7 (a) Ultrasonication density versus pretreatment time, energy delivered (b) Soluble COD (c) Soluble COD to total COD ratio for Thickened excess activated sludge.

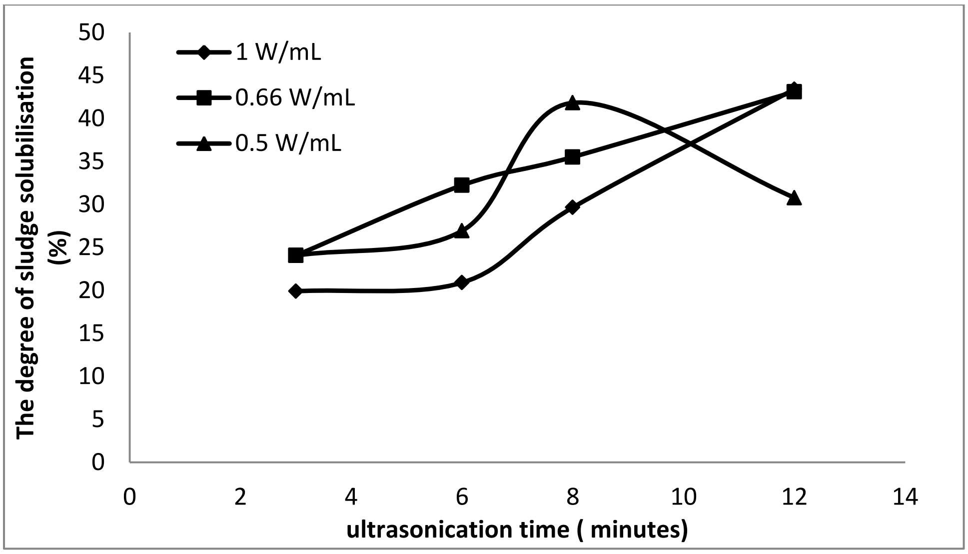


Figure 5.8 Degree of sludge solubilisation for varying ultrasonication time and density.

The specific energy input is proportional to sonication time. The longer sonication time means a higher specific energy input; thus resulting in higher SCOD release (Figure 5.9). Wang et al. (2005) investigated the release in SCOD concentration at three different sonication times of 5, 15 and 20 min at TS content of 3%, frequency of 20 KHz and ultrasonic density of 0.768 W/mL. This particular study shows the release of SCOD as a function of the specific energy input for ultrasonic densities of 1W/mL, 0.66 W/mL, 0.5 W/mL and total solid concentration of 45 g/L.

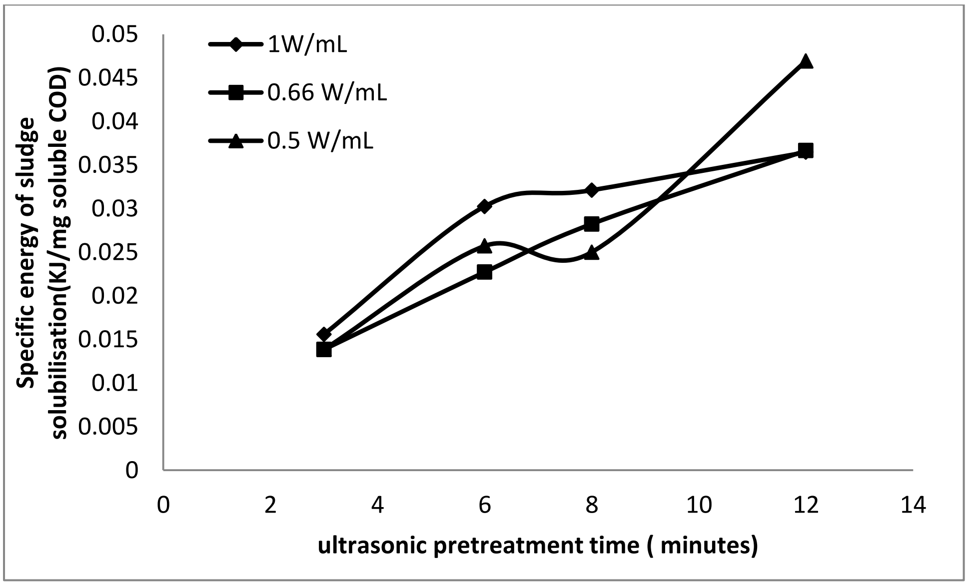


Figure 5.9 Specific energy consumption for sludge solubilisation for varying ultrasonication time and density.

5.4.3 Optimization of Anaerobic digester performance for combined microwave-ultrasonic pretreated sludge.

The digestion tests conducted for microwave-ultrasonic treated mixed sludge samples show that the highest methane production (164 ml) was achieved for ultrasonication power (intensity) of 100W (2.6 W/cm^2) as shown on Figure 5.11 for SRT of 25 days. Higher SCOD/TCOD ratio for this ultrasonication condition justifies the increased methane production (Figure 5.14). The methane production potential at SRT of 7-20 days for ultrasonication at 80W (2.1 W/cm^2) was relatively greater than the methane yield for the other sonication conditions. Moreover, the dewaterability of digested sludge for this condition (80W ultrasonication power) was the lowest (144 seconds) as measured by the capillary suction timer (Figure 5.20). Higher degree of disintegration of flocs and greater percentage of fine particles due greater power of ultrasonic treatment resulted in deteriorated dewaterability (Yu et al., 2009). The percentage reduction in TCOD was the highest for the sample with higher ultrasonication power (150W). The sludge sample pretreated at 100W had the least SCOD/TCOD ratio (17.9%) as the ultimate methane production was greater for the treatment at this particular condition. Likewise, the methane production potential for combined microwave-ultrasonic pretreated TEAS was higher for higher ultrasonication density (1W/mL) and longer pretreatment duration (8 min) as compared to ultrasonic density of 0.66W/mL and 0.5 W/mL for the same sonication time as shown in Figure 5.11 for an SRT of 28 days. Besides, the extracellular polymeric substances that may have played the role of floc formation have been disintegrated more at higher power and sonication time. The SCOD/TCOD ratio for the set of TEAS digesters confirm that higher sonication density and duration of pretreatment (1 W/mL, 8 min and 0.66 W/mL, 12min) associated to higher methane production (Figure 5.12). The volatile solid removal for these pretreatment conditions was significantly higher (60.77 for 1W/mL, 8 min. 69.28 for 0.66 W/mL, 12 min) than the other pretreatment conditions. Total solid reduction of combined microwave-ultrasonic pretreated sludge at the sonication condition of 0.66 W/mL, 12 min was the highest (Figure 5.13). Ultrasonication at intermediate power density, intensity and relatively longer duration favours enhanced methane production and solid removal (Liu et al., 2009).

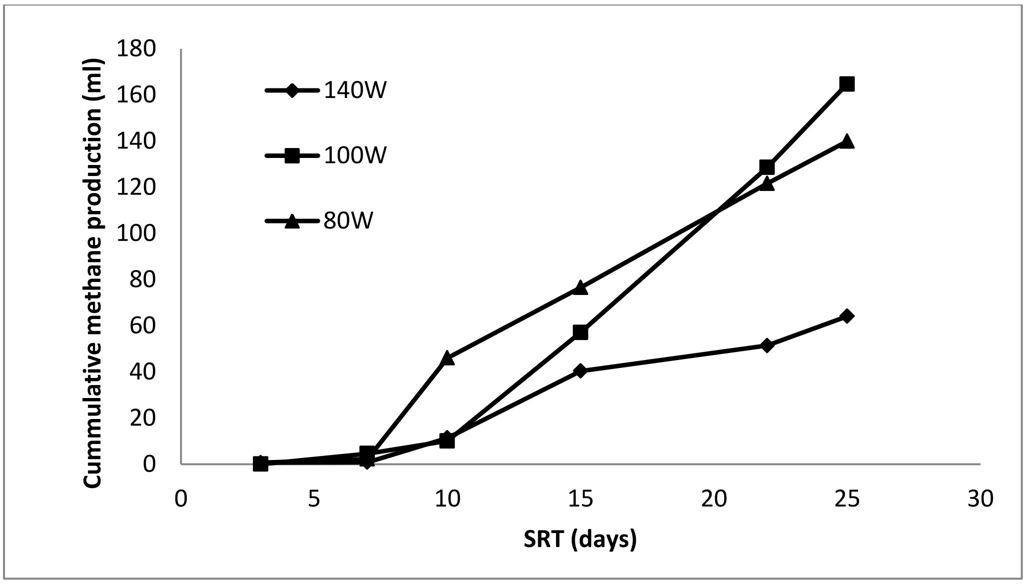


Figure 5.10 Cumulative methane production for microwave-ultrasonic pretreated mixed sludge for varying ultrasonic power. (150 W, 100W and 80 W correspond to ultrasonic intensities of 3.9 W/cm^{2,} 2.6W/cm^{2,} 2.1 W/cm^2 respectively).

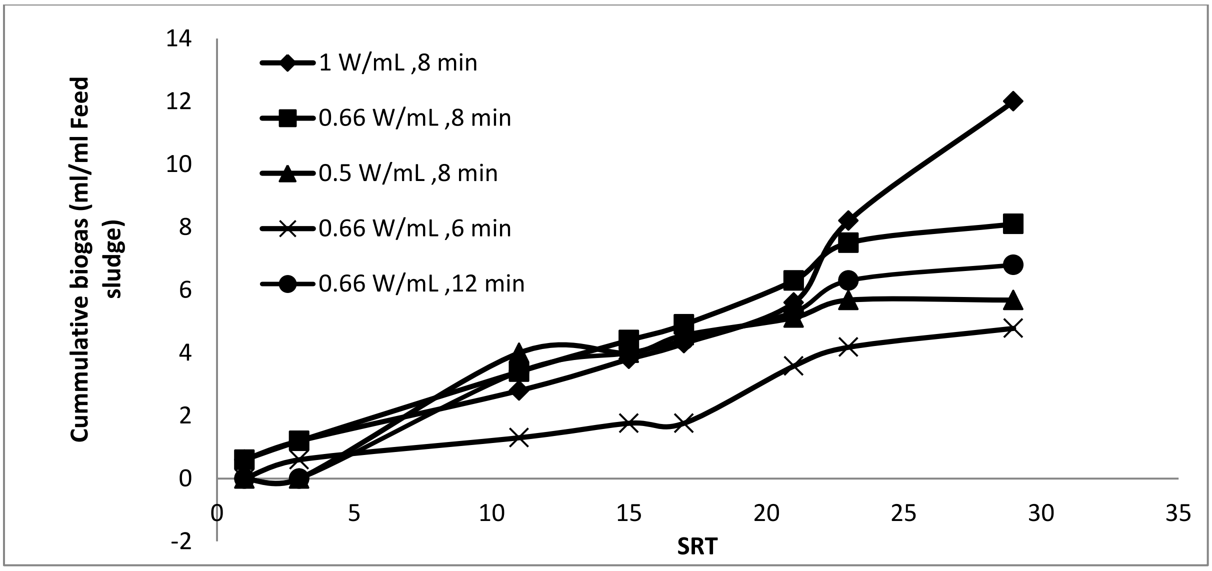


Figure 5.11 Cumulative biogas production of pretreated microwave-ultrasonic pretreated thickened excess activated sludge for varying ultrasonic density and time.

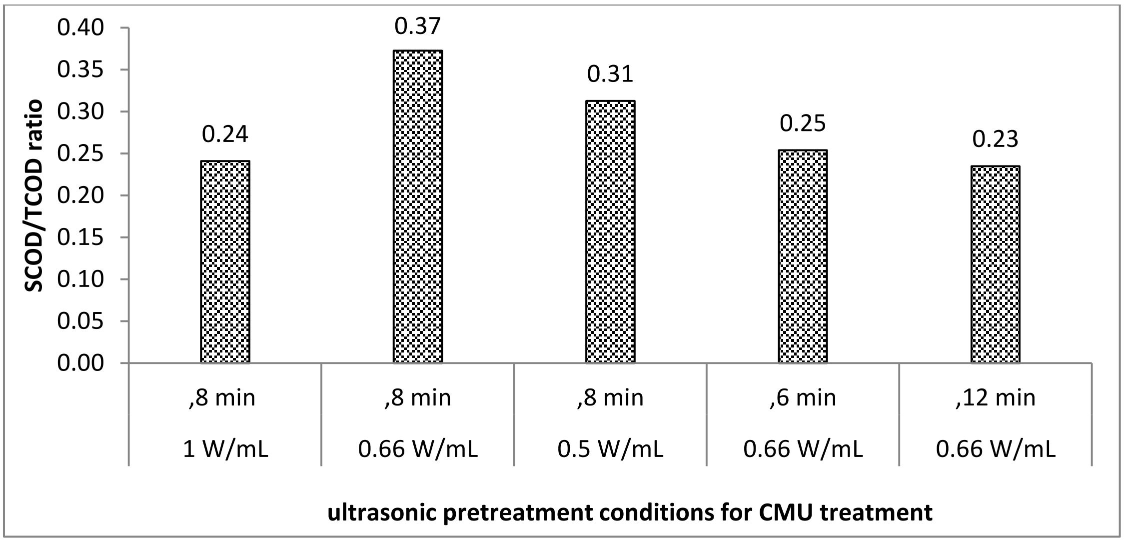


Figure 5.12 Effect of combined microwave-ultrasonic pretreatment for varying ultrasonic density and duration of pretreatment on anaerobic solubilisation of sludge.

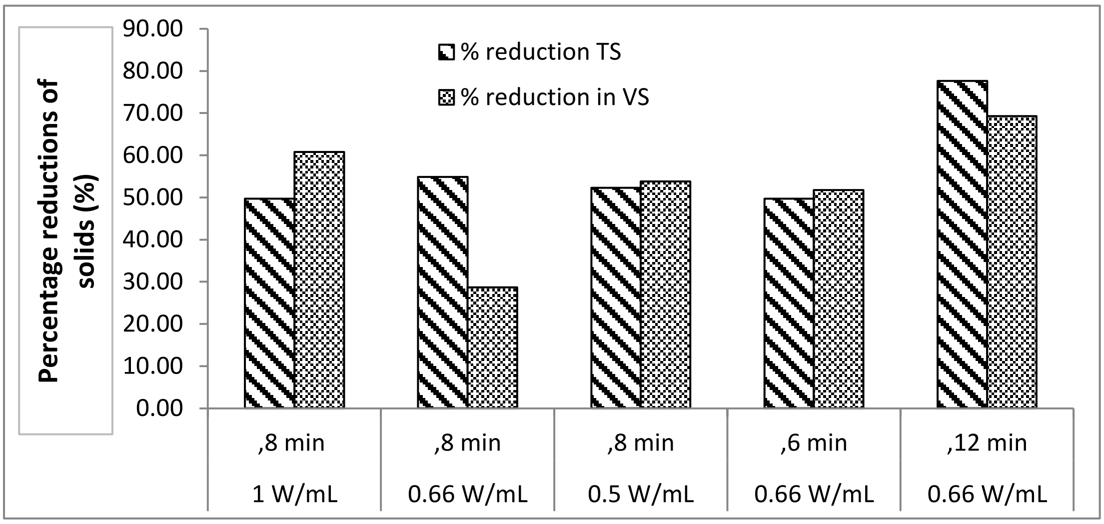


Figure 5.13 Percentage reduction of total and volatile solids for different ultrasonic density and time.

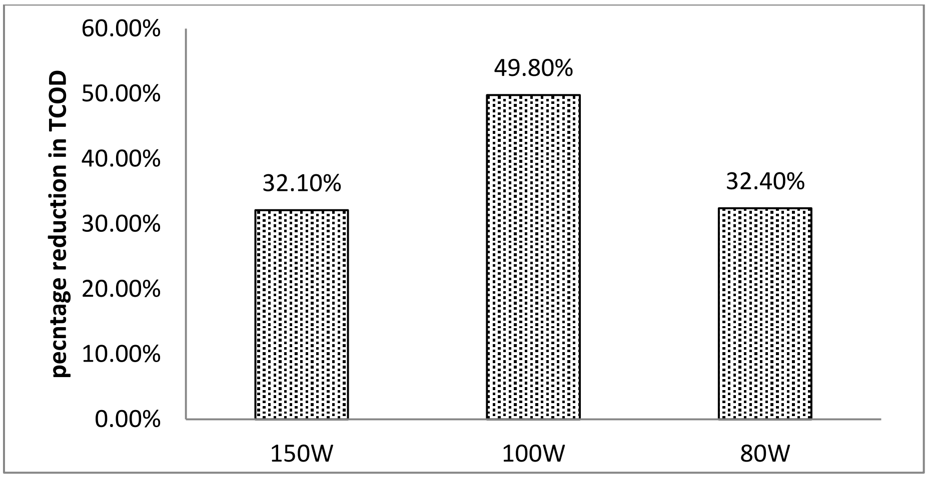


Figure 5.14 Percentage reductions in TCOD for microwave-ultrasonic pretreated sludge at different ultrasonication power.

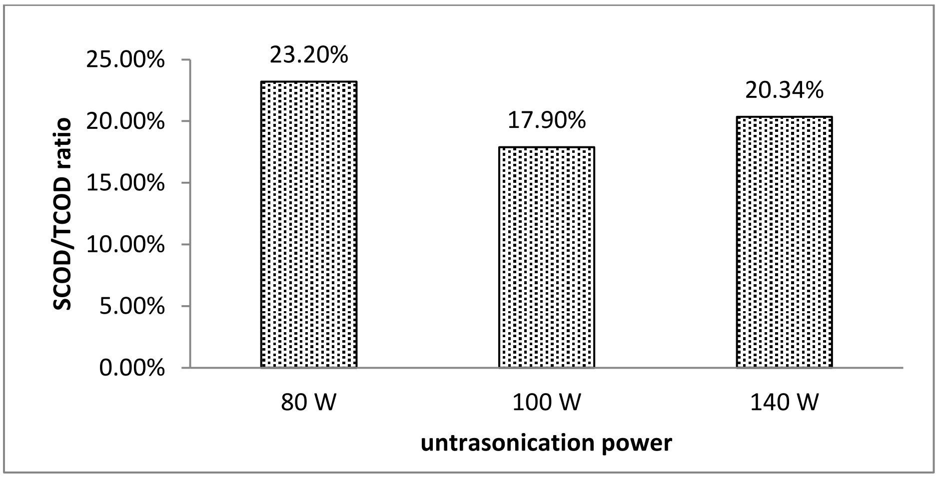


Figure 5.15 Soluble COD content of digested pretreated sludge at different ultrasonication power.

Figure 5.14 and 5.15 show that higher SCOD/TCOD ratio for the ultrasonic intensity of 80W (2.1 W/cm^2) relates well to the higher methane production in for SRT of 7-20 days. The higher TCOD percentage reduction shows how ultrasonic intensity and subsequent anaerobic digestion affects the reduction in TCOD.

The FTIR images show similar trends for sludge samples treated at 80W and 150W; bands around (1100-1000 cm-1 1070 cm-1 , 2925-2950 cm-1 1630-1650 cm-1) show increased polysaccharide, protein and fatty acid decomposition at 100 W as compared to the degradation at 80 W and 150 W (Figure 5.16).

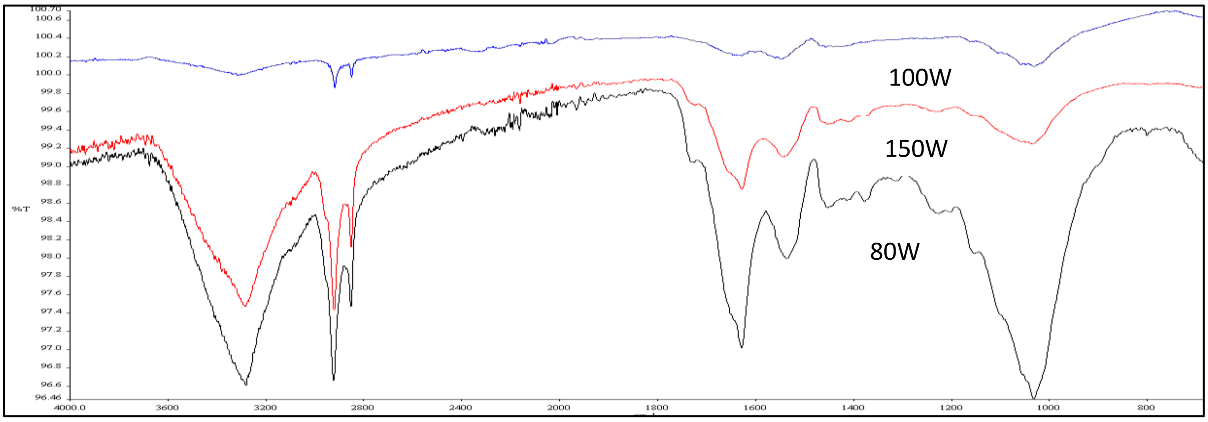


Figure 5. 16 FTIR image of digested sludge samples for the three reactors.

The summary of the overall performance for the digestion at different ultrasonication power in the combined ultrasonic microwave treatment is shown in Table5.3. The Rheological property for samples treated at different conditions is shown in chapter 9

Table 5.3 Summarized comparison on the Effect of ultrasonicaton power on combined microwave-ultrasonic treatment for mixed sludge biodegradability.

	CMU (150 W)	CMU (100W)	CMU (80W)
organic removal efficiency			
percentage reduction in TCOD	32.10%	49.80%	32.40%
biogas production and quality			
total volume of methane produced (ml/g TCOD)	12	44.5	27.9
average daily methane production (ml/day)	64.4	83.3	110
total volume of methane produced (ml/g VS)	26.9	54.8	48.1
Average SRT for stabilization (days)	25	25	25
dewaterability as CST (seconds)	239	286	144
SCOD/TCOD ratio after digestion	23.20%	17.90%	20.34%

5.4.4 Impact of microwave and ultrasonic pretreatment energy density and duration of pretreatment on sludge dewaterability.

Dewatering is an essential cost factor that greatly affects the performance of anaerobic digestion unit in a wastewater treatment plant. Yu et al. (2009) showed that microwave pretreatment with larger intensities and shorter period of time is more effective that pretreatment for a longer duration. Longer pretreatment duration increases the percentage of fine particles which are not required during sludge conditioning. Besides increased release of hydrophilic EPS that bound water contribute to the deterioration of the dewatering process. Greater level of microwave density of 6.4 W/ml and shorter duration of treatment of 3 min resulted in better dewaterability. Microwave pretreated sludge shows relatively better dewaterability than untreated or ultrasonic pretreated sludge. Eskicioglu et al, Yu et al. (2009) have reported that short duration and higher pretreatment density enhances dewaterability. Microwave pretreated TEAS for shorter duration of 1min (60s) had dewaterability of 18.6 seconds (measured in capillary suction time) which was comparatively better than the result for longer duration (Figure 5.18). Microwave pretreatment in such condition enhances sludge dewaterability and settleability by breaking the flocs into small fragments which will be reflocculated easily for improved dewaterability (Yu et al., 2009, (Tyagi et al., 2013).

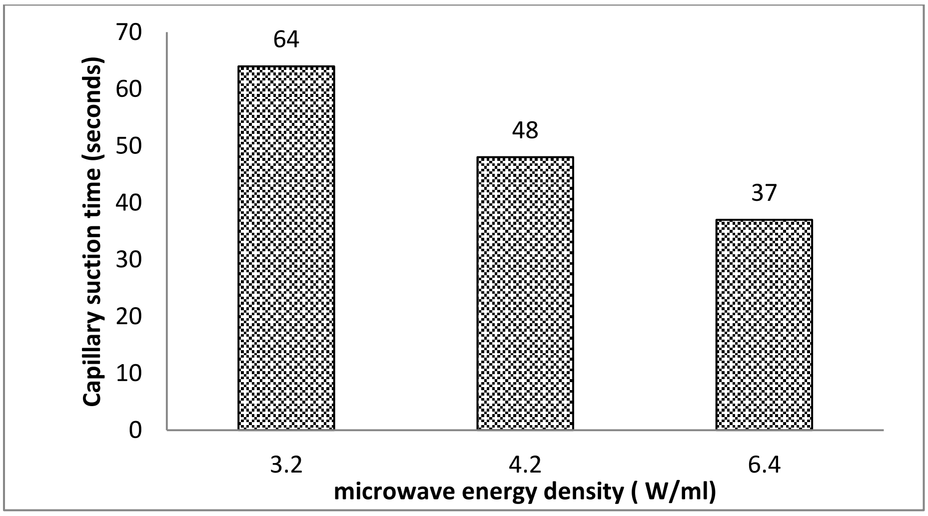


Figure 5.17 Effect of microwave energy density on dewaterability of TEAS.

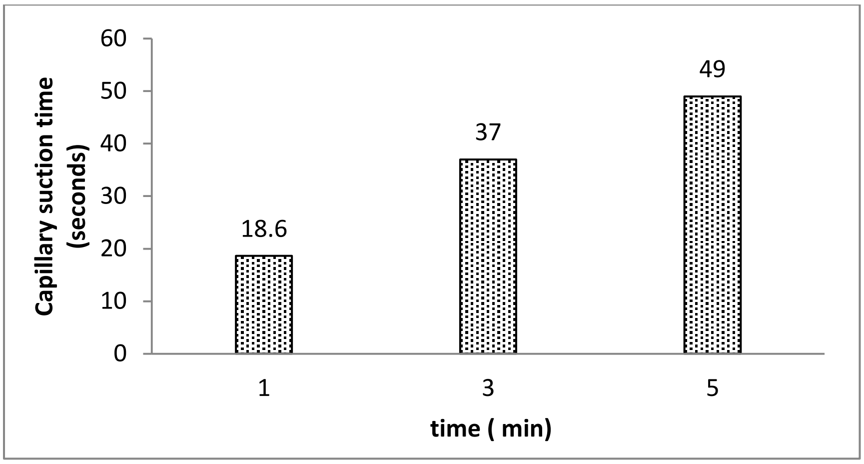


Figure 5.18 Effect of pretreatment time on dewaterability of TEAS

Ultrasonication has both negative and positive effects on sludge dewaterability. Lower ultrasonic power level with less sonication time enhances dewaterability (Pilli et al., 2011b). However the extent of solubilisation for lower ultrasonic power is limited. This shows that the selection of ultrasonic pretreatment time and power is a trade-off between sludge solubilisation and dewaterability. Figure 5.20 shows that combined microwave- ultrasonic pretreatment and anaerobic digestion of mixed sludge at lower ultrasonic density of 80 W resulted in better dewaterability, 144s (decreased CST). Dewaterability decreases with increasing ultrasonic intensity but anaerobic digestion improves dewaterability (Quarmby et al., 1999). Greater ultrasonic power increases bound water content and reduces particle size there by enhancing the surface area for the adsorption of more bound water (Chu et al., 2001). Higher ultrasonic density and intensity resulted in the deterioration of the dewaterability of sludge (Figures 5.19, 5.20, 5.21).

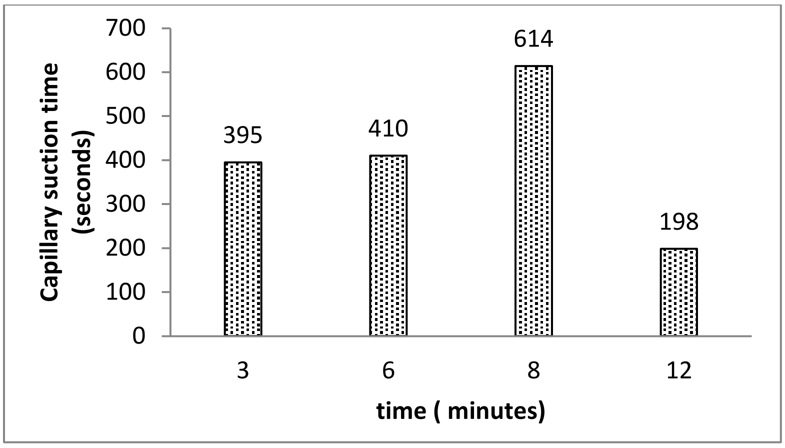


Figure 5.19 Dewaterability of ultrasonicated sludge at 1 W/ml energy density.

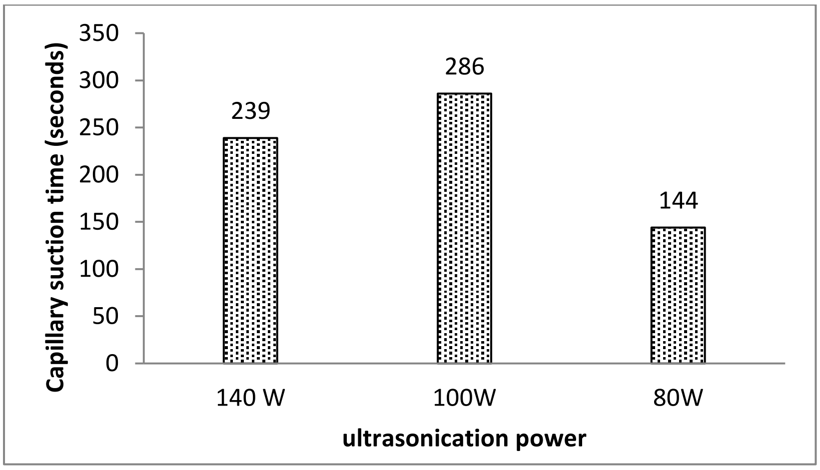


Figure 5.20 Dewaterability of microwave-ultrasonic pretreatment for ultrasonication power.

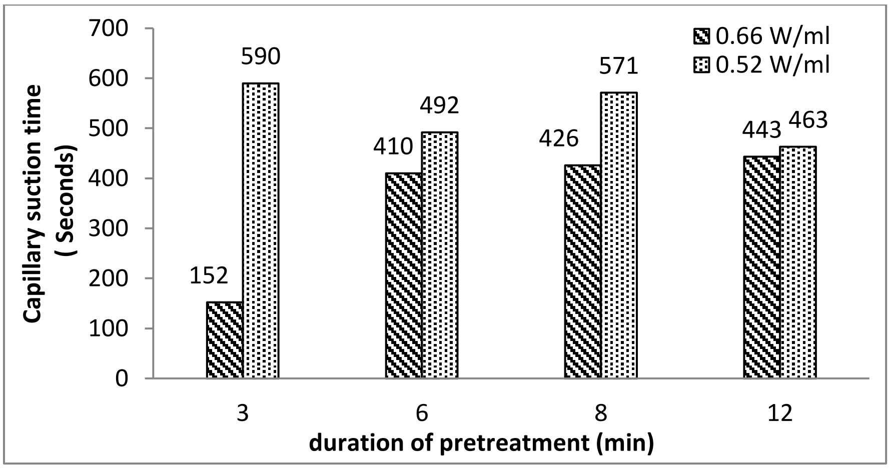


Figure 5.21 Dewaterability of Microwave-ultrasonic pretreated sludge at ultrasonic density of 0.5W/mL and 0.66 W/mL.

The increase in SCOD/TCOD ratio is associated to increased release of EPS. The increase in the concentration of EPS and soluble organics increases the viscosity of the sludge. Increased viscosity along with the thin film that EPS builds on the filter media, the dewaterability deteriorates (Oh, 2006). The relationship between SCOD/TCOD ratio and dewaterability can be represented by an exponential function. Dewaterability requires optimization of sludge disintegration for enhanced methane production and solid removal as optimum level disintegration contributes positively to the dewaterability.

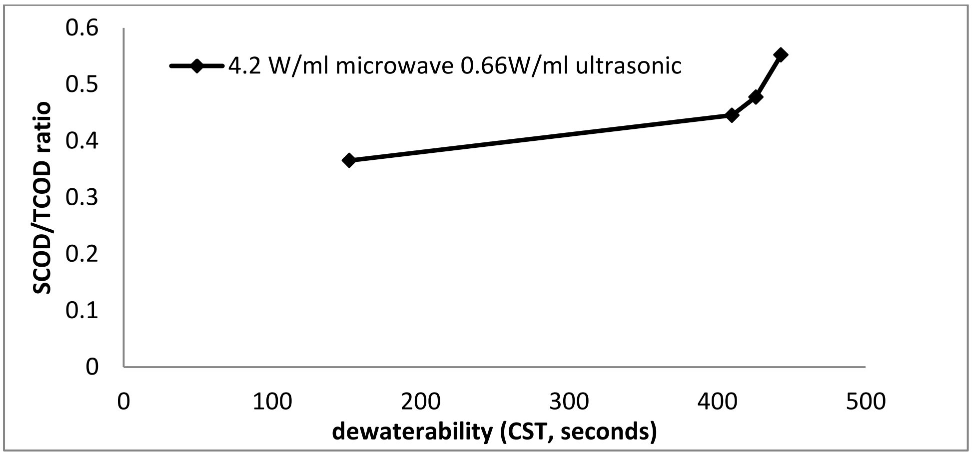


Figure 5.22 Microwave-ultrasonic pretreated sludge dewaterability versus sludge disintegration

In evaluating the effects of sonication conditions on sludge, disintegration factors such as pH also become very important. Figure 5.23 shows how pH drops down with increasing pretreatment duration. The release of soluble organics and EPS (mainly protein and carbohydrates) results in the decrease of pH. During anaerobic digestion lower pH will result in the growth of filamentous bacteria and a high pH results in build-up of unionized ammonia (Grady Jr et al., 1999).

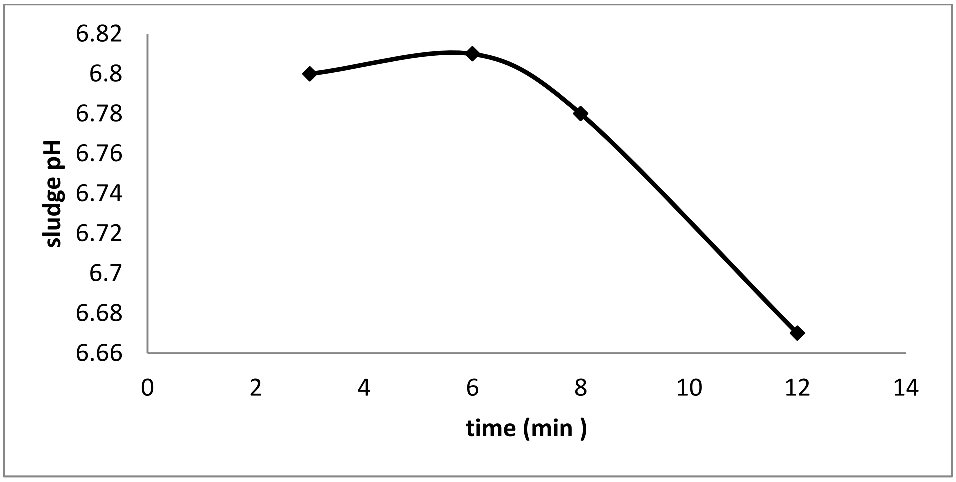


Figure 5.23 Microwave-ultrasonic pretreated sludge pH versus pretreatment duration (0.66W/ml ultrasonic + 4.2W/ml microwave

5.4.5 Microwave- ultrasonic pretreatment and dewaterability of digested sludge from BWWTP

The result of the study on the effect of microwave- ultrasonic pretreatment on the dewaterability of digested sludge from BWWTP is presented in this section. The results show that pH increases with increasing microwave pretreatment duration (Figure5.24). The dewaterability shows improvement in the first 3 minutes of the pretreatment; however, further pretreatment affects the dewaterability negatively (Figure 5.25). The percentage of nitrogen in the digested sludge is observed to be high resulting in deterioration of dewaterability due to water hold up by the protein as a nitrogenous biopolymer.

Table 5.4 Elemental analysis of digested sludge

	% carbon	**% Hydrogen**	**% Nitrogen**	**% Sulfur**
Digested Sludge (DS)	37.88	6.609	6.384	2.418

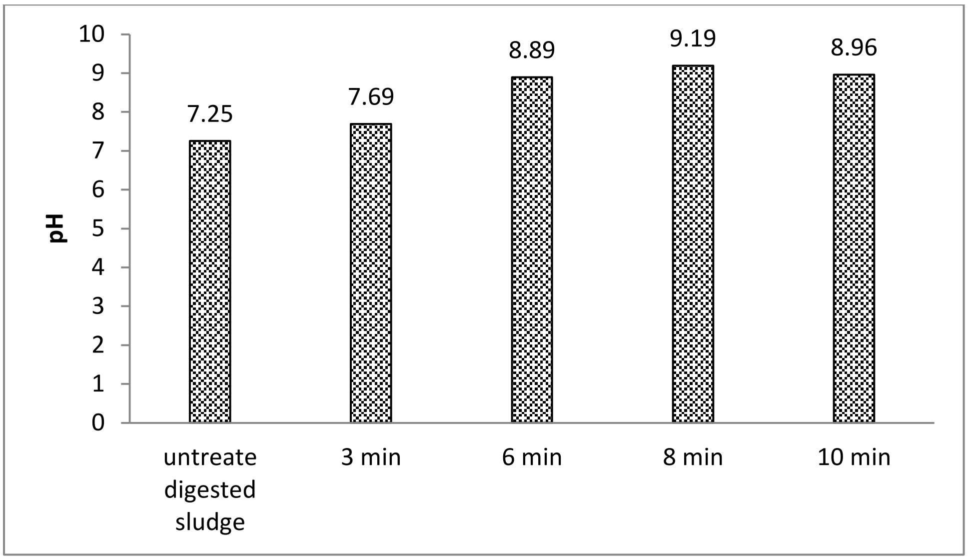


Figure 5.24 Effect of pretreatment on pH of digested sludge.

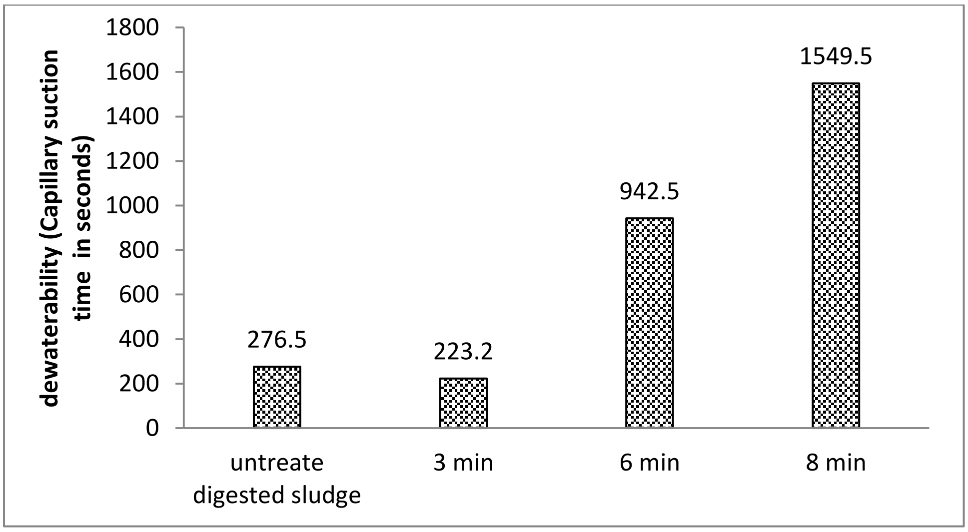


Figure 5.25 Effect of pretreatment on dewaterability of digested sludge

5.4.6 Microwave-ultrasonic pretreatment and kinetics of SCOD release

The disintegration kinetics during microwave, ultrasonic and combined microwave-ultrasonic pretreatment fit well to first-order kinetic equation for the short pretreatment duration considered in the study (Wang et al., 2005). For microwave and ultrasonic pretreatment the rate of release of SCOD becomes slow as microwave and ultrasonic disintegration time increases. For ultrasonic pretreatment a disintegration time of 12 min was chosen to establish the models. In this research, the disintegration of mixed sludge with solid content of 23.7 g/L and thickened excess activated sludge with solid content of 47.5 g/L were investigated. Ultrasonic and microwave density, pH, duration of treatment and intensity all have impact on the kinetics of sludge disintegration and dewaterability. Therefore, sludge concentration, microwave and ultrasonic density, pH and disintegration time are chosen as independent variables and SCOD+ and SCOD% are chosen as dependent variables. The mathematical forms are shown in equations 5.3, 5.4 (Fen Wang, 2005).

$$d(SCOD)/dt = K \tag{5.3}$$

$$d(SCOD\ \%)/dt = u \tag{5.4}$$

Where

$k = K_0 (I)^a (pH)^b (D)^c (X)^d$

$u = u_0 (I)^e (pH)^f (D)^g (C))^h$

K,u= rate constant

K_0, U_o = intrinsic arhenius constants = A exp ($-\Delta E^a/RT$)

D = Microwave-ultrasonic density

X= Sludge concentration

I = Microwave-ultrasonic Intensity

C = Concentration of Sludge

The values of a,b, c, d, f, g, h were determined from the plots of SCOD versus microwave and ultrasonic disintegration time, SCOD versus microwave and ultrasonic density and SCOD versus solid concentration. And the model equations were established using first order linear regression model. The kinetic model equations were established from equation 5.3 and 5.4 by integrating and taking the logarithm of the integral as given in equations 5.5 and 5.6.

$$d(SCOD)/dt = \ln(SCOD+) = \ln(k0) + \alpha \ln(I) + \beta \ln(pH) + \gamma \ln(D) + \delta \ln(C) + \ln t \tag{5.5}$$

$$\ln(SCOD\%) = \ln(k0) + \varphi \ln(I) + \nu \ln(pH) + \lambda \ln(D) + \varepsilon \ln(C) + \ln t \tag{5.6}$$

The output of the first order linear regression model provided the model expression given in equation (5.7) and (5.8) based on the experimental data for the relationship between change in soluble chemical oxygen demand to the change in pH, sludge concentration, change in pretreatment density and intensity and pretreatment time. Sludge concentration does not correlate well with SCOD% thus this factor was removed from the equation.

$$d(SCOD)/dt = 0.0954\ [D]^{0.689}\ [pH]^{1.153} [I]^{0.783} [C]^{0.858} \tag{5.7}$$

$$\ln(SCOD\%)/dt = 0.00972\ [D]^{0.584}\ [pH]^{1.205}\ [I]^{0.542} \tag{5.8}$$

5.5 Conclusion

The optimization study on microwave, ultrasonic and combined microwave-ultrasonic pretreatment has revealed that ultrasonication and microwave pretreatment power, intensity, density, duration of pretreatment and sludge concentration have significant impact on the performance on anaerobic digesters. Microwave pretreatment density of 3.2 W/ml for a duration of 5 min and ultrasonic pretreatment condition of 0.66 W/ml for a duration of 8 minute provided reasonably better result in terms of biogas production, dewaterability, solid removal and energy consumption for the pretreatment. The kinetics of pretreatment process shows that, sludge concentration, density and intensity of pretreatment and sludge pH have significant impact on the rate of the pretreatment process. Pretreatment tests for the experimental work presented in chapters 6, 7, 8 and 9 were conducted based on the optimum pretreatment conditions obtained from this chapter.

CHAPTER 6

EFFECT OF MICROWAVE AND COMBINED MICROWAVE-ULTRASONIC PRETREATMENT ON ANAEROBIC DIGESTION OF MIXED SLUDGE

Abstract

This chapter analyses the effect of Microwave (M) pretreatment in comparison to Combined Microwave-Ultrasonic (CMU) pretreatment on how the two techniques enhance anaerobic biodegradability of mixed sludge composed of 75% Primary sludge (PS) and 25% Thickened excess activated sludge (TEAS). 0.5 L of mixed sludge was subjected to microwave pretreatment at 2450 MHz, 640 W and 10 min and fed to semi-batch continuously stirred anaerobic digester at an organic loading rate of 2.75 gCOD/L day. 0.5 L of Mixed sludge sample in another digester was pretreated in two stages. The Microwave treatment took place under the same conditions stated above followed by ultrasonic treatment at a density of 0.4 W/mL, amplitude of 90%, Intensity of 3.2 kJ/g TCOD, pulse of 55/5 for 8 min. The removal of TS was 37.7 % for M pretreated sludge whereas the TS reduction for CMU pretreated sludge was 69.1%.The removal of volatile solids for CMU pretreated sludge was 21% higher than M pretreated one. The SCOD/TCOD ratio for both M and CMU pretreated sludge was 33% for 15 days of SRT however, percentage change in SCOD/TCOD ratio after 30 days of SRT for CMU pretreated sludge was 40.6% more than M pretreated sludge sample due to increased methanogenic disintegration of organics. Maximum percentage of methane produced was 71 % for CMU pretreated sludge while it was only 56 % for the M pretreated sludge. Nonetheless, the dewaterability measured in capillary suction time for M pretreated sludge was better (348 seconds) than that of CMU pretreated sludge (398 seconds) due to higher percentage of fine sludge particles in CMU pretreated sludge. The average particle size and floc size for microwave pretreated digested sludge was much bigger than that of combined microwave ultrasonic pretreated digested sludge.

6.1 Introduction

The chapter on optimization of microwave and ultrasonic pretreatment conditions (Chapter 5) has partially shown the benefits of the combined microwave-ultrasonic pretreatment over individual microwave or ultrasonic pretreatment techniques. This particular chapter provides further in-depth analysis and comparison between microwave and combined microwave-ultrasonic pretreatment techniques. Biochemical methane production potential, solid reduction capacity, gas quality, particle size distribution, dewaterability and structural sludge flocs of microwave and combined microwave ultrasonic pretreated sludge were compared. Microwave pretreatment has significant enhancement effect on anaerobic digestion performance and the quality of digested sludge produced from the process (Park and Jang, 2011). Increase in methane gas production capacity, improved solid removal, higher organic reduction and enhanced pathogen destruction rate are among the major benefits of the pretreatment or pre hydrolysis step recommended by many researchers (Eskicioglu et al., 2008a, Park et al., 2004, Park, 2011, Toreci et al., 2011, Zheng et al., 2009). More recent studies with a bench-scale industrial MW unit equipped with pressure-sealed vessels at 175 °C achieved 31% more biogas and dewaterability of pretreated municipal sludge after digestion was enhanced by 75% (Eskicioglu et al., 2009). So far, sonication of municipal biosolids have been studied at lab-scale (Bougrier et al., 2006), pilot-scale, and full-scale (Saha et al., 2011). It has been shown to be effective at solubilizing organic matter, as well as improving biogas production (Grönroos et al., 2005, Bougrier et al., 2005b). The proposed combined pretreatment technique, combined microwave-ultrasonic pretreatment (microwave coupled with ultrasonic pretreatment technique) was compared to microwave pretreatment technique which is reported by many researchers for its beneficial effects on sludge characteristics and anaerobic digester performance.

6.2 Materials and methods

The effect of Microwave pretreatment is compared to Combined Microwave-Ultrasonic pretreatment on how the two techniques enhance anaerobic biodegradability of mixed sludge composed of 75% primary sludge (PS) and 25% thickened excess activated sludge (TEAS). 0.5 L of Mixed Sludge was subjected to

Microwave pretreatment at 2450 MHz, 640 W and 10 min and fed to semi-batch continuously stirred anaerobic digester at an organic loading rate of 2.75 gCOD/L day. 0.5 L of Mixed sludge sample in another digester was pretreated in two stages. The sludge was subjected to microwave irradiation as detailed above, followed by ultrasonic treatment at a density of 0.4 W/mL, amplitude of 90%, Intensity of 150W, pulse of 55/5 for 8min. The sludge samples were characterized after the pretreatment as shown below in section 6.2.1 and introduced to the digesters.

6.2.1 Sampling and characterization

Primary sludge was collected from primary gallery underflow particularly from primary sedimentation tank No. 4 of BWWTP (Figure 3.1 and Section 3.2.1). Thickened excess activated sludge was collected from the discharge of the dissolved air floatation tank (DAFT) before mixing with primary sludge (Figure 3.1 and Section 3.2.2). Primary and thickened excess activated sludge samples were mixed with a ratio of 75% primary to 25% thickened excess activated sludge to serve as the mixed sludge to be pretreated before charging the samples to the jacketed digesters. Each of the sludge samples were characterised as shown in Table6.1 after pretreatment. The first digester was charged with microwave pretreated sludge while the second digester was fed with combined microwave-ultrasonic pretreated sludge. It can be seen from Table 6.1 that the increase in TCOD and SCOD in digester 2 is indicative of the enhancement due to the combined treatment.

Table 6.1 Characteristics of microwave and combined microwave-ultrasonic pretreated sludge

Characteristic parameter	**Digester1 (D1)**	**Digester 2 (D2)**
TS (%)	4.1	4.1
VS (%)	83	80
TCOD (mg/l)	25050	27750
SCOD (mg/l)	2200	3300
pH	7.1	7.1

6.2.2 Pretreatment and preparation of the sludge

The pretreatment and operational conditions of each digester are given in Table6.2. The first digester (D1) was subjected to microwave pretreatment while digester 2 was subjected to combined microwave-ultrasonic pretreatment at the conditions indicated in Table 6.2. Sample collection and pretreatment took place as discussed in chapter 3, Section 3.4.

Table 6.2 Pretreatment type and conditions for the mixed sludge samples fed to each of the digesters.

Digester	Pretreatment Conditions	Operational Conditions
Digester1	Microwave pretreatment: Frequency= 2450 MHz, time = 10 min, Power= 640 W, 64KJ/g	Temperature= 36- 36.5^oC pH = 6.8-7.1
Digester 2	Combined pretreatment Microwave: 2450 MHz, 640 W, 10 min (64KJ/g) ultrasonic treatment: 0.4 W/mL density, 90% amplitude (150W), 55/5 pulse, time: 9 min (3.2KJ/g)	Temperature= 36- 36.5^oC pH= 6.8-7.1

6.2.3 Experimental setup and digester operation

Mesophilic semi-batch anaerobic digestion took place in two digesters for a total SRT of 45 days. The digester working volumes were 500ml and the organic loading rate was 2.75 g TCOD/day for both digesters. The results from two continuously stirred, semi-continuous jacketed digesters from the four digester setup shown in section 3.4.3 Figure 3.7 were used for the microwave-ultrasonic versus microwave comparison test. Jacket heating system was applied to maintain the desired mesophilic digester temperature of 36.5°C. The digesters were placed on magnetic stirrers to maintain continuous mixing. Sludge was fed to the digesters through the sludge charging tube and the biogas produced passes through a 1L buffering bottle, placed outside the water bath heater, for removal of any condensates. Water displacement method was used to measure the gas volume and the biogas composition was measured using GA Plus 2000 Biogas Analyser as shown in Section 3.4

Digester 1 was fed with microwave pretreated sludge while digester 2 was charged with combined microwave-ultrasonic pretreated sludge. The digesters were continuously purged with N_2 at 25-40mL/min after the charging.

The size distributions of the flocs were determined by a Mastersizer 2000 with lens which enables the measurement of particles in the range 0.02-2000μm. This instrument measures the size of particles by means of light scattering. It utilizes dual-wavelength detection system. A short wavelength blue light source is used in conjunction with forward and backscatter detection. The sludge samples were exposed to He–Ne laser and a refractive index of 1.58 was used for the sludge test.

The microstructure of sludge flocs was examined by light microscopy and images were captured on Olympus LG-PS2 Optical Microscope equipped with an Olympus digital camera and image pro plus 5.1 software.

The digesters were operated for a total SRT of 45 days and the results in the first 20-25 days were used for the analysis. Periodically, the volume and composition of the gas produced was measured and recorded. The gas composition was measured by connecting the Gas Analyser probe to the inlet tube of the buffering bottle to pump out the biogas from the digesters.

6.2.4 Analytical methods

All the analysis required for the experimental work in this section including determination of TS, VS, SCOD, TCOD, pH, dewaterability (CST), elemental analysis, particle size analysis, optical microscope imaging, FTIR imaging and SEM imaging were based on the methods and techniques presented in sections 3.3.1 through 3.3.15.

6.3 Results and discussion

The performance of anaerobic digesters was studied for microwave and combined microwave-ultrasonic pretreated sludge samples. Biochemical methane potential and biogas quality test results are reported in this section. Solid reduction, sludge solubilisation and biodegradability tends, particle size distribution and dynamics were analysed for different sludge retention time. The appearance and structure of the sludge and its dewaterability is also reported in this part.

6.3.1 Biochemical methane production potential and biogas quality

The cumulative methane production (ml/g TCOD) for microwave-ultrasonic pretreated mixed sludge was greater than microwave pretreated sludge sample by 18% after a sludge retention time of 22 days as shown in Figure 6.1. Chu et al., (2001) reported 17% improvement in methane yield ml/g COD) for combined microwave-alkali pretreatment of thermophilic anaerobic digester feed sludge. The methane production improvement in this particular research for combined microwave-ultrasonic pretreatment is better than what others have reported for the combination of microwave irradiation with other pretreatment techniques (Tyagi and Lo, 2013a). (Chi et al., 2011) reported 14% increase in methane production for microwave-alkali pretreatment as compared to the control. Interestingly the cumulative biogas production for combined microwave-ultrasonic pretreated sludge is higher than that of untreated sludge by 43%. Digester kinetics and methane production trends for combined microwave-ultrasonic pretreated sludge show that the hydrolysis rate of combined microwave-ultrasonic pretreated sludge was faster proving the additional enhancement effect of ultrasonication. Moreover, quality of biogas is one essential factor showing the performance of anaerobic digesters.

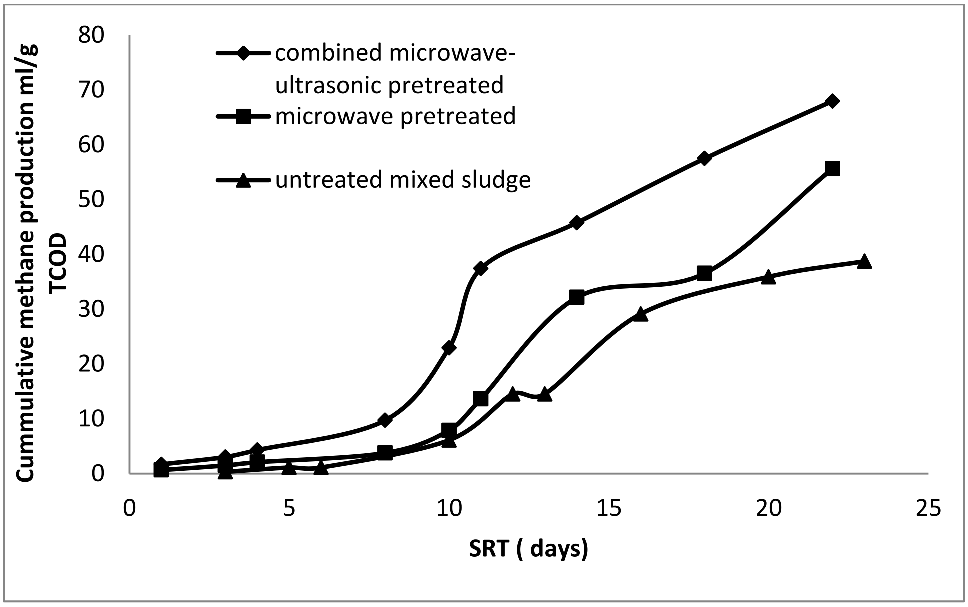
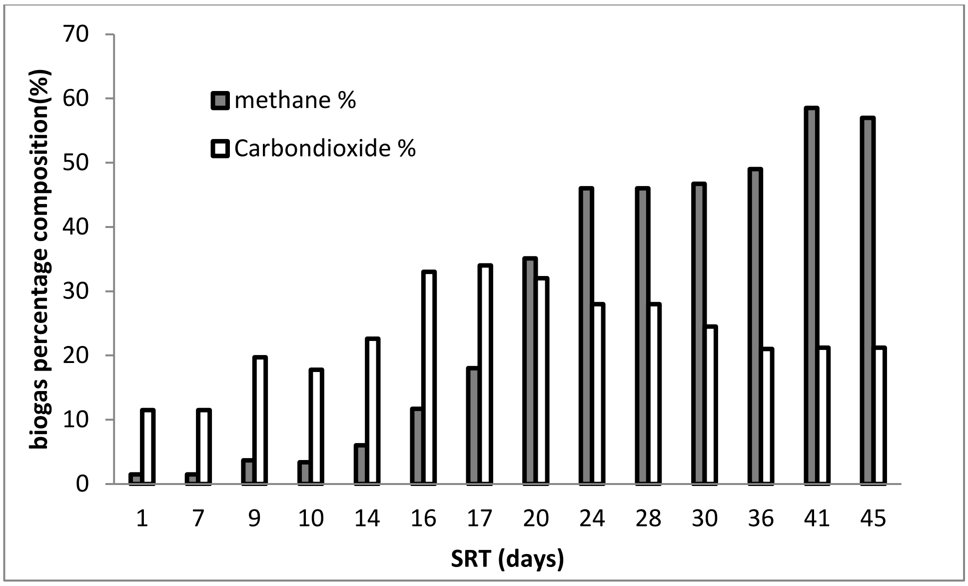
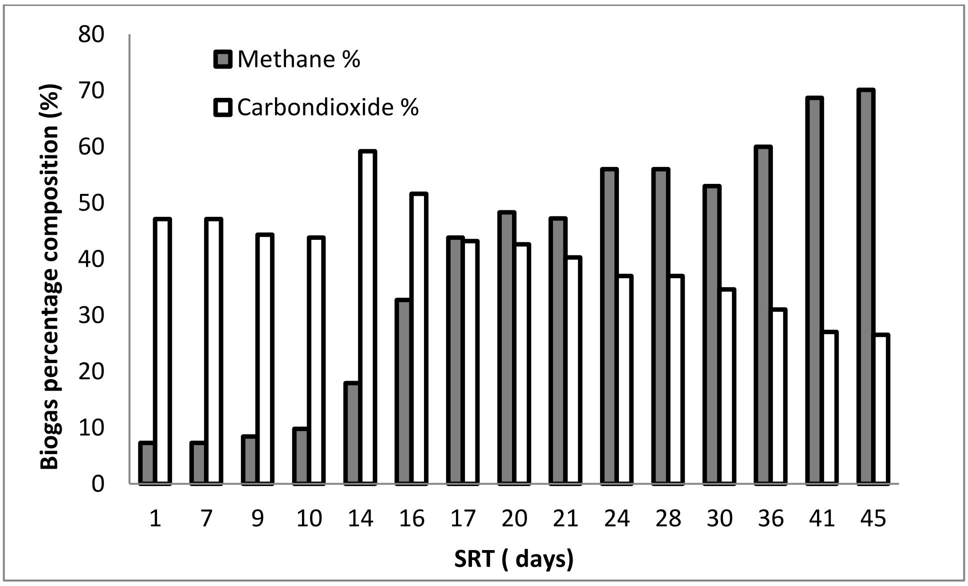


Figure 6.1 Cumulative methane production of untreated, microwave and combined microwave-ultrasonic pretreated mixed sludge

Methane/carbon dioxide ratio indicates how efficiently and healthily the digester is working. The methane/carbon dioxide ratio of both the combined microwave-ultrasonic pretreated and microwave pretreated sludge anaerobic digesters reached 1.5 after 24 days of SRT. Similarly, the percentage of methane for combined microwave-ultrasonic pretreated sludge was 56 % when the percentage composition of the microwave pretreated sludge was 51% with increasing sludge retention time the percentage composition for the combined microwave-ultrasonic pretreatment reached up to 70 % and that of the microwave pretreated 58.5 % (Figure 6.2). Combined microwave-ultrasonic pretreatment makes the digester achieve higher methane composition at shorter sludge retention time with the carbon dioxide concentration decreasing correspondingly (Figure 6.2 b). The enhancement in methane production and biogas quality shows the increased methanogenic activity. The carbondioxide concentration was progressively reduced as methane production increased contributing to better gas quality. The percentage of CO_2 from microwave pretreated sludge was slightly smaller (better quality) than the CO_2 composition of combined microwave ultrasonic pretreated sludge after 35 days of SRT. This is because of higher CO2 production at an earlier stage of the digestion of combined microwave-ultrasonic pretreated sludge due to enhanced microbial activity which takes time for removal (Figure 6.2 b).



(a)



(b)

Figure 6.2 Methane/carbondioxide ratio for different sludge retention time in (a) digester 1 and (b) digester 2

6.3.2 Sludge biodegradability in microwave and combined microwave ultrasonic pretreated digesters.

Soluble COD content is a measure of biodegradability or the extent of solubilisation of organics for anaerobic degradation. Percentage change in SCOD/TCOD ratio for combined microwave-ultrasonic pretreated digested sludge was 47 % higher than the ratio for microwave pretreated digested sludge after 32 days of SRT as shown in Figure 6.3. The extent of sludge solubilisation during the first 15 days was similar for both sludge types. Higher SCOD/TCOD ratio and enhanced SCOD reduction during anaerobic digestion in the case of combined microwave-ultrasonic pretreatment indicates increased solubilisation or release of organics achieved by the disintegration of flocs, disruption of cells and rapid internal heating due to microwave irradiation and further cavitation and floc disruption and organic reduction due to release of free radicals during ultrasonication. Figure 6.4 shows that the removal of TCOD for combined microwave-ultrasonic pretreated sludge was enhanced by 31.4% as compared to the TCOD reduction for microwave pretreated sludge.

Table6.3 shows that percentage reduction of total solids was 37.7% and 69.1% for microwave pretreated and combined microwave-ultrasonic pretreated sludge respectively. Similarly, the percentage reduction in volatile solids was 37.3% and 58.4 % for microwave pretreated and combined microwave-ultrasonic pretreated digested sludge (Table 6.3). An increase of 31 % in total solid removal and 21% in volatile solid removal for Combined pretreatment is mainly due to the disintegration of the complex floc structure of the sludge and enhanced hydrolysis as shown in section 6.3.4. The combined pretreatment has assisted to improve the biodegradability. Microwave treatment results in efficient and faster cell disruption due to rapid internal heating; whereas the ultrasonication causes cavitation, floc size reduction and encourages formation of highly reactive radicals that facilitate destruction of organics. The effect of enhancement in volatile solid removal and SCOD solubilisation can also be observed from the improvement in methane production and digestion kinetics.

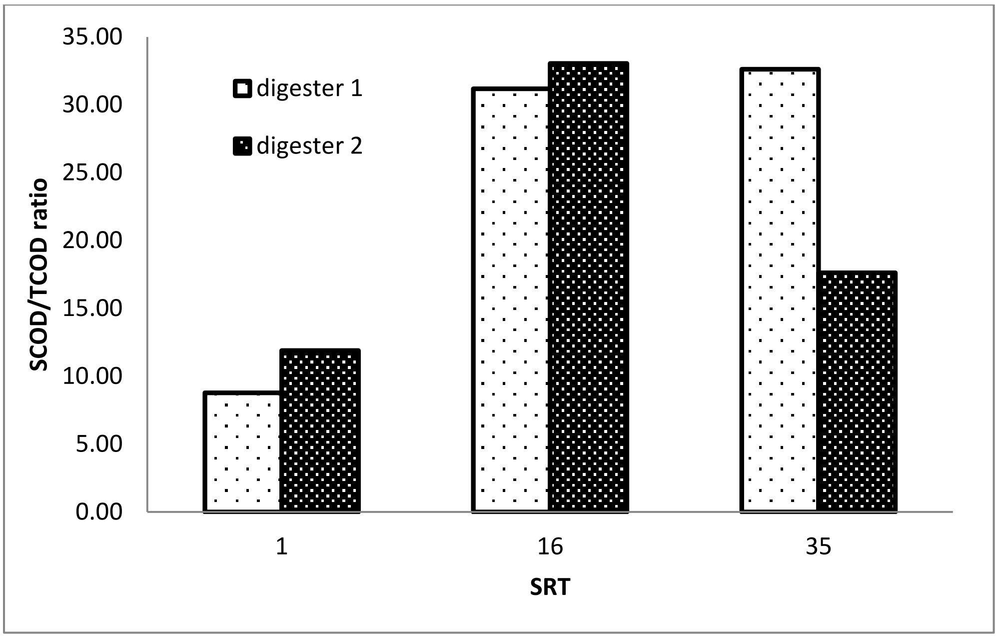


Figure 6.4 Soluble chemical oxygen demand to total oxygen demand ratio as a function of SRT

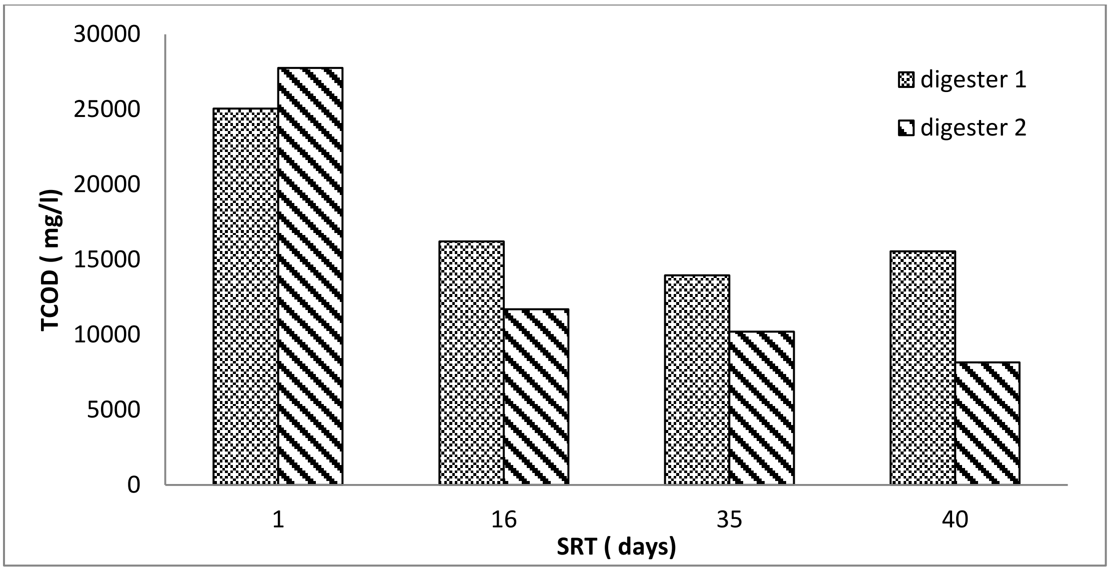


Figure 6.3 Reductions in TCOD for microwave treated (digester 1) and combined microwave-ultrasonic pretreated sludge (digester 2) as a function of SRT.

Table 6.3 Total and volatile solid content of feed and digested mixed sludge.

	Feed TS	**Effluent TS**	**Feed VS**	**Effluent VS**
digester 1	2.4	1.7	83	71.4
digester 2	4.1	1.9	79	61

6.3.3 Dewaterability of microwave and combined microwave ultrasonic-pretreated digested sludge.

Dewaterability is a function of particle size of the flocs and the hydrophilicty of biopolymers released due to the disintegration of microbial cells as discussed in chapter 7 section 7.7.3. studies show that the characteristics of the digested sludge flocs affect the dewaterability, especially the floc particle size distribution has a significant impact on dewaterability of sludge as shown in chapters 4 and 5 (Hanjie, 2010). Previous studies about the effect of flocculating ability of sludge flocs have shown that strongly flocculated particles have higher degree of compressibility of activated sludge determined as sludge volume index (Lay et al., 1999). They found out that flocculation mechanism or the internal forces produced by molecular and electrostatic interactions have the ability to enhance the water binding ability of the sludge flocs. This is an important factor affecting dewaterability. Figure 6.5 shows that microwave pretreated digested sludge (digester 1) shows better dewaterability (smaller CST value) as compared to combined microwave-ultrasonic pretreated digested sludge. The combined microwave-ultrasonic pretreatment resulted in the decrease of average size of flocs as shown in the optical microscope and SEM images and increases release of biopolymers which may trap water and limit the dewaterability. With increasing values of flocculating ability, hydrophobicity and negative surface charge, both bound water and CST tended to increase. The change in floc structure and colloidal charge because of the pretreatment may have also contributed to the reduction in dewaterability. However, the dewaterability shown here for both pretreatment conditions is better than the dewaterability of untreated and pretreated mixed sludge reported in chapter 7. Pretreatment to a limited extent enhances dewaterability, the deterioration in the combined pretreatment happened due to sonication which significantly decreases dewaterability (Eskicioglu et al., 2007b, Saha et al., 2011a).

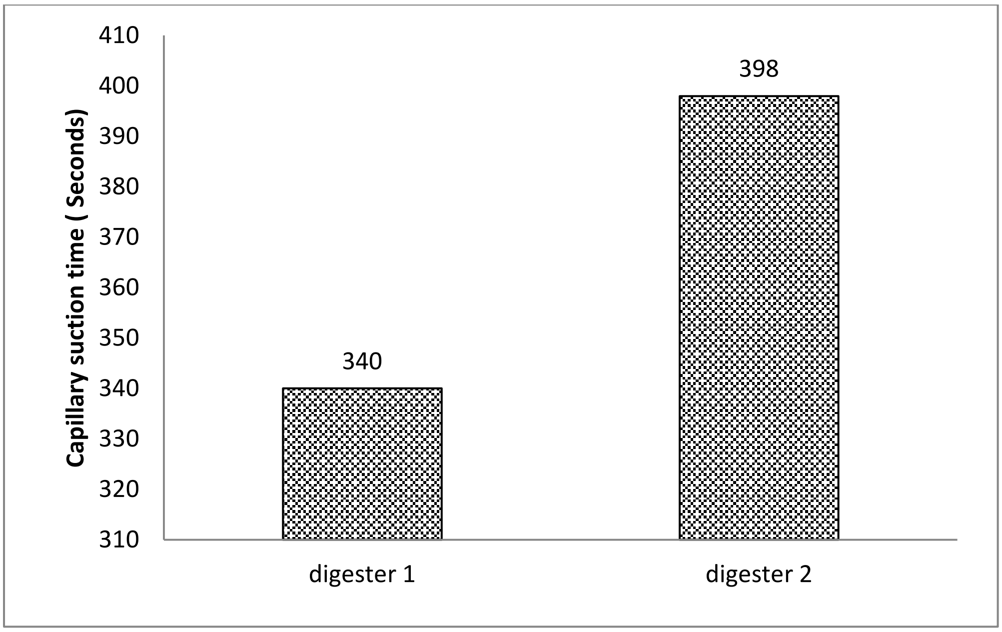
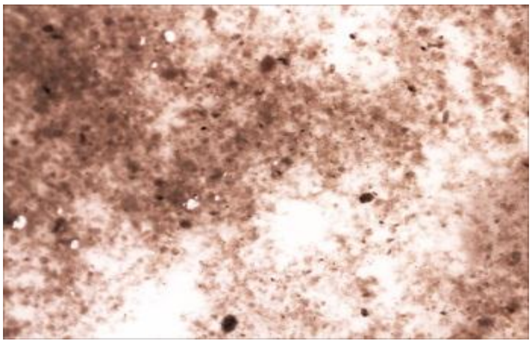


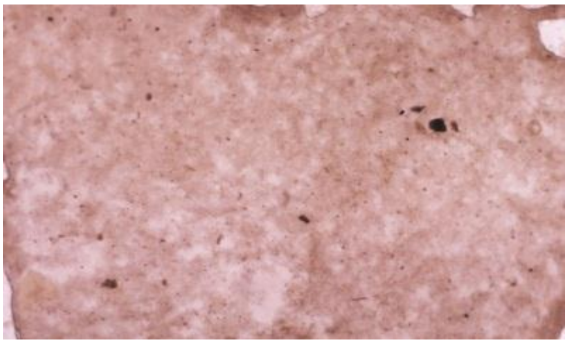
Figure 6.5 Dewaterability of microwave pretreated (digester 1) and combined microwave-ultrasonic pretreated (digester 2) digested mixed sludge.

6.3.4 Structure of microwave and combined microwave-ultrasonic pretreated sludge under optical and scanning electro-microscope.

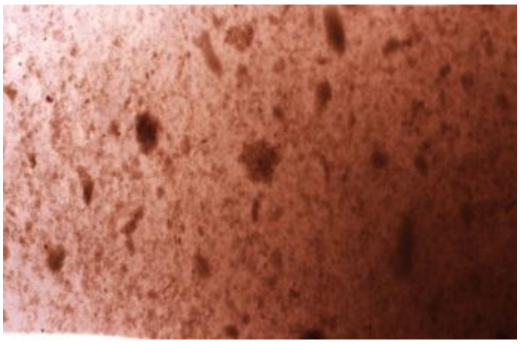
Figures 6.6 and 6.7 show optical and scanning electro-microscope images of microwave and combined microwave-ultrasonic pretreated sludge flocs. The optical microscopic images of Figure 6.6 show that combined microwave-ultrasonic pretreated sludge samples have undergone greater degree of sludge disintegration and floc disruption as compared to microwave pretreated sludge samples. Sonication in the combined pretreatment step has significant impact on the floc structure and particle size of sludge. Microwave pretreated sludge (Figure 6.6 a) shows bigger floc sizes and denser appearance as compared to microwave-ultrasonic pretreated sludge (Figure 6.6 b). Microwave pretreated digested sludge sample (Figure 6.6 c) resulted in further loosening effect due to the anaerobic digestion process and release of extra cellular Polymeric substances which breakdown the sludge floc due to enzymatic digestion effects. The votex mixing during digestion also contributes to the floc disruption process. Combined microwave-ultrasonic pretreated sludge samples in Figure 6.6 b and d show further enhancement to what was observed in the microwave pretreated sludge samples. The enhancement is due to the ultrasonication after the microwave pretreatment which resulted in significant disruption of the floc structure due to the hydrodynamic shear forces and the cavitation effect during sonication. The SEM images in Figure 6.7 a,b,c,d,e show floc structure and microscopic appearance of untreated mixed, microwave pretreated and digested, combined microwave-ultrasonic pretreated and digested sludge samples. The SEM images provide a more detailed view of the sludge flocs compared to the optical microscope images. Figure 6.8 shows that the degree of disruption and morphology of microwave pretreated sludge is less intense (coarser appearance and larger flocs) as compared to the disruption in the combined microwave-ultrasonic pretreatment. The effect of sonication on floc disruption and particle size is significant. The cell disruption in combined pretreatment is more significant because of the sonication impact in disrupting the microbial cells. The methanogenic archae is observed to have greater concentration in combined microwave-ultrasonic pretreated digested sludge (Figure 6.7 e).



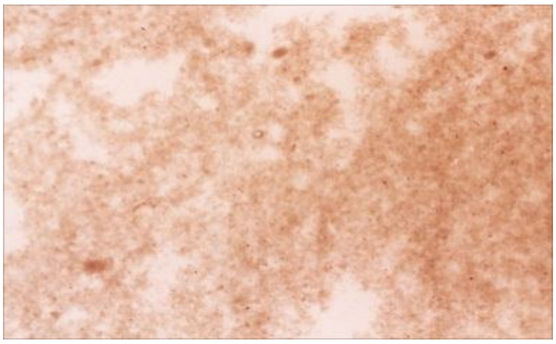
(a) Digester 1 microwave pretreated



(b)Digester 2 microwave-ultrasonic pretreated

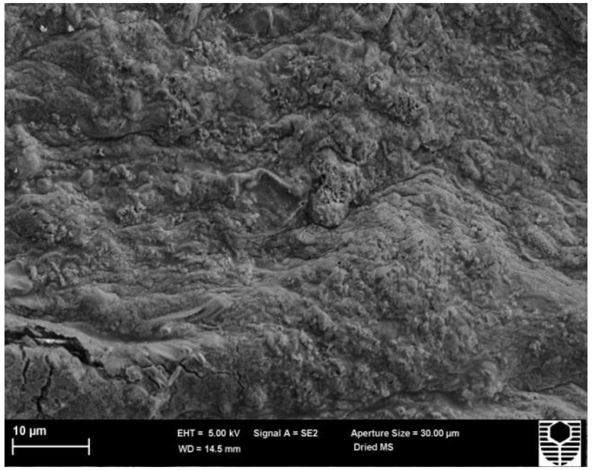


(c) Digester 1, microwave pretreated at 22 days of SRT

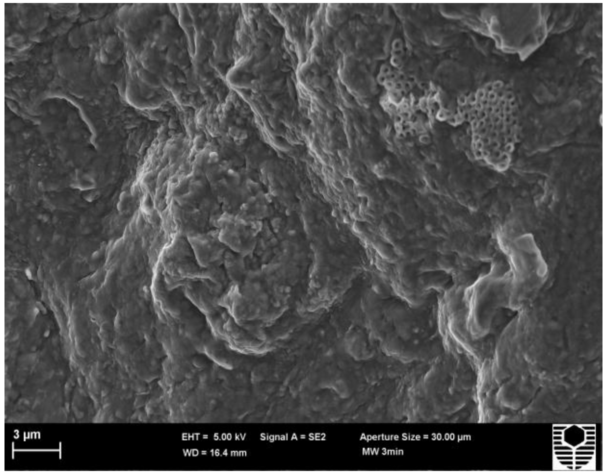


(d) Digester 2, combined microwave-ultrasonic pretreated at 22 days of SRT

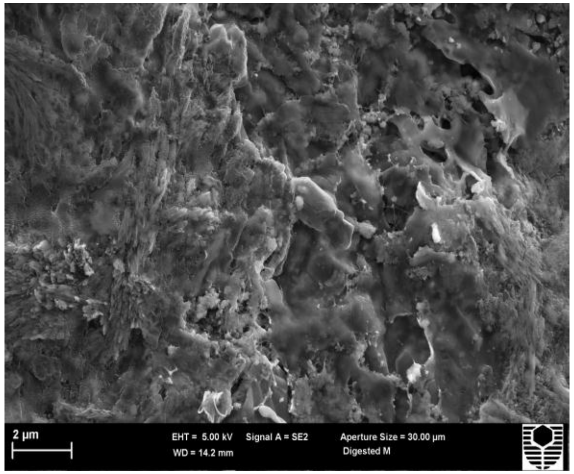
Figure 6.6 Optical microscope images of Sludge flocs for microwave pretreated and combined microwave ultrasonic pretreated sludge



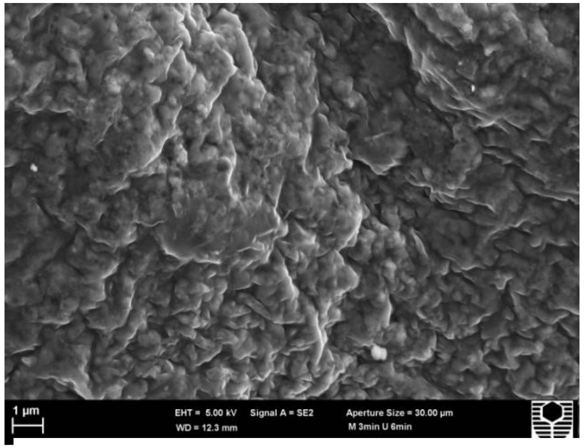
dried mixed sludge **(a)**



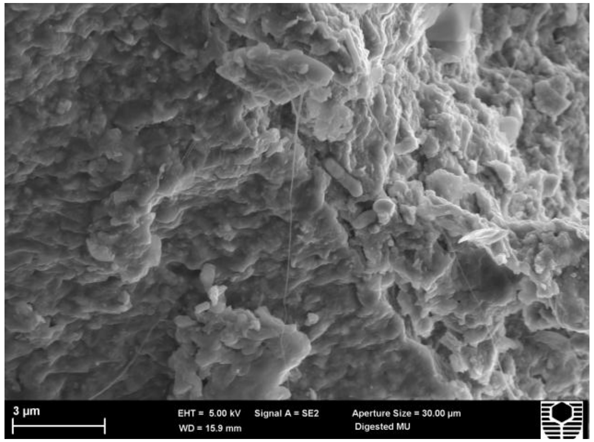
Microwave pretreated sludge **(b)**



Microwave pretreated digested sludge **(c)**



Microwave-ultrasonic pretreated sludge **(e)**



Microwave-ultrasonic pretreated digested sludge **(e)**

Figure 6.7 Scanning electromicro scope (SEM) images of untreated mixed sludge, microwave pretreated and combined microwave-ultrasonic pretreated sludge before and after digestion.

6.3.5 FTIR-ATR spectra of microwave and combined microwave-ultrasonic pretreated sludge.

Figure 6.8 shows FTIR-ATR images for microwave and combined microwave ultrasonic pretreated digested sludge. In the band range between 3600 to 3200 cm^{-1}, OH functional groups of carboxylic acids, alcohols, phenols and water are observed for both digested sludge types. Similarly, stretching aliphatic groups with very high degree of aliphaticity are represented in the FTIR bands around 2920-2930 cm^{-1} for both sludge types.

In the band range between 1600-1500 cm^{-1}, hydrogen bonded to C=O carbonyl groups of primary amides and a lower band of 1520-1540 cm^{-1} show NH_2 deformation amides for microwave treated, and combined microwave-ultrasonic treated digested sludge samples. The specific bands of 1630-1650 cm^{-1} show C=C bonds in aromatic groups and C=O in ketone, amide and Quinone groups for microwave treated and combined microwave-ultrasonic treated digested sludge samples.

FTIR bands around 1100-1000 cm^{-1} represent the occurrence of polysaccharides which are of reasonably lower intensities for microwave pretreated digested sludge samples confirming enhanced disintegration of polysaccharides. Weaker intensity of bands around 2920-2950 cm^{-1} for combined microwave-ultrasonic pretreated sludge as compared to microwave pretreated sludge shows greater decomposition of fatty acids and lipid components in case of the combined pretreatment. The weakening of bands of amide I and carboxylates around 1630-1650 cm^{-1} and primary and secondary amines around 1298 cm^{-1} particularly with combined microwave-ultrasonic treated digested sludge and amide III components around 1240-1250 cm^{-1} confirm the enhancement in the decomposition of proteins. Weaker bands of secondary amines around 1550-1560 cm^{-1} for combined microwave-ultrasonic pretreated digested sludge further justify the enhancement effect in protein degradation. Increased degradation of protein, humic acid and other organics in case of combined microwave-ultrasonic pretreatment is in agreement to higher sludge solubilisation and COD removal for the combined pretreatment. Moreover, enhanced protein degradation with greater percentage of combined microwave-ultrasonic pretreated TEAS presented in chapter 8 agrees well with the FTIR-ATR results (enas Shimidt, et al. 2011)

Proteins are believed to improve floc formation, but high concentrations may lead to poor settling and compaction properties (Show et al., 2007, Lay et al., 1999). The deterioration in dewaterability for combined microwave-ultrasonic pretreatment is partly due to increased solubilisation of proteins which may trap more bound water.

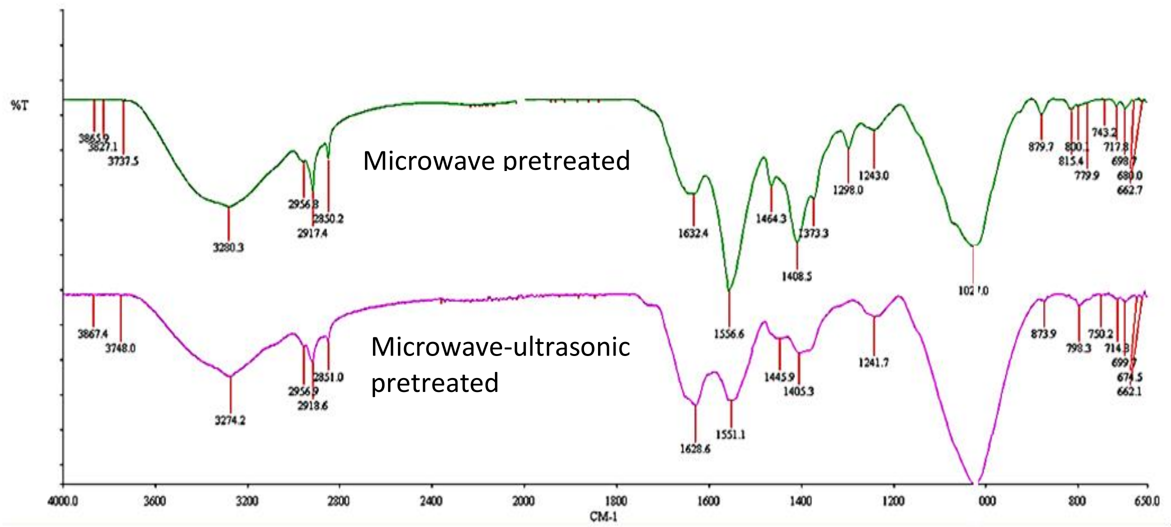


Figure 6.8 FTIR spectroscopic images of microwave pretreated and combined microwave-ultrasonic pretreated digested sludge.

6.3.6 Particle size distribution of microwave and microwave-ultrasonic pretreated sludge samples at different SRT in the digestion process.

The particle size of solids decreases and becomes more uniform after pretreatment based on the principle of disintegration of sludge (Saha et al., 2011a). In this particular study the effect of the two pretreatment techniques on the particle size distribution and average surface area of the particles were compared. The average particle size decreases with increasing SRT. For combined microwave ultrasonic pretreated sludge 50% of the sludge particles (d(0.5) were under the size range of 51.1 μm in the feed which decreased to 36.6 μm after 12 days of SRT and to 34.8 μm after 25 days of SRT. On the other hand, the average particle size d(0.5) of microwave pretreated sludge feed was 92 μm (much bigger than the CMU pretreated sludge (Table6.3). The particle size for the microwave pretreated sample reduced to 48 μm in 12 days of SRT and further down to 40.3 μm after 25 days of SRT. The greatest reduction in particle size took place in the first 12 days; further reduction in particle size after 12 days of SRT was less significant indicating the homogenization and stabilization of the sludge particles. Smaller sludge particles have greater surface area which enhances intimacy of contact and mass transfer between phases resulting in better sludge solubilisation, digester performance. The average surface area of both microwave and combined microwave ultrasonic pretreated particles increased with increasing SRT. To the contrary, reduction in particle size contributed negatively to the dewaterability of the sludge. Smaller sludge particles resulted in densification and higher resistance to the flow of water. The reduced dewaterability for CMU pretreated sludge with the smaller digested sludge particles shown in (Figure 6.5) justifies this. Reduction in particle size may also result in release of extracellular polymeric substances which may trap more bound water hindering the separation of water from the sludge during filtration.

Table 6.4: Particle size distribution of microwave pretreated and combined microwave-ultrasonic pretreated digested sludge samples.

Method of pre-treatment	d(0.1) (μm)	d(0.5) (μm)	d(0.9) (μm)	Specific surface area (m^2/g)	Surface-weighted mean (μm)	Volume-weighted mean (μm)
Microwave-ultrasonic treated feed	9.2	51.1	569.31	0.26	22.7	173
Microwave-ultrasonic treated (day 12)	9.2	36.6	163.72	0.29	20.7	77.3
Microwave-ultrasonic treated (day 25)	9.7	34.8	216	0.29	20.7	90.8
Microwave treated feed	23.1	92.0	2.5	0.12	49.5	139.0
Microwave pretreated (Day 12)	13.1	48.0	131.92	0.22	26.6	70.0
Microwave pretreated (Day 25)	12.8	40.5	710	0.24	24.8	171.3

6.4 Conclusions

Combined microwave-ultrasonic pretreatment significantly enhanced sludge biodegradability and anaerobic digestion process compared to microwave pretreatment. Microwave pretreatment was reported by many researchers to be very effective in terms of enhancing rate/extent of biodegradation, solids reduction and pathogen removal. The combination of microwave pretreatment with other techniques further enhances the beneficial effects by reducing the microwave pretreatment costs. In this chapter, a combination of microwave pretreatment with ultrasonic pretreatment was compared to microwave pretreatment. Interestingly, the combination of the microwave irradiation with sonication as a sludge pretreatment step has resulted in enhancement of hydrolysis rate, TCOD removal volatile and total solids reduction, methane production and biogas quality. The floc structure and particle size were smaller in the combined microwave-ultrasonic pretreatment due to the cavitation effects and hydro mechanical shear forces which reduce floc size and enhance release of organics and radicals important to improve the biodegradability. However, the dewaterability slightly deteriorated in case of combined microwave-ultrasonic pretreatment due to greater size reduction and floc disruption which causes compaction and increase in the amount of bound water trapped within solubilized organics and EPS. Analysis on combined microwave-ultrasonic pretreatment in section 5.3.6 shows the relative advantages of combined pretreatment over individual microwave only, ultrasonic only or other pretreatment techniques from literature. Hence, chapters 7, 8 and 9 will exclusively focus on combined microwave-ultrasonic pretreatment.

CHAPTER 7

EFFECT OF COMBINED MICROWAVE-ULTRASONIC PRETREATMENT ON ANAEROBIC DIGESTION OF PRIMARY, EXCESS ACTIVATED AND MIXED SLUDGES

Abstract

This chapter deals with the effect of combined microwave-ultrasonic pretreatment on the anaerobic biodegradability of primary, excess activated and mixed sludge. The characteristics and anaerobic biodegradation of raw primary, excess activated and mixed sludge were compared to combined microwave-ultrasonic pretreated primary, excess activated and mixed sludge. The effect of mixing ratio of primary sludge to excess activated sludge was also studied. Methane production in pretreated primary sludge was significantly greater (11.9ml/g TCOD) than the methane yield of the untreated primary sludge (7.9 ml/g TCOD). SCOD/TCOD ratio decreased by 48% for primary digested sludge due to the pretreatment. Pretreatment resulted in the reduction of SCOD/TCOD ratio of the digested mixed sludge by 58% compared to that of untreated digested mixed sludge. Cumulative methane production of pretreated Excess Activated Sludge (EAS) was higher (66.5 ml/g TCOD) than the methane yield from pretreated mixed sludge (44.1 ml/g TCOD). Furthermore, digested EAS showed significantly higher dewaterability (201s) than digested primary sludge (305s) or mixed sludge (522s). The average methane: Carbon dioxide ratio from EAS (1.85) was higher than that for mixed untreated sludge (1.24). VS reduction was also higher for EAS than the other two sludge types. However, pretreatment of EAS resulted in significant reduction in dewaterability due to higher percentage of fine floc particles in the pretreated EAS. Thickened excess activated sludge which has greater solid concentration resulted in a better digester performance after pretreatment.

7.1 Introduction

Different pretreatment technologies were found to enhance sludge hydrolysis and anaerobic digestion performance (Carrère et al., 2010a). Pretreatment of sludge through ultrasonic, mechanical, chemical or thermal techniques result in bacterial cell wall disruption, disintegration of EPS and release of enzymes which enhance the rate of hydrolysis and biodegradation (Tyagi and Lo, 2011, Eskicioglu et al., 2006). Primary sludge, excess activated sludge and mixed sludge have distinctively different biochemical composition, rheological property, response to pretreatment, biodegradability and methane potential, floc size and dewaterability. Studying effect of pretreatment technologies and biodegradability of each of the sludge types is beneficial for the selection of appropriate pretreatment technology and pretreatment condition, better design and operation of sludge treatment units (Zhang, 2010). This particular chapter focuses on understanding the effect of combined microwave-ultrasonic pretreatment on biodegradability, methane potential, dewaterability and characteristics of primary, excess activated and mixed sludge systems.

7.2 Materials and Methods

7.2.1 Sampling and Characterization

Primary sludge was collected from primary gallery underflow lines particularly from primary sedimentation tank No. 4 of BWWTP (Figure 3.1 and Section 3.3.1). Excess Activated Sludge was collected from Module 4 of the secondary treatment section of BWWTP (Figure 3.1 and Section 3.3.2). Thickened excess activated sludge was collected from the discharge of the DAFT unit before mixing with primary sludge (Figure 3.2 and Section 3.3.2). Mixed sludge was collected from Beenyup anaerobic digesters feed mixed sludge sampling point (Figure 3.1 and Section 3.3.3) and the Primary and Excess activated sludge samples were mixed with 3:1 ratio to prepare the mixed sludge before all the samples were charged to the jacketed digesters. Samples were withdrawn from each anaerobic digester for characterization purpose. The characteristics of sludge fed to the digesters are presented in Table 7.1. Elemental analysis results are shown on Table 7.2. Mixed sludge showed intermediate composition of carbon, hydrogen, nitrogen and sulfur. Higher nitrogen content of EAS/TEAS is because of higher amount of microbial biomass in this type of sludge compared to the other two. The percentage of carbon in the primary sludge is greater than the percentage of carbon in mixed or excess activated sludge.

Table 7.1. Characteristics of the sludge fed to the reactors

Parameter	TS (%)	VS (% TS)	COD (g/l)	pH
Raw primary sludge	2	88.8	30.5	7.2
Primary pretreated sludge	2	88.8	32.8	7.1
Excess activated sludge	1	90	18.9	6.9
Pretreated thickened excess activated sludge	2.7	83	40	7
Untreated Mixed Sludge	1.5	87.5	22.9	7.1
Mixed Pretreated Sludge	1.5	87.5	24.9	7.1

Table 7.2 Elemental analysis results of the composition of different sludge samples from BWWTP

	% Carbon	% Hydrogen	% Nitrogen	% Sulphur
Digested Sludge (DS)	37.88	6.609	6.384	2.418
TEASludge (TEAS)	41.001	6.84	8.098	2.463
Mixed Sludge (MS)	42.66	7.006	5.116	2.305
primary Sludge (PS)	43.394	7.413	3.338	2.187

7.2.2 Analytical methods

The standard analytical methods discussed in Chapter 3, Section 3.4 were used to measure pH, total and soluble chemical oxygen demand, $CH_4/CO_2/O_2$ composition, ammonia, dewaterability and other characteristic and operational parameters.

7.2.3 Combined Microwave-Ultrasonic Pretreatment

Primary, excess activated and mixed sludge samples were pretreated according to the conditions shown in Table 7.2. Initially each of the sludge samples was homogenized and pretreatment was carried out in the sequence of microwave treatment first followed by ultrasonic pretreatment at the conditions specified in Table2. Pretreatment conditions were selected based on the treatment power and time optimization experiments presented in chapter 5.

Table 7.2. Different conditions of pre-treatment

Pre-treatment method	**Conditions**
Microwave- ultrasonic treatment (MU)	Microwave: 2450 MHz, 800 W, 3 min, Ultrasonic: 0.4 W/mL, 48,000 Joules, 90% amplitude, 55/5 pulse , 6 min-8 min

7.2.4 Experimental setup for methane potential and sludge biodegradability tests.

The tests for methane potential were conducted in batch continuously stirred 1L jacketed digesters. All the digesters were kept at a mesophilic temperature of 36.5^{0}C by means of a water bath heater. 50 ml of digested sludge was introduced to each of the digesters for acclimation. The digesters were inoculated with the digested sludge for a period of 3 days and sludge feeding to the digesters was carried out after adjusting the pH and purging the digesters with nitrogen gas. The effective digester volume was 500 ml for each of the reactors after charging the feed sludge. The pH was maintained between 6.8-7.3 using sodium hydroxide and hydrochloric acid. The biogas generated was allowed to pass through buffer tanks to remove any condensate before the gas volume was measured in inverted cylinders by water displacement technique. The biogas composition and other parameters were continuously monitored until biogas generation reached SRT of 25 days.

7.3 Result and Discussion

7.3.1 Methane production potential of different kinds of sludge

Methane production in pretreated primary sludge (11.9ml/g TCOD) was 33.6 % greater than the methane yield of the untreated primary sludge (7.9 ml/g TCOD) as shown in Figure 7.1. SCOD/TCOD ratio for pretreated primary sludge was 48% less than the ratio for untreated primary sludge as it is consumed due to increased organic disintegration and methanogenic activity in the anaerobic digestion process. In case of untreated primary sludge, the biopolymers and organics are dominantly present in the solid phase than in the soluble liquid phase. Pretreatment enhances destruction of complex floc structure of secondary sludge and biopolymers in primary sludge and promotes the transfer of organics to the soluble phase (Eskicioglu et al., 2008a). Specific methane yield of pretreated mixed sludge was 12.6 % greater than untreated mixed sludge as shown in Figure 7.2. Excess Activated Sludge (EAS) showed less methane production (20.7ml/gTCOD) as compared to Pretreated Excess Activated Sludge (PEAS) (66.5ml/g TCOD) as shown in Figure 7.3. The thickening process in the dissolved air floatation tank (DAFT) has significantly increased the solid concentration and the pretreatment further enhanced the methane production and the kinetics of the digestion process.

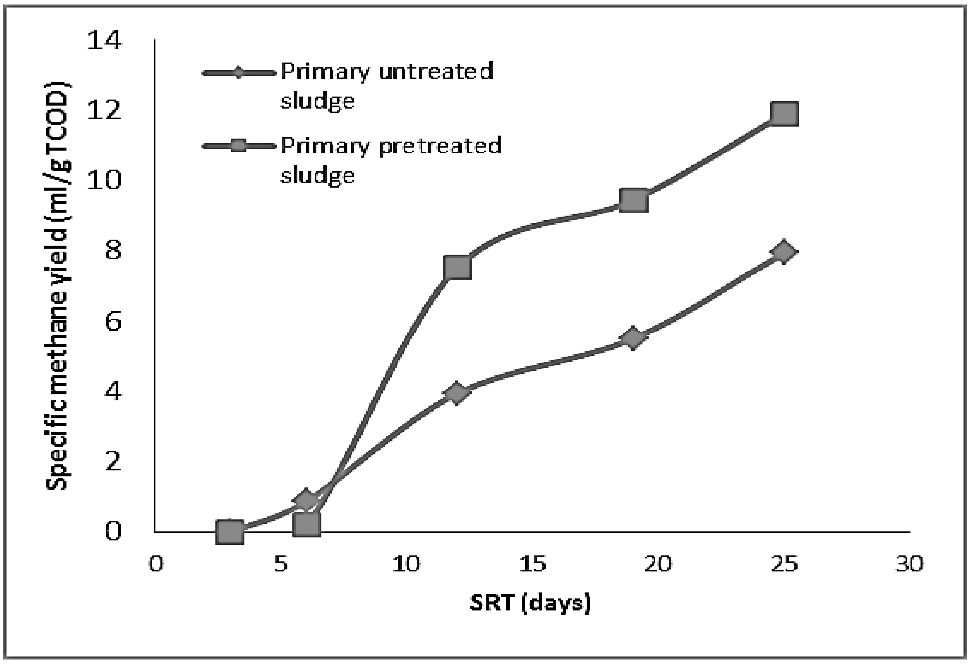


Figure 7.1. Specific methane yield from pretreated and untreated primary sludge

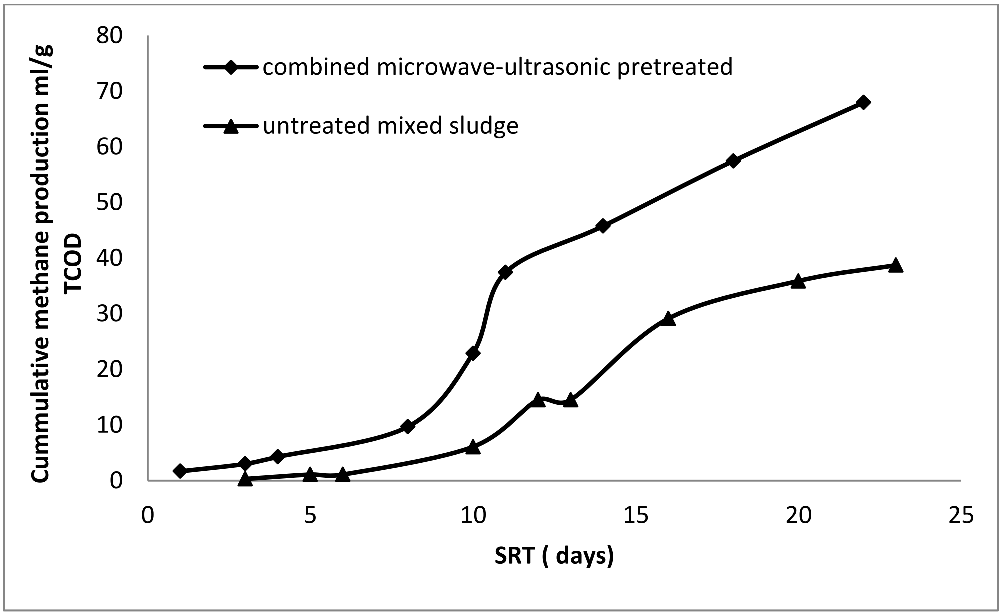


Figure 7.2. Specific methane yield from untreated and pretreated mixed sludge

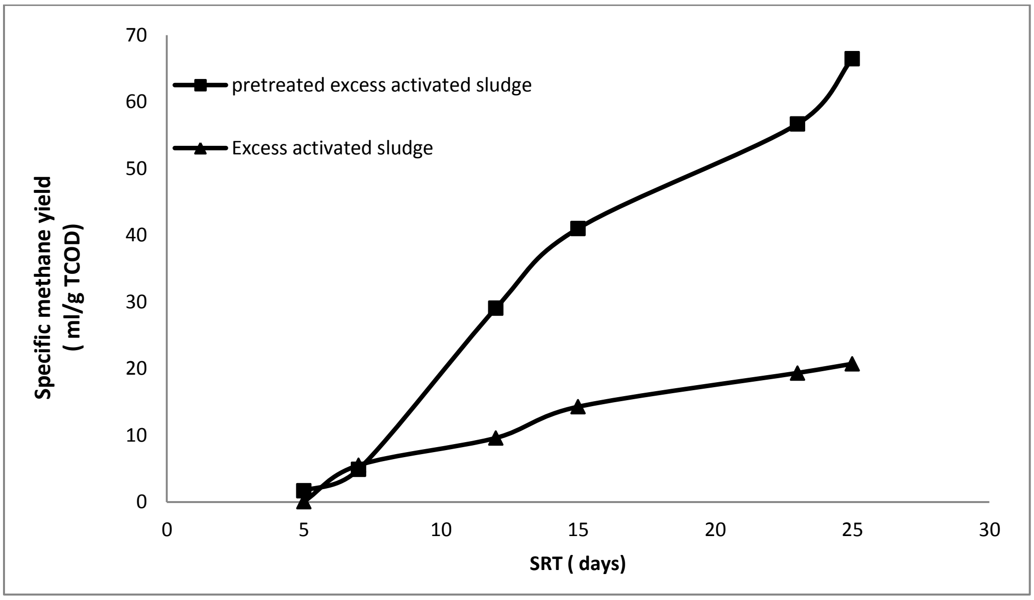


Figure 7.3. Specific methane yield from untreated and pretreated excess activated sludge.

7.3.2 Effect of pretreatment on sludge biodegradability (COD and VS removal)

Soluble COD content is a measure of biodegradability or the methane potential of the sludge after anaerobic digestion. SCOD/TCOD ratio for pretreated primary sludge after 25 days of SRT was 48% less than the ratio for untreated primary sludge as shown in Figure 7.5. SCOD/TCOD ratio for pretreated mixed sludge was 58% less than that of the untreated mixed sludge. Higher reduction in SCOD in the case of mixed sludge indicates increased solubilisation or release of organics achieved by the disintegration of flocs, disruption of cells and rapid internal heating due to microwave irradiation. Consumption of the hydrolysable organics by anaerobic bacteria during methanogenesis resulted in increased methane production. Figure 7.4 shows that the reduction in TCOD was equal for primary and mixed sludge types after 25 days of SRT while that of EAS was slightly greater. Combined microwave-ultrasonic pretreatment enhanced the TCOD removal from Excess activated and mixed sludge by 33.2% and 32.7 % respectively. The volatile solids reduction after anaerobic digestion of EAS was 42.7%. The volatile solid reduction achieved for primary and mixed sludge types was 26% and 30 % respectively. Combined pretreatment technique disintegrates the complex floc structure of EAS. The combined pretreatment in all the three sludge types improved biodegradability. Microwave treatment assisted efficient and faster cell disruption due rapid internal heating; whereas the ultrasonicaton assists cavitation, floc size reduction and encourages formation of highly reactive radicals that facilitate destruction of organics.

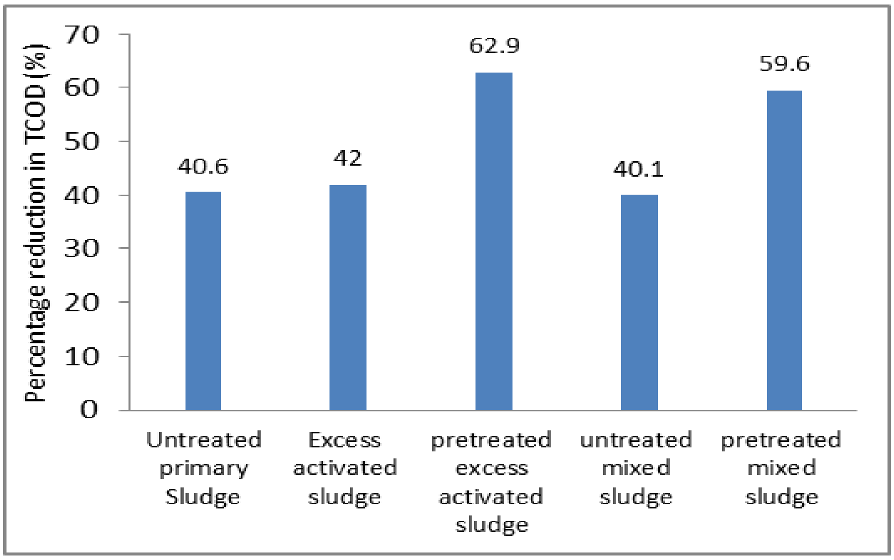


Figure 7.4. Reduction in TCOD for different kinds of sludge

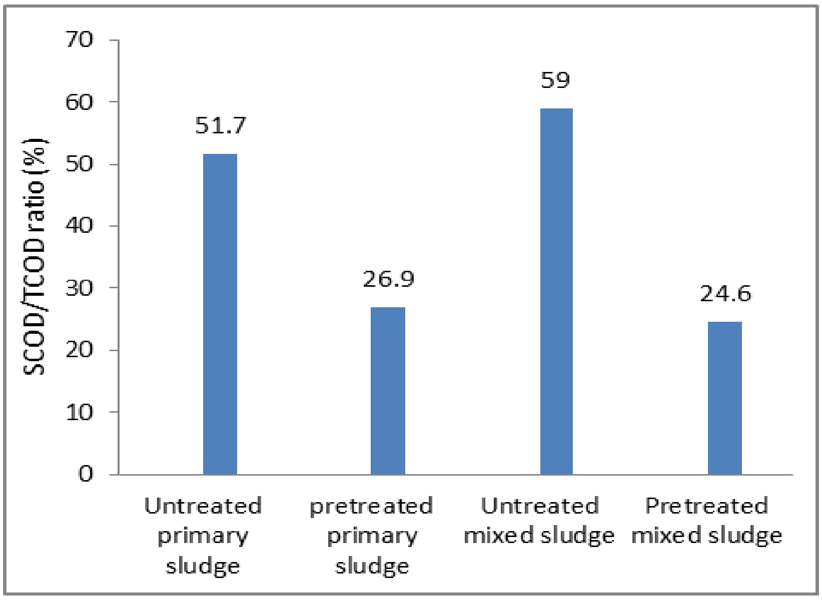


Figure 7.5. Soluble COD/ Total COD ratio of various digested sludge samples.

7.3.3 Effect of pretreatment on the dewaterability of different kinds of sludge

The dewaterability of Excess activated sludge was significantly better than primary or mixed sludge as the total solid in EAS was less than the other two sludge types Figure 7.6. However, combined microwave-ultrasonic pretreatment resulted in the deterioration of the dewaterability of excess activated or slight improvement in case of mixed sludge. Dewaterability is a function of particle size of the flocs and the hydrophilicty of biopolymers released due to the disintegration of microbial cells. Pretreatment decreases average size of flocs and increases release of biopolymers which may trap water and limit the dewaterability. The change in floc structure and colloidal charge may have also contributed to the reduction in dewaterability. Ultrasonication is known to have effects of changing the surface charge.

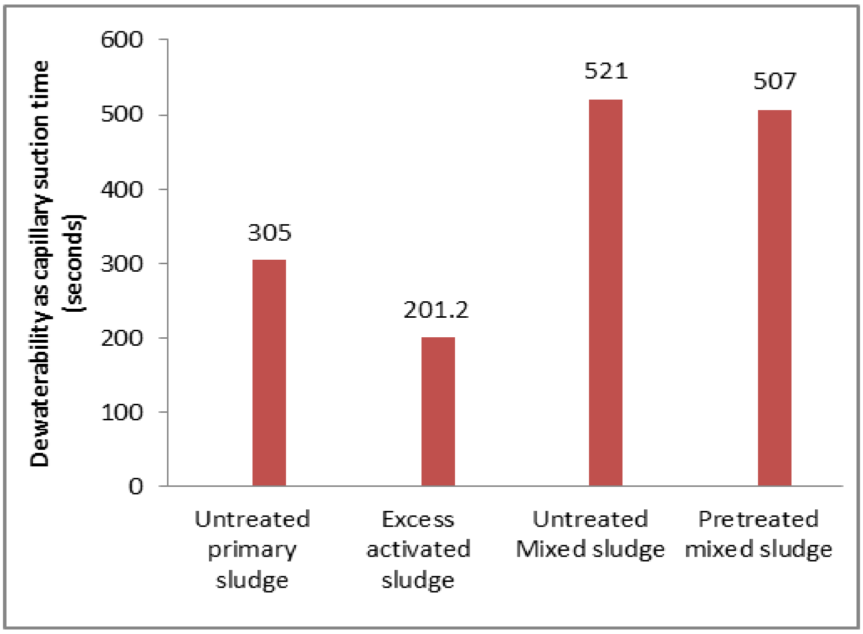


Figure 7.6. Dewaterability of different sludge samples.

7.3.4 Effect of pretreatment on biogas composition and CH4/CO2 ratio

The maximum CH_4/CO_2 ratio for EAS was 1.85 and pretreatment enhanced the quality of the biogas by 7.5 %. The CH_4/CO_2 ratio for mixed untreated sludge was 1.24 and the enhancement in gas quality due to combined microwave-ultrasonic pretreatment was 18.9 %. The effect of pretreatment on biogas quality was much greater in mixed sludge system than excess activated sludge. In the initial phase of the digestion process, the CH_4/CO_2 ratio was relatively lower for all sludge types; it progressively increased due to the conversion of CO2 to CH4 through hydrogenotrophic methanogenesis reaching the maximum CH4/CO2 ratio after 25 days of SRT as shown in Figure 7.7.

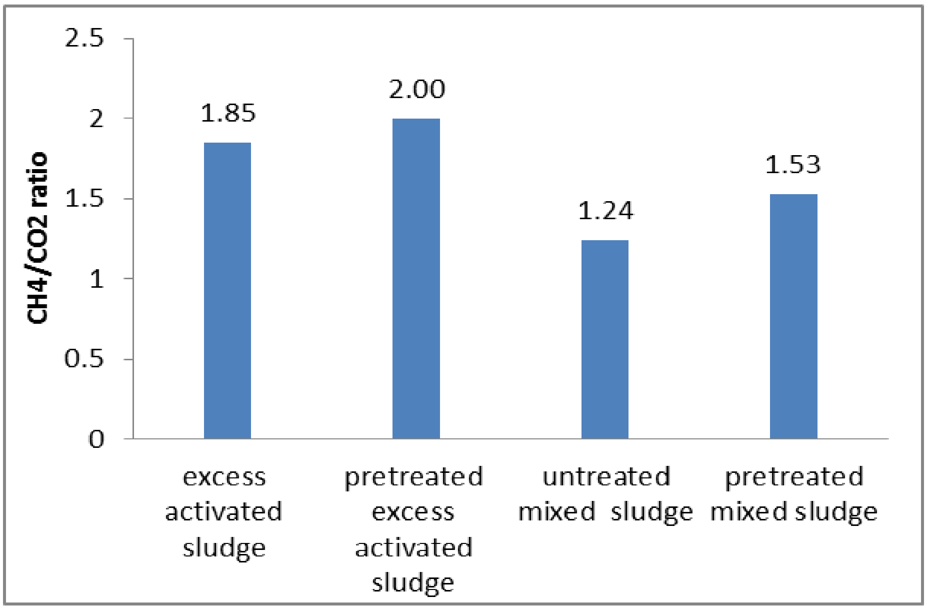


Figure 7.7. Maximum CH4/CO2 ratio in the biogas after 25 days of SRT for untreated and pretreated sludge samples.

7.3 Conclusion

Combined microwave-ultrasonic pretreatment improved sludge solubilisation, biogas production and anaerobic digester performance and biodegradability of primary, EAS and mixed sludge. This Combined pretreatment technique disintegrates the complex floc structure of EAS and macromolecules in primary sludge. The degree of sludge solubilisation after pretreatment for different sludge types was different. The combined Pretreatment resulted in comparatively greater improvement of methane production and biogas quality (CH4/CO2 ratio) and VS destruction in EAS. The increase in digestion efficiency is proportional to the degree of sludge disintegration. Sludge disintegration and increased biodegradability is due to rapid internal heating of microwave radiation and the floc destruction achieved by ultrasonic treatment. EAS also showed better dewaterability compared to other sludge types. However, dewaterability deteriorated with pretreatment due to higher percentage of fines and greater availability of biopolymers which increased the amount of bound water. It can be understood from this chapter that the anaerobic digestion enhancement is much greater when combined microwave–ultrasonic pretreatment is applied on Excess activated and thickened excess activated sludge. Chapter 8 presents the effect of pretreatment of thickened excess activated sludge mixed with untreated primary sludge. Subsequent chapters focus on effect of operational parameters on anaerobic biodegradation of mixed sludge system.

CHAPTER 8

ANAEROBIC BIODEGRADABILITY OF COMBINED MICROWAVE-ULTRASONIC PRETREATED THICKENED EXCESS ACTIVATED SLUDGE MIXED WITH UNTREATED PRIMARY SLUDGE

Abstract

The findings of the previous chapters confirm that combined microwave-ultrasonic pretreatment of thickened excess activated sludge enhances anaerobic digestion more than the effect on mixed or primary sludge. In this chapter, anaerobic biodegradability of Combined Microwave-Ultrasonic pretreated thickened excess activated sludge (PTEAS) mixed with untreated primary sludge (PS) was investigated. Two continuously stirred mesophilic anaerobic digesters were charged with a mixture of PTEAS and PS. Digester 1 was charged with 75% PTEAS + 25% PS while digester 2 was fed with 25% PTEAS + 75% PS. The working volume was 0.5 L for both digesters. The pretreatment of TEAS was carried out at microwave irradiation condition of 2450 MHz at a density of 36.92KJ/L g SCOD followed by ultrasonic treatment at a density of 0.66 W/mL, amplitude of 90%, and pulse of 55/5 for a period of 8min. The anaerobic digestion was conducted in the two continuously stirred batch anaerobic digesters for a sludge retention time of 32 days. The specific methane yield was 122 ml CH_4/g TCOD for digester 1 and 101 ml CH_4/ g TCOD for digester 2 after sludge retention time of 20 days. The amount further increased to 187 ml CH_4/g TCOD for digester 1 and 116 ml CH_4/g TCOD for digester 2 after SRT of 27 days. The CH_4/CO2 ratio reached 2.2:1 and 1.1:1 after SRT of 20days for digester 1 and digester 2 respectively. The percentage reduction in TCOD after 13 days of SRT for digester 1 is 7% more than the percentage reduction in digester 2. The percentage composition of methane produced was 73.1% for digester 1 and 55 % for digester 2 after 20 days of SRT. The dewaterability measured in capillary suction time for the anaerobic digester with higher TEAS content (digester 1) was less than that of digester 2. Furthermore, higher percentage of the pretreated TEAS

increases the digestion kinetics, the methane production capacity and the biogas quality.

8.1 Introduction

The effect of combined microwave-ultrasonic pretreatment on anaerobic digestion of thickened excess activated sludge and mixed sludge was addressed in chapters 5, 6 and 7. This particular chapter focuses on intensive investigation of combined microwave-ultrasonic pretreatment when the pretreatment is applied only on the thickened excess activated sludge part before it is mixed with primary sludge. This investigation is motivated by the significant enhancement effect combined microwave-ultrasonic pretreatment brought on the digestibility and organic matter removal for thickened excess activated sludge. Sludge disintegration effect and methane production potential of waste activated sludge is more than that of primary sludge after ultrasonic pretreatment (Pilli et al., 2011a). Besides, the significance of the technique on the cost of operation of digesters is quite significant.

8.2 Materials and Methodology

Thickened excess activated sludge and primary sludge samples were collected from BWWTP for the pretreatment and digestion study. The samples were stored at 4°C. Digested sludge from previous experiments was utilized to inoculate the digesters. Digester acclimation was done for over one month. In this experiment, TEAS was pretreated under optimum combined microwave-ultrasonic pretreatment conditions and the TEAS was mixed with untreated primary sludge before the mixture was fed into the anaerobic digesters.

800 mL sample of TEAS was microwave irradiated at a frequency of 2450 MHz and density of 36.92 KJ/L g SCOD. The TEAS sample was then mounted on to Sonics Vibrocell ultrasonication unit for sonication at 48000 J, 55/5 pulse, 90% amplitude, and for 8 minutes. 200mL of the sample was transferred to a 250mL plastic storage bottle, labelled, and stored at 4^{o}C for the digestion test.

The pretreated TEAS was mixed in two ratios by volume with untreated primary sludge: 25% PS and 75% pretreated TEAS (Digester 1), and 75% PS and 25% pretreated TEAS (Digester 2). The percentage compositions were selected based on the results on the study of mixing ratio (Chapter 9) and the significant enhancement by pretreatment technique obtained for TEAS (chapter 7). The digester feed samples were characterized as shown in Table 8.1.

Table 8.1: Characteristics of digester feed sludge

Parameter	**25% PS + 75% TEAS Sludge**	**75% PS + 25% TEAS Sludge**
pH	6.95	7.1
TCOD (mg/L)	36850	26750
SCOD (mg/L)	13000	7800
SCOD/TCOD	0.35	0.29
TS (%)	2.9	2.6
VS (%)	84.7	86.6

8.2.1 Experimental digester set-up

Two continuously stirred, semi-continuous jacketed digesters from the four digester setup shown in section 3.4.3 Figure 3.7 were used for the anaerobic digestion test. Jacket heating system was applied to maintain the desired mesophilic digester temperature of 36.5°C. The digesters were placed on magnetic stirrers to maintain continuous mixing. Sludge samples were fed to the digesters through the sludge charging tube and the biogas produced will exit through another tube to a 1L buffering bottle, placed outside the water bath heater, for removal of any condensate. Water displacement method was used to measure the gas volume and the biogas composition was measured using GA plus 2000 biogas analyser as shown in section 3.3.

The digested sludge used for inoculation accounts for 20% of the digester volume; therefore in each of the digesters, 100ml of the inoculum was mixed with 400 mL of the feed sludge samples.

Digester 1 was fed with 75% pretreated TEAS and 25% primary sludge and digester 2 was charged with mixed sludge with composition of 25% pretreated TEAS and 75% primary sludge. The digesters were continuously purged with N_2 at 25-40mL/min after the charging.

The digesters were operated in semi-continuous mode with 25mL of digested sludge being removed from the digester periodically and 25mL of previously stored sludge was introduced at the same time. The digesters were operated for a total SRT of 32 days and the results in the first 20-27 were used for the analysis.

Periodically, the volume and composition of the gas produced was measured and recorded. The gas composition was measured by connecting the Gas Analyser probe to the inlet tube of the buffering bottle to pump out the biogas from the digesters.

8.2.2 Analytical methods

All the analysis required for the experimental work in this section including determination of TS, VS, SCOD, TCOD, pH, dewaterability (CST), elemental analysis, particle size analysis, Rheology, protein analysis and microbial analysis were based on the methods and techniques presented in sections 3.2.1 through 3.2.13.

8.3 Results and Discussion

The samples from the two continuously stirred anaerobic digesters were collected and analysed using the methods discussed in Section 3.3.1 through 3.3. 13 The results from the analyses are presented in this section of the thesis. The results are discussed in terms of the effect of combined microwave-ultrasonic pretreatment at the optimum condition and mixing ratio on the performance of the anaerobic digesters.

8.3.1 Effect of Combined microwave-ultrasonic pretreatment on biochemical methane potential and biogas composition.

The results obtained from the biochemical methane potential test conducted during the 32 day of SRT are presented in this section. Figure 8.1 shows the specific methane yield for both digesters where the yield from digester 1 was significantly greater than that of digester 2 after the lag phase of the hydrolysis stage of the digestion process is completed.

The slow lag phase at the start is the result of the rate limiting hydrolysis process where sludge disintegration and solubilisation of complex organic molecules takes place followed by the rapid acidogenesis and acetogenesis stages. Substantial amount of methane with a maximum of 205 mL CH_4/g TCOD was produced in digester 1 after 8 days in the lag phase. The maximum methane yield of digester 2 was 157 mL CH_4/g TCOD which was produced after a lag phase of 13 days. The difference in sludge yield is attributed to the percentage composition of pretreated TEAS in the two digesters which determines the rate of sludge disintegration. Digester 1 contains significantly greater amount of pretreated TEAS, which has more readily degradable organics. On the other hand, higher percentage of primary sludge in digester2 prolongs the period of hydrolysis to disintegration unlike the case in digester 1.

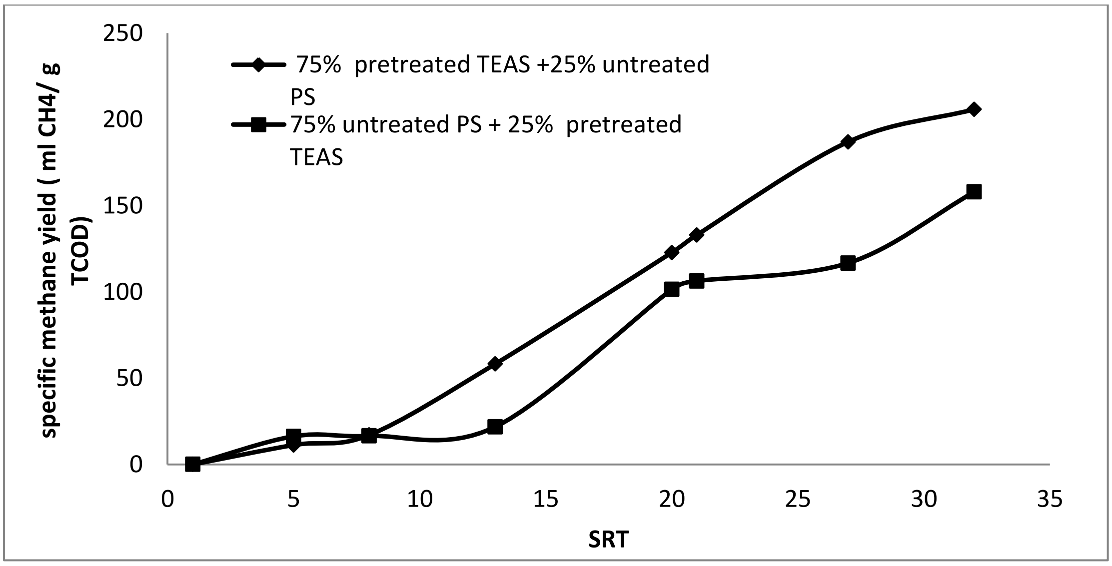


Figure 8.1: Specific Methane Yield in digester 1 and 2

The results obtained in this study evidently show the benefit of the combined pretreatment of TEAS when comparing the results to other studies based on microwave or other pretreatment methods on waste activated sludge. Saha et. al. (2011a) pretreated samples of 40% waste activated sludge (WAS) and 60% PS with microwave (2450 MHz, 0-1250 W, 25-260°C) and ultrasonic (20 kHz, 1 W/mL, 15-90 mins) pretreatment methods separately. The maximum specific methane yield achieved at a digestion time of 20 days was 80 mL CH_4/g TCOD for microwave and ultrasonic pretreatment. Digester 2 in this study produced 100 mL CH_4/g TCOD for SRT of 20 days. This clearly shows the effectiveness of the combined microwave-ultrasonic pretreatment method in terms of enhancing anaerobic digestion processes.

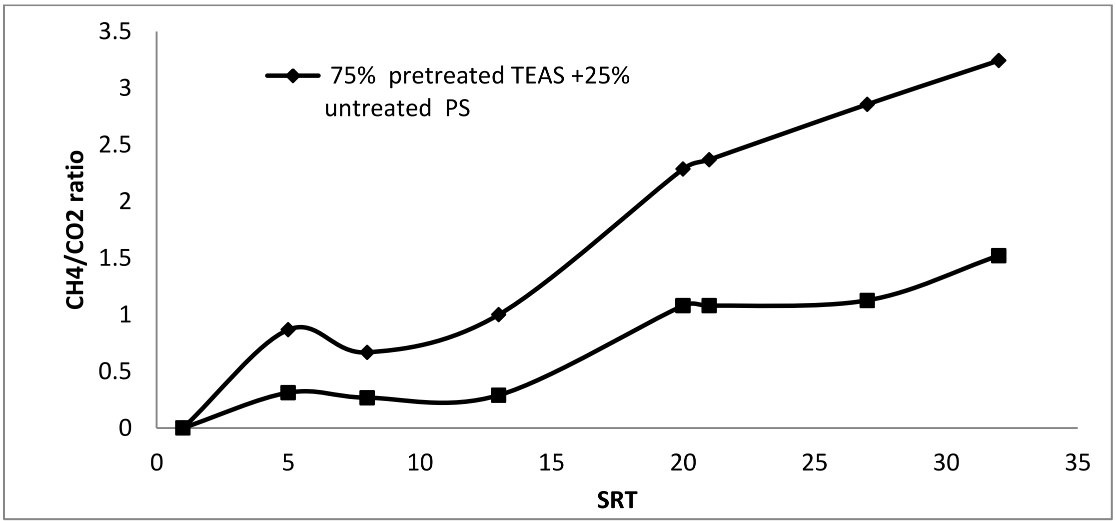


Figure 8.2: Methane /Carbondioxide Production Ratio in digesters 1 and 2

Figure 8.2 represents the methane production of each digester in the form of methane to carbondioxide ratio. The methane/carbondioxide ratio (biogas quality) indicated the level of methanogenic activity in the digesters. High CH_4/CO_2 ratio justifies how healthy the digesters are to convert all VFA into acetate and ultimately to methane (acetogenic methanogenesis) or CO_2 and H_2 to methane (hydrogenotrophic methanogenesis).

It can be observed that digester 1 produced significantly larger amount of methane to carbon dioxide in a short duration of time, whereas digester 2 produced more carbon dioxide than methane up until an SRT of 20 days. Methanogenesis in digester 1 happened faster and more readily due to higher degree of degradation and availability of soluble organics and due to the pretreatment which makes the hydrolysis and acidogenesis stages much shorter, whereas digester 2 which contains more primary sludge showed slow degradation rate. Primary sludge contains a large fraction of lignin and cellulose, with polysaccharides present in the cell walls of organic structures (Saha et al., 2011a). The reduction of such compounds to the soluble phase requires a large amount of energy; this can explain the slow hydrolysis step in digester 2.

In terms of environmental sustainability of the process, reducing the carbon dioxide produced is very beneficial as greenhouse gas emissions is a nuisance to the environment. The carbon dioxide generated in digester 1 is reduced after 5 days to the value digester 2 achieved at an SRT of 20 day. This is a substantial difference, which demonstrates the benefits of combined microwave-ultrasonic pretreatment of the TEAS portion of the mixture.

Figure 8.3 and 8.4 demonstrate the biogas compositions for both digesters separately. It can be observed that the methane/carbon dioxide ratio became 1:1 in an SRT of less than 10 days for digester 1. This shows high methanogenic activity in digester 1 at a reasonably short retention time as discussed earlier. The most important feature of this graph is the maximum methane production of 71%, which was achieved at 15 day SRT. Other studies, applying alternative pretreatment methods to mixed sludge feed achieved significantly lower methane percentages. For microwave pretreatment of a 1:1 ratio of primary to secondary sludge mixture, a methane composition of 59% was achieved after 20 days (Park and Ahn, 2011a). Applying ultrasonic pretreatment on secondary sludge (activated sludge) feed methane percentage of 65.9% was achieved a for an ultrasonication pretreatment duration of 30 minutes (Tiehm et al., 2001). This indicates the benefits of combined microwave-ultrasonic pretreatment in terms of enhancing organics solubilisation that are more readily available for reduction, hence increasing methane production.

Figure 8.4 shows that, for digester 2, the steps discussed above take significantly longer time, due to the large percentage of primary untreated sludge available in the digester. Methane production reached a steady percentage composition of 55% CH_4 after 15 days of SRT. As 75% of the digester feed is raw primary sludge in digester 2, 55% methane production in 15 days is an interesting result compared to the results from other studies presented above.

It is also important to note the oxygen content recorded in both digesters was very low at all stages of digestion indicating that appropriate anaerobic condition was maintained throughout the process. Exposing anaerobic bacteria to oxygen can result in the formation of toxic radicals, which result in the destruction of the anaerobic environment, causing digester inhibition (Rolfe et al., 1978).

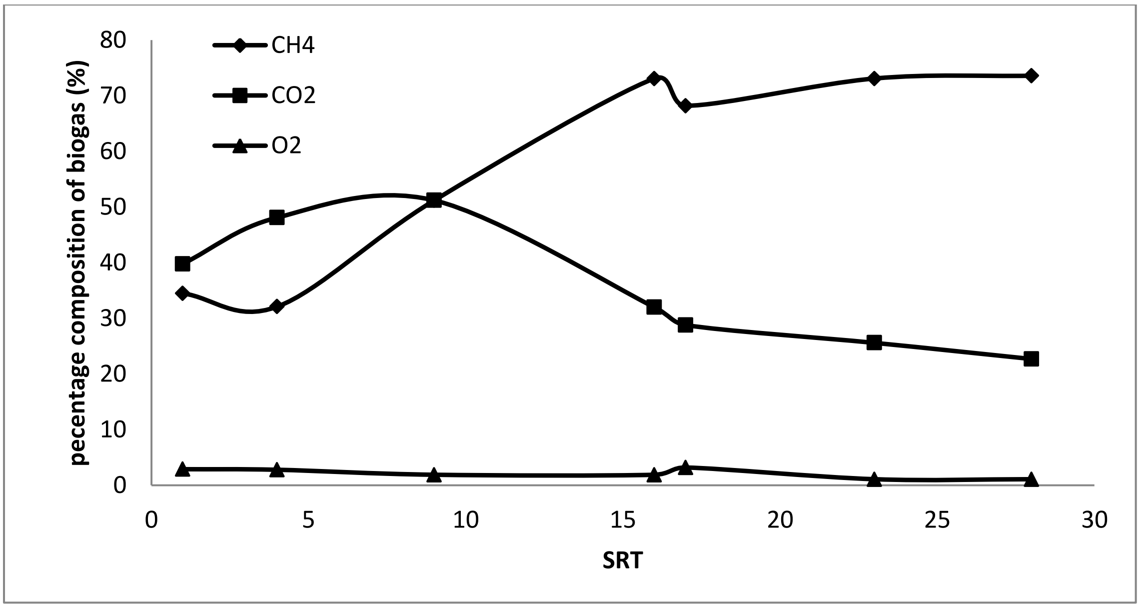


Figure 8.3: Biogas composition for digester 1

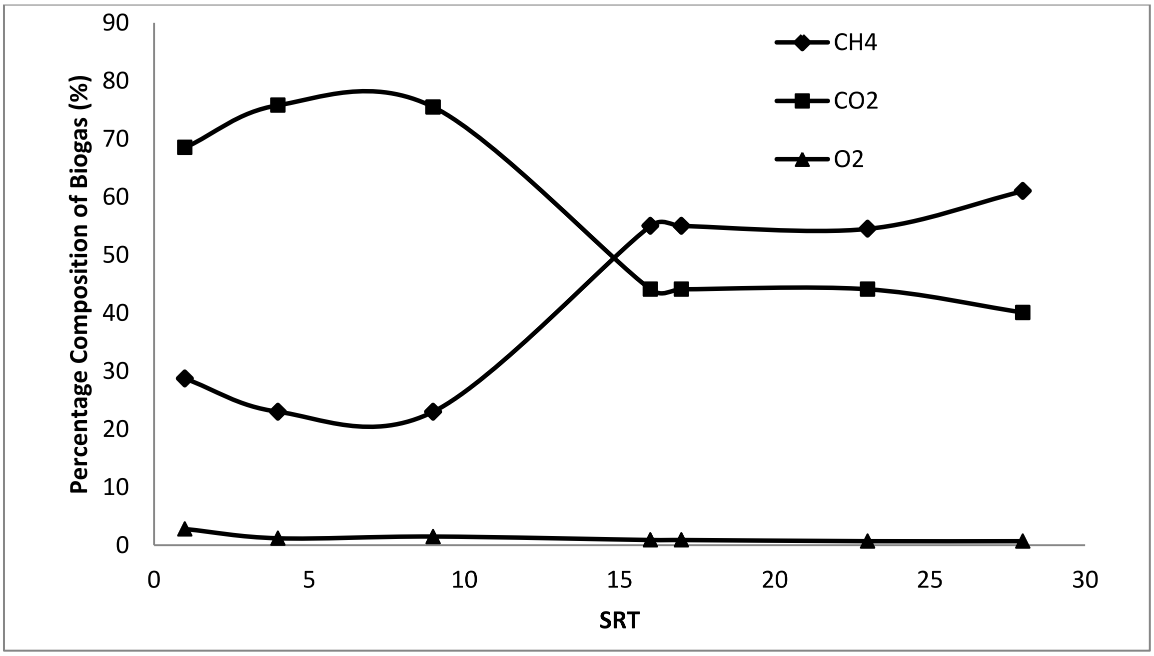


Figure 8.4: Biogas composition for digester 2

8.3.2 Effect of pretreatment on solids removal and sludge disintegration

Total and volatile solids reductions are used to directly measure the degree of biodegradation that has occurred in the sludge through the anaerobic digestion process. Figure 8.5 shows the total solid content of the sludge at different times in the 32 day SRT. Digester 1 achieved a 43% total solids reduction over a 20-day SRT, while digester 2 experienced a 46% reduction in the same amount of time. The most important feature to note from the Figure is the rate of solids reduction in the first 5 days of the SRT. Total solids percentage in digester 1 was reduced more significantly in the first 5 days of digestion as compared to digester 2. This is due to the larger percentage of pretreated TEAS present in digester 1, as pretreatment aims to alter the feed characteristics of the sludge, hence making it more susceptible to reduction by hydrolysis. This enables the organic complexes originally available in the solids phase to be solubilised faster facilitating the availability of soluble organics for the later stages of anaerobic digestion.

Interestingly, Figure 8.5 after SRT of 20 days shows that total solid in both digesters does not decrease further which confirms that anaerobic digestion for sludge retention time longer than 20 days is not required which is beneficial for large-scale anaerobic digestion applications. significant cost saving can be achieved due to shorter retention time required to meet sufficient sludge degradation, and greater amount of methane production (Lee et al., 2011).

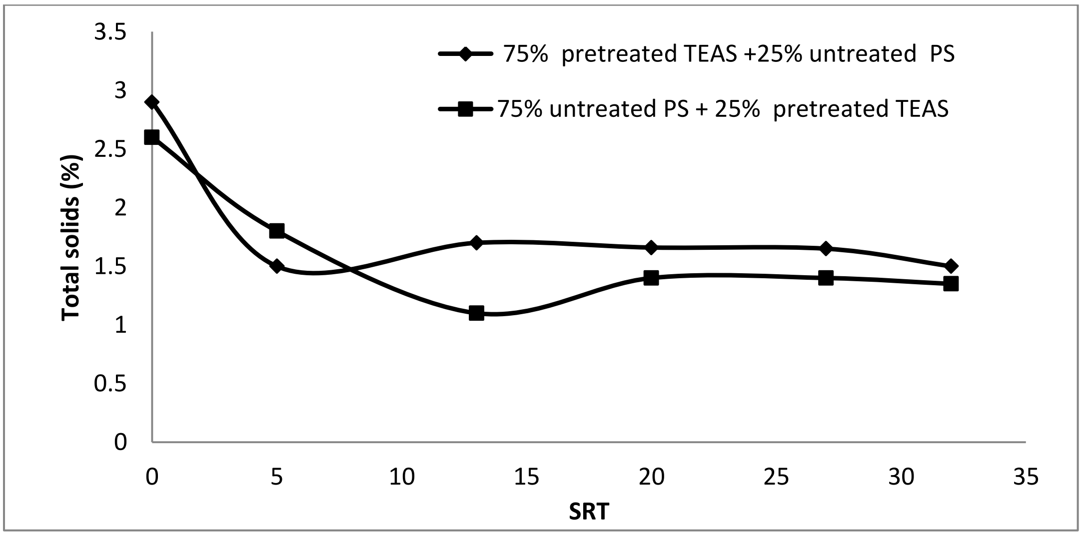


Figure 8.5: Total solids percentage as a function of sludge retention time

The total solids reduction achieved in this research was compared to the findings of other researchers for other kinds of pretreatment which demonstrated the effectiveness of combined microwave-ultrasonic pretreatment for efficient sludge solubilisation. For two digesters charged with a 40:60 WAS to PS feed, maximum total solids reductions of 25% and 20% were achieved for microwave (2450 MHz, 0-1250 W, 25-260℃) and ultrasonic (20 kHz, 1 W/mL, 15-90 mins) pretreatments respectively (Saha et al., 2011a). significantly lower solids reductions was achieved for similar mixed feed sludge from this experiment proving that combined microwave-ultrasonic pretreatment appears to be advantageous in terms of solids removal.

Volatile solids as a percentage of the total solids in the sludge are represented graphically in Figure 8.6. The graph depicts the volatile solids reduction achieved in the system. Volatile solids reduction of 57.8% and 76% was achieved for digesters 1 and 2 respectively for an SRT of 20 days. Volatile solid measures the amount biodegradable and non-biodegradable organic components present in the sludge, and therefore, it can be inferred from the graph that digester 2 contains a significantly larger portion of reducible organics. This is due to the high percentage of primary sludge in the digester feed stock, it is well known that primary sludge digestion results in a higher volatile solids reduction (Grönroos et al., 2005). Figure 8.6 shows that digester 1 has volatile solids percentage almost equal to that of digester 2 but the reducible organic fraction is more predominant in the digester with more primary sludge.

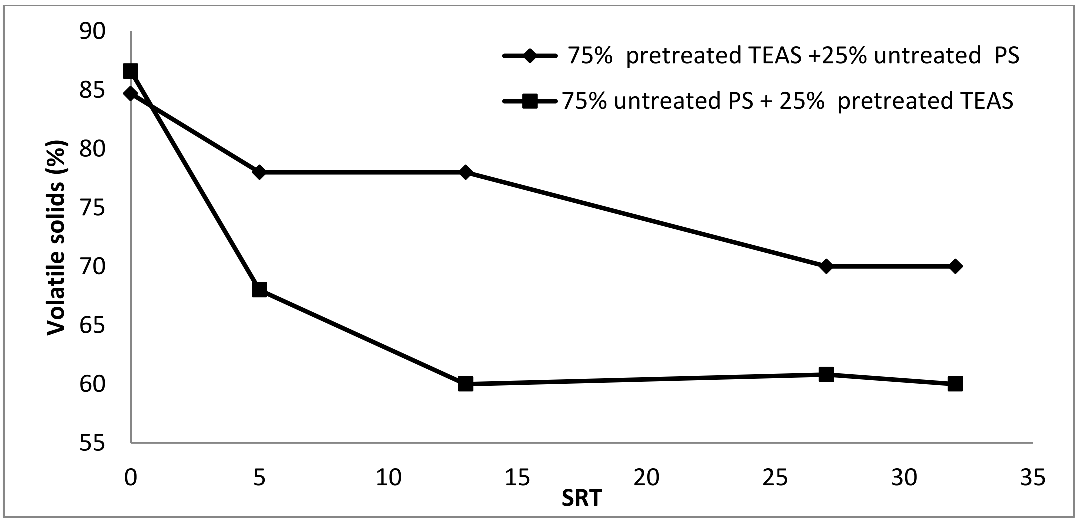
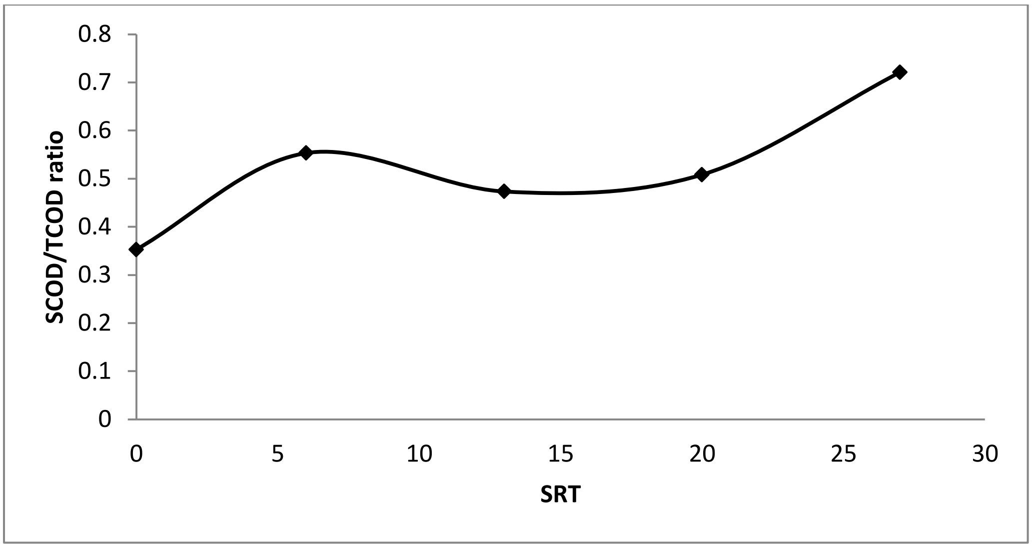


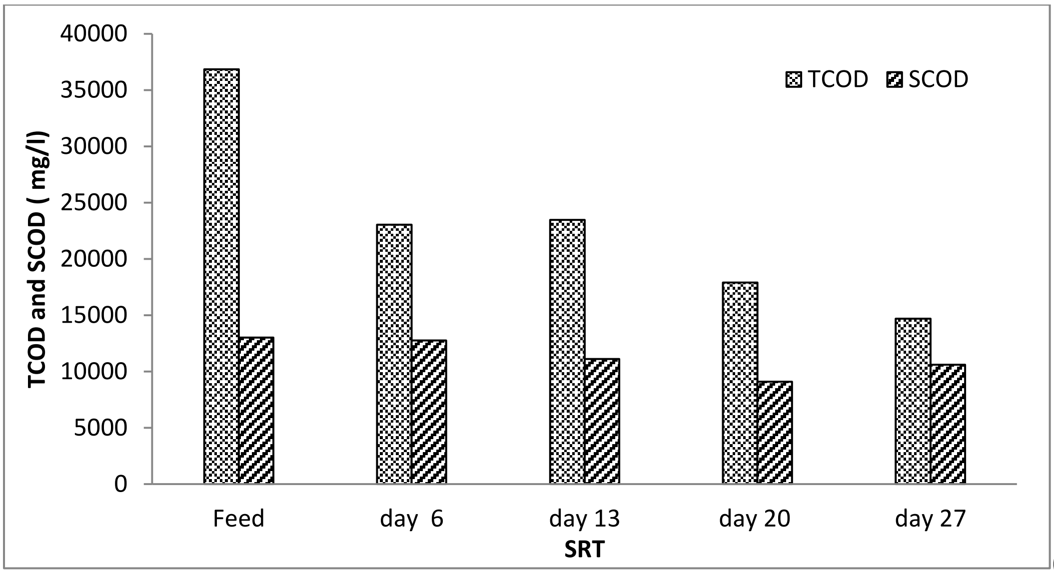
Figure 8.6: Volatile solids percentage as a function of sludge retention time

Total chemical oxygen demand (TCOD) and soluble chemical oxygen demand (SCOD) are other indicators of the degree of sludge biodegradability. These parameters show the amount of organic and inorganic species and organisms that can be chemically oxidised in the sludge, Figure 8.7 and 8.8 shows that the TCOD fraction decreased more significantly in digester 1 than digester 2. 37.5% reduction in TCOD was observed in the first 6 days of the SRT for digester 1, whereas a 16.8% reduction is achieved in digester 2 for the same SRT. This justifies the advantage of applying combined microwave-ultrasonic pretreatment on the activated sludge portion of the mixture before digestion. It also shows that the higher the percentage of pretreated TEAS the greater will be solid disintegration and biodegradation. Microorganisms required for sludge oxidisation can be disrupted to a greater extent by pretreatment.

The SCOD trend showed in Figures 8.7 and 8.8 displays the SCOD fraction changing marginally over the total SRT for digester 1, whereas digester 2 shows an increase up to 6 days of digestion and then decreases progressively until SRT of 27. The fact that the SCOD fraction in digester 1 does not change can be explained by faster mixing and solubilisation of SCOD after mixing of the feed that no discernible decrease in the SCOD was observed over the digestion period. This can be supported by the rapid rate of TCOD reduction observed in the digester containing more pretreated TEAS. The results shown in digester 2 for the SCOD removal are more typical of anaerobic digestion process, as the SCOD fraction increases in the original phase of digestion due to increased solubilisation of organics in the hydrolysis stage, and then a decrease is observed due to the consumption of organics with increasing SRT. SCOD Reduction in digester 2 took place at a slower rate because of the lower fraction of pretreated TEAS in the feed. Pretreating the sludge increases the fraction of soluble organics available for oxidisation which will, in turn, increase percentage reduction in SCOD and enhances biogas production and solid removal. Furthermore, the maximum reduction achieved in the first 6 days of operation confirms advantage of pretreatment for the reduction.



(a)



(b)

Figure 8.7: (a) SCOD/TCOD ratio , (b) Changes in TCOD and SCOD in digester 1

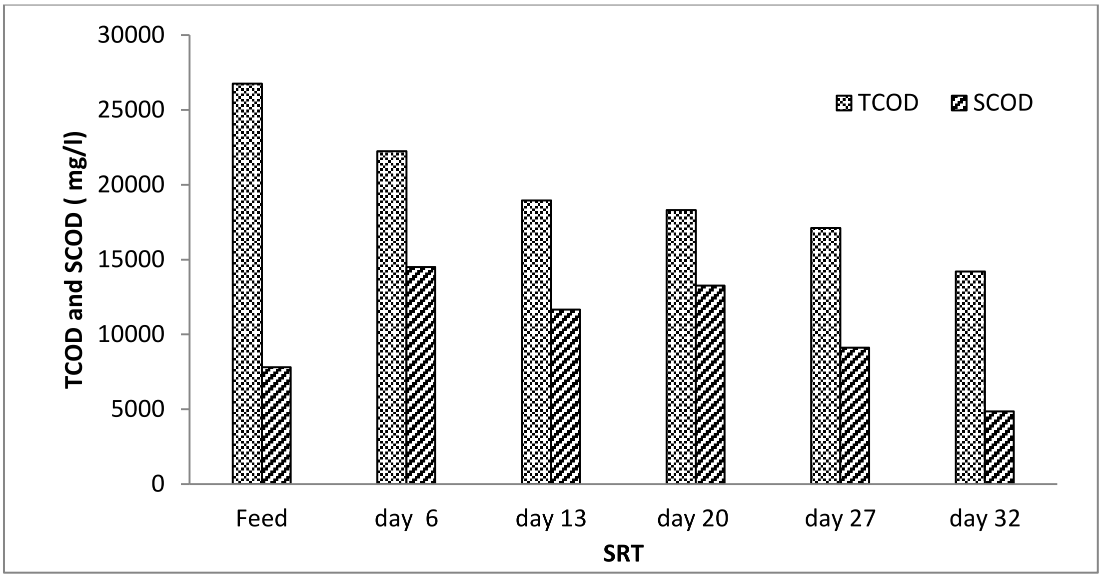


Figure 8.8: Changes in TCOD and SCOD in Digester 2

8.3.3 Effect of pretreatment on bacterial reduction

The total coliform content of sludge after digestion is one important factor determining effluent quality. Coliforms are bacteria that are associated with faecal matter and the degree of coliform and E. coli (pathogen) removal is an important target of the anaerobic digestion process (Lafitte-Trouqué and Forster, 2002). The bacterial analysis of the two digesters at the beginning and end of a 20 day SRT was completed, and the results are displayed in Table 8.2 below.

Table 8.2: Results of bacterial cell count

	25% PS + 75% TEAS Sludge (R1)			**75% PS + 25% TEAS Sludge (R2)**		
	Coliform	E.coli	Total Coliform	Coliform	E. coli	Total Coliform
Feed (number of coliform/100 mL)	135000	20000	155000	200000	55000	255000
Product (number of coliform/100 mL)	25000	0	25000	145000	0	145000

The results portray an 84% reduction in the total coliform count for digester 1, while digester 2 resulted in a lesser reduction of 44%. This is again attributed to the larger percentage of pretreated TEAS in digester one, as the pretreatment disturbs the cell membranes of the bacteria prior to digestion, which results in an improved destruction of microbial bacteria through the anaerobic digestion process. It can also be noted that complete destruction of the E. coli bacteria was observed in both digesters, which is very beneficial as these pathogens and the major nuisances which should not be present in the bio solids or the liquid effluent after the digestion process.

8.3.4 Effect on protein solubilisation

Protein is an organic compound that occupies approximately 50% of the total organics present in waste activated sludge (Shao et al., 2013).The reduction of protein can be used to measure sludge degradation in terms of the total organics removal achieved throughout the anaerobic digestion process. Figure 8.9 displays the amount of total protein available in a 500 mL of sludge at different stages of the digestion process both in digester 1 and digester 2.

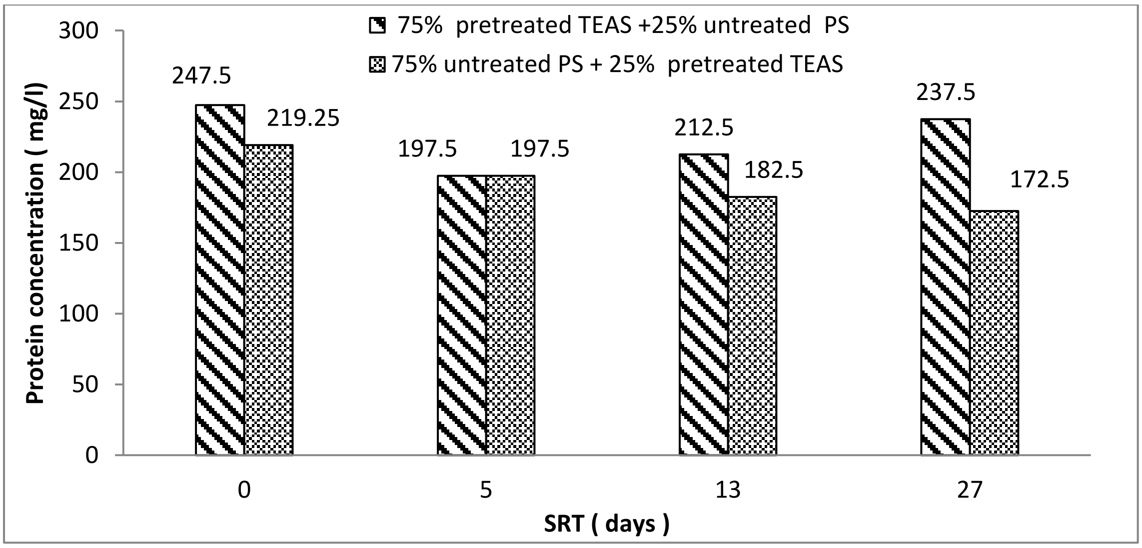


Figure 8.9: Results of protein solubilisation analysis of a 500 mL sludge sample

Figure 8.9 displays that the protein content decreases continuously in digester 2 over the total SRT considered in the study. However, the rate of reduction in the first 5 days is higher in digester 1, which is attributed to the higher fraction of pretreated TEAS in the digester feed. Greater proportion of PTEAS increases the availability of proteins for reduction by hydrolysis in the early stages of digestion. However, an increase in the protein content was observed in digester 1 after the initial period of faster protein reduction. This can be explained by the original biomass in the sludge being reduced in the first stage of digestion, and due to the solubilisation of more proteins from the original sludge and new microorganisms (methanogens) which grew more in the later stage.

8.3.5 Particle size distribution

The particle size distribution for the two digesters are shown in Tables 8.3 a and b. The average particle size of the feed sludge is different from the average particle size of digested sludge at different SRT. The particle size distribution of the two digesters generally shows similar trend, yet there is a difference in average particle size and specific surface area at different SRT. This is because of the differences in biodegradability of the different types of feed sludge. The d(0.1), d(0.5) and d(0.9) values indicate that 10%, 50% and 90% of the particles measured were less than or equal to the size stated. According to the distributions shown in Tables 8.3 a and b, particle size distribution of digester 1 appeared to have smaller particles as compared to the distribution of particles in digester 2 in the first 13 days . This is related to increased disintegration of the TEAS particles which accounts for 75% of feed in digester 1 than the case in digester 2 after the combined microwave ultrasonic pretreatment. The sludge specific surface area was derived from the particle size distribution. The specific surface area data quoted in Table 8.3 a and b illustrate that the smaller particles contributed more in terms of specific surface area than the larger size fractions and specific surface area increases with increasing SRT. Table 8.3b shows greater specific surface area of particles for digester 2 after long SRT . The smaller particle sizes are indicative of the disintegration that happened because of pre-treatment and the digestion process which has ultimately assisted the release of organic matter and biogas production.

Table 8.3 (a) Particle size distribution of sludge in digester 1 at different SRT

SRT (days)	**D (0.1) um**	**d(0.5) um**	**d(0.9) um**	**surface area (m^2/g)**	**surface weighted mean diameter (um)**	**volume weighted mean diameter (um)**
Feed	0.252	1	88.524	9.9	0.606	23.175
13	0.078	0.199	1.96	35.9	0.167	2.736
27	0.097	0.374	34.442	25.4	0.236	7.07
32	0.08	0.217	2.254	34.2	0.176	3.015

Table 8.3 (b) Particle size distribution of sludge in digester 2 at different SRT

SRT (days)	**D (0.1) um**	**d(0.5) um**	**d(0.9) um**	**surface area (m^2/g)**	**surface weighted mean diameter (um)**	**volume weighted mean diameter (um)**
Feed	0.252	1	88.524	9.9	0.606	23.175
13	0.081	0.213	1.729	34.5	0.174	2.436
27	0.081	0.195	1.191	35.9	0.167	0.629
32	0.079	0.186	1.087	37.5	0.16	0.466

8.3.6 Calculation of the hydrolysis rate constant

The hydrolysis rate constant can be calculated by using biochemical methane potential data from the digesters. The methane yield is a function of the reduction of organic material achieved during anaerobic digestion, which reflects on the hydrolysis rate. The hydrolysis rate constant is an indicator of the speed of hydrolysis achieved in the digesters. Enhancing the hydrolysis rate constant is an important factor.

The model used to determine the hydrolysis rate constant was the Gompertz equation (Gadhamshetty et al., 2010). This model represents cumulative methane production as a function of the methane production potential, maximum methane production rate and duration of the lag phase. The equation is shown below.

$$M = P \times \exp\left\{-exp\left|\frac{R_m \times e}{P}(\lambda - t) + 1\right\}\right. \qquad (8.1)$$

where: M is the cumulative methane production (mL),

P is the methane production potential (mL),

Rm the maximum methane production rate (mL/d), λ is the duration of the lag phase (d), and

t is the duration of the assay in which cumulative methane production M is calculated (d).

Using the non-linear regression model, the Gompertz equation was used to develop the predictive model curves based on the experimental results achieved for the cumulative methane production in both digester 1 and 2. These results are shown in Figures 8.10 and 8.11.

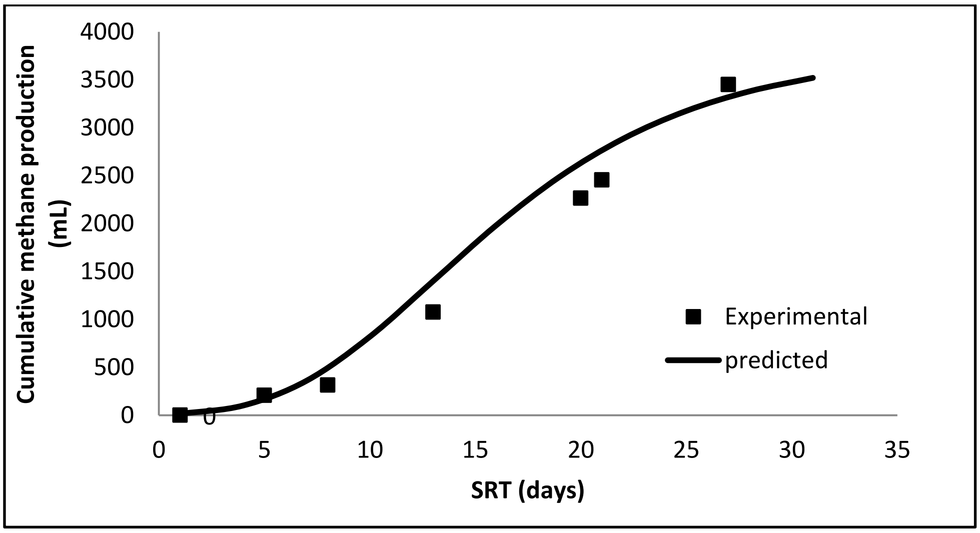


Figure 8.10: The results of modelling the Gompertz equation to the methane production for digester 1

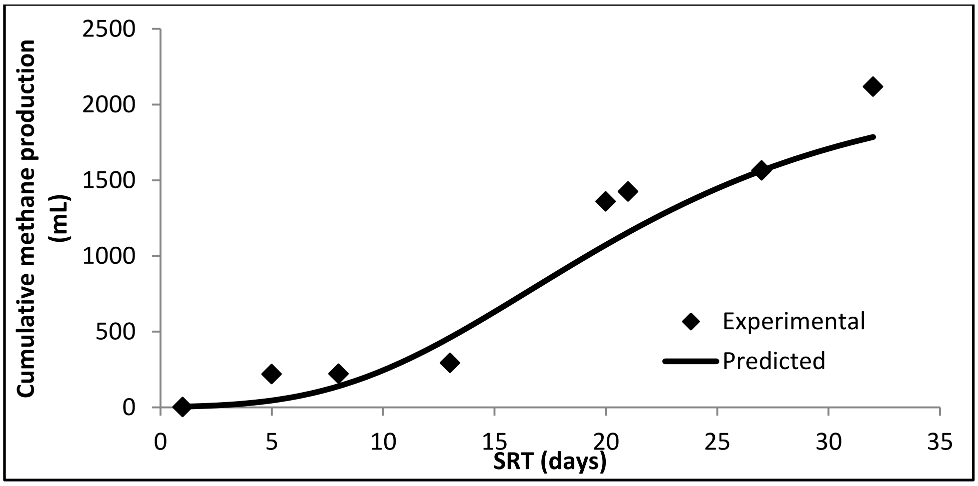


Figure 8.11: The results of modelling the Gompertz equation to the methane production for digester 2

Subsequent to fitting the equation to the experimental results, the values of the methane production potential (P), maximum methane production rate (Rm), and the duration of the lag phase (λ) can be calculated by applying a least squares regression fit to the experimental data obtained.

Table 8.4: Results for the methane potential, daily rate and lag time

	25% PS + 75% TEAS Sludge (R1)	**75% PS + 25% TEAS Sludge (R2)**
Methane Potential, P (mL)	3451.58	1868.22
Maximum methane production rate, Rm (mL/day)	180	100
Lag time, λ (days)	6	8
Correlation Coefficient (R^2)	0.993	0.98

Table 8.5 below displays the values for these parameters determined for each digester and the correlation coefficient (R^2), which indicated the fitting between the experimental and theoretical models. Furthermore, Table 8.4 depicts the experimental and predicted values of the cumulative methane production after an SRT of 20 days.

Table 8.5: Predicted and experimental methane production at SRT = 20 days

Cumulative methane (mL)	**25% PS + 75% TEAS Sludge (Digester 1)**	**75% PS + 25% TEAS Sludge ((Digester2)**
Predicted	2561.7	1268.5
Experimental	2455.2	1424.5

From the data presented in the tables above, it can be observed that the Gompertz equation is suiTablefor estimating the methane production trend observed in the experiment. High daily methane production potential and shorter lag time calculated for digester 1 as compared to digester 2 correlates well to the experimental results discussed in the above section of this chapter. Higher daily methane production rate and shorter lag time in digester 1 is attributed to the higher proportion of combined microwave-ultrasonic pretreated sludge in the digester feedstock, which reduces the time taken to reach the methanogenesis stage of digestion.

The hydrolysis rate constant k can be determined from this data using first-order rate equation 8.2 shown below. The value for this constant helps to understand the kinetics of the digestion process, and evaluate the effect that the combined microwave-ultrasonic pretreatment method and mixing ratio has on the performance of the anaerobic digestion of sludge.

$$\mathrm{M} = \mathrm{P}(1 - \exp(-\mathrm{kt})) \quad (8.2)$$

Where: M = cumulative methane production (mL) at time, t (day),

P = methane production potential (mL) which was assumed to be equal to the final cumulative methane volume.

By linearizing the results calculated from the above equation, the relationship between the cumulative methane production and time is determined. Figure 8.12 displays these results graphically and Table 8.6 shows the values for the hydrolysis rate constant for each digester.

Table 8.6: Anaerobic digestion rate constants determined for digester 1 and 2

	25% PS + 75% TEAS Sludge (R1)	**75% PS + 25% TEAS Sludge (R2)**
Kinetic constant	0.0922	0.0599
Correlation Coefficient	0.9	0.9

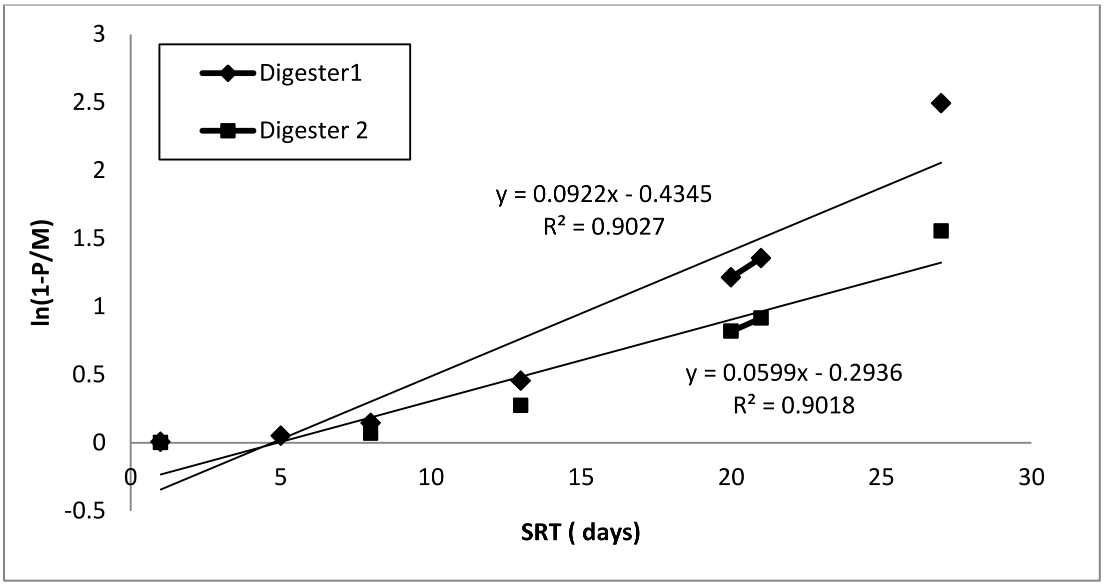


Figure 8.12: Determination of the first order hydrolysis rate constant

Table 8.6 shows that the rate of hydrolysis observed in digester 1, containing a larger portion of pretreated TEAS is faster than that of digester 2. This perfectly agrees with the experimental results discussed throughout this section about the effects of the combined microwave-ultrasonic pretreatment method in reducing the time required for the completion of the hydrolysis stage of digestion. Moreover, higher percentage of PTEAS (greater PTEAS mixing ratio) resulted in greater enhancement in the kinetics of the anaerobic digestion process.

8.4 Conclusion

Separate pretreatment of TEAS before mixing with primary sludge resulted in substantial improvement in the biodegradability, solid reduction, methane production kinetics and biogas quality, protein removal, microbial destruction and overall performance of anaerobic digestion process. Furthermore, higher percentage of the pretreated TEAS increases the digestion kinetics, the methane production capacity and the biogas quality. Whereas, greater volatile and total solid removal was achieve for the digester with greater percentage of primary sludge. Great percentage of primary sludge in the anaerobic feed sludge mixed with pretreated activated sludge can be easily digested unlike the digestion kinetics and performance of raw primary sludge. The significance of the findings of this study in large scale wastewater treatment plants is enormous in terms of reducing the sludge treatment and handling costs. It will also help to enhance anaerobic digestion kinetics and overall performance.

CHAPTER 9

EFFECT OF MIXING RATIO AND ORGANIC LOADING RATE ON ANAEROBIC DIGESTER PERFORMANCE AND SLUDGE BIODEGRADABILITY

Abstract

In this chapter, the effect of mixing ratio of primary sludge (PS) to excess activated sludge (EAS), organic loading rate (OLR) and sludge retention time (SRT) on biogas production capacity and sludge biodegradability was investigated. Primary sludge /excess activated sludge ratios of 65/35 v/v, 50/50 v/v, 35/65 v/v were assessed for a sludge retention time of 23 days at mesophilic temperature of 36.5^0C. The performance of the anaerobic digestion process was also tested for the organic loading rates of 0.7 g VS/L- 2 gVS/L and HRT of 5, 10, 15 and 20 for combined microwave pretreated and untreated sludge. The sludge with the mixing ratio of 65/35 v/v produced the highest amount of methane (485 ml of CH4 or 36.5ml/g TCOD) for the 500 ml reactor volume considered in the study. The kinetics of the digestion process was faster for this mixing ratio. The methane/ carbon dioxide ratio was found to be highest (2.5 -3.1) for the sludge sample with the mixing ratio of 65/35 v/v. While considering biodegradability, TCOD reduction of 46.6%, 53.7%, 72.3% and volatile solid removal of 32.6%, 25.8%, and 34% was achieved for mixing ratios of 65/35, 50/50 and 35/65 respectively. The highest reduction in TCOD and VS was achieved for the sample with more EAS (35/65) as the microbial biomass for this sample is greater. Moreover, for an increased organic loading rate and shorter HRT (5 days), combined microwave-ultrasonic pretreated sludge resulted in higher reduction in VS and COD compared to the untreated sludge. Furthermore, the microbial content and the Rheology of the digested sludge samples were analysed. In general, mixed sludge with higher proportion of raw primary sludge has better effects on biodegradability and quality of digested sludge.

9.1 Introduction

The objective of this chapter is studying effect of mixing ratio, organic loading rate and HRT on methane production capacity, solid reduction capacity and sludge dewaterability experimentally. Determining the optimum mixing ratio and organic loading rate enhances methane production, effluent sludge quality, dewaterability and pathogen removal. The behavior of digesters under changing organic loading rates (OLR) is unpredictable. Hence, thorough investigation on the effect of operational parameters is essential for all different sludge types (Noutsopoulos et al., 2013). The effect of mixing ratio between primary sludge and excess activated sludge on digester performance is discussed in this chapter. The effect of organic loading rate and sludge retention time on anaerobic digester performance for pretreated feed sludge is also presented. The last part of the chapter provides a general overview of the rheological characteristics of untreated and pretreated sludge and the effect of pretreatment conditions and solid concentration on sludge rheology is also included in this section.

9.2 Materials and methods

9.2.1 Sampling and characterization

Primary sludge was collected from primary gallery, primary sedimentation tank No. 4 and Excess Activated Sludge (EAS) was collected from Module 4 of the secondary treatment section of BWWTP. Primary and excess activated sludge samples were mixed with ratios of 65/35, 50/50 and 35/65 v/v and were introduced to the jacketed digesters. Samples were withdrawn from each anaerobic digester for characterization purpose. The characteristics of sludge fed to the three digesters are presented in Table 9.1.

Table 9.1 Characteristics of feed sludge with different mixing ratios

Parameter	**Reactor 1 (65/35)**	**Reactor 2 (50/50)**	**Reactor 3 (35/65)**
TS (%)	2.1	1.4	1.5
VS (% TS)	89.1	86.2	87
TCOD (g/l)	53.3	44.4	40.1
pH	7.1	7.1	7.2

9.2.2 Experimental setup for methane potential and sludge biodegradability tests.

In the experimental setup, the biochemical methane potential tests were conducted in 1L continuously-stirred batch anaerobic digesters. These simultaneously operating three single-stage digesters were kept at a mesophilic temperature of 36.5^0C and were first fed with 50 ml digested sludge as seed for inoculation purpose. The digesters were acclimated with the digested sludge for 5 days and were separately fed with equal 450 ml of mixed sludge samples with the characteristics as given in Table 9.1. The pH in each digester was adjusted to 7.0 using sodium hydroxide and hydrochloric acid. The methane generated was allowed to pass through buffer tanks to remove any condensate before the gas volume was measured in inverted cylinders by water displacement technique. The biogas composition and other parameters were continuously monitored using the methods described in section 3.4 until biogas generation ceased at SRT of 23 days.

9.2.3 Effect of organic loading rate and sludge retention time on sludge biodegradability and digester performance.

The experiment on the effect of organic loading rate and sludge retention time was performed in the continuous mode by increasing the organic loading rate progressively. Hydraulic retention time was decreased correspondingly from 20 day down to 5 days as shown in Table 9.2. Steady state operation and digester stabilization was achieved at each stage before changing the OLR or HRT.

Table 9.2: Analysis of effect of SRT and OLR for digestion of pretreated samples

Parameter to be tested for the pretreated mixed sludge feed	**Ranges and conditions for the experiment**
Effect of organic loading rate (OLR)	3.96 -15.6 gTCOD/l/Day)
Effect of hydraulic retention time (HRT)	5, 10, 15, 20 days of SRT

9.2.4 Rheological investigation on different sludge types and pretreatment conditions

Raw primary, excess activated sludge thickened excess activated sludge and mixed sludge samples were homogenized on a magnetic stirrer for duration of 10 minutes. Sludge samples pre-treated under different ultrasonication and microwave pretreatment conditions were prepared in the same manner for the rheological tests. Homogenised samples of untreated and pretreated sludge were subjected to rheological measurement on HAAKE MARS Rheometer from Thermo SCIENTIFIC for the rheological tests during anaerobic digestion. All tests were conducted at a temperature of 25^{0}C. The temperature was controlled by water bath heater connected to the Rheometer. The shear stress versus shear rate and viscosity versus shear rate curves were plotted for raw untreated sludge samples and microwave-ultrasonic pretreated sludge samples at various treatment conditions from the other experiments in the study. The plots were assessed as to which model they correspond or fit better, visco-plastic model like Bingham and Herschel-Buckley model or shear thinning model like Ostwald model.

9.3 Result and discussion

9.3.1 Effect of various sludge mixing ratios on methane production

Figure 9.1 shows that anaerobic digester with mixing ratio, 65/35 v/v had the highest average daily methane production rate of 69.3 ml/day followed by daily rates of 47.2 ml/day and 37 ml/day for mixing ratios of 50/50 v/v and 35/65 v/v respectively. Higher primary sludge content favoured higher methane production. Total volume of methane produced was the highest at 36.5 ml/g TCOD for the sludge sample with greater proportion of primary sludge followed by 26.2 ml/g TCOD and 25.9 ml/g TCOD for the 50/50 v/v and 35/65 v/v mixtures. The kinetics of biogas production was also much higher for PS: EAS= 65/35. It has been stated that biogas production from primary sludge is higher unless the sludge contains less digestible complex organics like cellulose and lignin (Hanjie, 2010).

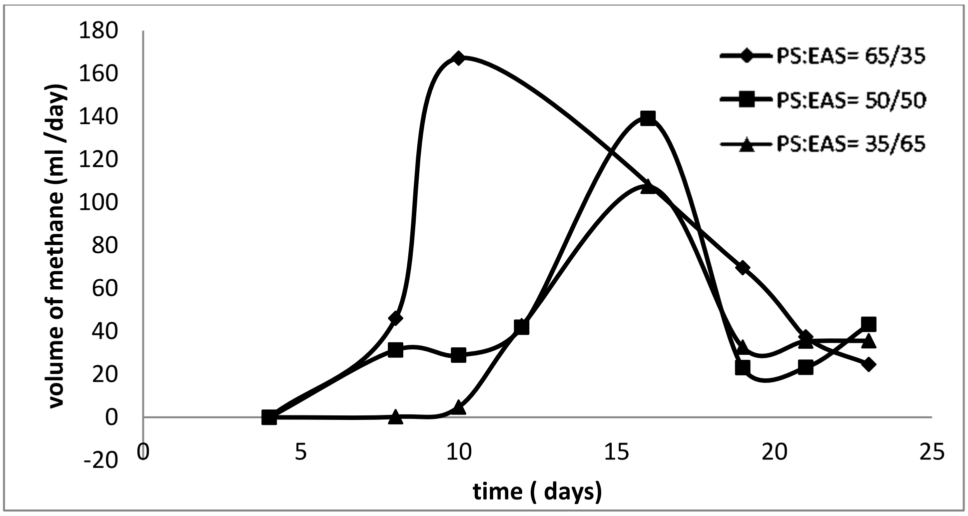


Figure 9.1 Daily methane productions for the three different sludge compositions

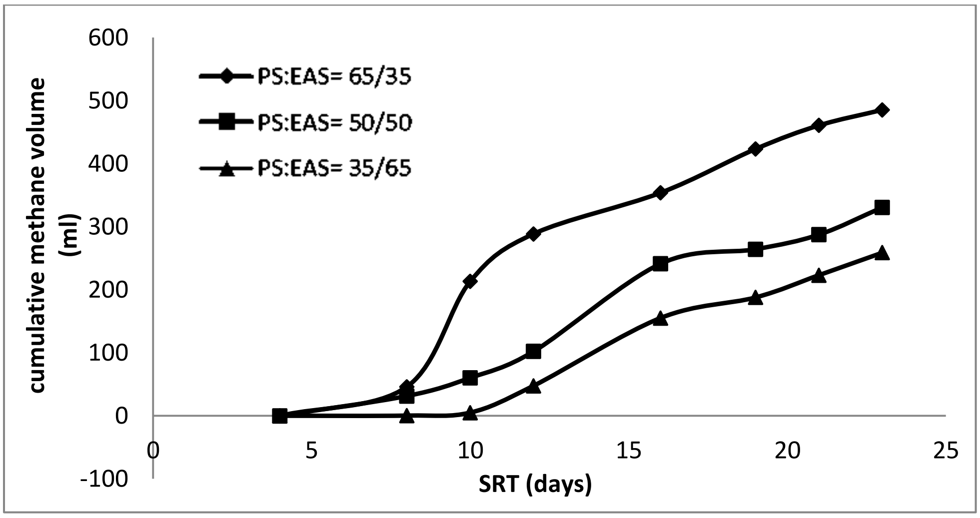


Figure 9.2 Cumulative methane productions.

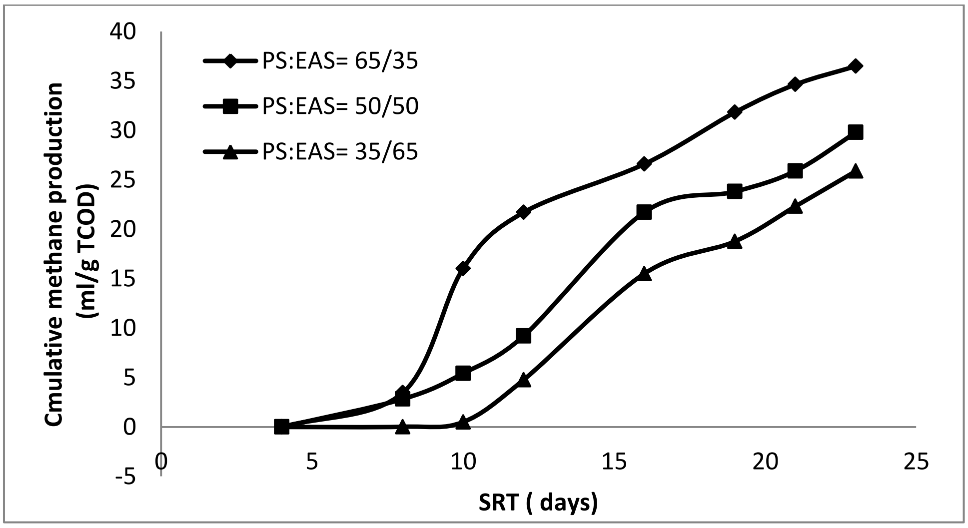


Figure 9.3 Specific methane productions.

9.3.2 Effect of mixing ratio on sludge biodegradability (COD and VS removal)

TCOD reduction of 46.6%, 53.7%, 72.3% and volatile solid removal of 32.6%, 25.8%, and 34% was achieved for mixing ratios of 65/35, 50/50 and 35/65 respectively as shown in Figure 9.4 and 9.5. The highest reduction in TCOD was achieved for the sample with more EAS (35/65) as the microbial biomass and biodegradable organic for this sample is greater. In terms of VS removal, greater percentage reduction was also obtained for this mixing ratio as shown in Figure 9.6. However, the sample with mixing ratio of 65/35 had the highest methane production of 36.5 ml /g TCOD. The hydrolysis and biodegradation rate for this process was also faster than the other two combinations. Greater methane gas quality was achieved for the sludge with mixing ratio of 65/35 v/v as shown in Figure 9.7.

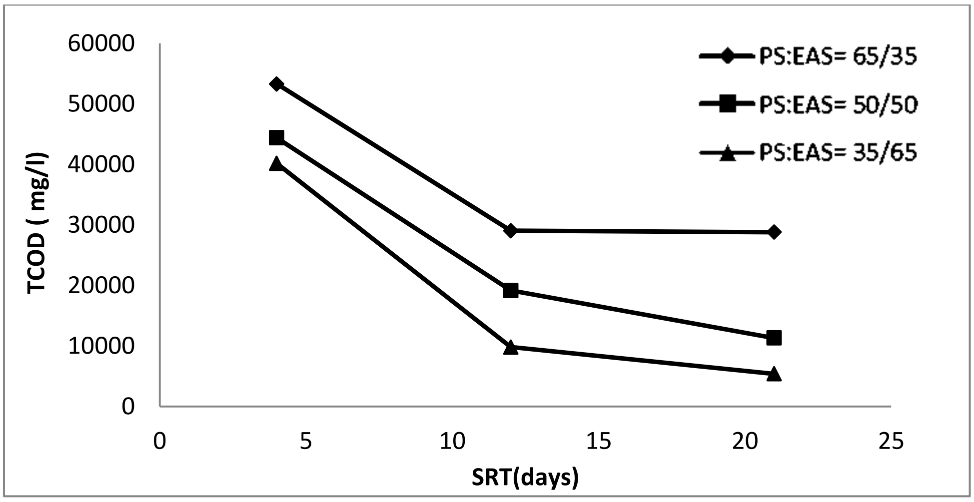


Figure 9.4 Reduction in TCOD during the anaerobic digestion process

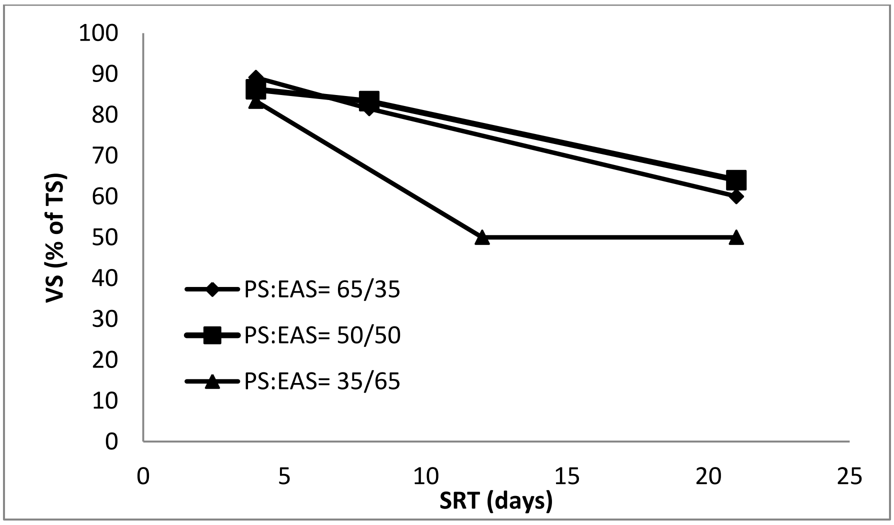


Figure 9.5 Reductions in VS during the anaerobic digestion process.

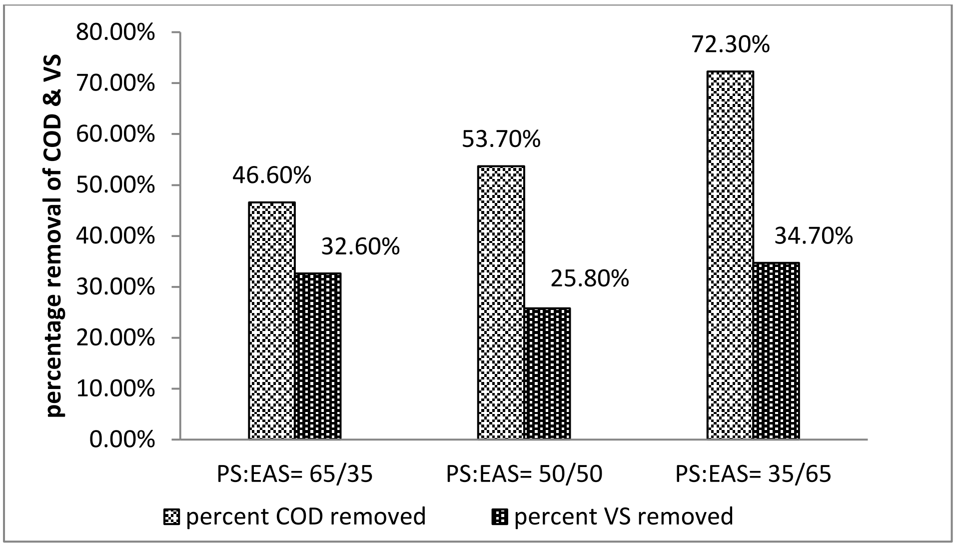


Figure 9.6 Percentage reductions in COD and VS content.

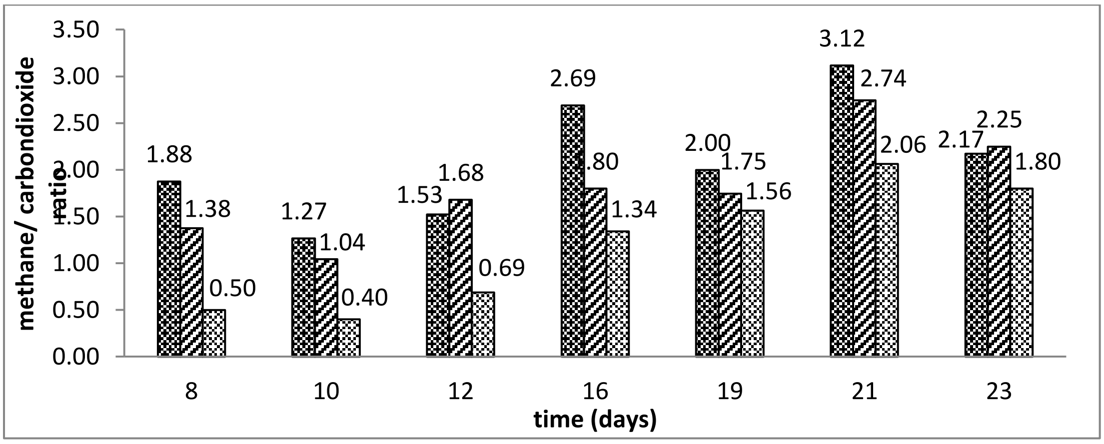


Figure 9.7 Average methane/carbondioxide ratio in biogas generated during anaerobic digestion of different kinds of sludge.

9.3.3 Effect of mixing ratio on sludge dewaterability

In this study, the CST results showed that the sample with more EAS had better dewaterability. The lesser the concentration of EAS the bigger was the CST value in seconds (reduced dewaterability) (Figure 9.8). This is due to Extracellular polymeric substances that are present more in activated sludge than the primary sludge, such polymeric substances assist floc formation and result in subsequent improvement of dewaterability (Eskicioglu et al., 2006).

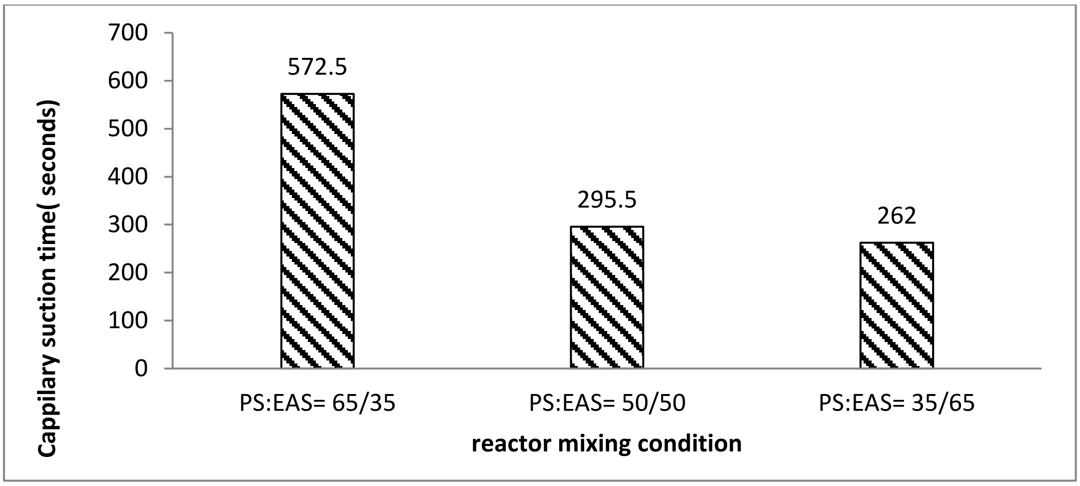


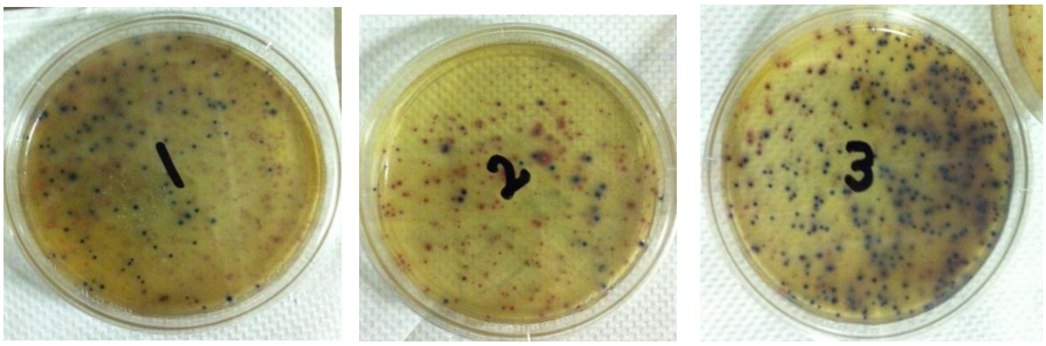
Figure 9.8 Dewaterability of digested sludge samples.

9.3.4 Microbial content and sludge mixing ratio

The microbial biomass content of digested sludge from the three digesters was estimated using the bacterial count method shown in section 3.4.11. The digested mixed sludge sample with more EAS contained more E coli and coliform as shown in Figure 9.9. The destruction of pathogens and microorganisms was also one of the targets. Hence, the pathogen removal for the mixed sludge with 50/50 mixing ratio was greater than the others as shown on Table 9.3.

Table 9.3 Microbial count for the digested sludge with different mixing ratio

Reactor type	E. coli	Coliform	Total
PS:EAS= 65/35	10100	11000	21100
PS:EAS= 50/50	1700	14700	16400
PS:EAS= 35/65	22500	4100	26600



(a) **(b)** **(c)**

Figure 9.9 Test for microbial content in the mixed sludge samples with different mixing ratios: (a) 65/35 v/v, (b) 50/50 v/v and (c) 35/65 v/v.

9.3.5 Determination of hydrolysis rate constant, lag time and daily methane production based on Gompertz equation

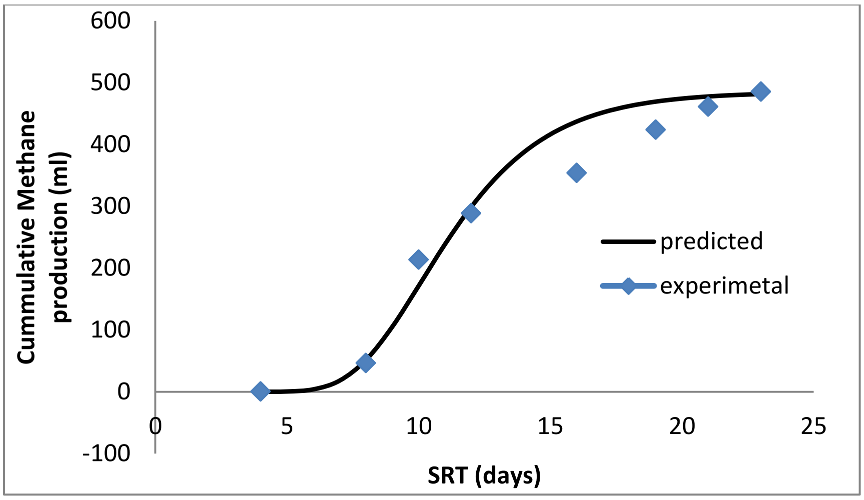
Methane production can be used to represent hydrolysis rate of particulate organic matter when there is no accumulation of intermediary products. There are several model equations used for the determination of the hydrolysis constant. In this work, the lag-phase before the start of methane production, the methane production potential and the maximum methane production rate were determined using the Gompertz equation.

$$M = P \times \exp\left\{-exp\left|\frac{Rm \times e}{P}(\lambda - t) + 1\right.\right\} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \text{(9.1)}$$

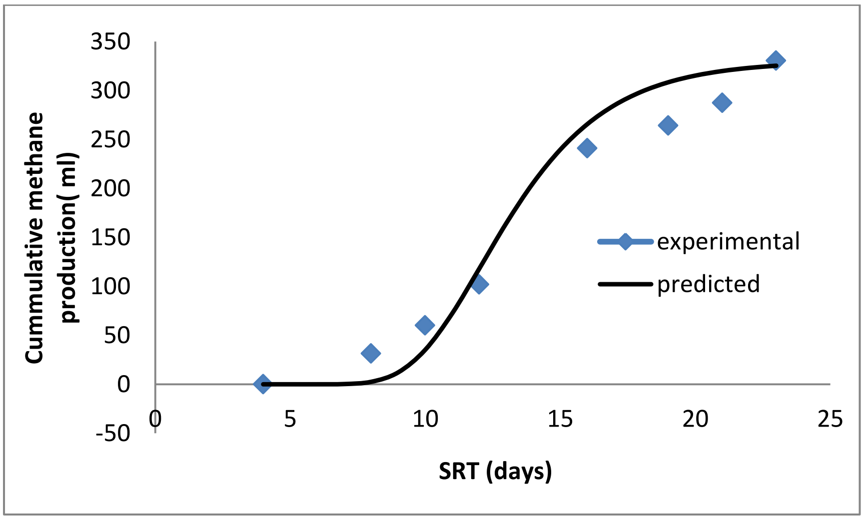
where M is the cumulative methane production (mL), P is the methane production potential (mL), Rm the maximum methane production rate (mL/d), λ is the duration of the lag phase (d), and t is the duration of the assay in which cumulative methane production M is calculated (d).

9.3.6 Preliminary prediction of methane production based on Gompertz model

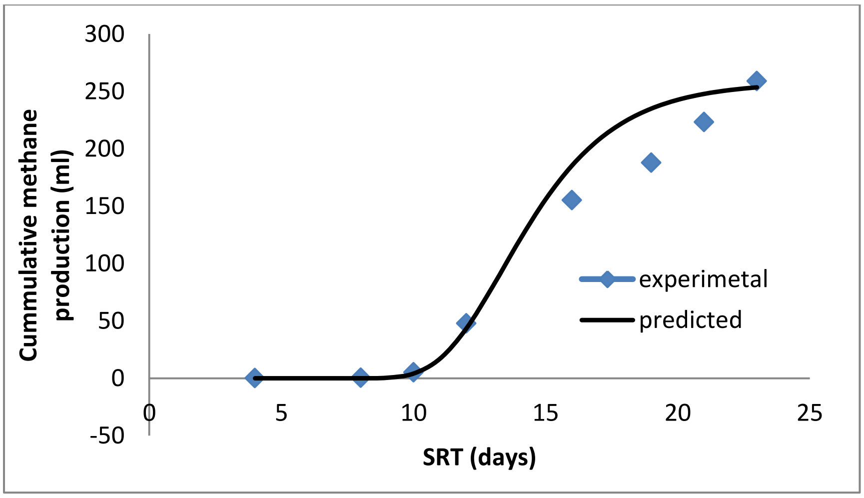
The experimental date from anaerobic digestion study that was carried out to investigate effect of sludge mixing ratio was employed for the model fitting and prediction. Gompertz equation was applied to predict the methane potential, lag time, daily and cumulative methane production by the non-linear regression method.



(a)



(b)



(c)

Figure 9.10: Prediction of methane production by Gompertz model. (a) PS:EAS= 65/35, (b) PS:EAS= 50/50, (c) PS:EAS= 35/65

The parameters P, λ, and Rm from the equation were estimated by applying a least squares fit of the above equation to the experimental data set. The results from this model were compared to those obtained from experimental investigation for the effect of mixing ratio.

Table 9.4: methane potential, daily rate and lag time for anaerobic digestion experiment for different mixing ratios.

sludge mixing ratio	**methane potential (P)**	**Max. Daily rate (Rm)**	**lag time (λ)**	**R^2**
PS: EAS=65/35	481.5	68.5	8.5	0.985
PS: EAS=50/50	325.32	47.2	10.5	0.989
PS: EAS=35/65	253.5	40	12	0.989

Table 9.5: Predicted and experimental methane production

Cumulative methane (ml)	**PS: EAS=65/35**	**PS: EAS=50/50**	**PS: EAS=35/65**
Predicted	481.6	325.32	256
Experimental	485.42	330.67	258.81
standard deviation	1.91	2.675	1.405

It can be observed from Tables 9.4 and 9.5 that the predictions made based on Gompertz model fit well to the experimental data with very high correlation coefficient of 0.99. The methane production potential and daily rate for the mixed sludge sample with higher proportion of primary sludge was higher with shorter lag time. This confirms well the higher anaerobic and methanogenic activity achieved for the anaerobic digestion with greater proportion of primary sludge.

9.3.7 Hydrolysis rate constant determination

The rate of hydrolysis is the key step in anaerobic digestion process that determines the methane production rate, sludge retention time and overall performance of the digester. Determination of the hydrolysis rate constant helps to quantitatively understand the kinetics of the process. Hydrolysis rate constant K for the anaerobic digestion experiment on effect of mixing ratio was described as first order rate kinetics. Thus the production of methane was assumed to follow the equation:

$$M = P.(1 - \exp(-kt)) \quad \text{..(9.2)}$$

Where M represents the cumulative methane production (ml) at time t (day), P is the methane production potential (ml) and was assumed to be equal to the final cumulative methane volume. The estimation of the first order hydrolysis constant was made by linearizing equation (9.2) and the linearized plot given in Figure 9.11. As the pH in the experiment was always in the range of 6.8-7.2, the consumption of volatile fatty acid was significant that there was no accumulation of VFA.

Table 9.6: Determination of hydrolysis rate constant

Sludge Mixing ratio	kinetic constant (-d)	Correlation coefficient
PS: EAS= 65/35	0.256	0.86
PS: EAS= 50/50	0.279	0.89
PS: EAS= 35/65	0.233	0.82

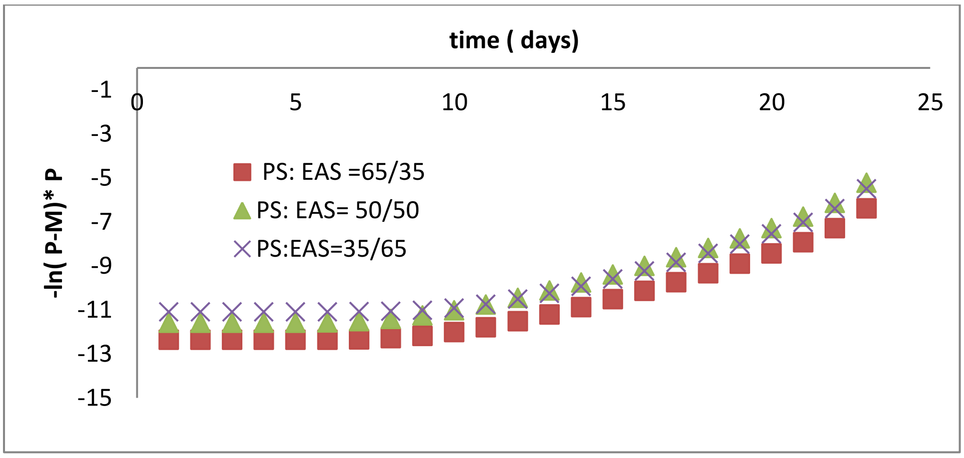


Figure 9.11. First order rate constants.

9.4 Effect of organic loading rate and Hydraulic retention time on sludge biodegradability

Based on the SRT tests presented in previous chapters, combined microwave ultrasonic pretreatment resulted in significant improvement of the process kinetics that higher degree of sludge solubilisation and biogas production can be achieved at shorter retention time. As shown in section 8.3.6, the lag phase of the hydrolysis was completed in less than 6 days and the process reached steady state at SRT of less than 15 days. Likewise study on organic loading rates for hydraulic retention time of 5, 10, 15, 20 days shows that Methane production for combined microwave ultrasonic pretreated thickened excess activated sludge was significantly higher at organic loading rate 3.96 gTCOD/l day which corresponds to 5 days of HRT. The maximum percentage of methane recorded for pretreated TEAS was 71% with 26% carbon dioxide. The removal of VS was improved by 50% due to the pretreatment and the release of organics and their disintegration increased the SCOD/TCOD ratio to 66% and the reduction in SCOD/TCOD ratio was 12 % higher for pre-treated TEAS resulting in increased average daily methane production rate of 782 ml/day. The average daily methane production was 592 ml/day for the untreated TEAS at the specified organic loading rate. The COD reduction achieved was 68.7 % and the volatile solid removal achieved was 71.5 %.

9.5 Rheological study on untreated and pre-treated sludge

The rheology of raw primary, excess activated and mixed sludge was studied before and after pretreatment. Homogenised samples of different digested and feed sludge were mounted on to the rheometer and the shear stress versus shear rate and viscosity versus shear rate curves were plotted for raw untreated sludge samples and microwave-ultrasonic pretreated sludge samples at various treatment conditions from the other experiments in the study. Sludge has a very dynamic and versatile character (Eshtiyaghi, 2013). The rheograms for different kinds of sludge or similar sludge samples subjected to different pretreatment conditions were found to be very different from each other. Sewage sludge is categorized under the class of non-Newtonian fluids and it manifests shear thinning behaviour. The flow patterns and rheology of sludge in wastewater treatment plants particularly before and after the anaerobic digestion process affects the pumping costs and the dewaterability of digested sludge. In this section, the rheology of raw primary, thickened excess activated, mixed and digested sludge were studied for different microwave-ultrasonic pretreatment conditions.

The shear stress versus shear rate and viscosity versus shear rate curves were plotted for raw untreated and microwave-ultrasonic pretreated sludge samples at various pretreatment conditions. The effect of solid concentration on the rheological properties of thickened excess activated sludge was also studied. The rheograms for primary, thickened excess activated, mixed and digested sludge samples subjected to different pretreatment conditions were found to be very different from each other. Increasing ultrasonication time improved sludge rheology.

9.5.1 Rheology of various untreated and pre-treated sludge samples

Rheological measurement can be a very useful tool for the characterising sewage sludge. But, flocculation and formation of aggregates makes rheological measurements a bit difficult. Generally, sludge shows liquid, Plastic and solid behaviours. The rheograms presented in Figures 9.12-9.20 show the shear stress versus shear rate for untreated and pre-treated primary and mixed sludge systems for a shear rate (1/S) ranging from 0 to 500. Sludge flow seems to be obstructed due to the size of the internal structure of the sludge flocs. Solid concentration is another major factor affecting sludge rheology. This can be well confirmed from the difference in the rheograms for primary and mixed sludge samples as shown in Figures 9.12 and 9.13.

Besides, the total solid content and the amount of polymeric substances is responsible for the significant variation in the rheological property among different sludge samples. Pretreatment has the effect of flocs destruction or deflocculation either my mechanical action or alteration of the composition and physico-chemical characteristics of the sludge. Particle size and dewaterability progressively decreased and specific surface area increased and the rheology improved during the digestion process for all pretreatment conditions as discussed in previous chapters. The shear stress versus shear rate (strain) plots for different ultrasonication times shows how ultrasonication condition affects the flow properties (shear rate and viscosity).

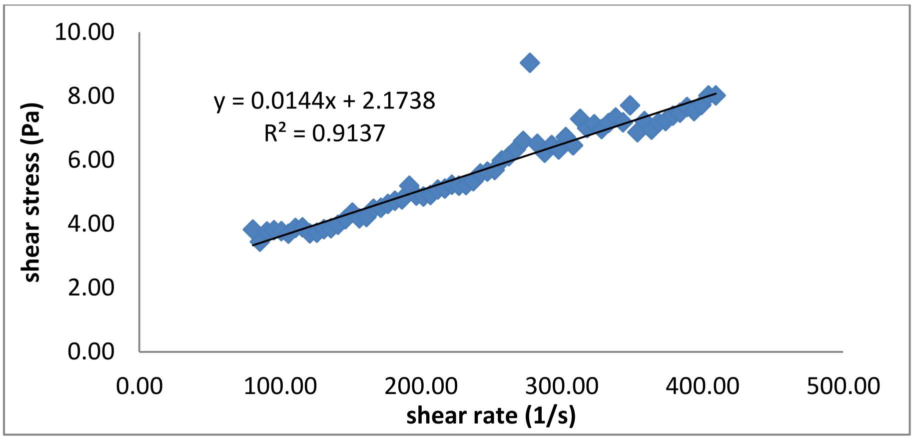


Figure 9.12: Shear stress as a function of shear rate for feed mixed sludge (70 primary: 30 excess activated sludge) before treatment.

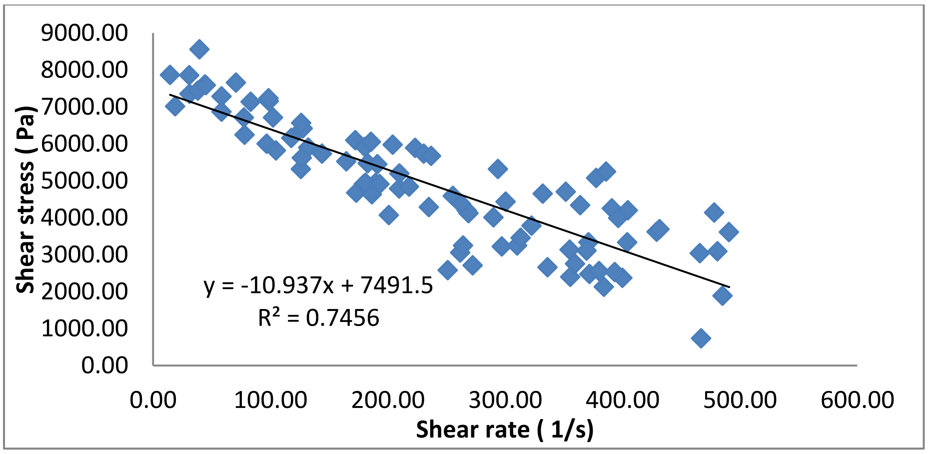


Figure 9.13: Shear stress as a function of shear rate for feed primary sludge before treatment

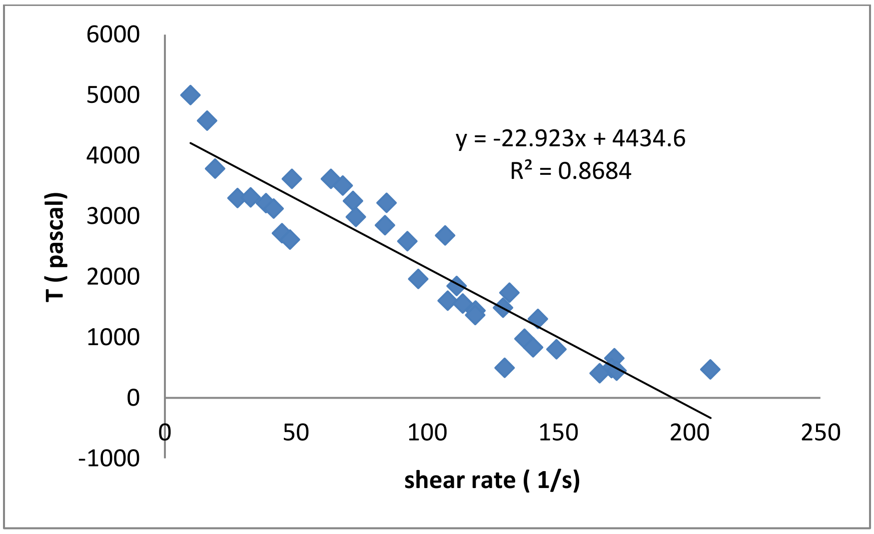


Figure 9.14: Rheology of microwave pretreated primary sludge

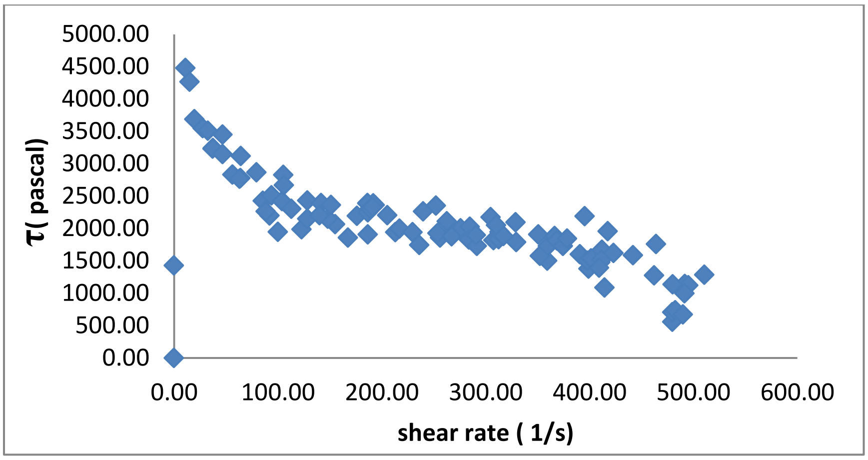


Figure 9.15: Rheology of mixed sludge (75% primary sludge)

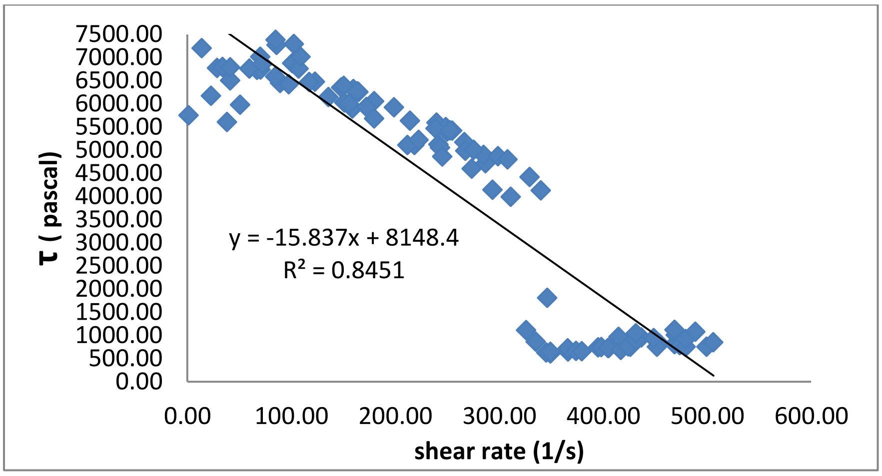


Figure 9.16: Rheology of mixed sludge after ultrasonic-microwave pretreatment (140W)

9.5.2 Effect of microwave-ultrasonic pretreatment on sludge rheology

The Rheology test for microwave-ultrasonic treated sludge samples at 4 min, 6 min and 12 min of ultrasonication time and 150W of ultrasonication power showed a decrease in viscosity for increasing shear rate making the trend more and more logarithmic. Shear thinning increased with increasing shear rate (Figure 9.17). A decreasing trend in shear stress was obtained for 6 minute of ultrasonication time (Figure 9.19) while the trend was slightly increasing for other ultrasonication times (Figure 9.18, 9.20).

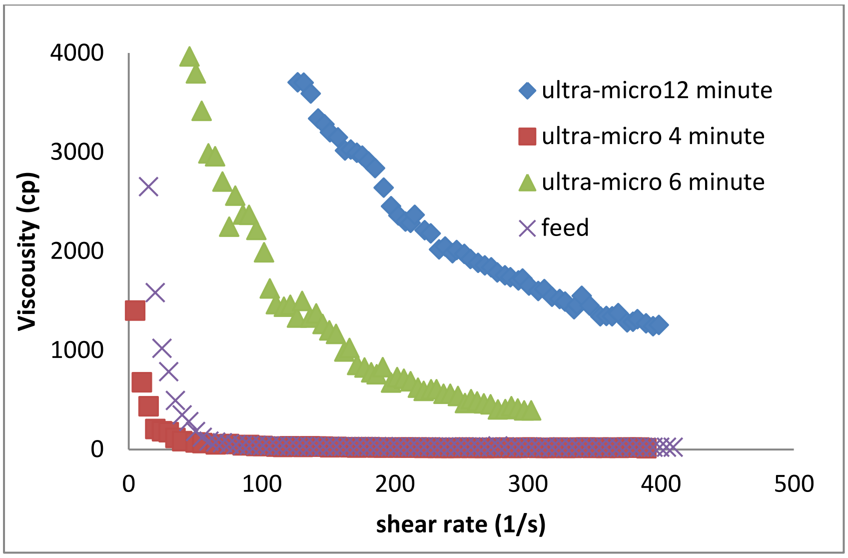


Figure 9.17: Viscosity as a function of shear rate for microwave-ultrasonic pre-treated sludge at varying ultrasonication time

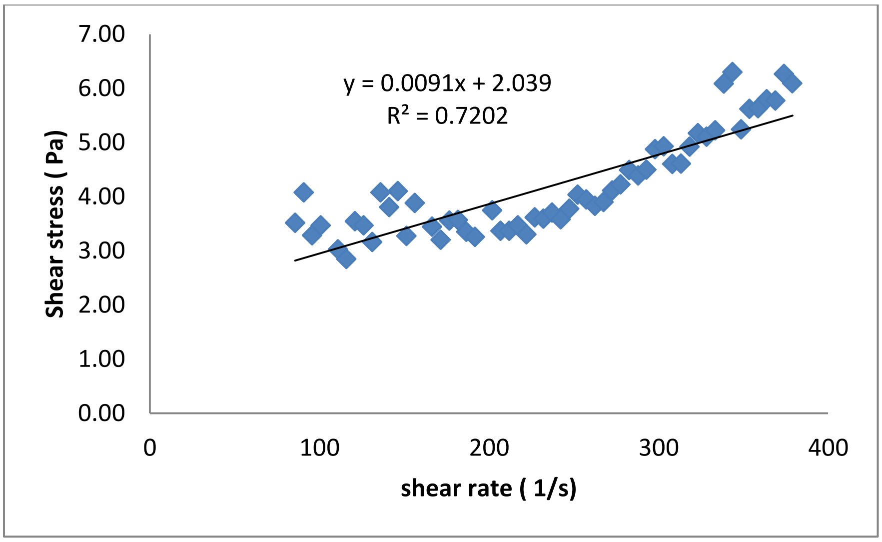


Figure 9.18: Shear stress as a function of shear rate for microwave- Ultrasonic treated sludge (4 minutes of ultrasonication time)

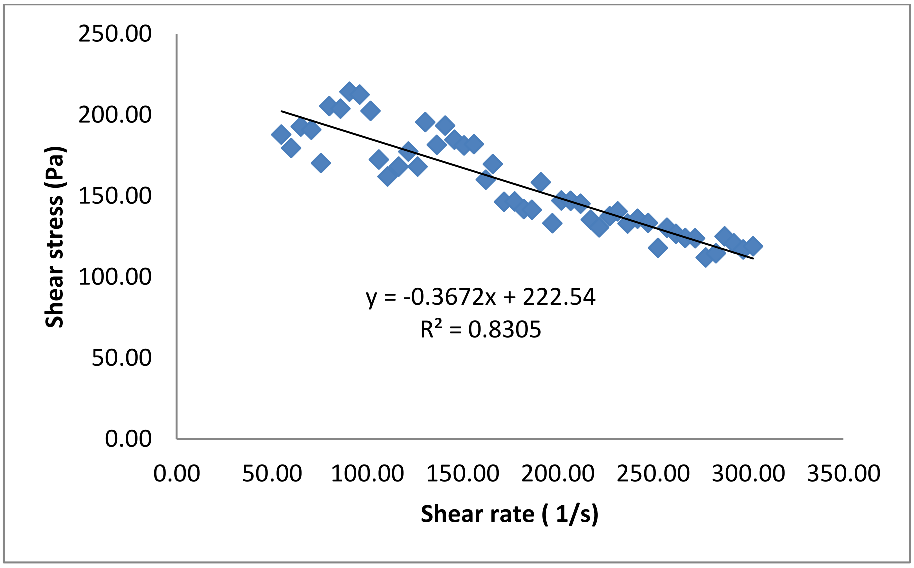


Figure 9.19: Shear stress as a function of shear rate for microwave- Ultrasonic treated sludge (6 minutes of ultrasonication time)

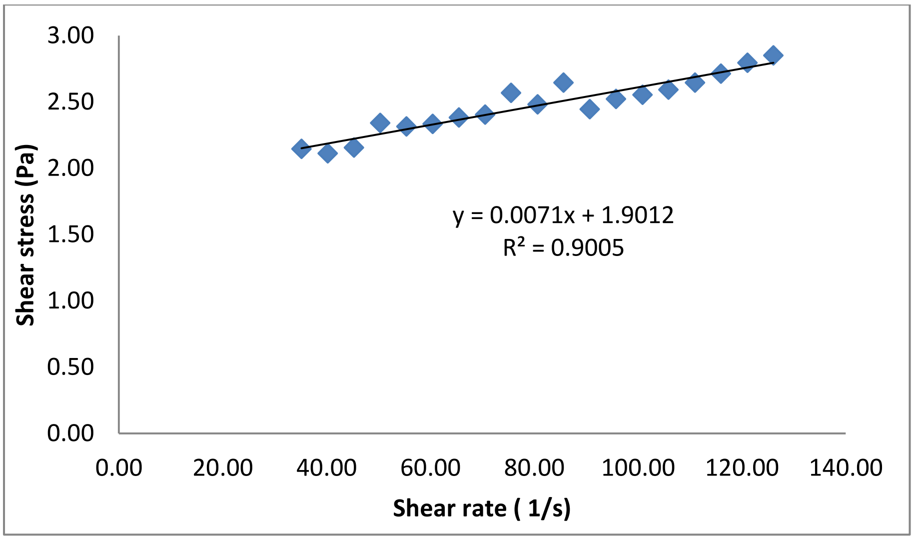


Figure 9.20: Shear stress as a function of shear rate for microwave- Ultrasonic treated sludge (8 minutes of ultrasonication time)

9.5.3 Rheology of thickened excess activated sludge (TEAS) against solid concentration at 25^{o} C

The total solid concentrations of TEAS considered in the study were 1.6%, 2.3% and 3.1 % TS. The rhograms generated using HAAKE MARS Rheometer from Thermo SCIENTIFIC for each of the thickened excess activated sludge samples are presented in Figures 9.20-9.27. Total solid concentration has significant impact on the viscosity and shear stress of all sludge samples. The rheology of thickened excess activated sludge at higher solid concentration is represented by Herschel-Buckley model which combines the power law before the sludge yields and begins to flow and the yield stress term which quantifies the amount of the stress on the sludge at the yielding point. The Bingham plastic model described well the rheology at lower solid concentration. The shear stress versus shear rate plot on Figure 9.21 can be represented by two models. The plot fits to the power law in the shear rate range of 1- 160 1/s as shown in Figure 9.22, and it fits to the Bingham plastic model in the shear rate range of (160-5001/s) as shown in Figure 9.23. The fluid consistency index (K) is 24.43 and the flow behavior index (n) is -0.347 for the first part of the plot which fits to the power law as shown in Figure 9.22. In the second part of the plot, the critical shear stress (Ty) is 3.88 as shown in Figure 9.23.

Viscosity versus shear rate curves obeyed the power law for all concentrations considered in the study (Figure 9.24, 9.26 and Figure 9.28).

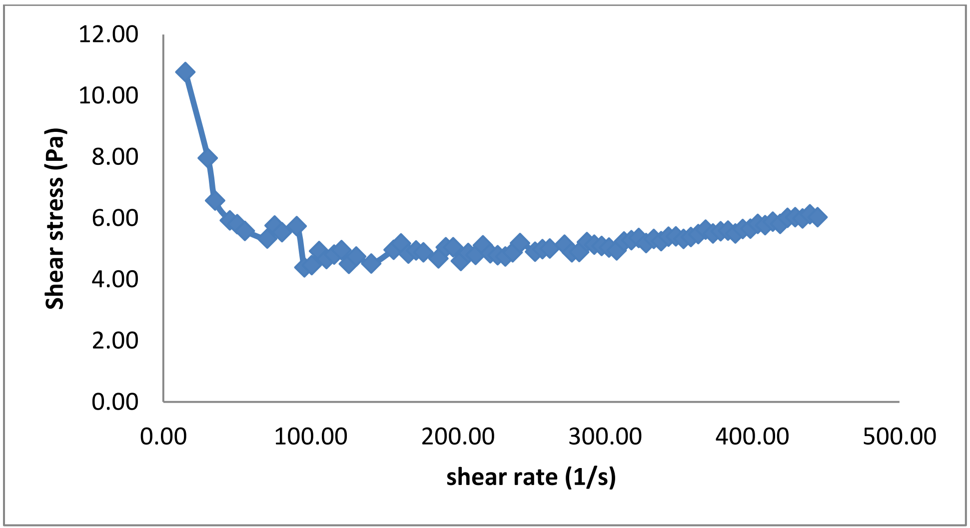


Figure 9.21. Shear stress versus shear rate in the range of (0-500 1/s) for Thickened excess activated sludge (TS= 3.1 %)

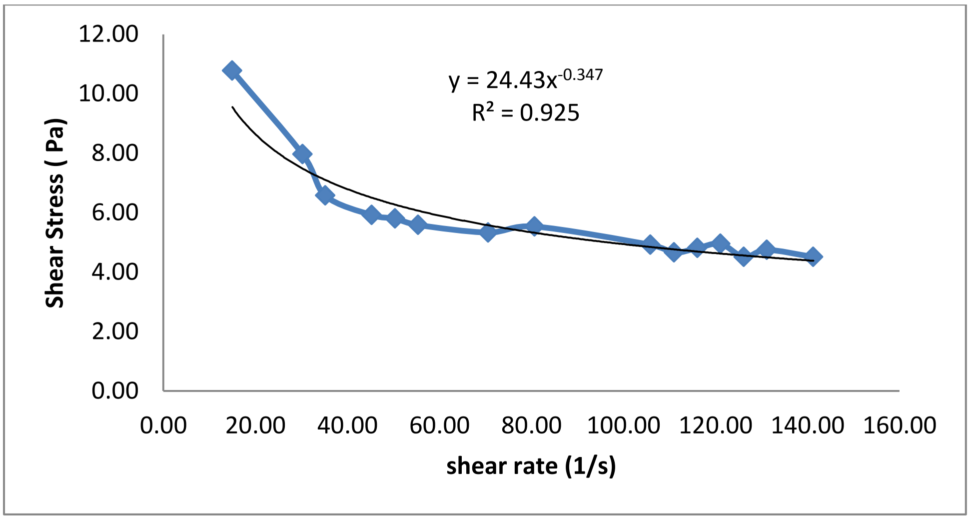


Figure 9.22. Shear stress versus shear rate curve first part (0-160 1/s) for thickened excess activated sludge (TS= 3.1%)

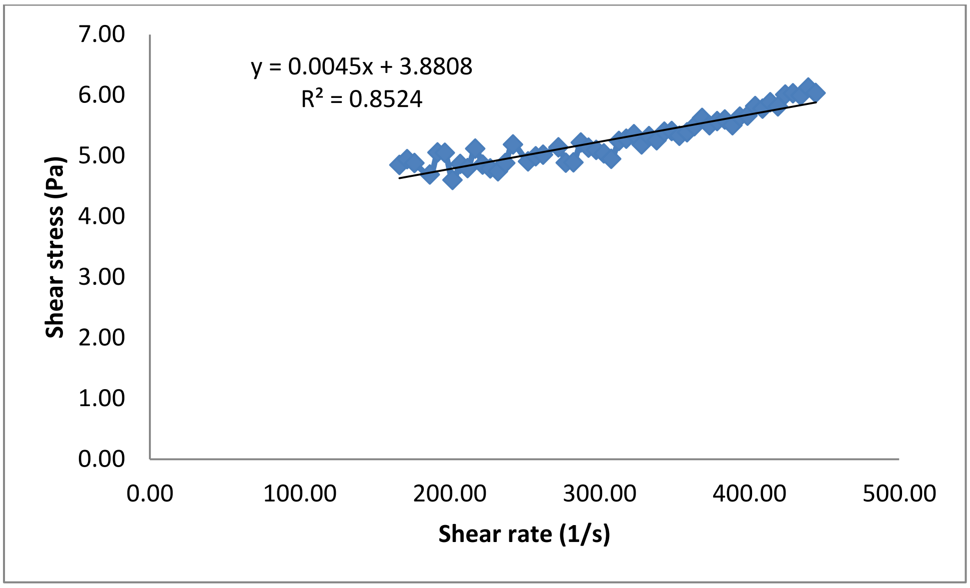


Figure 9.23. Shear stress versus shear rate curve second part (160- 500 1/s) for thickened excess activated sludge (TS = 3.1 %)

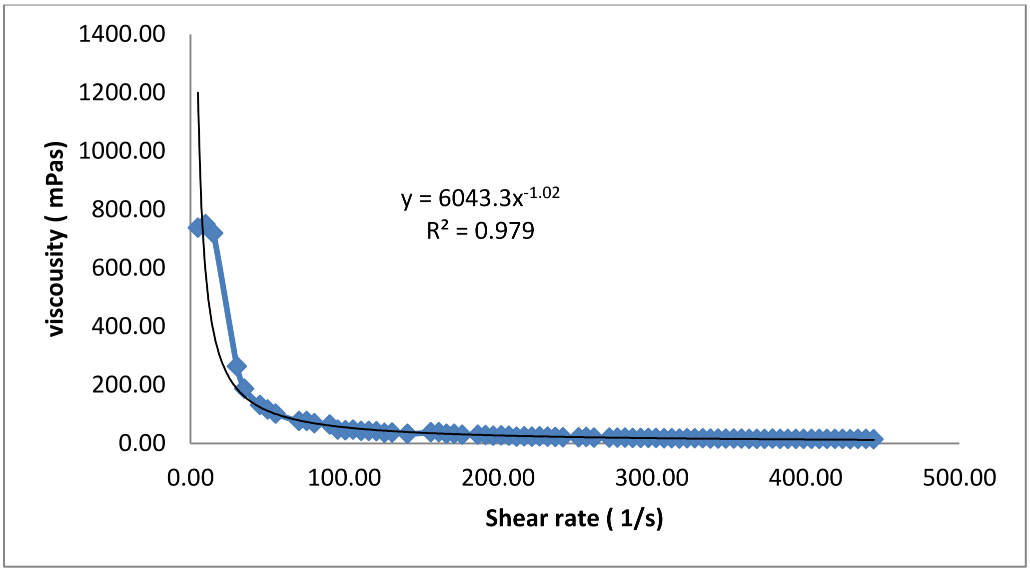


Figure 9.24. Viscosity versus shear rate in the range of (0-500 1/s) for Thickened excess activated sludge (TS= 3.1 %)

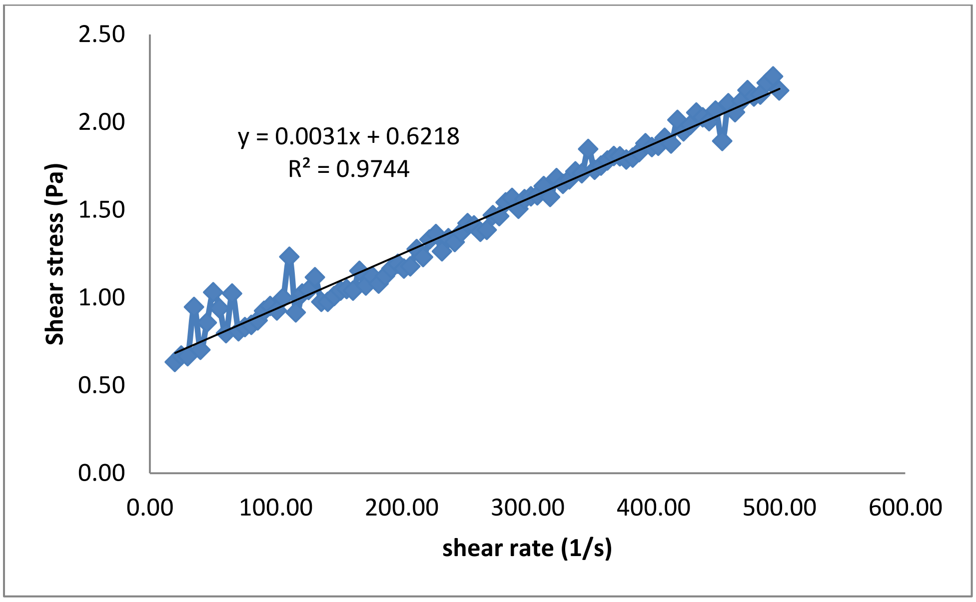


Figure 9.25. Shear stress versus shear rate curve for Thickened excess activated sludge (TS= 2.3 %)

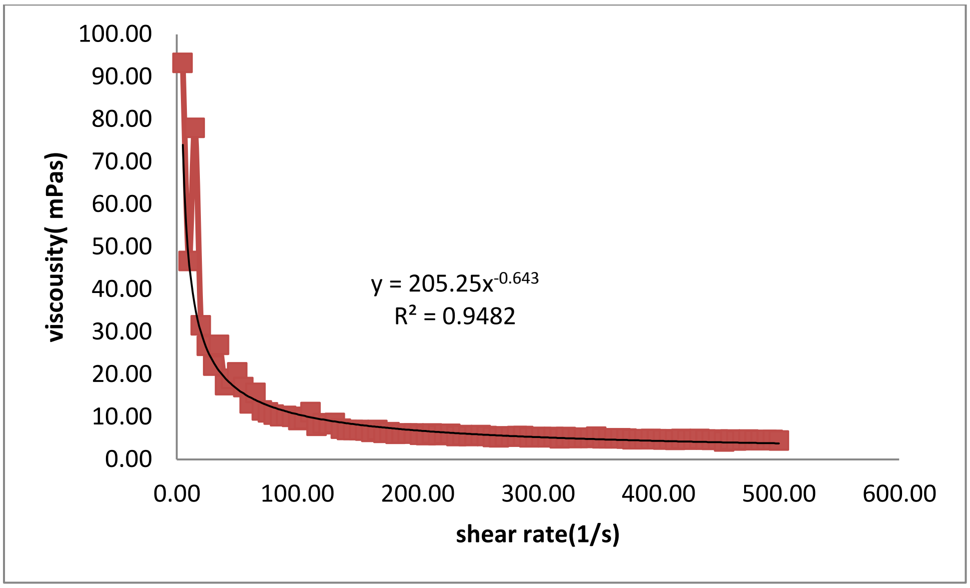


Figure 9.26. Viscosity versus shear rate curve for Thickened excess activated sludge (TS= 2.3 %)

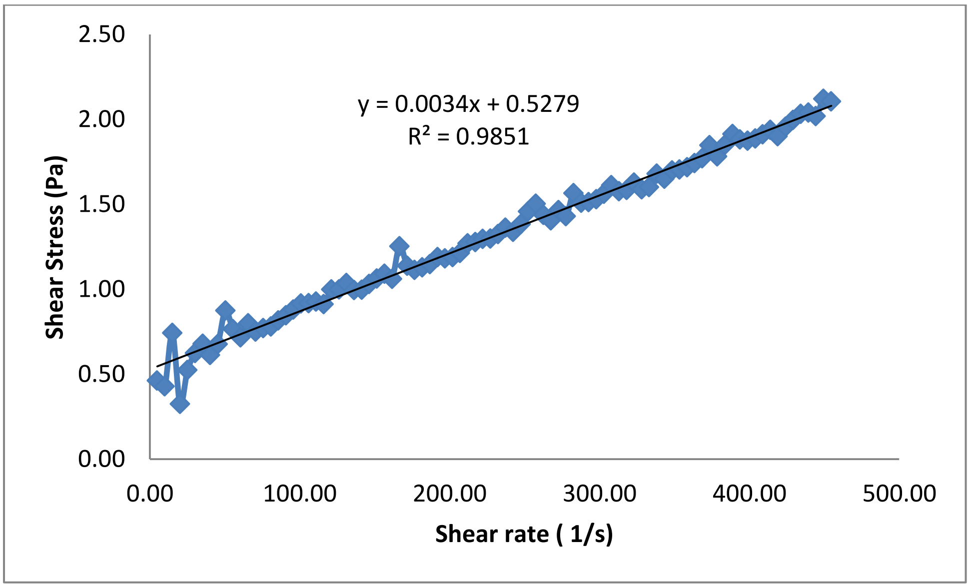


Figure 9.27. Shear stress versus shear rate for thickened excess activated sludge. (TS = 1.6 %)

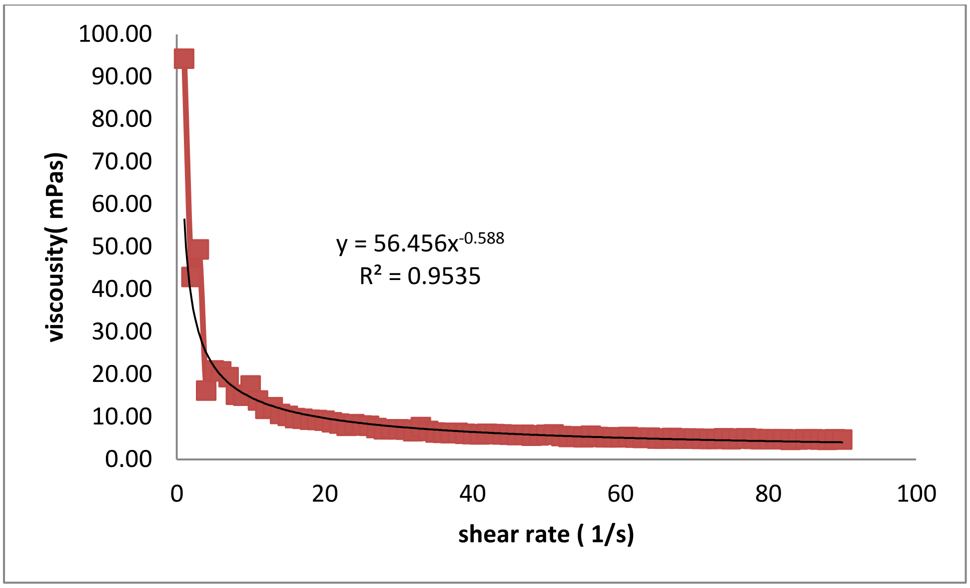


Figure 9.28. Viscosity versus shear rate for thickened excess activated sludge. (TS = 1.6 %)

9.6 Conclusion

The study on the effects of mixing ratio, organic loading rate, and hydraulic retention time shows that higher methane production was achieved for the mixture with greater percentage of primary sludge. The solid removal and COD reduction was significant for the sludge with larger percentage of EAS, the dewaterability was found to be better for the sludge with greater concentration of excess activated sludge. Organic loading rate for shorter HRT produced higher amount of methane for combined microwave-ultrasonic pretreated thickened excess activated sludge compared to the untreated TEAS sludge. This confirms the availability of more readily available organics for microbial attach which helps to maintain higher biogas production and higher reduction of COD and VS at a lower HRT with high organic loading rate. The rheograms for primary, thickened excess activated, mixed and digested sludge samples subjected to different pretreatment conditions were found to be very different from each other. Increasing ultrasonication time improved sludge rheology. Total solid concentration had significant impact on the viscosity and shear stress of thickened excess activated sludge.

CHAPTER 10

OPTIMIZATION OF ANAEROBIC DIGESTER INPUT AND OUTPUT PARAMETERS AND PREDICTIONS USING ADAPTIVE FUZZY LOGIC INFERENCE SYSTEM (ANFIS): A CASE STUDY

Abstract

Anaerobic digestion system converts organic matter to intermediate products like organic acids before methanogenesis. pH and alkalinity are frequently monitored parameters in the influent, process and effluent streams to control the conversion of the organic acids to methane gas. COD, VS and VFA content can also directly affect the performance of the digester and methane yield. Hence, understanding the relationship between such input variables and their effect on methane yield and effluent COD, VS and pH helps to determine the optimum operating conditions. In this chapter, one year operational data collected from Beenyup wastewater treatment plant of Water Corporation was utilized for model based predictions on Adaptive neuro-fuzzy application in MATLAB. All the key parameters affecting digester performance were used for training and testing the ANFIS model after normalization. The results obtained from back propagation and hybrid algorithms by fitting training data to the neural network helped to arrive at sound predictive approximations. The type and number of input membership functions in the ANFIS model were selected by minimizing the errors. mean square error (MSE), root mean square normalized error (RMSE) and mean absolute percentage error (MAPE) were used to compare model training dataset with the FIS generated output. The models were validated using model checking dataset.

The optimum methane potential, sludge feed flow rate (organic loading rate), pH and alkalinity were determined and the parameters that affect digester performance most were selected and optimized, the surface responses for the correlation between input and output variables were also developed.

10.1 Introduction

Adaptive Neural Fuzzy Inference Systems (ANFIS) the artificial intelligence techniques is widely preferred for modelling input and output parameters of anaerobic digester. It interprets the values in the input vector and assigns values to the output by means of some sets of fuzzy IF- then rules (Tay and Zhang 1999). Tay and Zhang applied the ANFIS in wastewater treatment and anaerobic digestion processes to predict effluent quality. Adaptive neuro-fuzzy inference system (ANFIS) involves first-order sugeno-fuzzy model which is based on back propagation or hybrid learning algorithm where the adaptive capabilities of neural network are integrated to the fuzzy logic qualitative nature (Piero et al. 2002 and Yazdi et al. 2010). ANFIS was applied in various areas with reasonably good prediction and approximation of nonlinear relationship among multiple inputs and outputs. The ANFIS approach was used to predict the off-line effluent parameters from important on-line input variables which are not available for the essential parameters in biological processes. Knowledge-based fuzzy inference systems (FIS) are more frequently adopted to describe biological behaviour despite the very complex and time consuming structure development which requires adoption of new rules that accommodate the complexity. ANFIS models are simpler to construct compared to FIS as the rules are adopted based on the available database which exists widely for anaerobic treatment systems. The model can be trained with new data or seasonal changes providing flexibility to the user to adapt or update the model continuously. It is based on non-linear functional dependency between input and output variables. In this research work, the purpose of this ANFIS application is to develop set of rules that relate inputs like (pH, alkalinity, TS, VS, Sludge feed flow rate and VFA) to outputs like biogas production and methane yield for actual industrial scale data shown in Figure 10.3. In this chapter, the operational data from the plant was normalized for the model training and testing before running the simulation for prediction. The model output was compared against the actual training data and the error was minimized to obtain the optimum operational points.

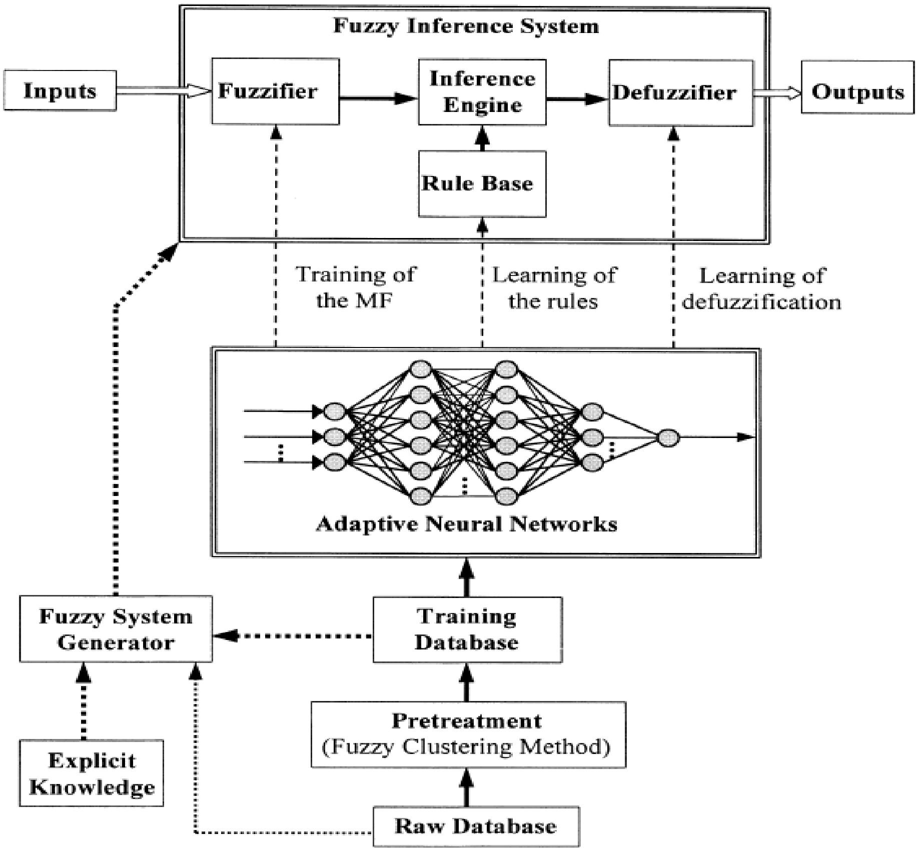


Figure 10.1 Architecture of conceptual adaptive neural fuzzy model of anaerobic wastewater treatment system (Tay and Zhang, 2000)

10.1.1 Model architecture and model components

The schematic architecture of the conceptual neural fuzzy model is depicted in Figure 10.1. It consists of the key components: inputs and outputs database and preprocessor, a fuzzy system generator, a fuzzy inference system, and an adaptive neural network representing the fuzzy system. The fuzzy inference system and its associated adaptive network are a Sugeno fuzzy inference system (Sugeno and Kang, 1986) and an adaptive network-based fuzzy inference system (ANFIS) (Jang, 1993). The input and output parameters are selected or generated from the parameters commonly used for system description. Generally, it is developed by collecting regularly monitored parameters.

For the liquid phase these parameters include pH, volatile fatty acids (VFA), alkalinity, organic loading rate (sludge feeding rate), VS reduction. In the gas phase, the parameters include biogas and methane production rates (Hickey et al.1991).

The quality of the training database is critical for the model to produce correct information about the system. In order for the model to describe the system accurately, the data-base should contain adequate and correct information on the system. On the other hand, it is common for a raw database to contain some redundant data. Thus, sometimes it is necessary for the raw training database to be pretreated to remove redundancies in the data.

As anaerobic systems convert organic matter to organic acids as intermediate products, pH and alkalinity are frequently monitored parameters in the influent, process and effluent streams. Besides, COD, VS and VFA content can directly affect the performance of the digester. Hence, understanding the relationship between such input variables and relating this to the methane yield and effluent values of VS and TS helps to determine the optimum operating conditions and to understand the relationship among the key performance parameters. The architecture and conceptual frame work of the anfis model is illustrated against the FIS method in Figure 10.1 with each unit shown in boxes.

Input General Information Base Unit; involves the input variables affecting the considered event and all the information related to these variables. The “general database” term is used due to the possibility of having information in numerical and/or text formats (pH, Temperature, COD, BOD, SS in this study). The model arrangement and configuration given in Table10.2 was selected based on the application. The selections given in Table10.2 provide a basis for the development of rule bases.

The Fuzzy Maker; is a processor assigning numerical input values to membership grades in fuzzy sets characterized with text (Common membership functions are triangular, bell curved, trapezoidal and Gaussian functions which are discussed in the next Section 10.2.1. The Gaussian function was selected as the main membership function for this study and some comparisons were made to other membership functions where ever necessary.

Fuzzy Rule Base Unit; contains all of the rules writeable in logical IF – THEN expression connecting input variables to output variables in the database. In writing these rules, all possible intermediate (fuzzy set) connections between inputs and outputs are taken into consideration. The fuzzy system can be applied in two ways each having different rules. The rule base was formed after assignment of the memberships.

Fuzzy Inference Motor Unit; is a mechanism covering the group of processes providing the single output behaviour of the system by gathering the separate relations built between the input and output fuzzy sets in the fuzzy rule base. This motor is used to determine what kind of an output will be obtained as a result of the input of the whole system by collecting all the rule inferences together.

Defuzzifier; transforms the fuzzy inference solutions obtained as a result of fuzzy processes into definite numerical output values. The results of the rules were combined and defuzzified via centroid method.

The Output Unit; expresses the group of the output values obtained at the end of the interaction performed between information and fuzzy rule bases by the help of the fuzzy inference motor.

In this chapter, one year historical data was classified systematically and all the data points were normalized. The normalized data was used for training and testing of the ANFIS model. Based on the training the set of rules (equations) important to draw the relationship between the input and output variables were determined. The membership functions (Gaussian type) that provide the best training data with minimum error were selected. And the surface responses were thoroughly investigated.

10.2 Materials and methods

10.2.1 Model equations and modelling tools used for analysis of anaerobic digester

Modelling is an essential tool in both design and operation of biological treatment plants. It can also be used for optimization purposes (Turkadogan et al. 2010). In this specific chapter, operational data for the year 2011-2012 was collected from

BWWTP for all the parameters as shown in Table10.2. Sludge feed flow rate and biogas production data collected from daily measurement was organized with the weekly data for the rest of the parameters. Such operational data can be used for training purpose in the adaptive neuro-fuzzy applications to arrive at sound predictions. Identification of parameters that could be used for monitoring anaerobic treatment system is an important factor for efficient operation of the anaerobic digesters. The experimental studies presented in chapters 4-9 were used to select the most essential parameters influencing biogas production and digester performance. In this chapter, the selected parameters were evaluated using the ANFIS model.

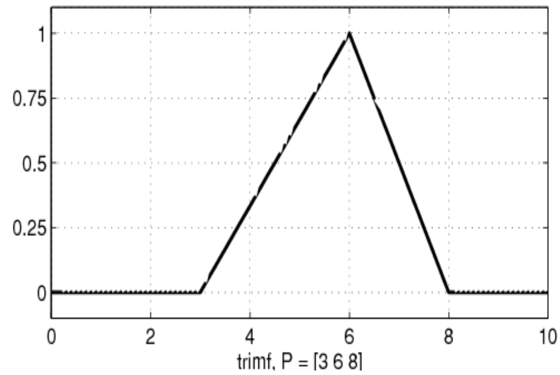
ANFIS uses a hybrid learning algorithm to identify the membership function parameters of single-output, Sugeno type fuzzy inference systems (FIS). A combination of least-squares and back propagation gradient descent methods are used for training FIS membership functions to model a given set of input/output data.

A general fuzzy system has four basic components as shown in section 10.1.1. The steps include fuzzification, fuzz rule base, fuzzy output engine and defuzzification (Akkurt et al. 2004). In the fuzzification step, the input and outputs are converted into one or more of the membership functions. Fuzzy inference engine transforms the inputs into the corresponding outputs, mostly minimization and product operator (prod) are employed in this step. The prod technique is selected because of its performance.

There are several membership functions used for the development of the adaptive neuro-fuzzy logic inference system. These include triangular, trapezoidal, generalized bell type, gaussian type and s-shaped functions. In this particular study, Gaussian type membership function was mostly found to provide minimum error and better represents the data set as compared to the other membership functions shown in equations 10.1 – 10.5 (Perendeci et al., 2007).

Triangular membership function depends on three scalar parameters a,b and c and it is given by equation 10.1 and the geometry of the plot is shown in Figure 10.2A

$$f(x,a,b,c) = \max\left(min\left(\frac{x-a}{b-a}, \frac{c-x}{c-b}\right), o\right)$$

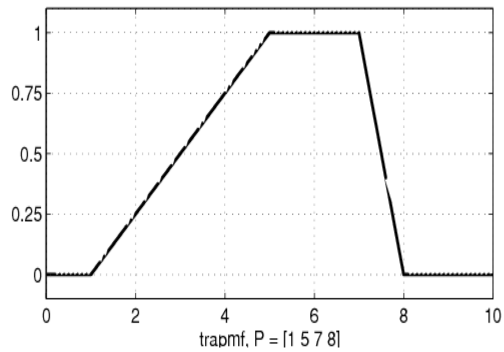


(10.1) Figure 10.2A

In equation (10.1) the parameters a and c locate the "feet" of the triangle and the parameter b locates the peak.

Trapezoidal membership functions involve the parameters *a* and *d* which show the "feet" of the trapezoid and the parameters *b* and *c* that show the "shoulders." of the trapezoid as shown in Figure 10,2B and the functions are represented by equation

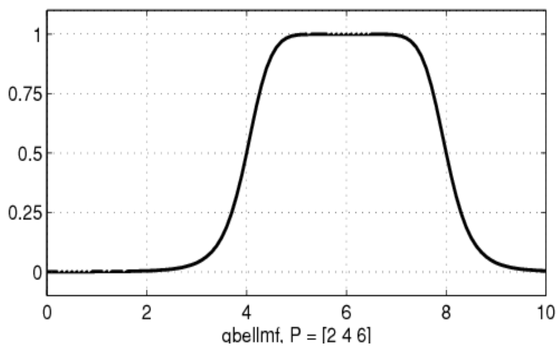
$$f(x,a,b,c,d) = \begin{cases} 0, & x \le a \\ \frac{x-a}{b-a}, & a \le x \le b \\ 1, & b \le x \le c \\ \frac{d-x}{d-c}, & c \le x \le d \\ 0, & d \le x \end{cases}$$



(10.2) Figure 10.2B

The generalized bell function depends on three parameters a, b, and c as given by equation (10.3) and Figure 10.2C the parameter b has a positive value. The parameter c locates the centre of the curve.

$$f(x,a,b,c) = \frac{1}{1 + \left|\frac{x-c}{a}\right| 2b}$$



(10.3) Figure 10.2C

The sigmoid curve plotted for the vector x depends on two parameters *a* and *c* as given by equation (10.4) and Figure10.2D. It is simply the product of two such curves plotted for the values of the vector x. $f_1(x; a_1, c_1) \times f_2(x; a_2, c_2)$. The parameters are listed in the order $[a_1\ c_1\ a_2\ c_2]$.

$$f(x, a, c) = \frac{1}{1 + e^{-a(x-c)}}$$

(10.4)

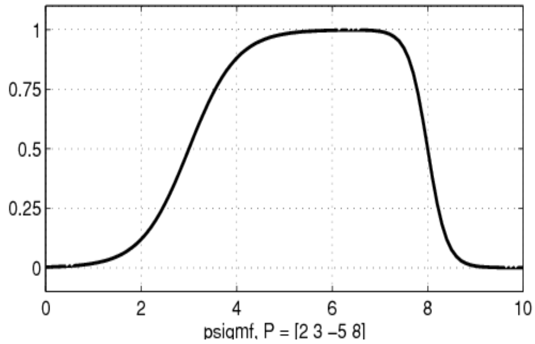


Figure 10.2D

In case of gaussian combination membership function the symmetric Gaussian function depends on two parameters σ and *c* as shown in equation (10.5) and Figure 10.2E

$$f(x, a, c) = e^{\frac{-(x-c)^2}{2\sigma^2}t}$$

(10.5)

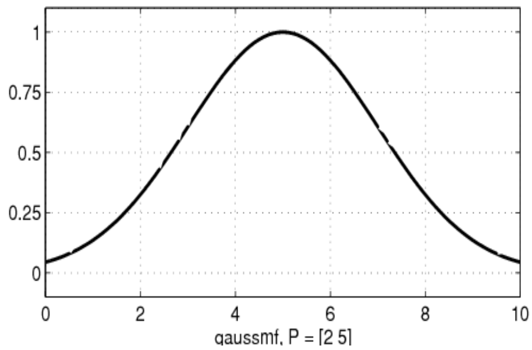


Figure 10.2E

10.2.2 Statistical evaluation

The prediction capacity of ANFIS model has to be evaluated and tested. There are several performance indicators to validate and test the model. Mean square error (MSE), root mean square normalized error (RMSE) and correlation coefficients are commonly used as performance indicators to evaluate the prediction capability of ANFIS trained by each data set. The MSE performance index was defined as

$$\text{MSE} = \frac{1}{n}\sum_{i=1}^{n}(y^* - y)^2 \qquad (10.6)$$

The RMSE performance index was defined as

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^{n}(y^{*}-y)^{2}}{n}} \tag{10.7}$$

where y is the measured values, y* the corresponding predicted values and n is the number of samples. The RMSE is used to calculate the errors during the prediction tests.

The architecture of the ANFIS model used in this study for the prediction are discussed in Section 10.3.1- 10.3.11. The types and numbers of MFs in ANFIS including Gaussian, generalized bell-shaped, triangular and trapezoidal shaped functions, and the parameters were tested to determine an appropriate ANFIS model. The selection criteria of the best final architecture were based on the values of RMSE and R between the model output values and observed values. Back propagation or hybrid learning algorithms were implemented and the final architectures of the ANFIS models were determined for each case after many trials. Most ANFIS models used generalized gaussian MFs for each input variable as these membership functions provided optimum results. The models were used to predict the biogas yield, VFA generated, effluent TS and VS and the corresponding percentage reductions in TS and VS based on input parameters.

The validity of the model training data was checked by using testing data set. The extent to which the training data fits to the FIS generated data was tested using the ANFIS editor GUI using this testing dataset. Besides, model over-fitting was controlled by using checking data set. The FIS models were selected to have parameters associated with the minimum checking data model error.

10.3 Results and discussion

This particular section presents the results obtained from the modelling and simulation tests conducted using the ANFIS tool in MATLAB. The training of the FIS was performed and the output was generated for different number and types of input and output parameters. The prediction errors were minimized to make the ANFIS output as representative as possible. The relationships between different input and output parameters were determined. The trends and optimum working conditions were identified for the ranges considered in the analysis.

The model predictions were based on normalized data set for one year operational data from BWWTP. The range of data, maximum and minimum values of each variable, used in the study is presented in Table10.2. The normalized data set according to equation (10.3) is shown in Figure 10.2.

Table 10.2 Minimum and maximum values of both input and output parameters used for normalization.

	alkalinity (mg/l)	**PVS (%)**	**PTS (%)**	**pH**	**ETS (%)**	**EVS (%)**	**VFA ppm**	**BG (m3/day)**	**SF (m3/day)**
minimum	**1816.67**	**83**	**2.3**	**7.03**	**1.08**	**73.00**	**4.50**	**13424.15**	**961.38**
Maximum	**2787.50**	**9.1**	**4.3**	**7.30**	**1.63**	**79.00**	**78.25**	**32728.91**	**1608.32**

(**PVS**: feed volatile solid content, **PTS**: feed total solid content, **ETS**: effluent total solid content, **EVS**: effluent volatile solid content, **VFA**: volatile fatty acid, **BG**: biogas production, **SF**: sludge feed flow rate).

All the operational and characteristics data were normalized after determining the maximum and minimum values for each parameter to avoid discrepancies during the model training and prediction. The normalization was carried out using Equation 10.3.

$$\text{Normalized value} = \frac{(\text{actual value} - \text{minimum value})}{(\text{maximum value} - \text{minimum value})} \qquad (10.3)$$

In order to generate an effective estimation model that can provide accurate predictions of the output parameters, a pre-processing may be helpful in input data selection as well as engineering judgment (Erdirencelebi et al. 2011). Despite some inconsistencies, the patterns of predicted and measured values were parallel. Considering the fluctuating characteristics of the influent sludge to the digesters, the prediction performances for each parameter were evaluated separately and for different combinations.

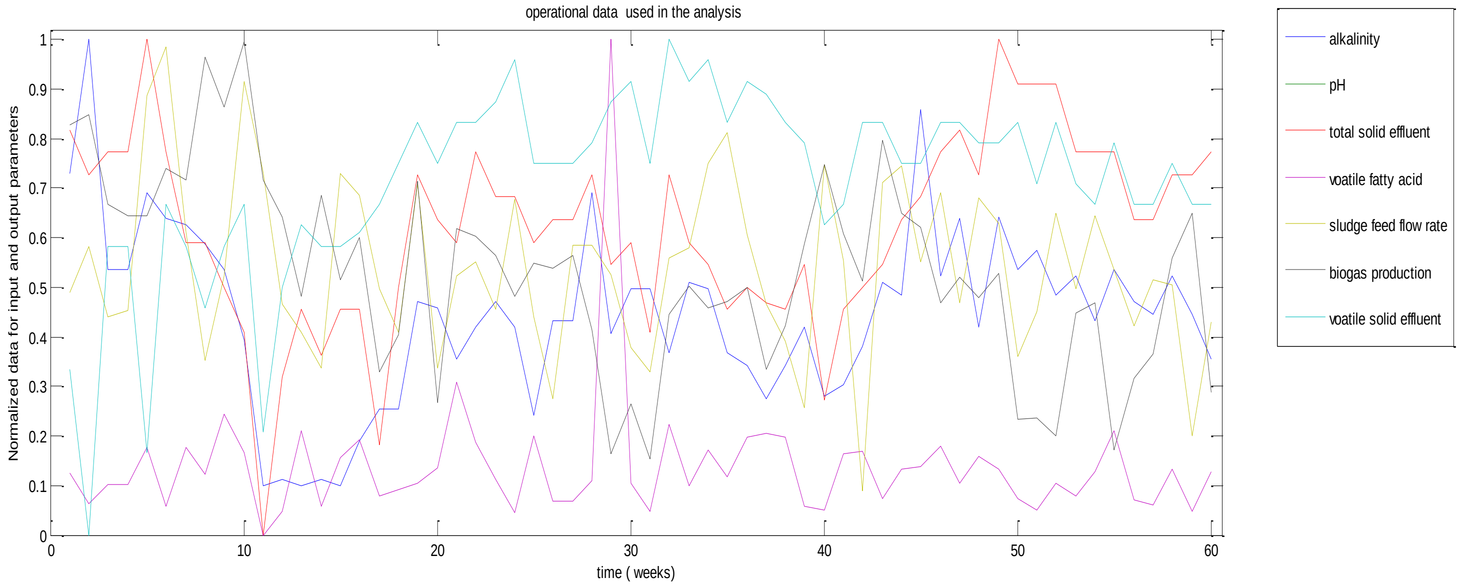


Figure 10.2 Normalized input and output operational data used in the modelling study.

The statistical distribution of the normalized input and output parameters from the operational data of BWWTP is given in Table 10. 3. These parameters were divided into input and output parameters systematically to predict the influence of one parameter on the other one. The normalized results for the effect of sludge feed flow rate on biogas production, effect of feed total and volatile and effluent total and volatile solid concentration and effect percentage reduction in volatile solids on biogas production are shown in Figures 10.3-10.8.

Table 10.3 Statistical distribution of the normalized operational data used in the building of the ANFIS model.

Characteristic parameter	**Min**	**Max**	**Mean**	**Mode**	**Standard deviation**	**Median**
Alkalinity	0.098	1	0.44	0.53	0.18	0.45
pH	0	1	0.41	0.27	0.22	1
Feed volatile solids	0	1	0.4	0.4	0.19	0.4
Effluent Volatile solid	0	1	0.71	0.83	0.19	0.83
Feed total Solid	0	1	0.4	0.4	0.19	0.4
Effluent Total solid	0	1	0.619	0.77	0.19	0.63
Volatile fatty acid	0	1	0.14	0.11	0.13	0.12
Sludge feed flow rate	0.089	0.98	0.54	0.09	0.17	0.53
Biogas production	0.15	0.99	0.52	0.15	0.19	0.52
Methane percentage	0	1	0.63	0.69	0.19	0.69

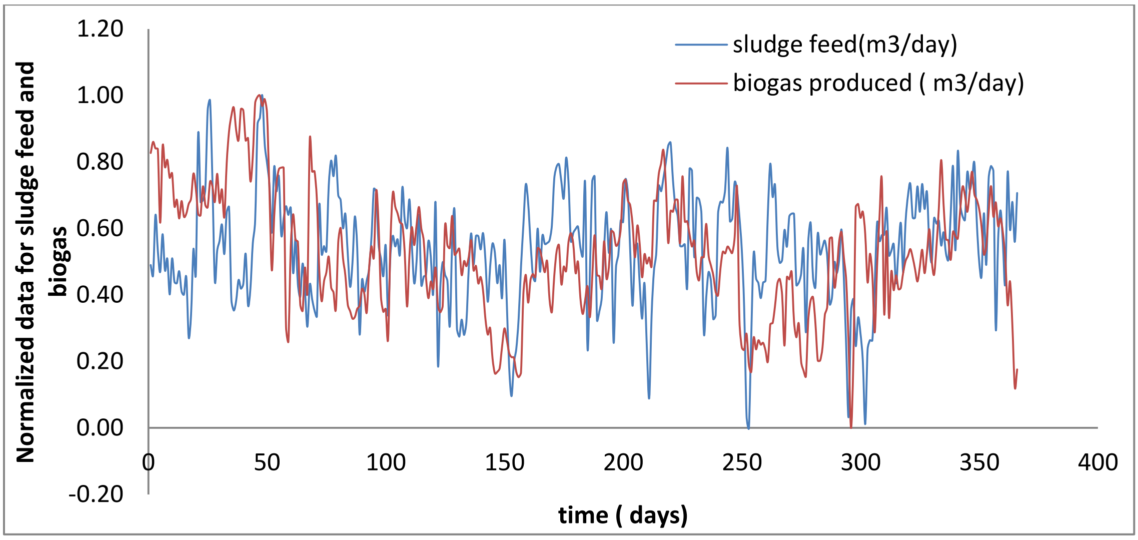


Figure 10.3 Normalized operational data sludge feed flow rate and biogas production

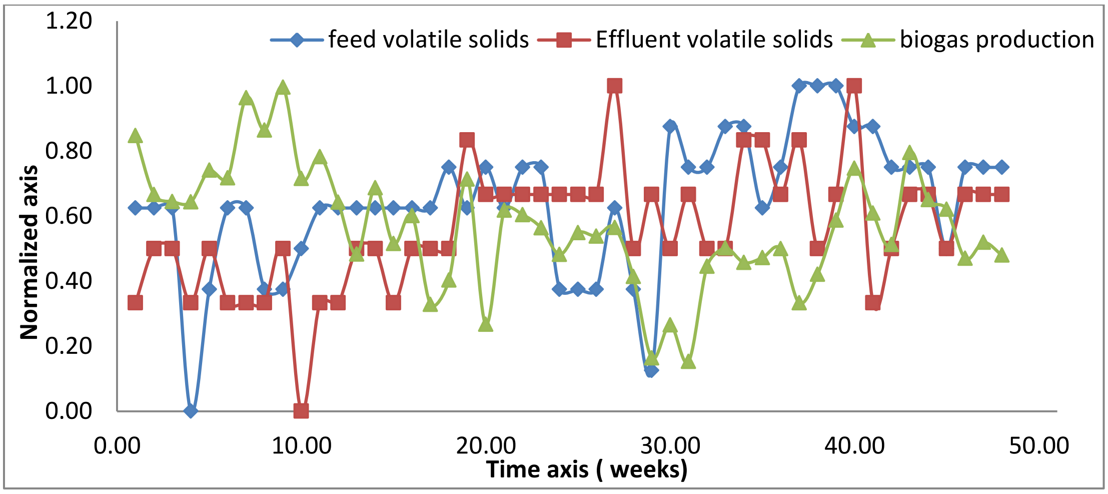


Figure 10.4 Normalized data for Feed and volatile solids with biogas production.

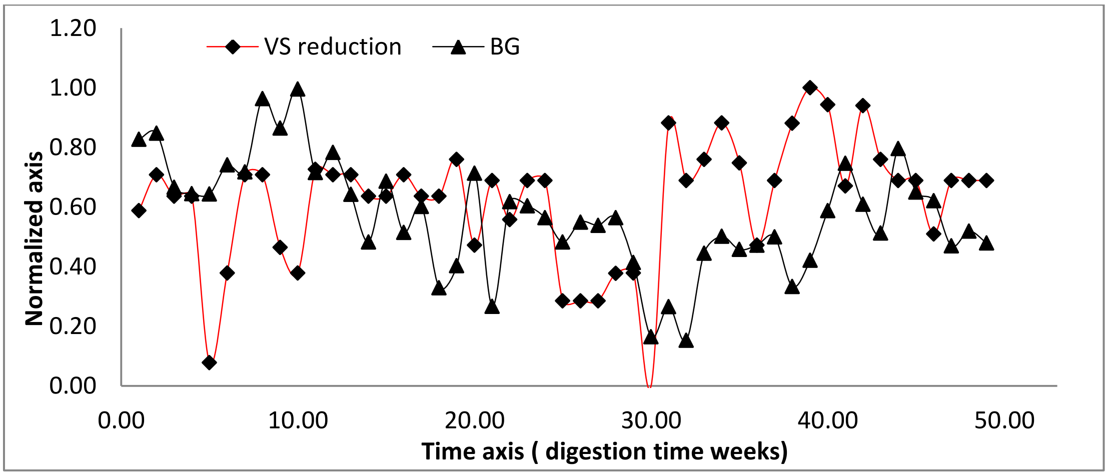


Figure 10.5 Normalized operational data percentage reduction on volatile solids versus biogas production.

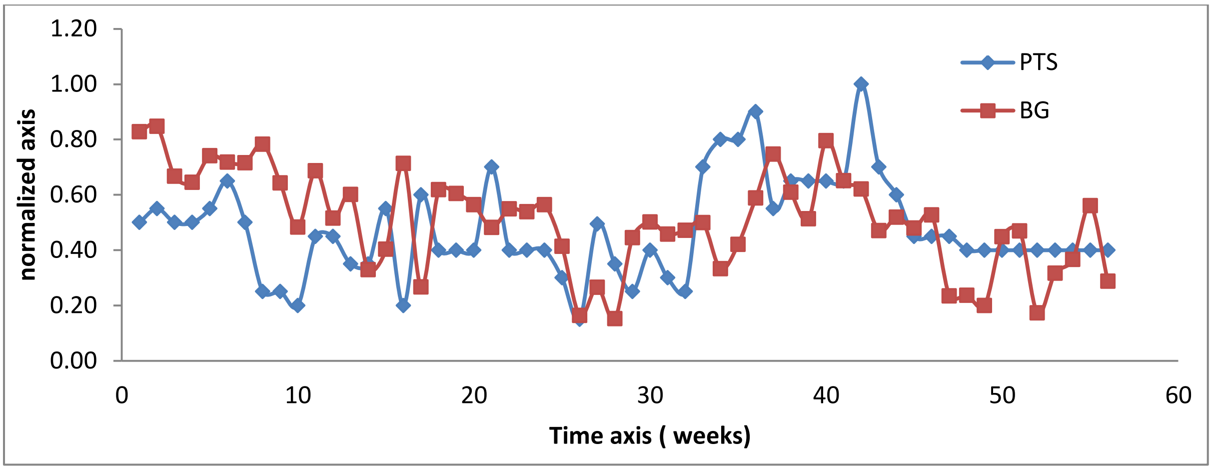


Figure 10.6 Normalized operational data feed total solid concentration and biogas production

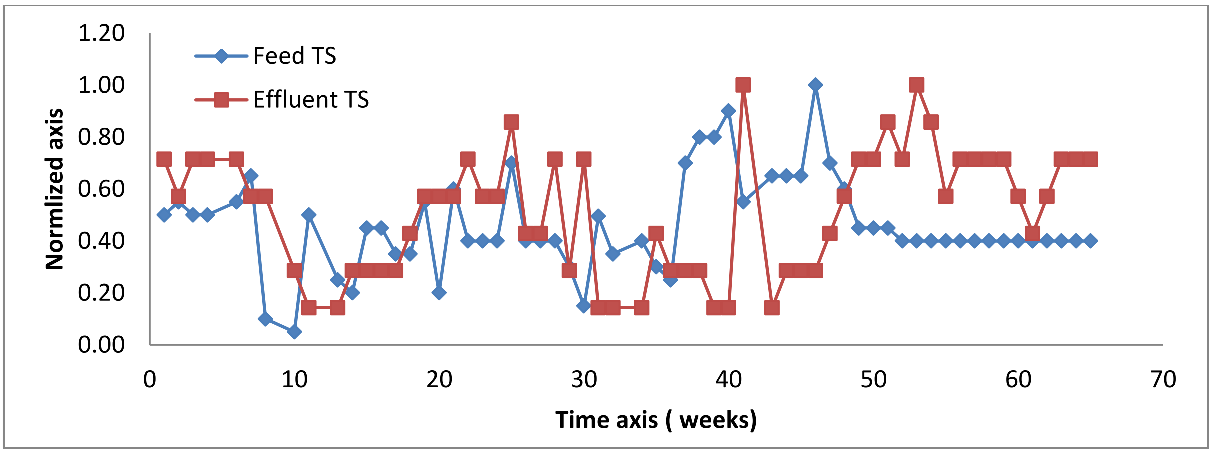


Figure 10.7 Normalized operational data for feed total solid and effluent total solid concentration.

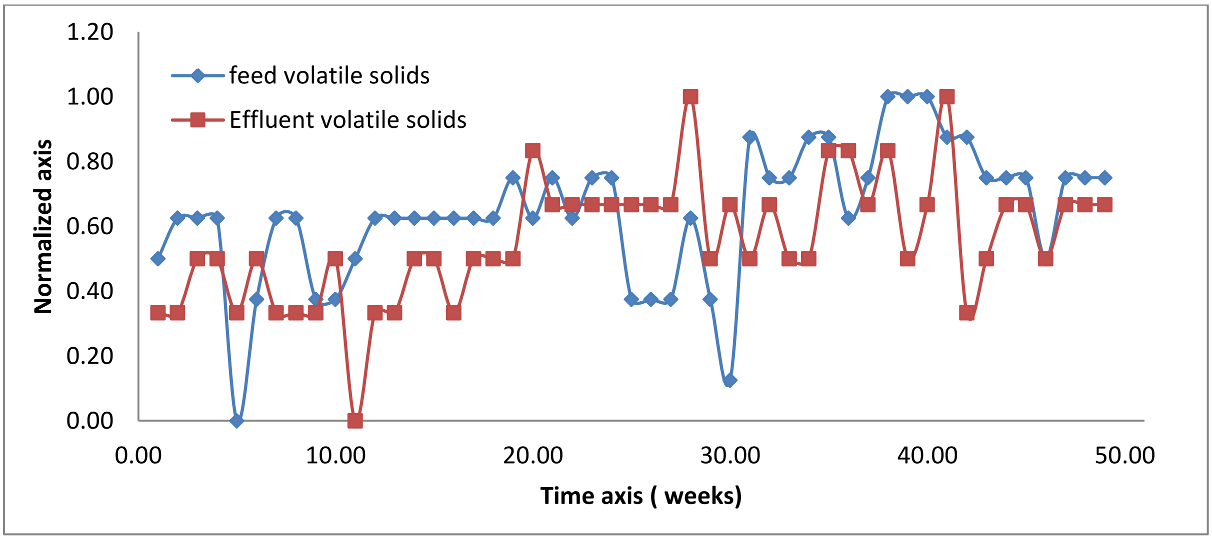


Figure 10.8 Normalized operational data for feed and effluent volatile solid concentration.

10.3.1 Prediction on effect of sludge feed flow rate on biogas production

Sludge feed flow rate was taken as input variable and biogas production was the output variable in this model prediction. Organic loading rate and sludge feed flow rate affect biogas production and overall performance of digester significantly**.** It is essential to determine the optimum sludge feed flow rate that maximizes biogas and methane production and performance of the anaerobic digestion system. The model structure has 20 nodes with 8 linear and 8 nonlinear parameters. Four Gaussian type membership functions were used to establish the fuzzy rules as shown in Figure.10.9. The number and type of member functions were selected after several trails on different membership functions for the ANFIS prediction. The number of model training data used for this specific prediction was 210 and the model validation and testing was performed using 153 data points. The FIS training was conducted using the hybrid algorithm. The minimum average testing error obtained when fitting the ANFIS model training data to the FIS generated output was 0.1517 as shown in Figure 10.10 and the model validation error was 0.2068 as shown 10.11. As shown in Figure 10.12 the ANFIS predictions and the model fitting show that an increase in sludge feed flow rate increases the biogas production until biogas production level of 0.6 (25000 m^3/day) and sludge feed flow rate of 0.33 (1155.46 m^3/day) Sludge feed from rate between 1155.46 m^3/day and 1466 m^3/day does not result in any significant change in biogas production. Sludge feed flow rate greater than 1466 m^3/day again results in a substantial increase in biogas production. Higher sludge feed flow is advantageous for the operation of the plant enabling higher biogas production at higher throughput.

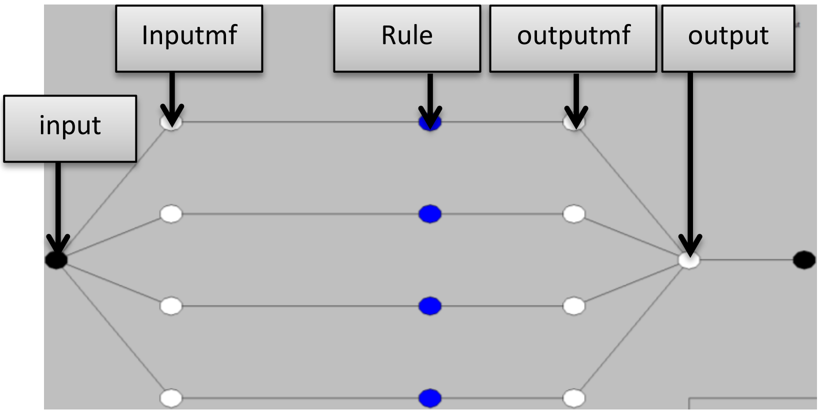


Figure 10.9 Model structure for the prediction of biogas production based on sludge feed flow rate

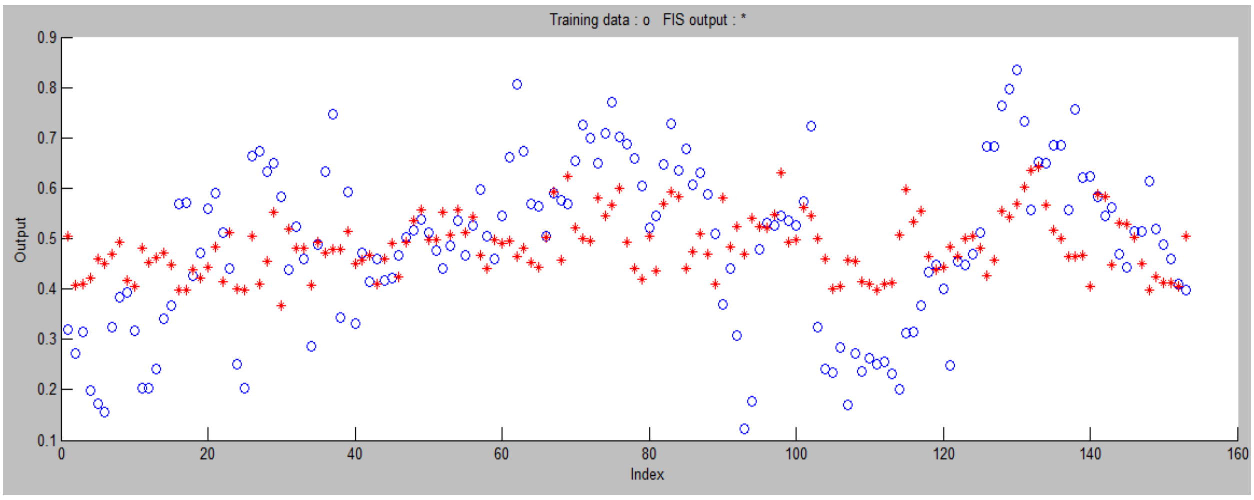


Figure 10.10 Plot of the training data (o) along with FIS generated output (*)

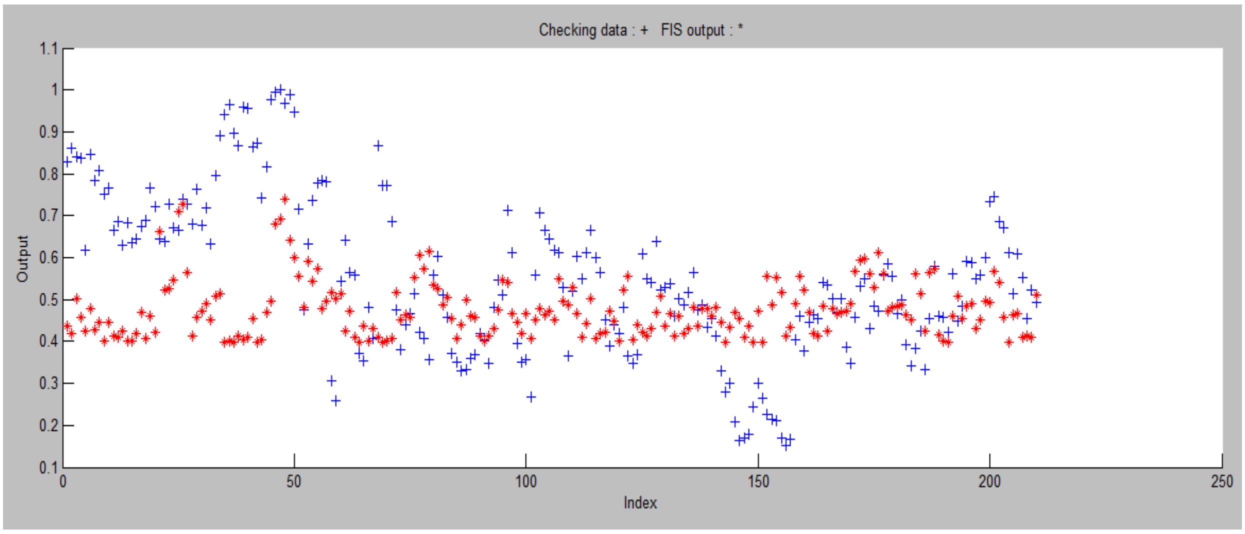


Figure 10.11 Time plot of the validation dataset to the FIS output.

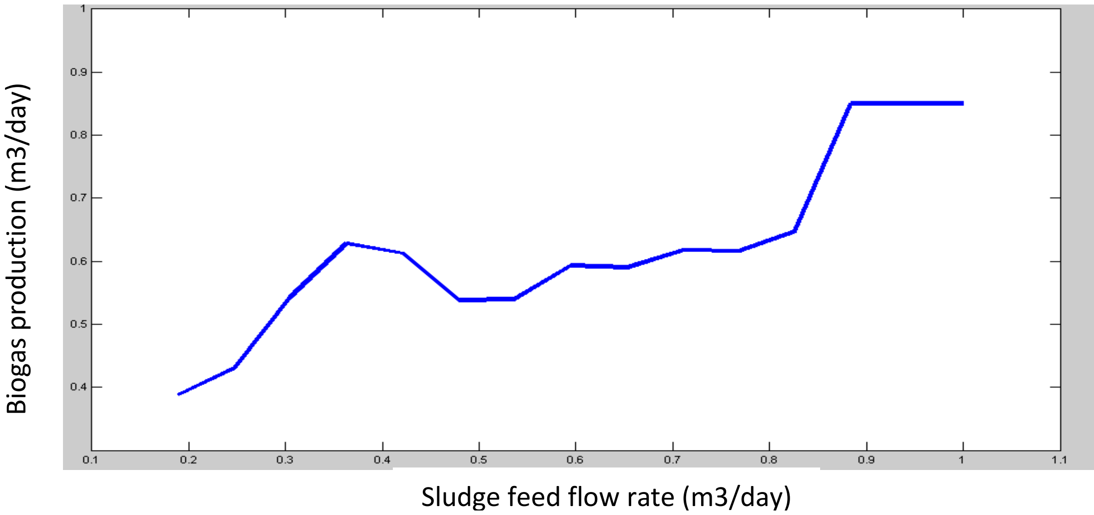


Figure 10.12 Predictions on biogas production as a function of sludge feed flow rate.

The actual sludge feed flow rate and biogas production were calculated from the normalized graph using equations 10.6 and 10.7

Actual SF=SF in graph*646.94 + 961.38 (10.6)

Actual BG=BG in graph*19304.76+ 13.424.15 (10.7)

The optimum sludge feed flow rates of 1597.97 m^3/day result in biogas production of 35,238.52 m^3/day.

10.3.2 Predictions on effect of feed volatile solid concentration on biogas production.

In this section, feed volatile solids concentration was analysed against biogas production. The organic content of the feed sludge is directly dependent on volatile solid concentration and biogas gas (methane gas). As biogas (methane) is the metabolic product of the methanogenic degradation of the organic feed, relating input volatile solid and volatile solid reduction to the biogas production directly indicates the performance of the digester. Feed volatile solid concentration and volatile solid reduction affect biogas production and overall performance of digester significantly**.** It is important to optimize feed volatile solid concentration and volatile solid reduction to maximize biogas and methane production. The model structure for the feed volatile solid concentration versus biogas production has 12 nodes with 4 linear and 4 nonlinear parameters. Two Gaussian type membership functions were used to establish the fuzzy rules as shown in Figure.10.13. The number of model training data points used for this specific prediction was 28 and the model validation and testing was performed using 17 data points. The FIS training was conducted using the hybrid algorithm. The minimum average testing error obtained when fitting the ANFIS model training data to the FIS generated output was 0.1521 as shown in Figure 10.10 and the model validation error was 0.1694 as shown in Figure 10.11. Another prediction on effect of Feed volatile solid concentration on effluent volatile solid concentration provided the input to output plot shown in Figure 10.17. The model structure has 3 Gaussian type membership functions and the error after training for 100 epochs was 0.16196. Feed volatile solid concentration from 0.1 (83.8%) to as high as 0.6 (87.8%) is related to higher effluent volatile solid concentration beyond which the effluent volatile solid concentration increased significantly. Maintaining the input around 0.6 (83.8 %) is observed to be reasonable as shown in Figure 10.17. The model prediction on biogas production as a function of volatile solids gives the maximum biogas production of 26.937.48 m3/day at sludge feed volatile solid concentration of 87%. Feed volatile solid concentration around 87% was found to be the optimum for the range considered in the study. On the other hand a similar ANFIS model was developed to study the effect of volatile solid reduction on methane production. Volatile solid reduction is directly proportional to biogas or methane production that maximizing the reduction in Volatile solid enhances solid removal and biogas production. The number of nodes was 16 with 3 fuzzy rules and 6 linear and non-linear parameters for this testing. The model training and validation (checking) errors were 0.1610 and 0.1965 respectively. The prediction of biogas production after the training and validation tests show that maximum biogas production of 0.995 (32632.39 m^3/day) was achieved at volatile solid reduction of 0.617 as shown in Figure 10.18 and 10.19. Volatile solid reduction between 0.4 and 0.8 resulted in biogas production of above 0.8 (28867.97 m^3/day) as shown in Figure 10.18. Biogas production was as low as 0.3 (19215.62 m^3/day) when volatile solid reduction was 0.0109 as shown in Figure 10.19b. Figure 10.19c shows that high volatile solid reduction of 0.985 resulted in a biogas production of 0.685 (26647.93 m^3/day). Based on grid partitioning and background propagation algorithm generated by ANFIS, the model training error was reduced to 0.1526**.**

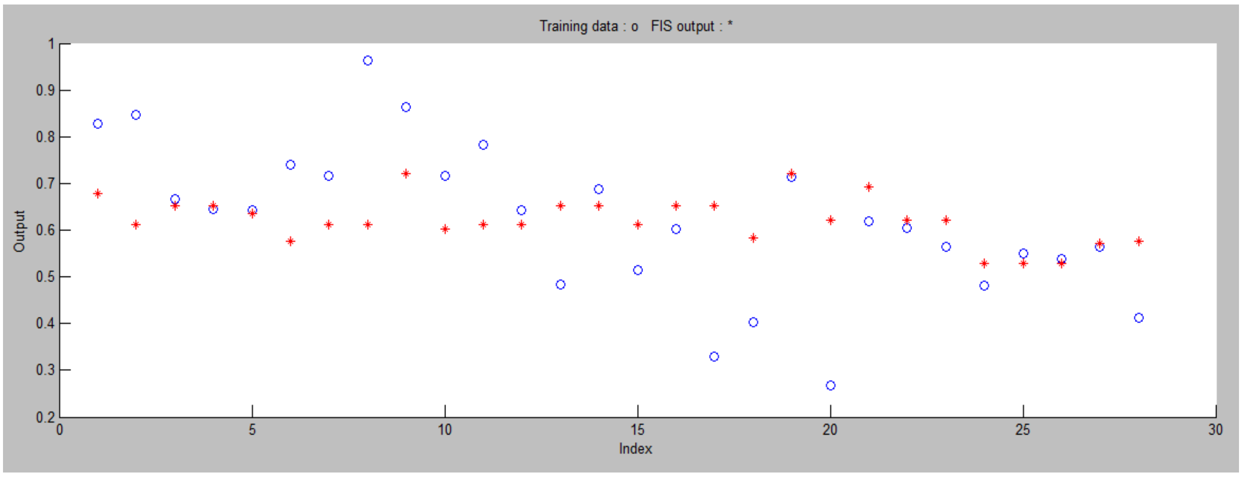


Figure 10.13 Model training for ANFIS based biogas prediction as a function of feed volatile solid concentration.

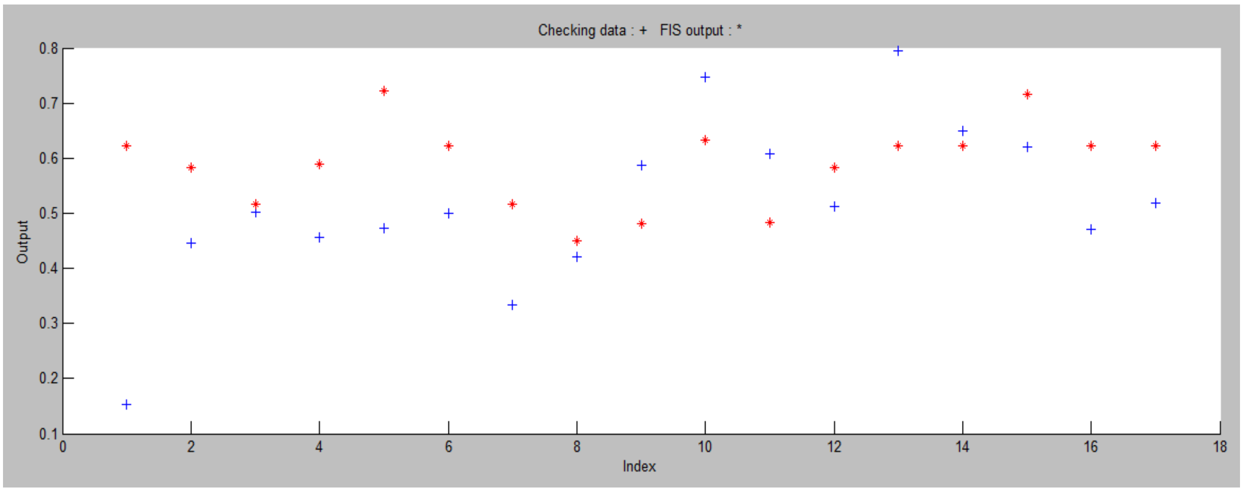


Figure 10.14 Model validation for ANFIS based biogas prediction as a function of feed volatile solid concentration.

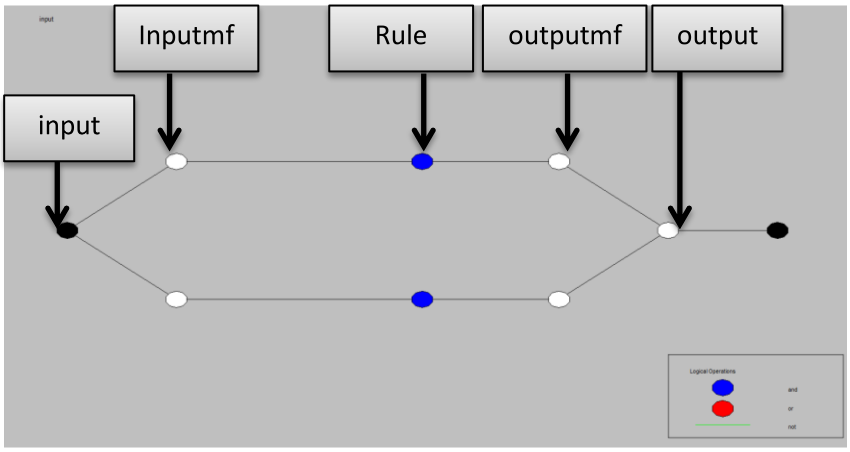


Figure 10.15 ANFIS structure of for the input, rule base and output variables.

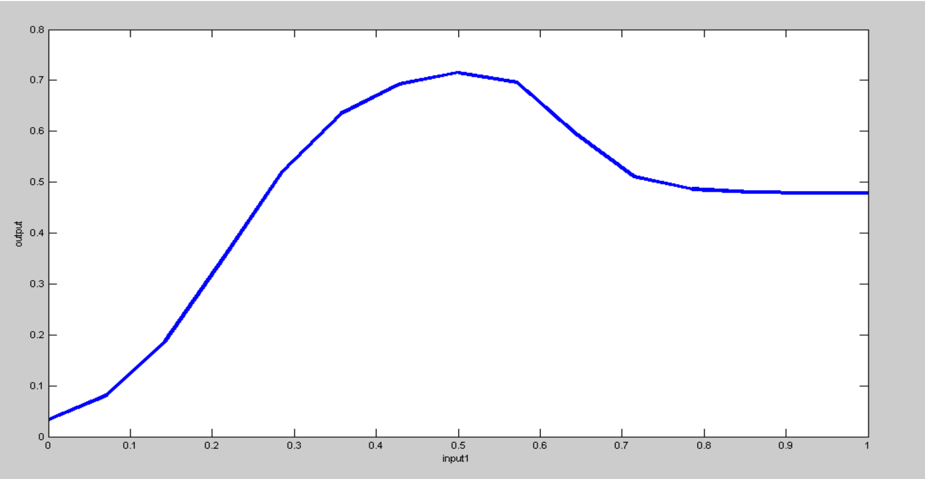


Figure 10.16 Prediction of biogas production as a function of feed volatile solid concentration.

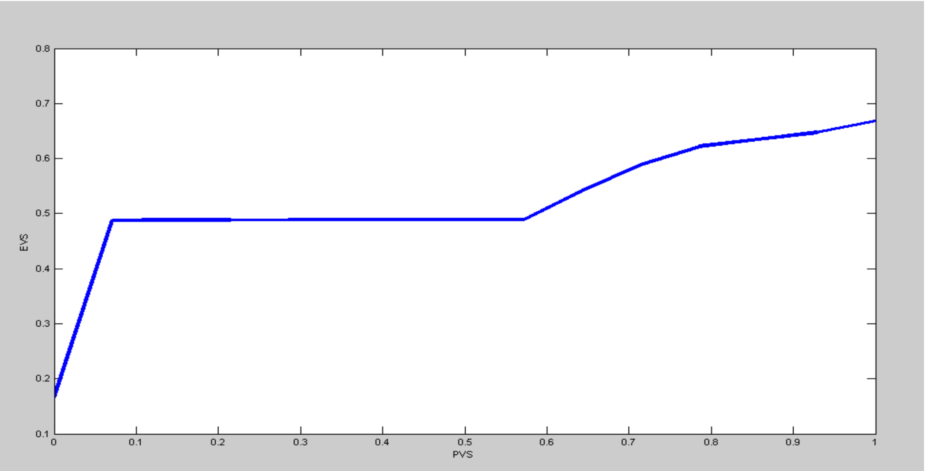


Figure 10.17 Feed volatile solid (PVS) as an input and Effluent volatile solid (EVS) as an output (effect of volatile solid content of the feed on the effluent volatile solid content)

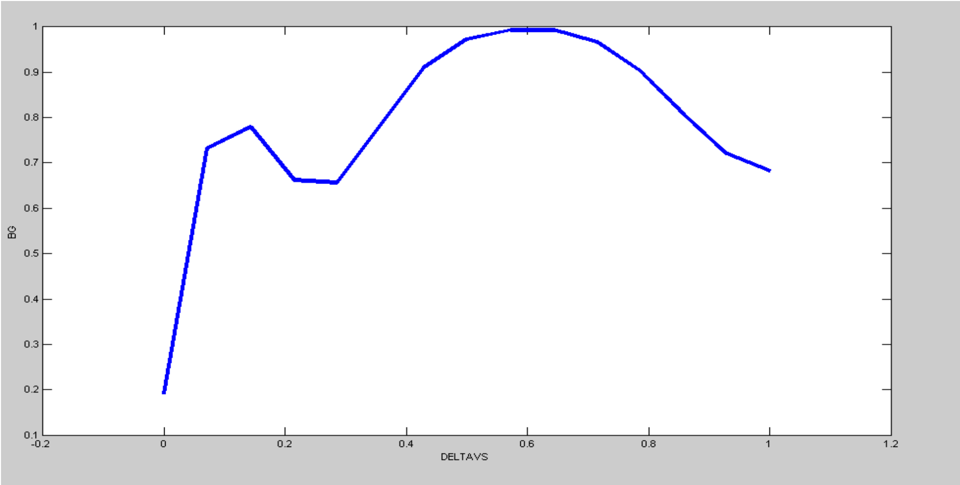
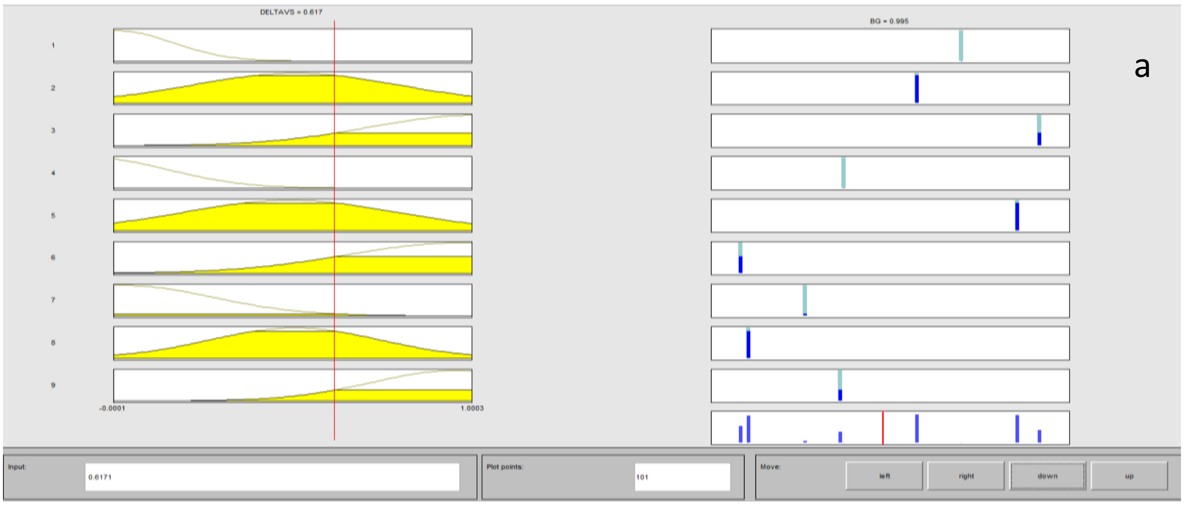


Figure 10.18 Correlation between percentage reductions in volatile solid and biogas production.

Table 10.4 ANFIS based prediction on biogas production (BG) as a function of volatile solid reduction (DELTAVS).

	Volatile solid reduction		Biogas production	
1	0.617	53%	0.995	32632.39 m^3/day
2	0.0109	32.4 %	0.316	19215.62 m^3/day
3	0.985	65.5 %	0.685	26647.93 m^3/day

Volatile Solid reduction =0.617 Biogas production =0.995



a

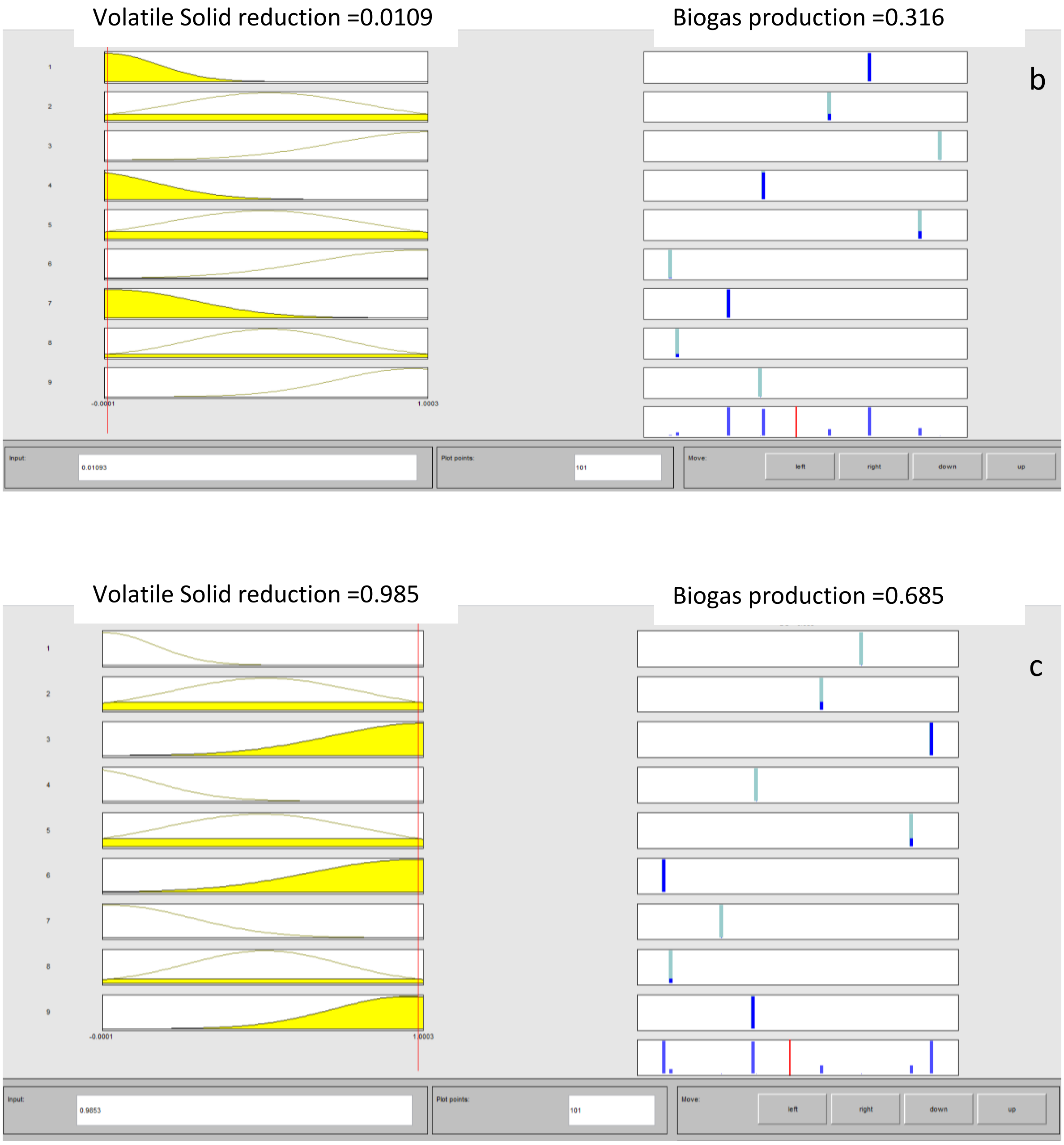


Figure 10.19 (a), (b), (c) ANFIS based prediction on biogas production (BG) as a function of volatile solid reduction (DELTAVS).

10.3.3 Predictions on Feed total solid concentration on biogas production

In this test, Biogas production was predicted against Feed total solid concentration. Total solid concentration is one of the major factors affecting biogas production by determining the total amount of available total organic matter for the action of the microorganisms, methanogens. It is essential to determine the optimum total solid concentration that maximize biogas and methane production and enhance performance of the anaerobic digestion system. The model structure has 20 nodes with 8 linear and 8 nonlinear parameters. As shown in Figure.10.9, four Gaussian type membership functions were used to establish the fuzzy rules. The number and type of membership functions were selected after many trails on other types and number of member functions for the ANFIS prediction. There were four fuzzy rules for this modelling test. The number of model training data used for this prediction was 40 and the model validation and testing was performed using 20 data points. The FIS training was conducted using the hybrid algorithm. The minimum average testing error obtained when fitting the ANFIS model training data to the FIS generated output was 0.1971 as shown in Figure 10.18 and the model validation error was 0.274 as shown in Figure 10.19.

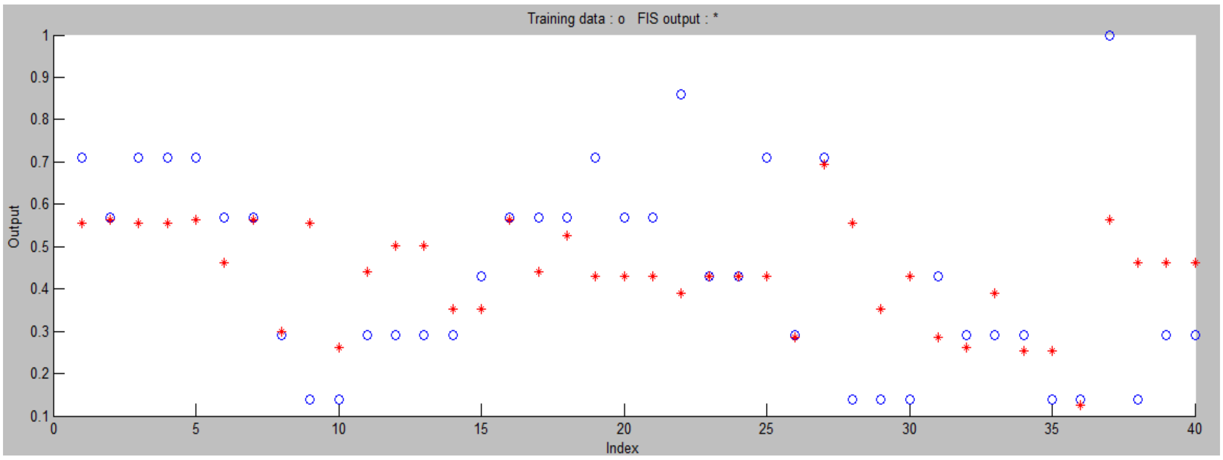


Figure 10.20 Model training plot for the prediction of biogas production as a function of feed total solid concentration.

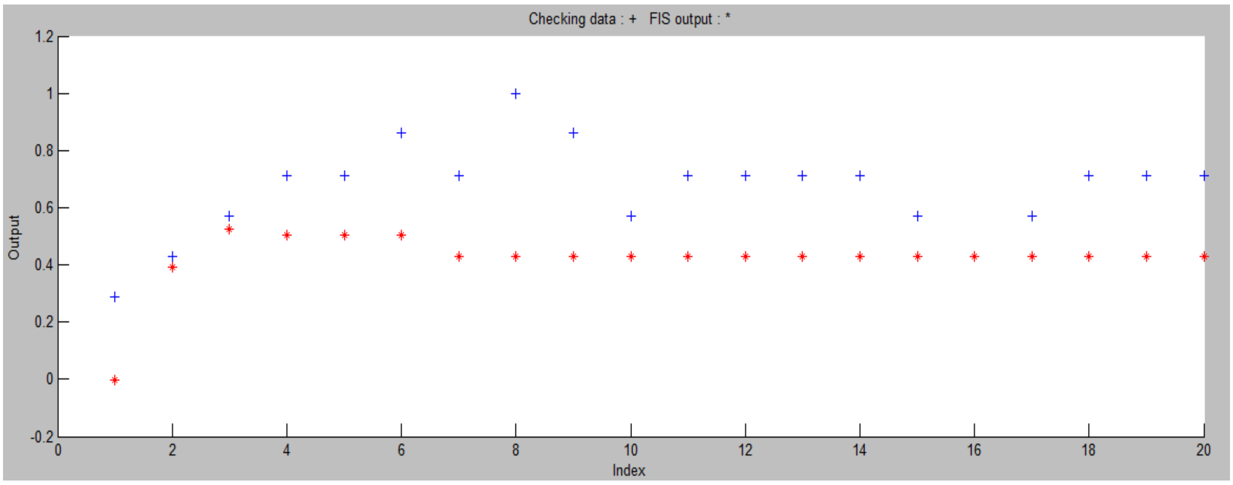


Figure 10.21 Model validation (checking) plot for the prediction of biogas production as a function of feed total solid concentration.

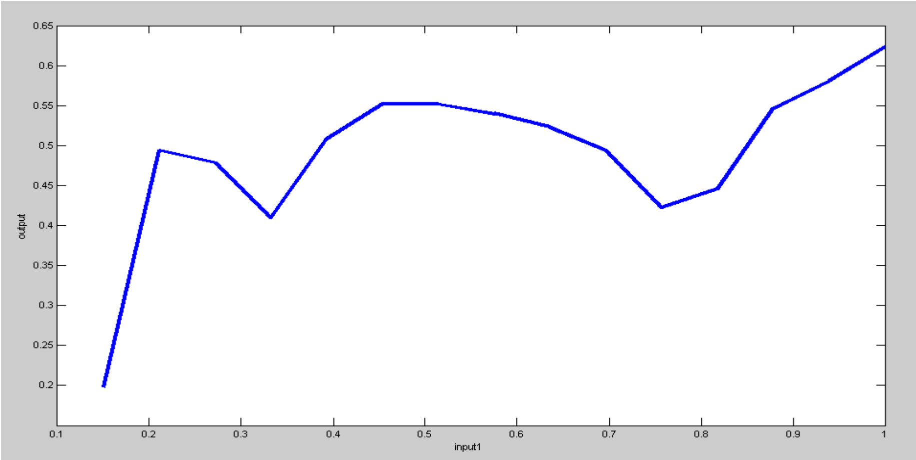


Figure 10.22 Digester feed total solid composition as input and Biogas production as out put

To calculate actual operational Figures in % for TS and m3/day for biogas production from the normalized data

Actual TS = (TS in graph) * (2) + 2.3 (10.8)

Actual BG= (BG in graph)*19304.76+ 13.424.15 (10.9)

10.3.4 Predictions on effect of pH and volatile fatty acid on biogas production

In this test, two input parameters, pH and volatile fatty acid (VFA) are taken as an input with biogas production as output. The hydrolysis and acedogenesis of the feed sludge results in the production of organic acids which will be consumed by methanogens for methane production**.** Volatile fatty acid is accumulation at the acedogenic phase of the digestion process which will eventually be reduced during the methanogenic stage. However, the pH reduction due to VFA accumulation may affect the activity of methanogens. Hence, studying the effect of pH and VFA on and biogas production will assist to understand the relationship and optimize the process for enhanced performance. Two ANFIS tests were performed to evaluate the effect of pH and VFA on biogas production. The first test is based on volatile fatty acid as an input and biogas production as an output. The model structure has 4 gaussian type membership functions with four fuzzy rules and hybrid algorithm was applied. The training and checking errors for the model training and testing datasets were found to be 0.1551 and 0.2795 respectively.

The model structure has 20 nodes with 8 linear and 8 nonlinear parameters. Two gaussian type membership functions were used for each input to establish the fuzzy rules as shown in Figure.10.23. The number of model training data used for this specific prediction was 40 and the model validation and testing was performed using 17 data points. The FIS training was conducted using the hybrid algorithm. The minimum average testing error obtained when fitting the ANFIS model training data to the FIS generated output was 0.1552 as shown in Figure 10.24 and the model validation error was 0.2795 as shown in Figure 10.25. The average errors for training and testing were obtained after 40 epouch of iteration for two gaussian type membership function. The ANFIS model output fits well to the training data. Higher pH in the effluent is associated to lower biogas production as shown in Figure 10.26. Likewise, higher VFA in the effluent beyond 0.6 (48.75 mg/L) indicates lower methane and biogas production (Figure 10.27). Higher VFA shows accumulation of VFA which would have been converted to biogas through methanogenesis.

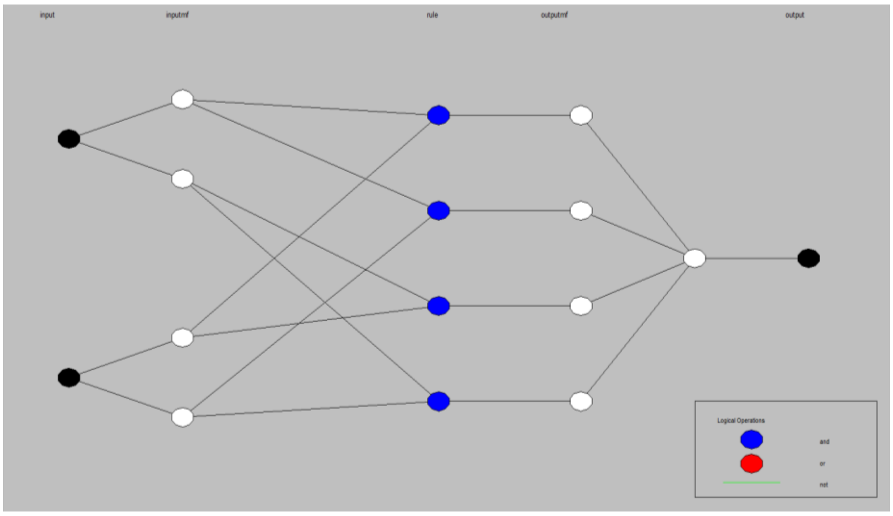


Figure 10.23 ANFIS input output and fuzzy rule base structure.

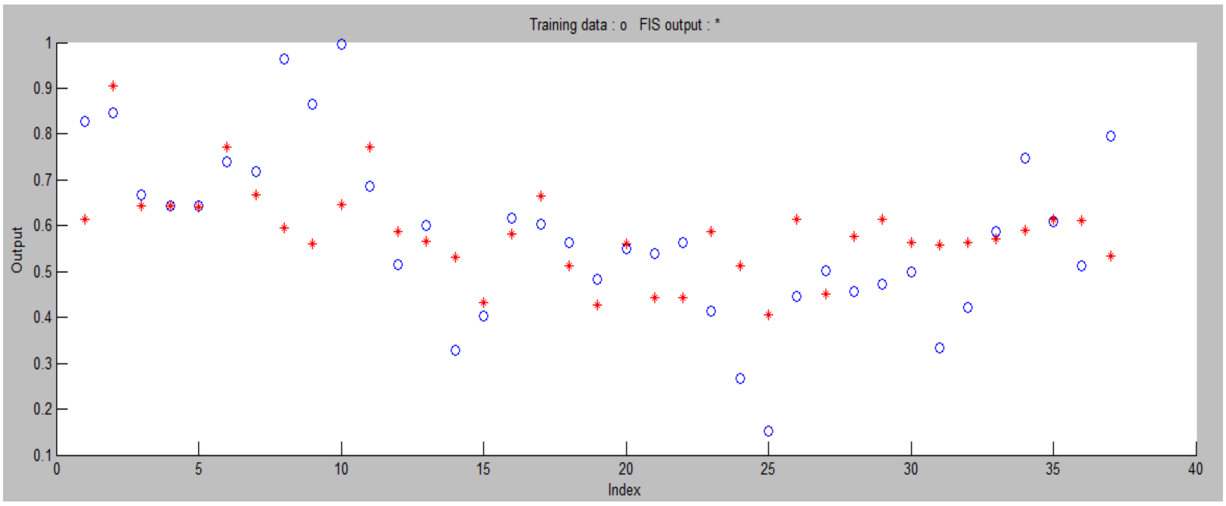


Figure 10.24 Fitting of training data set to FIS generated model output.

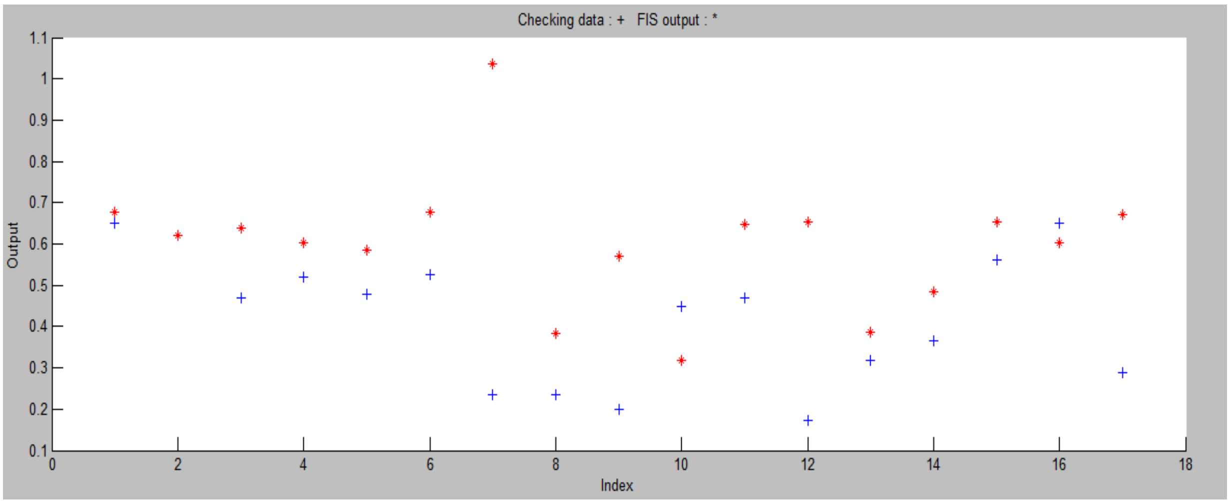


Figure 10.25 Model validation plot for the effect of pH and VFA on biogas production.

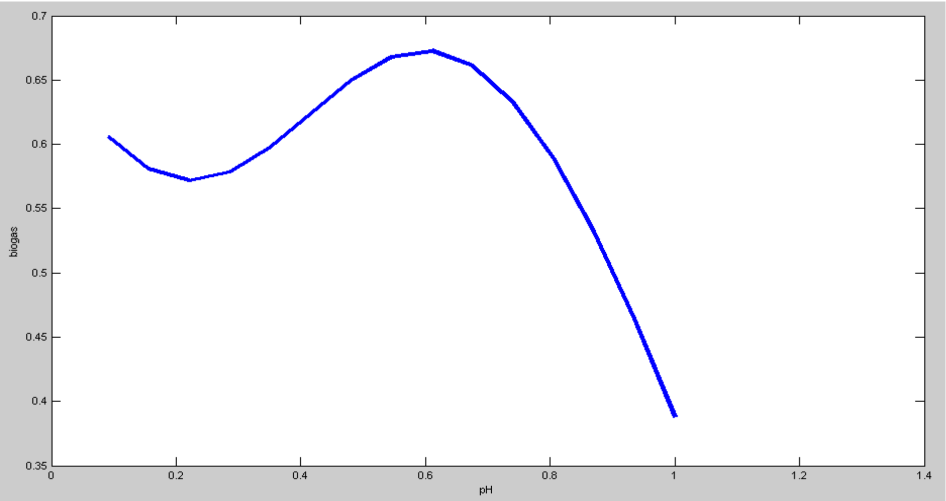


Figure 10.26 Model predictions on biogas production with respect to pH

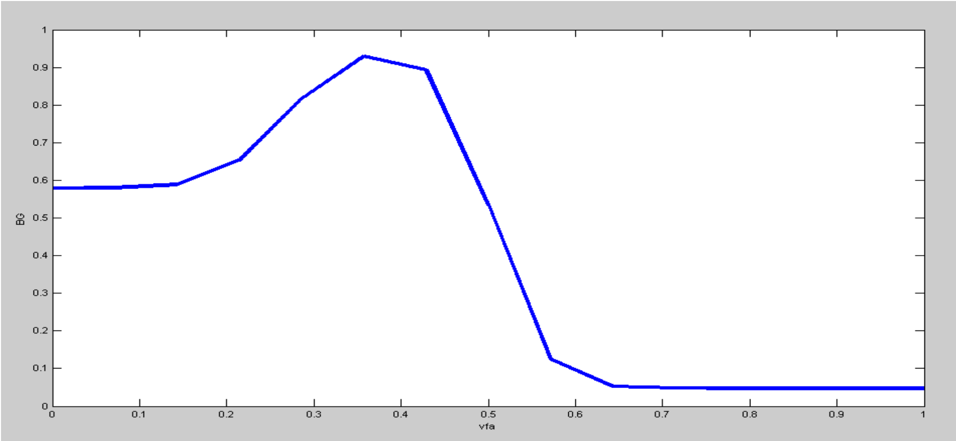


Figure 10.27 Total Volatile fatty acid (VFA) as an input and biogas production (BG) as output.

10.3.5 Predictions on effect of alkalinity on biogas production

Alkalinity is another essential factor affecting digester stability and performance. The buffering capacity of the digester to control fluctuation in pH during operation depends on the alkalinity**.** Hence, it is essential to determine the optimum alkalinity that helps to ensure maximum biogas and methane production and enhance performance of the anaerobic digestion system. Alkalinity was modelled against biogas production using the hybrid algorithm first-order sugeno-fuzzy model. The ANFIS structure constitutes 12 nodes with 4 linear and 4 nonlinear parameters. Two gaussian type membership functions were used to establish the fuzzy rules. The number of model training data used for this specific prediction was 37 and the model validation and testing was performed using 17 data points. The minimum average training error obtained when fitting the ANFIS model training data to the FIS generated output was 0.1576 and the model validation error was 0.2097 for two gaussian membership functions. Alkalinity versus biogas production plot has the profile shown in Figure 10.29 (a) and (b) for two gaussian membership functions. The optimum alkalinity that maximizes biogas production was at point 0.633(2431.21 mg/l) and the corresponding biogas production was 0.75 (27902.74 m^3/day) as shown on Figure 10.29(a). The average error was 0.14989 for training and 0.2469 for checking data sets respectively in case of three gaussian member functions. The optimum alkalinity that maximizes biogas production was at point

0.578 (2377.81 mg/l) and the corresponding biogas production was 0.75 (27902.74 m^3/day) for two gaussian member functions as shown on Figure 10.29(b).

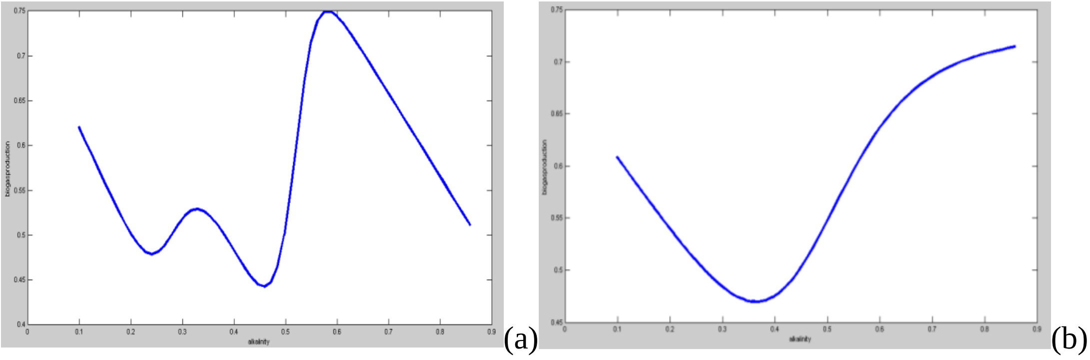


Figure 10.29 ANFIS model output for alkalinity versus biogas production (a) three membership functions (b) two membership functions.

10.3.6 Volatile solids, volatile fatty acid, sludge feed flow rate as input and biogas production as an output

Volatile solid, volatile fatty acid and sludge feed flow rate were used as an input in this study to investigate the surface response and combined effect of these parameters on biogas production. Volatile fatty acid is directly proportional to the biogas production capacity of anaerobic digestion system. Similarly, VFA concentration is an important indicator of methanogenic activity in a digester. The sludge through put (feed flow rate) is aother key operational parameter which is analysed in this study along with the other two input parameters. The model structure has 78 nodes with 108 linear and 27 nonlinear parameters. 27 fuzzy rules were used to establish the model. Three Gaussian member functions were used in the analysis. The number of model training data used for this specific prediction was 59 and the model validation and testing was performed using 15 data points. The minimum average testing error between model training data and the FIS generated output was 0.0362 as shown in Figure 10.30 and the model validation error was 0.0489 as shown in Figure 10.31. The training and validation (testing) errors were evaluated at 20 epoch. The average testing error for the training and checking data was reasonably good. The architecture of the ANFIS test used in this particular case is given in Figure 10.32 (a) and (b). The ANFIS model in this study with three inputs was also evaluated for a different ANFIS structure with two Gaussian member functions, 8 fuzzy rules, 34 nodes, 32 linear parameters, 12 nonlinear parameters, 15 checking data pairs and 59 training data pairs as shown in Figure 10.32(b). The model training and validation errors were 0.10003 and 0.14208 for this particular ANFIS structure as shown in Figure 10.33. Three Gaussian membership functions, three input parameters, 30 epoches and linear output were considered for this model.

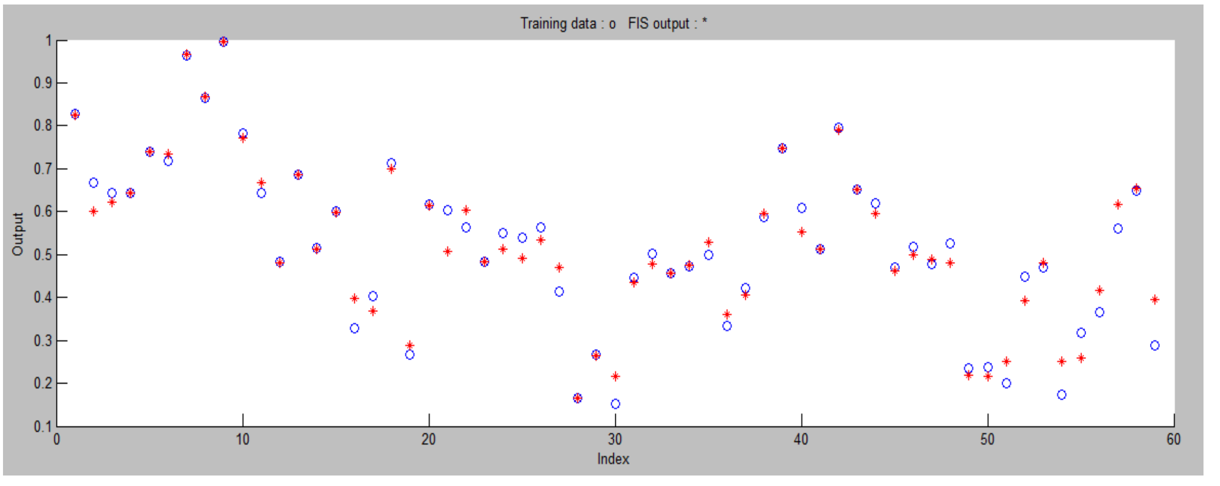


Figure 10.30 Model training data plotted with FIS generated output for the study with three inputs parameters.

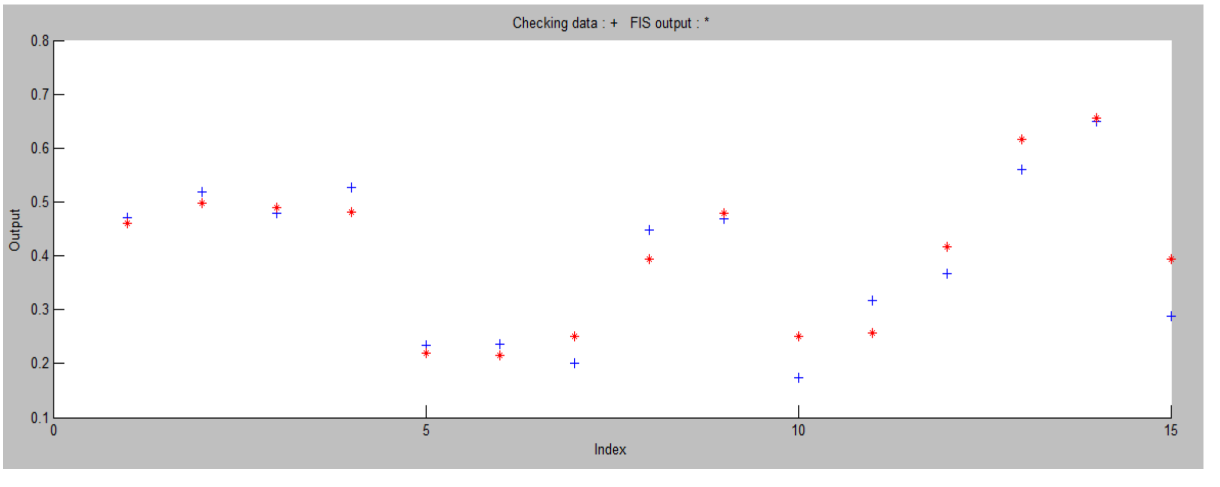
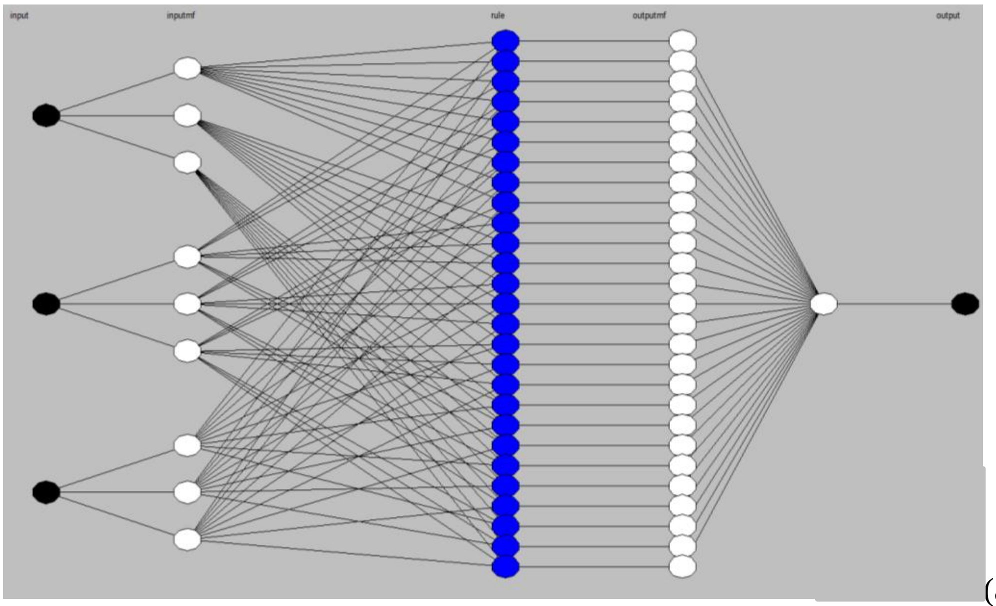
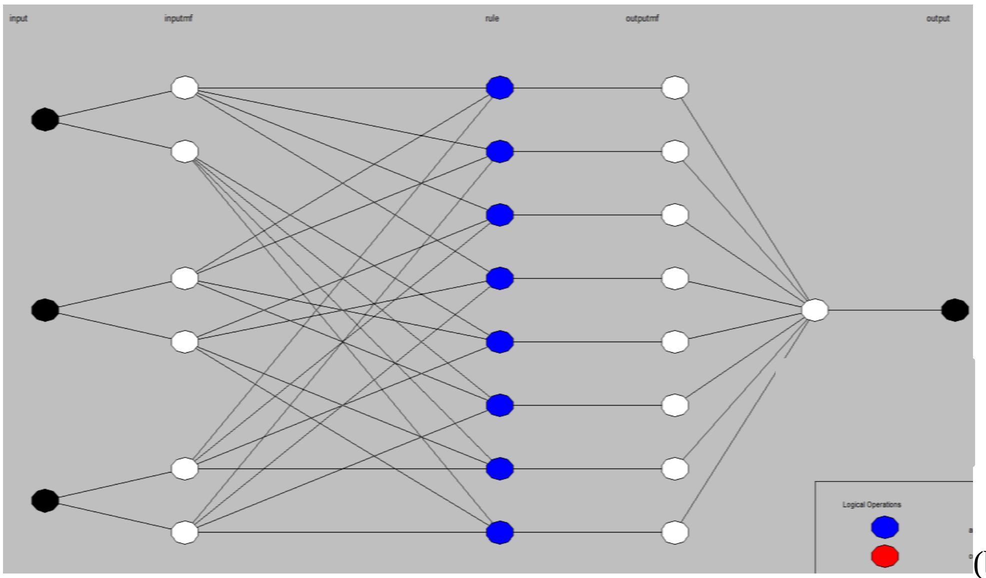


Figure 10.31 Model testing (validation) data for the study with three inputs parameters.



(a)



(b)

Figure 10.32 ANFIS structure with of the model with (a) 27 rules and three gaussian member functions (b) two gaussian membership functions and 8 rules bases.

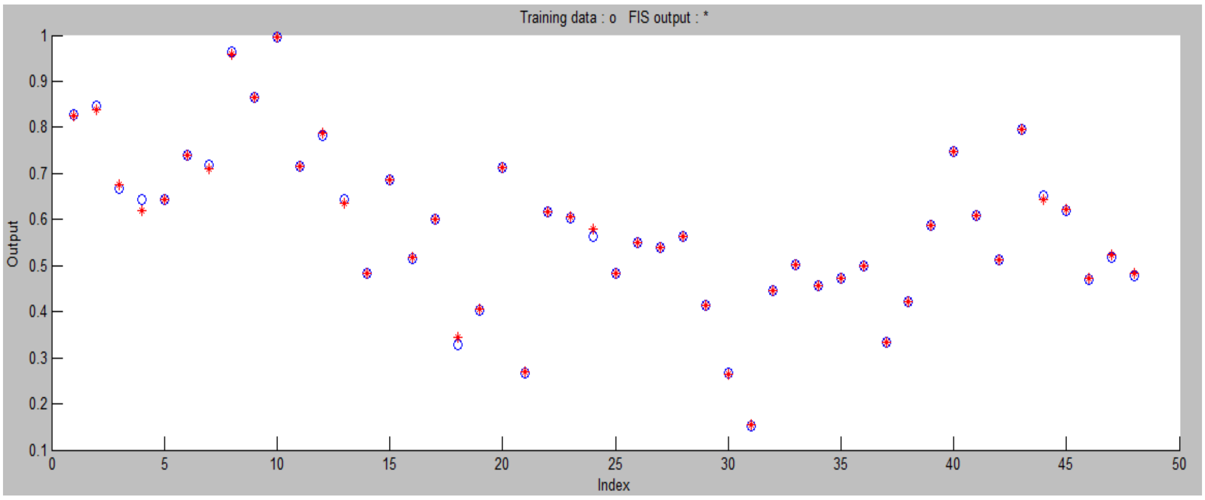


Figure 10.33 Model plot for three gaussian membership functions with 3 input, 30 epouch and linear output model.

10.3.7 ANFIS model on Effect of alkalinity, pH, VFA and sludge feed flow on biogas production.

In this particular case, four input variables which were investigated individually in the previous subsections are introduced as input together into the model with biogas production as an output. The model structure has 193 nodes, 405 linear and 24 nonlinear parameters. Three gaussian type membership functions were used to establish 81 fuzzy rules as shown in Figure 10.35. The number of model training data used for this specific prediction was 40 and the model validation and testing was performed using 17 data points. The FIS training was based on hybrid algorithm for 20 epochs. The minimum average testing error obtained when fitting the ANFIS model training data to the FIS generated output was 0.0004 as shown in Figure 10.36 and the model validation error was 0.2614 as shown in Figure 10.37. Figure 10.38 shows that an increase in alkalinity increases biogas production whereas pH is negatively related to biogas production, the highest production happening near the minimum pH value in the range (7.03) as shown in Figure 10:39. VFA concentration in the effluent stream is directly proportional to biogas production up to VFA level of 0.4 (34 mg/l). Further increase in VFA results in a decrease of biogas production. Figure 10.41 shows surface response and interaction effect among the input variables. Figure 10.41 (a) shows how high alkalinity and low VFA favor high biogas production. As alkalinity/VFA ratio is the best way to control stability of a digester, it also shows the correlations between alkalinity, VFA and biogas production. Likewise, intermediate VFA and low pH enhances biogas production as shown in Figure10.41 (b), low VFA in effluent stream and high sludge flow rate result in higher biogas production as shown in Figure 10.41 (c), besides VFA at 0.3 (26.6 mg/l) and sludge feed flow rate of 0.57 (1605 m^3/day) results in maximum biogas production. Combination of the four input variables in Table 10.5 (a) and (b) show input conditions which result in minimum biogas production (item (a)) and maximum biogas production (Item (b)).

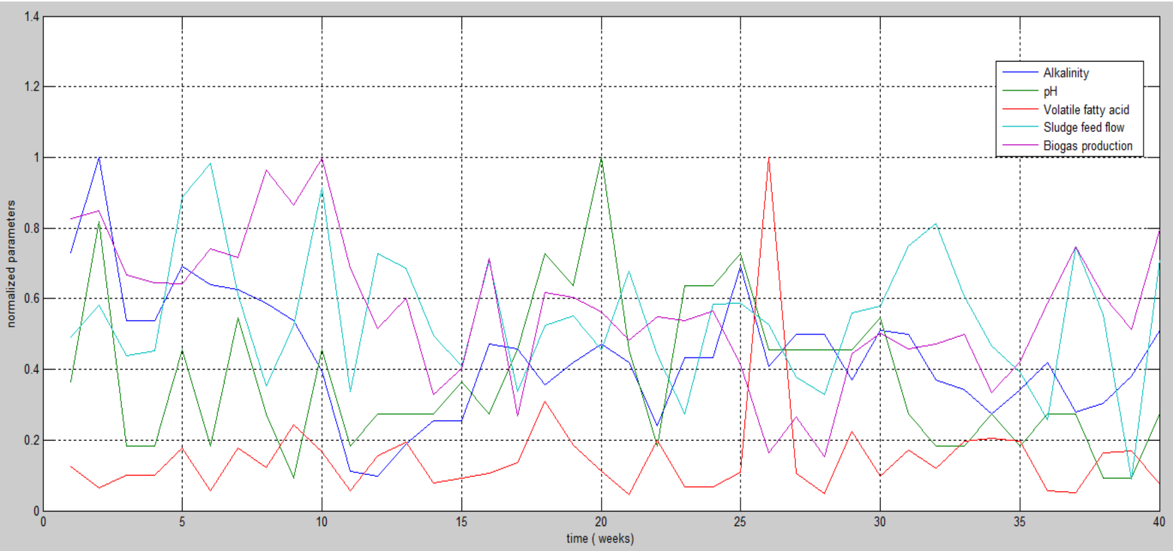


Figure 10.34 Normalized input and output data used for the training of the ANFIS model

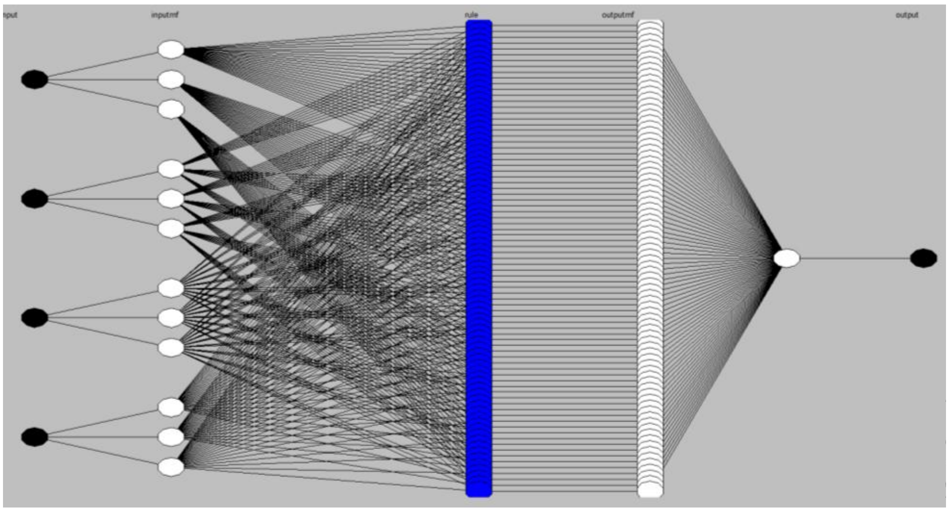


Figure 10.35 ANFIS structure for four inputs and 81 based rules.

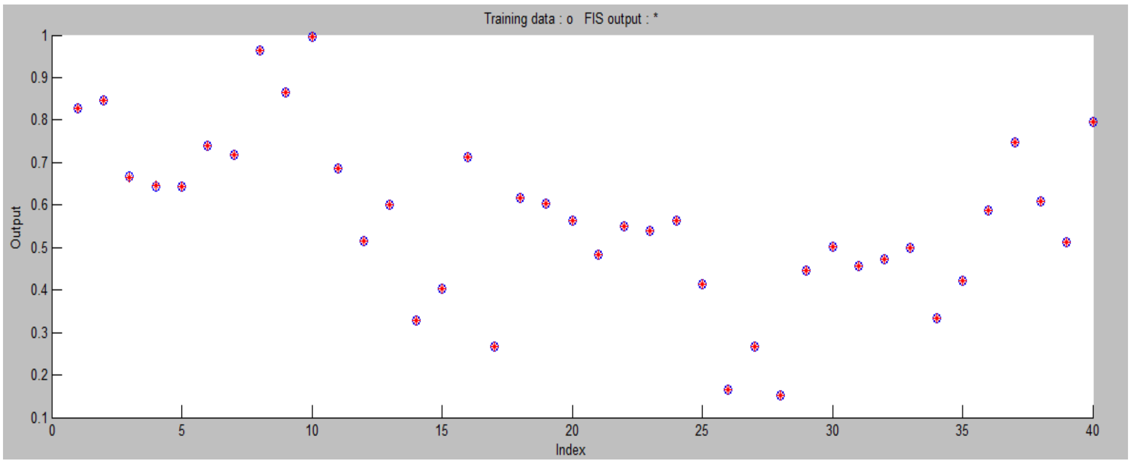


Figure 10.36 predicted FIS output with model training data.

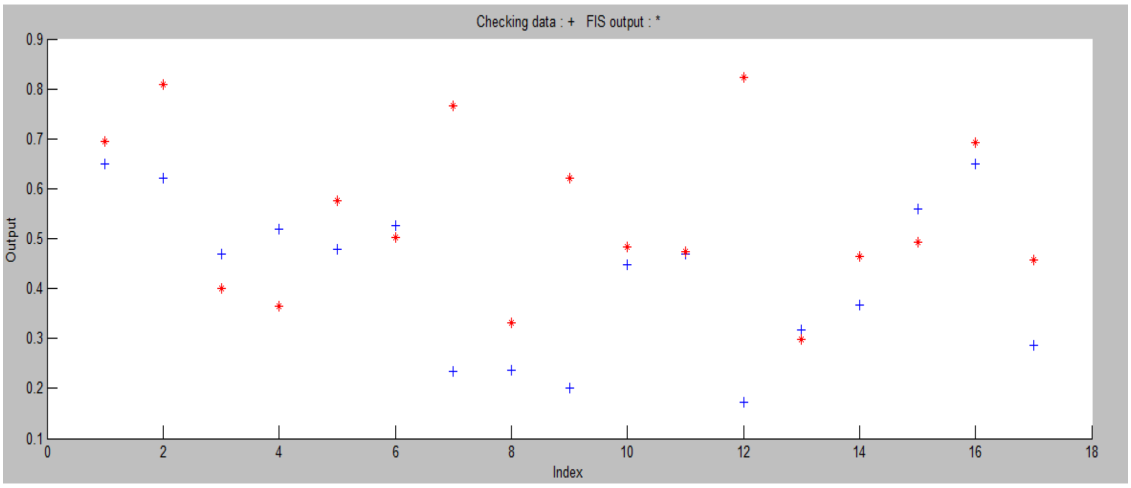


Figure 10.37 Model validation data with FIS output.

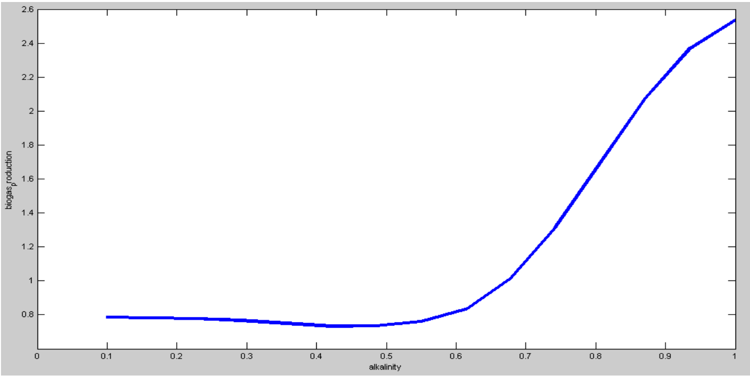


Figure 10.38 Model predicted relationship between alkalinity and biogas production

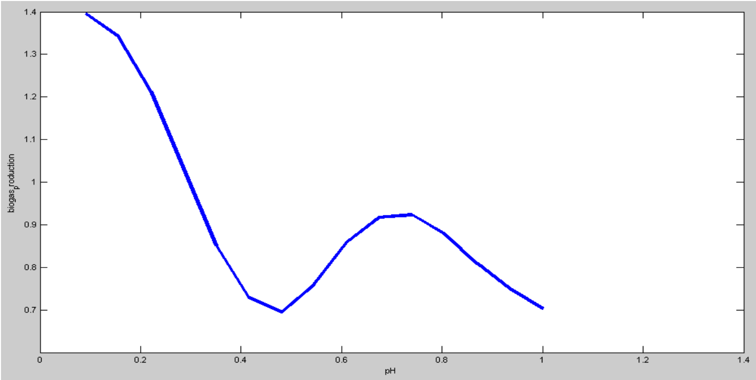


Figure 10.39 Predictions on effect of pH on biogas production

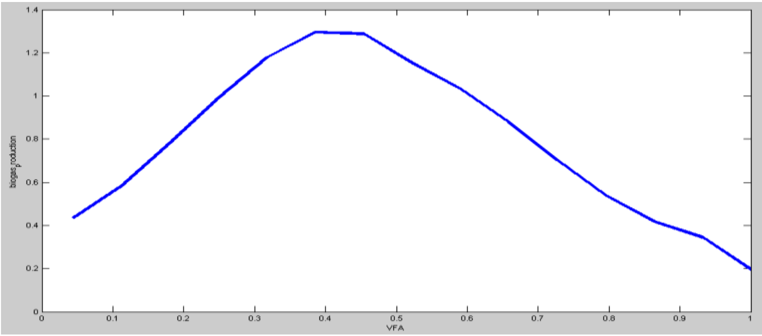
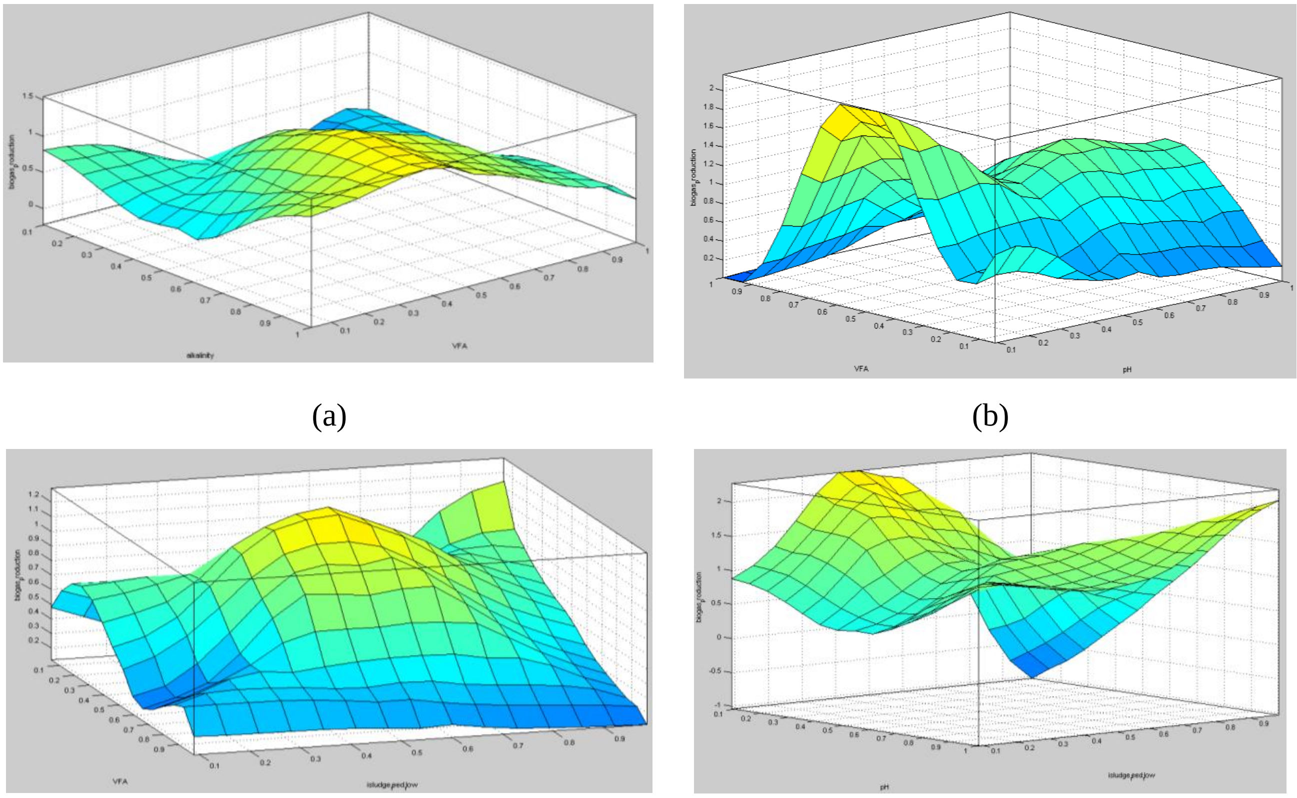


Figure 10.40 Predictions on effect of volatile fatty acid on biogas production



(a) (b)

(c) (d)

Figure 10.41 Surface responses on the effect of two input parameters on biogas production

Table 10.5 Selected FIS based predictions for different combinations of input and the impact on biogas production (a) normalised data (b) actual data

Table 10.5(a)

Item	Alkalinity	pH	Volatile fatty acid	Sludge feed flow	Biogas production
1	0.296	0.126	0.068	0.11	0.141
2	0.526	0.27	0.061	0.984	1.13

Table 10.5(b)

Item	alkalinity	pH	Volatile fatty acid	Sludge feed flow	Biogas Production
1	2104.04	7.06	9.52	1032.54	16146.17
2	2327.33	7.10	9.00	1597.97	35238.52

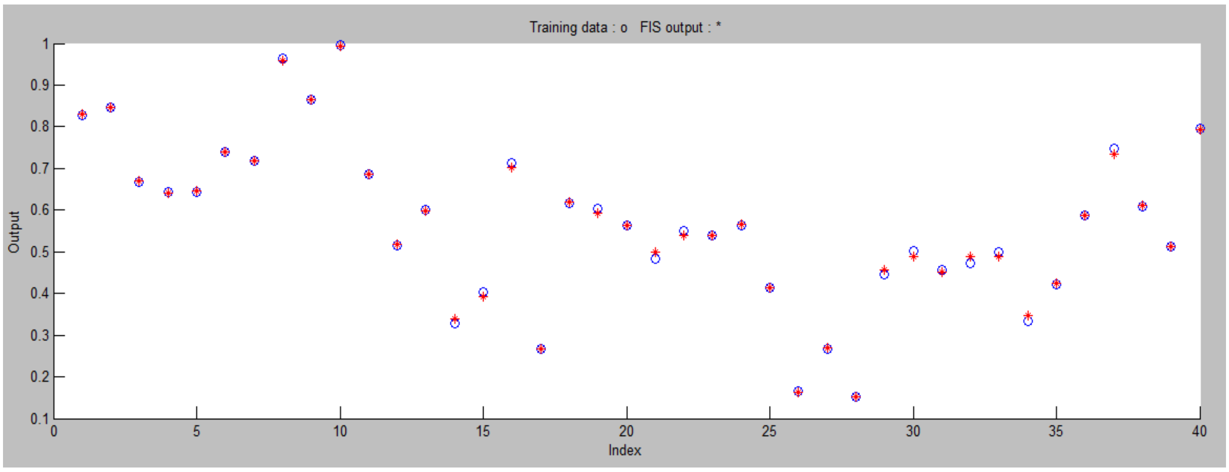


Figure 10.42 FIS based predictions with training dataset for four input parameters

10.3.8 ANFIS predictions for seven input parameters with methane percentage (biogas quality) as an output

In this particular case seven input variables which were investigated individually in the previous subsections are introduced as an input together into the model with methane percentage as a linear output as shown in Figure 10.43. The model structure has 294 nodes, 128 linear and 28 nonlinear parameters. Three gaussian type membership functions were used to establish 128 fuzzy rules as shown in Figure 10.51. The number of model training data used for this specific prediction was 61 and the model validation and testing was performed using 15 data points. The FIS training was conducted using the hybrid algorithm for 30 epochs. The minimum average testing error obtained when fitting the ANFIS model training data to the FIS generated output was 0.0009 as shown in Figure 10.48 and the model validation error was as shown in Figure 10.48. The historical data from BWWTP and the FIS based predictions show that high effluent alkalinity (2787.50 mg/l), higher effluent total solid concentration (1.6 %) and high sludge feed flow rate (1597 m^3/day) correlate to high methane percentage in the range of 54-62 % as shown in Figure 10.47. Figure 10.46 (a),(b),(c),(d) show surface responses for different combinations of input to predict methane percentage.

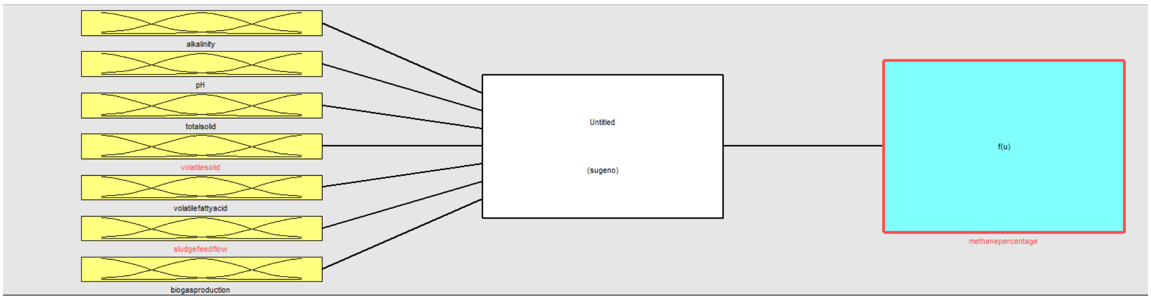


Figure 10.43 ANFIS structure on the FIS editor screen.

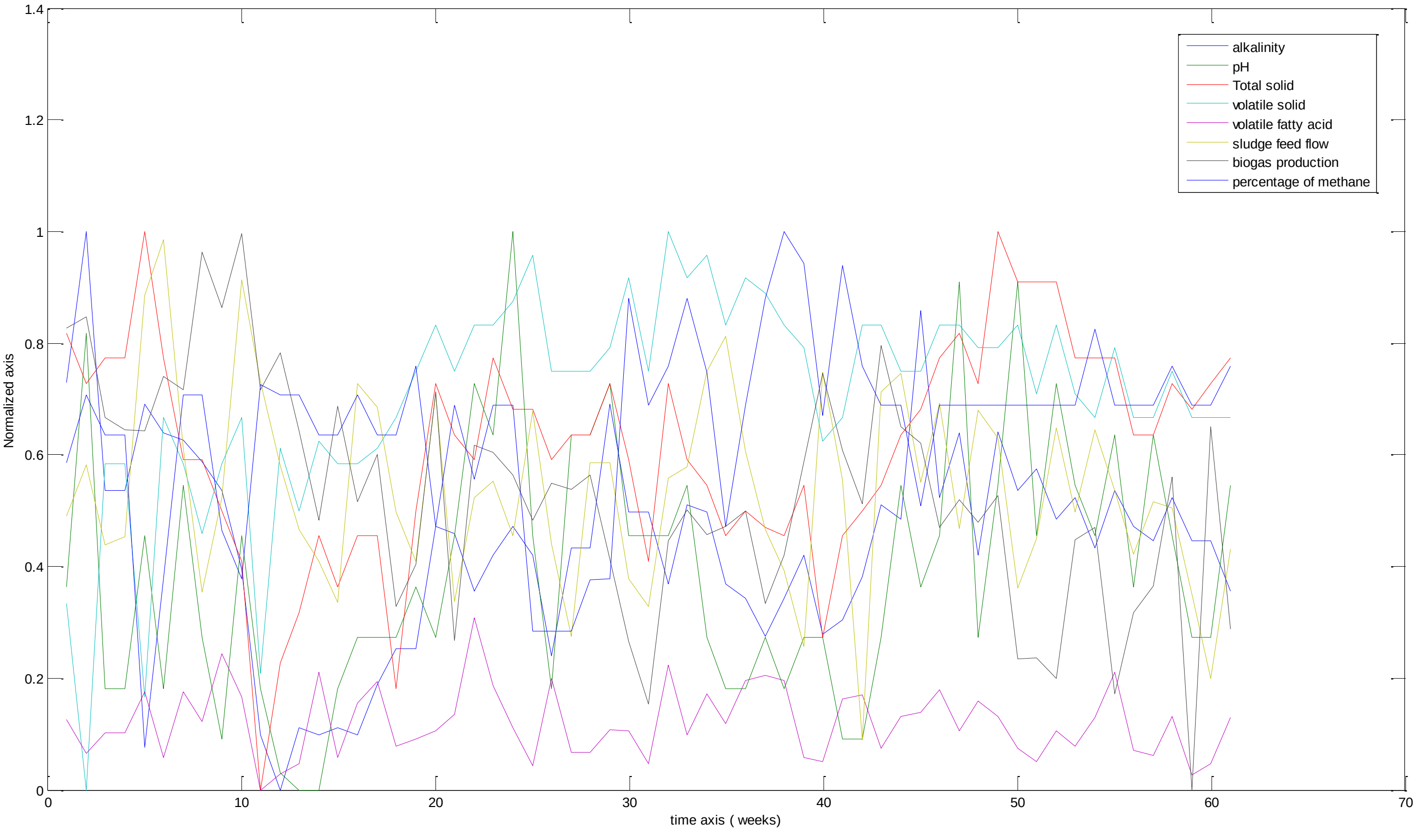


Figure 10.44 Training dataset used for the model prediction with input and methane percentage as output parameter.

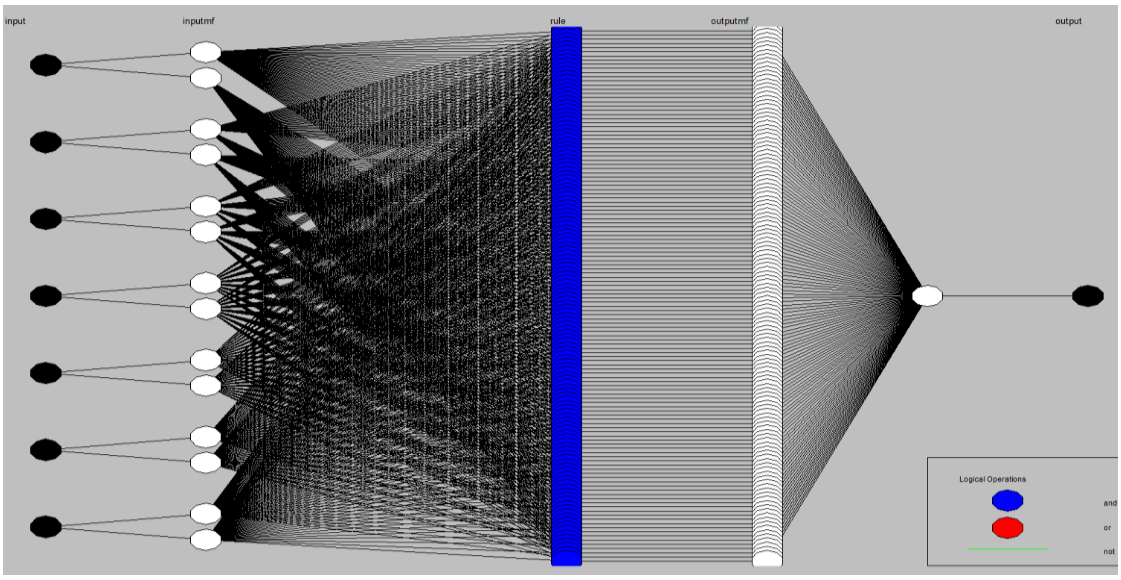


Figure 10.45 ANFIS model structure for 7 input variables

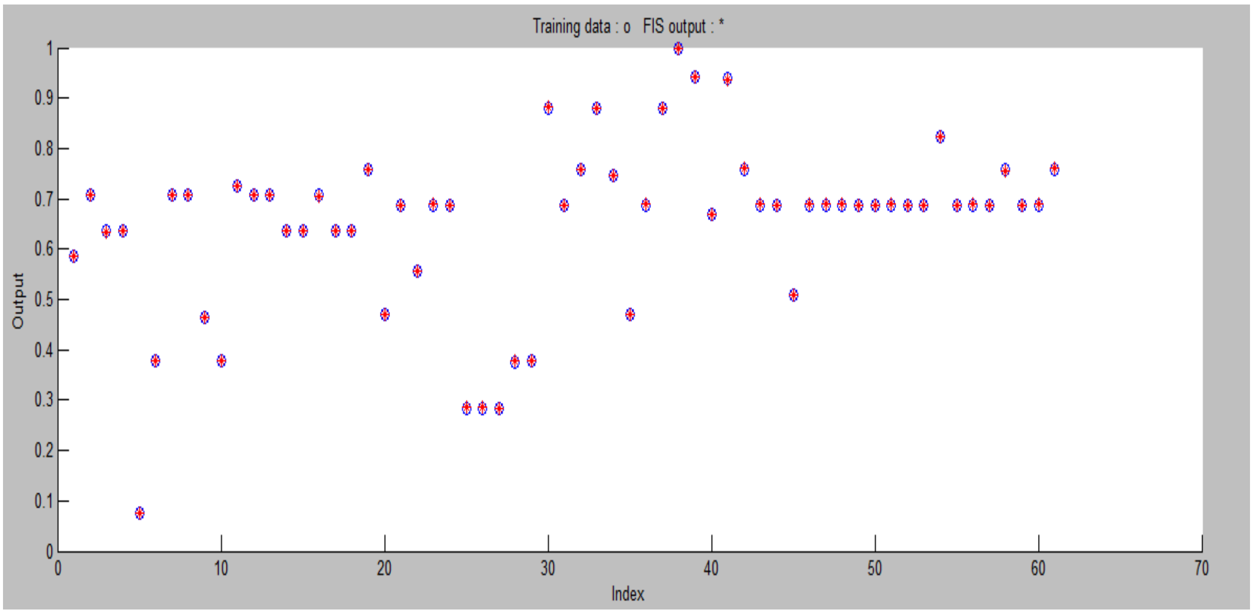


Figure 10.45 Training data set with FIS output for four input parameters

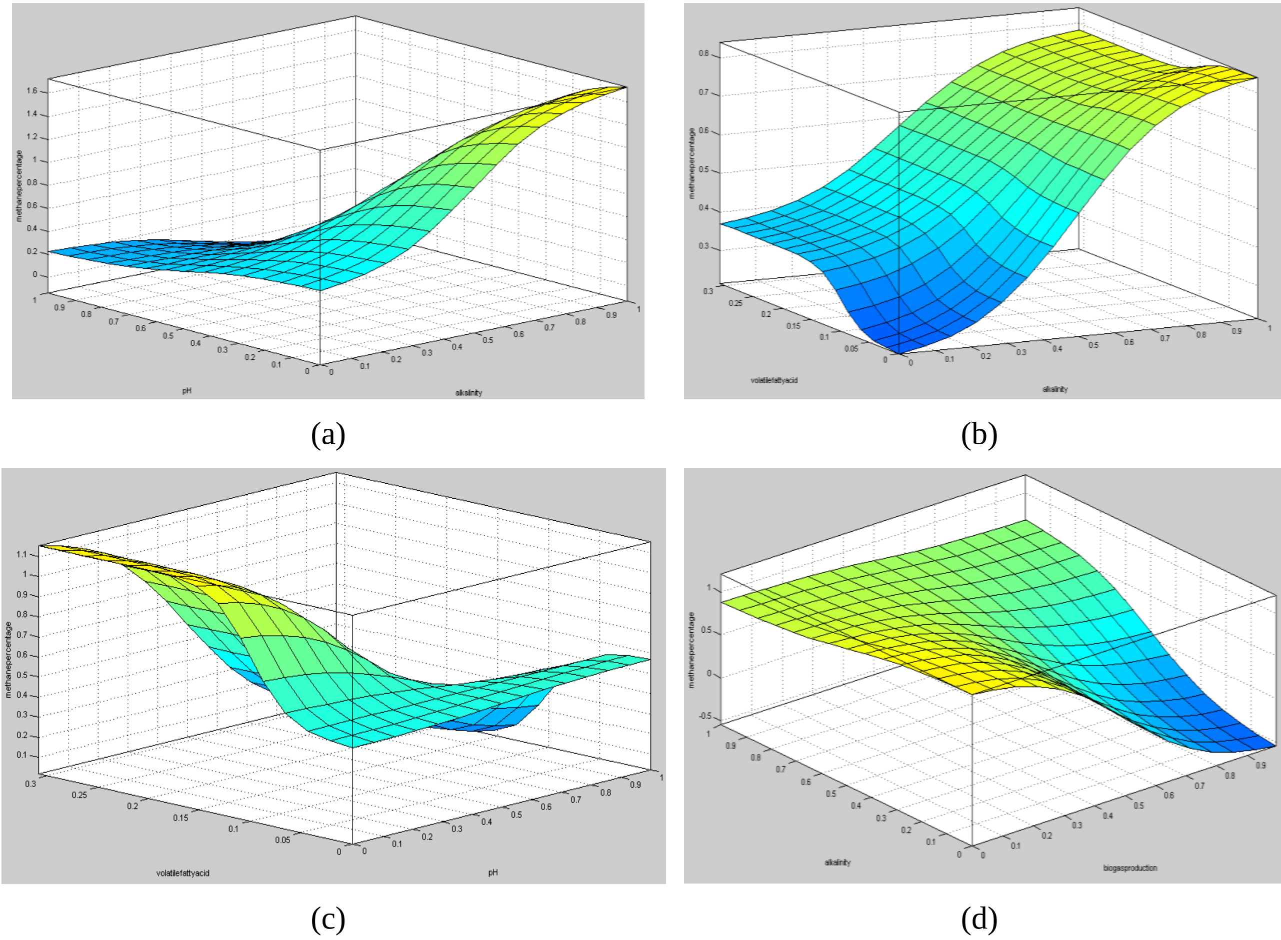
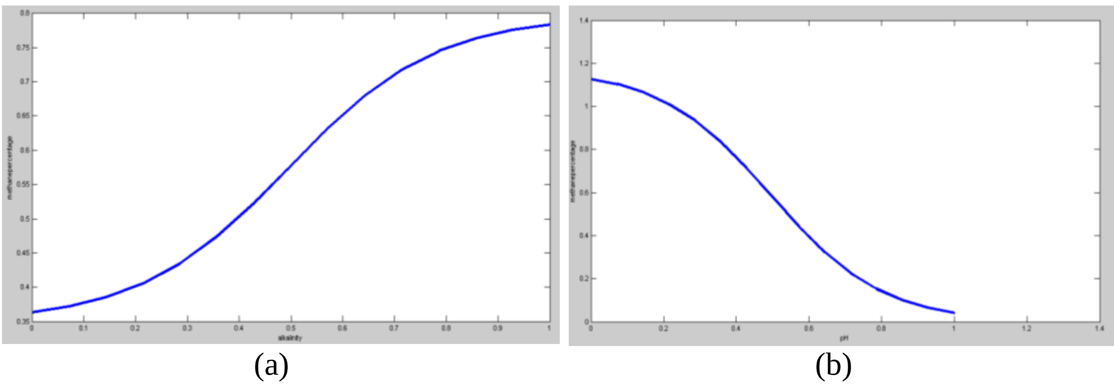
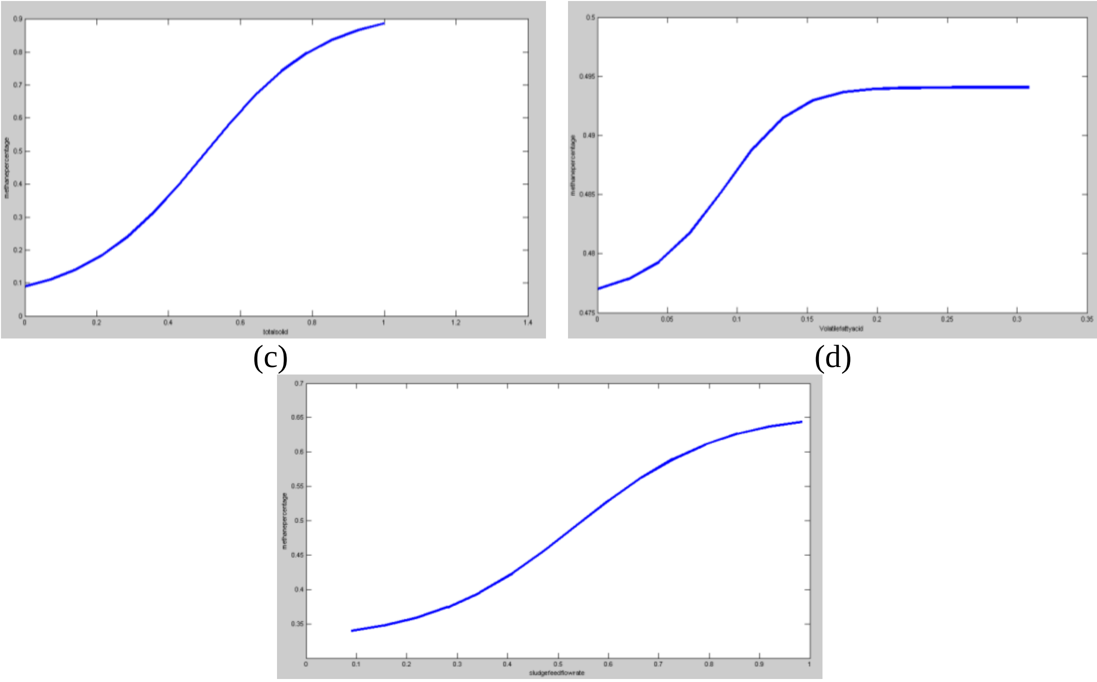


Figure 10.46 Surface responses for different combinations of input to predict biogas production. (a) pH and alkalinity, (b) VFA and Alkalinity, (c) VFA and pH, (d) biogas production and effluent volatile solid and (e)alkalinity and biogas production





(c) (d)

(e)

Figure 10.47 FIS model outputs for each input variable against methane percentage.

10.3.9 ANFIS predictions for seven input parameters with biogas production as an output

The ANIFS model was also tested for a different structure with 294 nodes, 128 linear and 28 nonlinear parameters. Three gaussian type membership functions were used to establish 128 fuzzy rules. The number of model training data used for this specific prediction was 61 and the model validation and testing was performed using 15 data points. The FIS training was conducted using the hybrid algorithm for 30 epochs. The minimum average testing error obtained when fitting the ANFIS model training data to the FIS generated output was 0.0002 as shown in Figure 10.48. There were 30 epouchs of training for 7 input parameters (alkalinity, pH, total solids, volatile solids, volatile fatty acid, sludge feed flow rate, methane percentage) and biogas production was the output variable, the error for the ANFIS test was 0.0055 as shown in Figure 10.50.

The structure of the ANFIS consisted of two gaussian type membership functions with one constant output. Designated epoch number reached stability and ANFIS training completed at epoch 2 with an error of 0.0055. In case of gaussian bell type member function with linear output, the ANFIS model prediction error was 0.0001.

Figure 10.49 shows that as the number of gaussian member functions is two the FIS output are simplified gaussian s- shape curves that minimize the error. An increase in alkalinity, total solid, volatile solid and sludge feed flow result in increase of biogas production. An increase in volatile fatty acid and pH to the contrary results in a decrease in biogas production. Selected model prediction points are tabulated in Table 10.6a for normalized input and output values and Table 10.6b for the actual values. Optimum selected combinations that maximize methane production are shown in these tables. High alkalinity, lower pH, intermediate VFA, higher sludge feed flow and total solid content in the effluent streams are correlated to high biogas production. A decrease in alkalinity and sludge feed flow rate significantly reduces methane production. Volatile solid reduction efficiency is proportional to the methane production that higher volatile solids in the effluent relate to lower biogas production as shown in Table 10.6b.

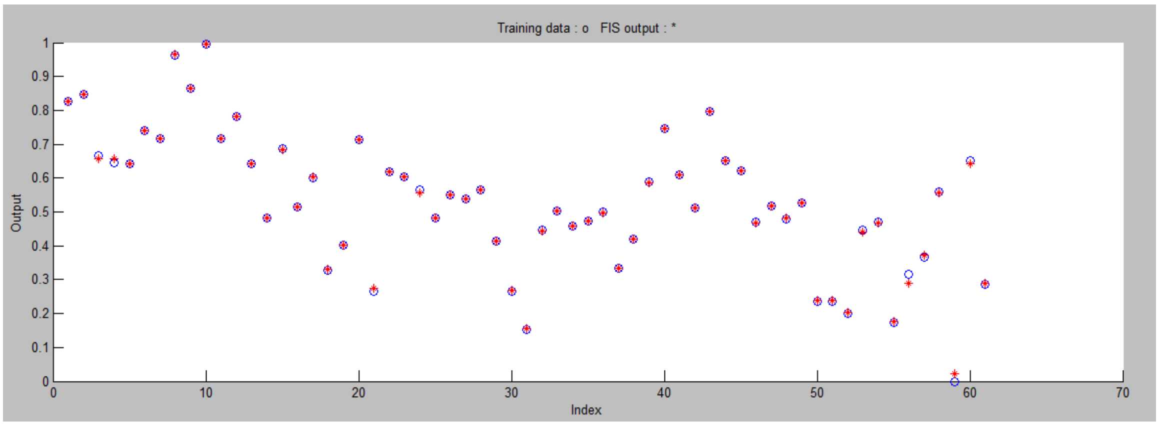
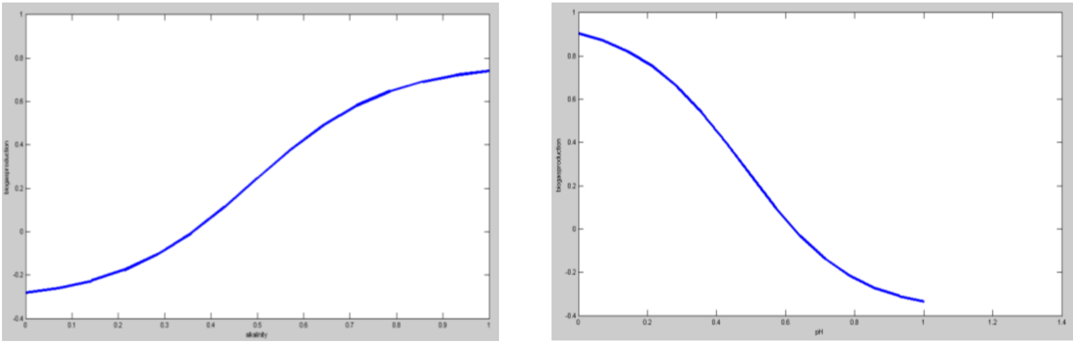
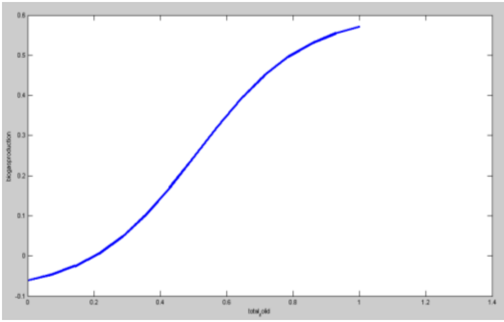


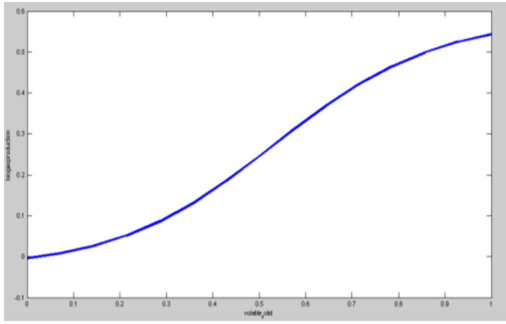
Figure 10.48 training data and FIS model predictions with methane percentage as output.



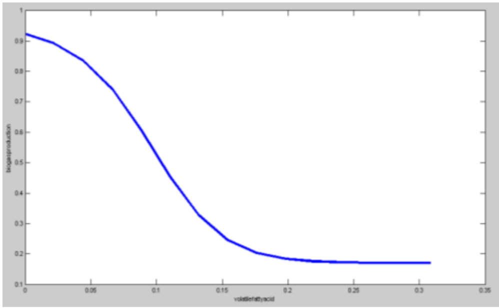
(a) (b)



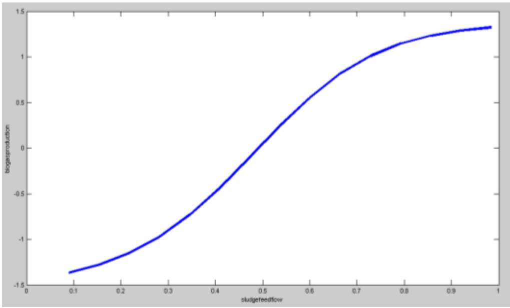
(c)



(d)



(e)



(f)

Figure 10.49 FIS model outputs for each input variable against biogas production.

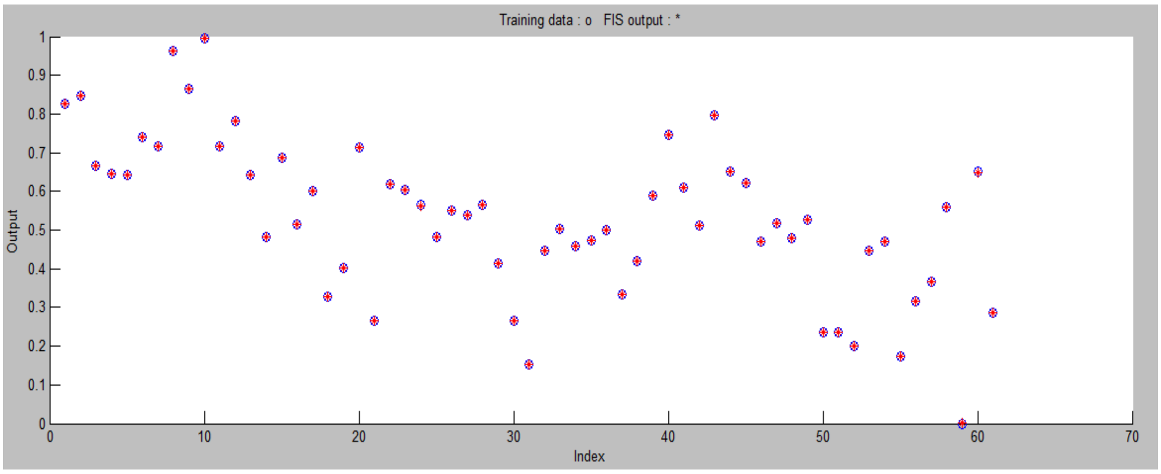


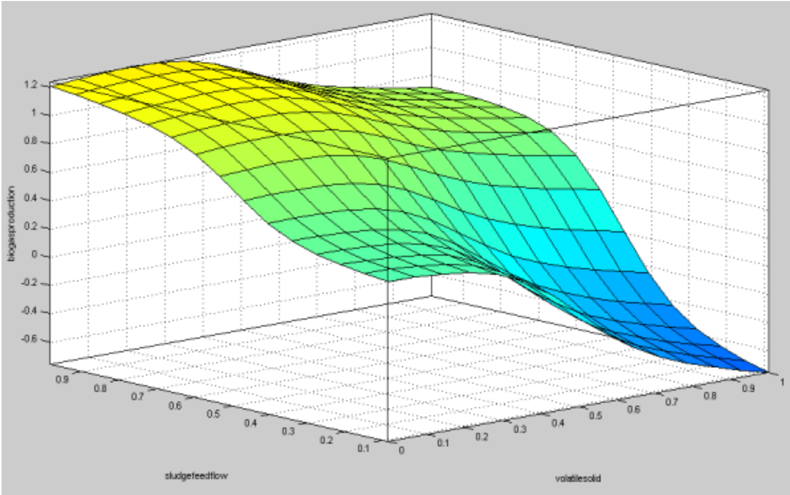
Figure 10.50 FIS model output versus training data for the prediction with methane percentage as an output normalized.

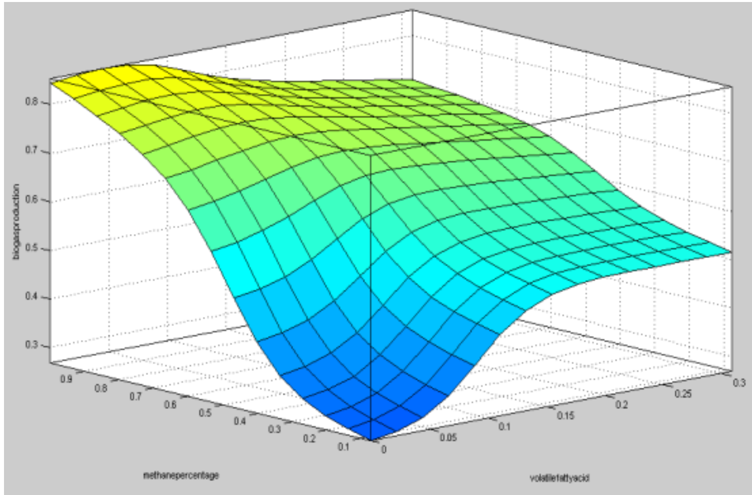
Table 10.6a FIS based predictions for different combinations of input and the impact on biogas production (Normalized data).

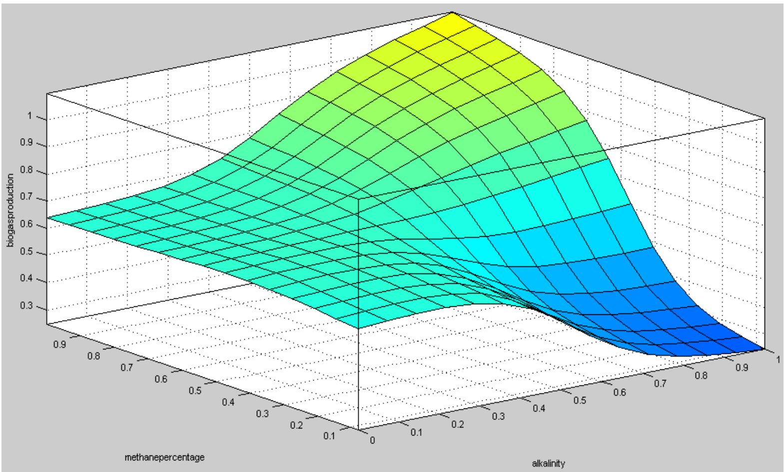
Case	Alkalinity	pH	TS	VS	VFA	Sludge Feed Flow	Methane percentage	Biogas production
1	1	0.354	0.939	0.589	0.308	0.704	0.93	1.32
2	0.484	0.858	0.939	0.026	0.508	0.516	1.2	0.882
3	0.0447	0.183	0.069	0.142	0.009	0.166	0.186	0.463
4	0.0447	0.809	0.114	0.045	0.009	0.923	0.936	1.11
5	0.0447	0.972	0.004	0.045	0.009	0.923	0.936	0.0156

Table 10.6b FIS based predictions for different combinations of input and the impact on biogas production (actual data converted from the normalized values).

Case	alkalinity (mg/l)	pH	ETS (%)	EVS (%)	VFA ppm	SF (m3/day)	BG (m3/day)
1	2787.50	7.13	1.60	76.53	27.22	1416.83	38906.41
2	2286.55	7.26	1.60	73.16	41.97	1295.20	30450.96
3	1860.07	7.08	1.12	73.85	5.15	1068.77	22362.29
4	1860.07	7.25	1.14	73.27	5.15	1558.51	34852.43
5	1860.07	7.29	1.08	73.27	5.15	1558.51	13725.36


biogasproduction
sludgefeedflow
volatilesolid
(a)


biogasproduction
methanepercentage
volatilefattyacid
(b)


biogasproduction
methanepercentage
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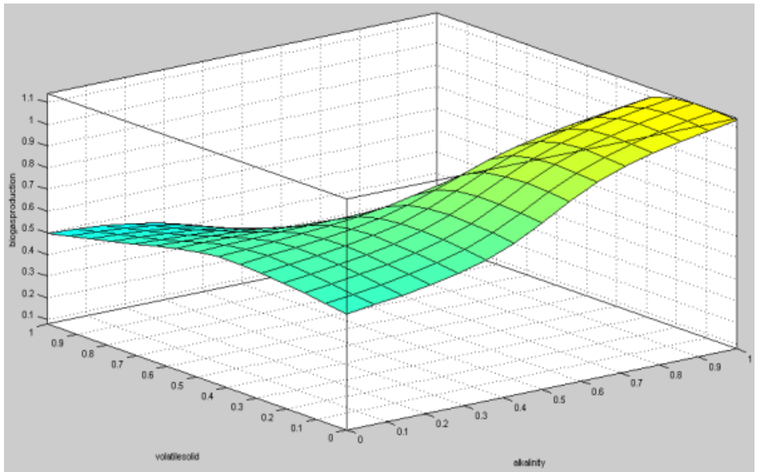
(c)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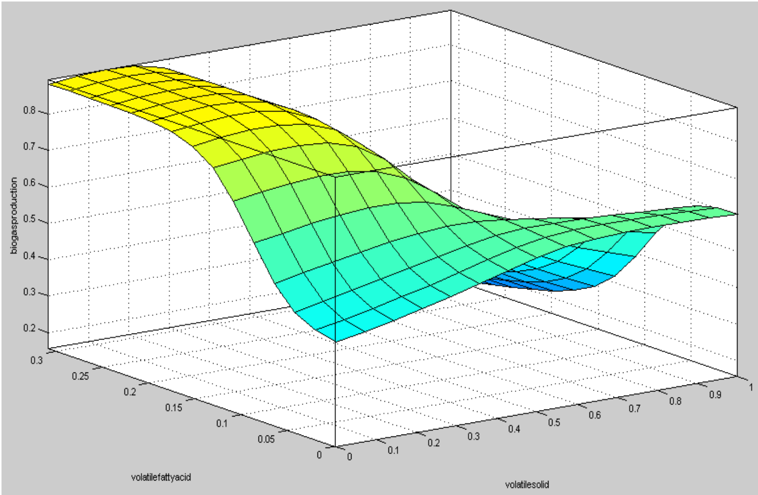
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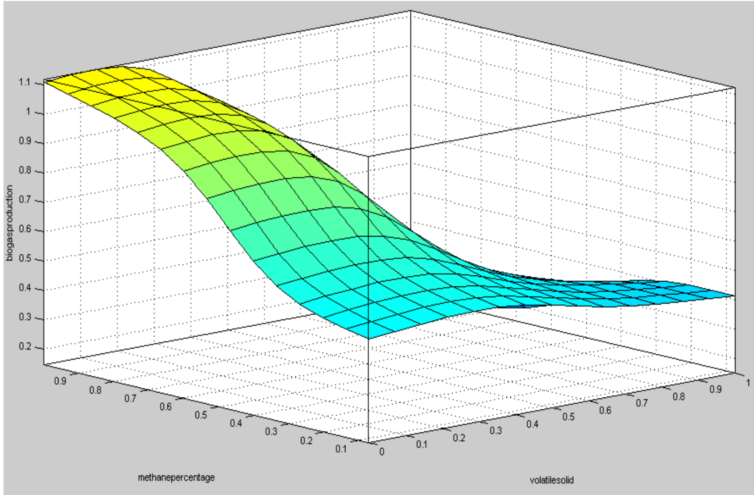
(e)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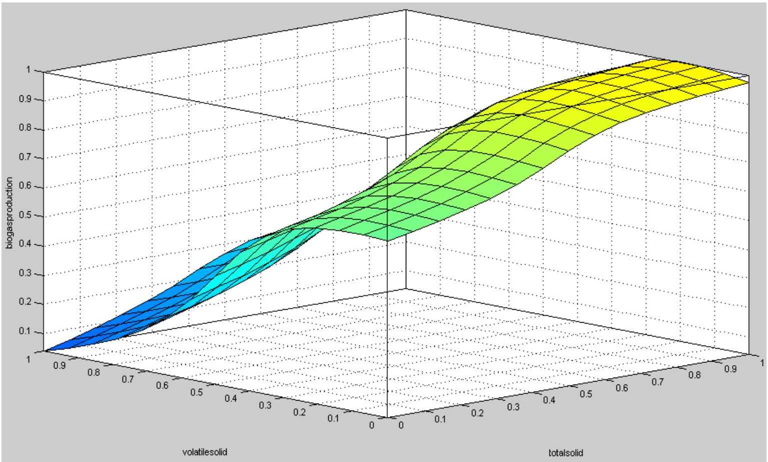
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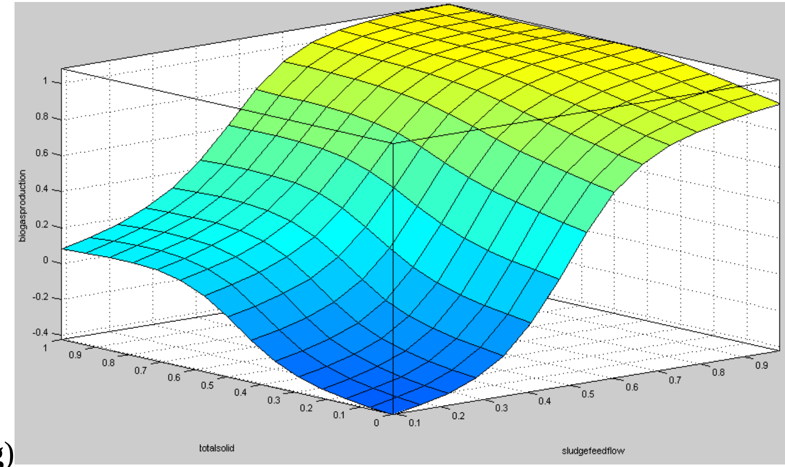
(g)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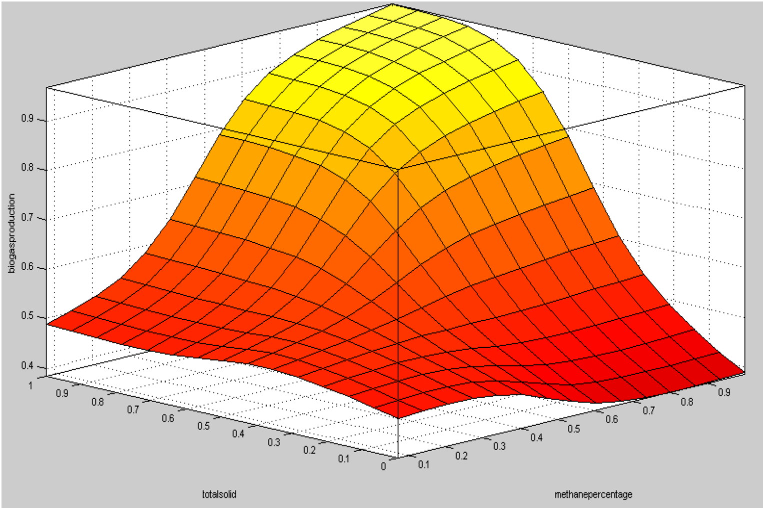
sludgefeedflow

(h)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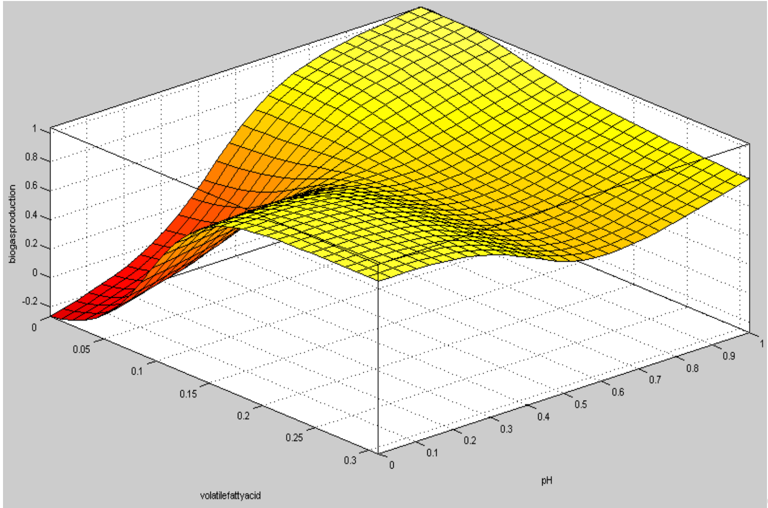
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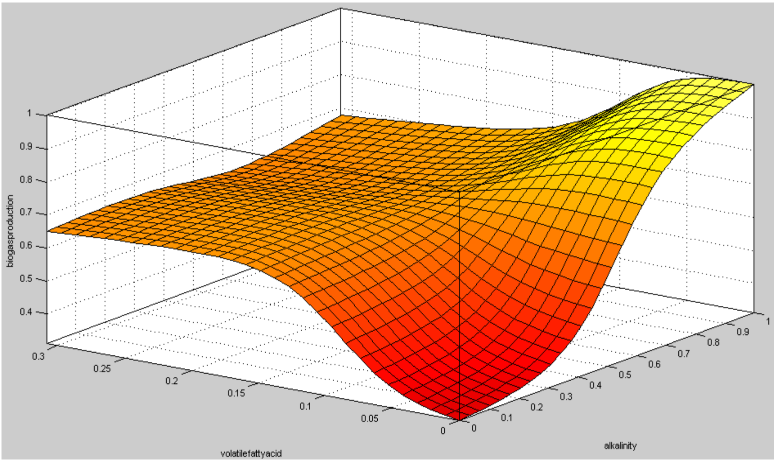
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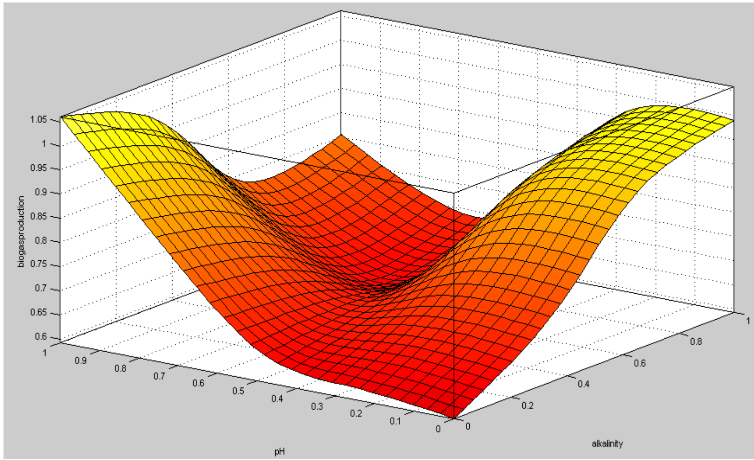
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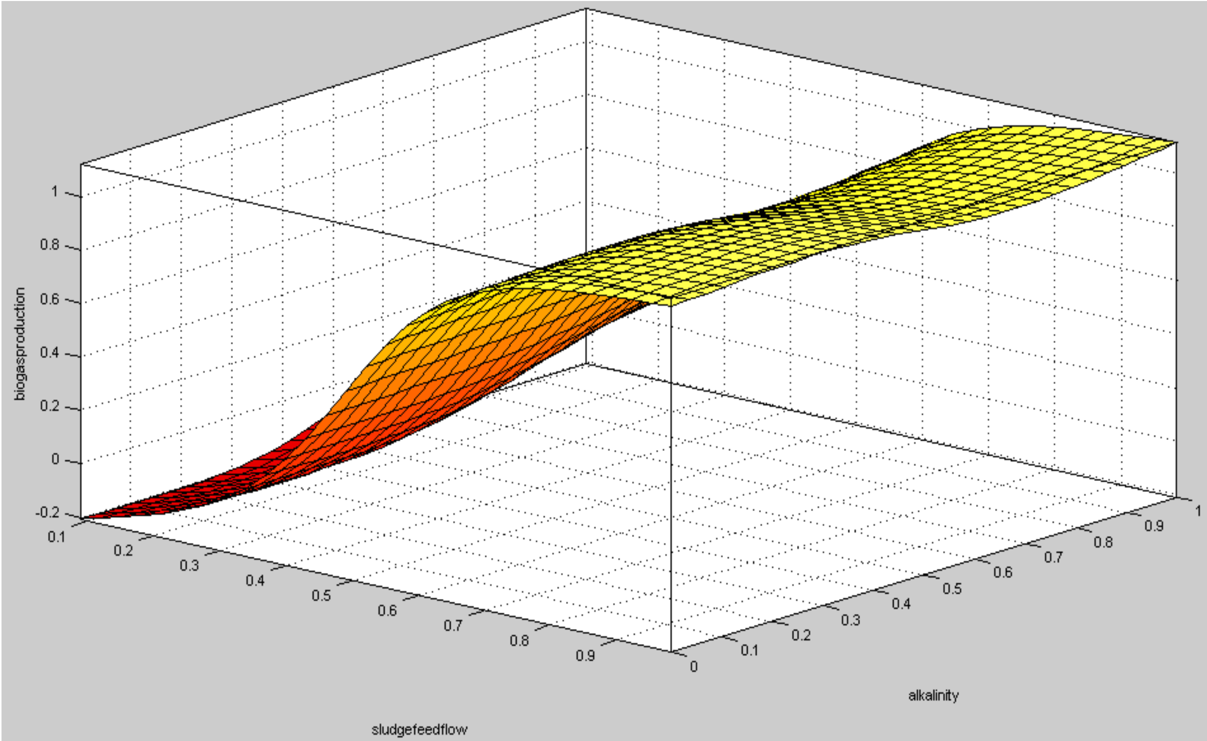
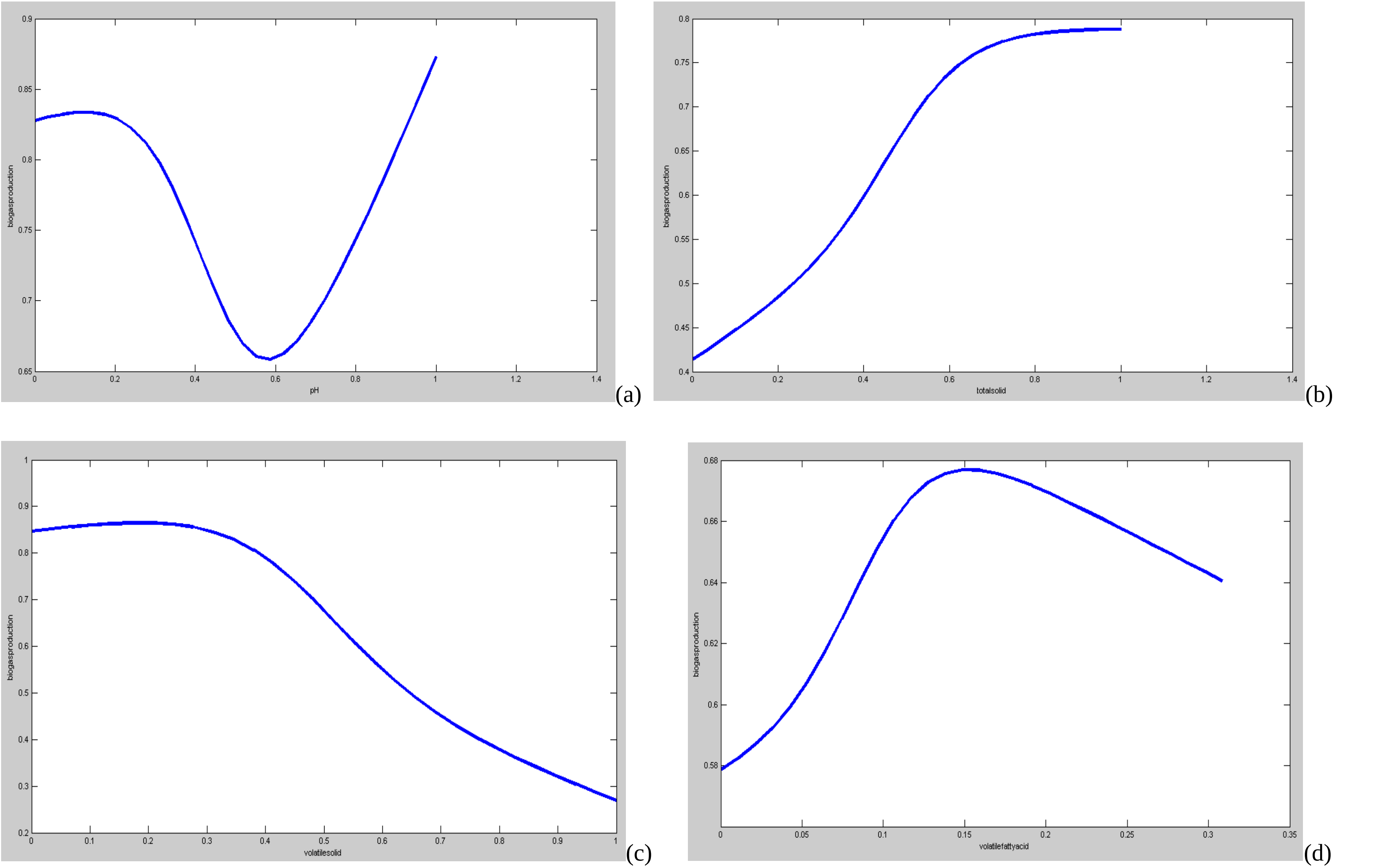


(m)

Figure 10.51 Surface responses for different combinations of input parameters to predict biogas production.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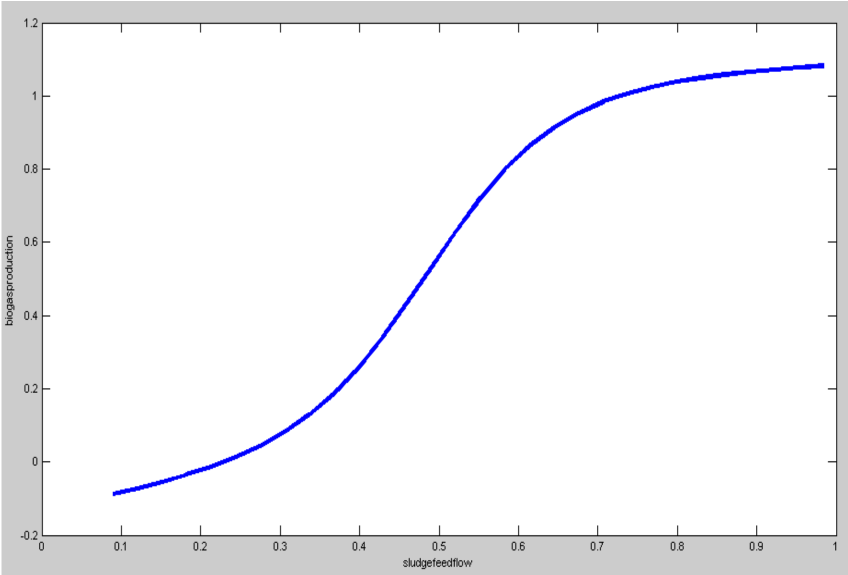
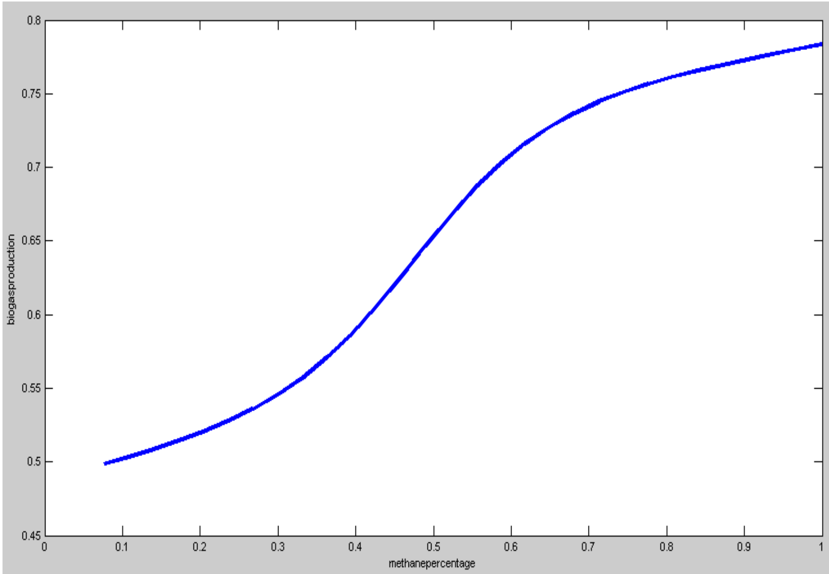


(e)



(f)

Figure 10.52 FIS model outputs for each input variable against biogas production

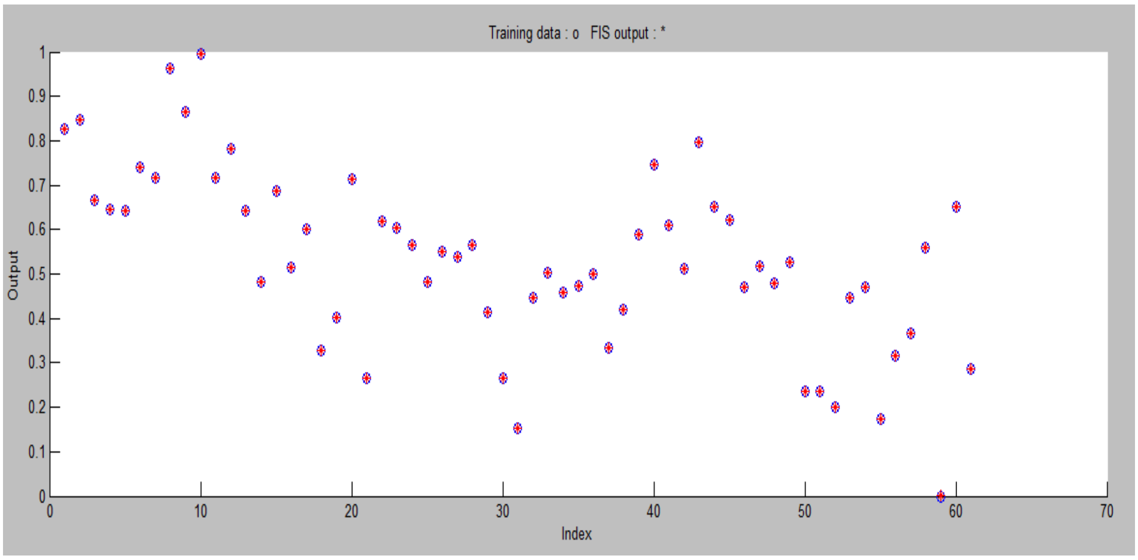


Figure 10.53 FIS based predictions with training data for seven inputs with biogas production as an output.

10.4 Conclusions.

ANFIS based predictions help to understand the impact of each key input parameter affecting anaerobic digestion process on output parameters like biogas production. In this chapter, historical data of BWWTP was used to train the model to make the predictions. The predictions based on the FIS model show that alkalinity, Sludge feed flow rate, pH and solid content of the sludge correlate well to biogas production. The optimum surface responses for alkalinity and VFA, pH and VFA and sludge feed flow and volatile solid content for optimum biogas production and higher methane quality were identified. The ANFIS structure that minimizes the error and makes predictions better was selected after many trials for each scenario. Sugeno-fuzzy model, hybrid training algorithm, gaussian type membership function and linear output variables were used for most of the ANFIS predictions. Increasing the size of data for the model development and prediction enhances the prediction power minimizing the error. Biogas production and anaerobic digester performance can be enhanced by monitoring and maintaining the key operational parameters.

CHAPTER 11

CONCLUSION AND RECOMMENDATION

11.1 Conclusion

The following general conclusions were made from the intensive experimental investigations on the effects of combined microwave-ultrasonic pretreatment and other operational parameters on the characteristics of sludge and the performance of the anaerobic digestion process.

- Combined microwave-ultrasonic pretreatment significantly enhanced sludge biodegradability, methane production, solid and COD removal at the optimum pretreatment and operating conditions compared to individual microwave and ultrasonic pretreatment techniques due to enhanced sludge solubilisation and higher degree of disintegration.

- The optimization study on microwave, ultrasonic and combined microwave-ultrasonic pretreatment has revealed that ultrasonication and microwave pretreatment power, intensity, density, duration of pretreatment and sludge concentration have significant impact on the performance of anaerobic digesters. Optimum Microwave and ultrasonic pretreatment durations are relatively shorter. The impact of this on the operational cost and feasibility of the technology is promising.

- The kinetics of pretreatment process shows that, sludge concentration, density and intensity of pretreatment and sludge pH have significant impact on the kinetics of the pretreatment process. The combined Pretreatment resulted in the improvement of biogas quality (CH_4/CO_2 ratio) and VS destruction during the digestion of all sludge types. The increase in digestion efficiency is proportional to the degree of sludge disintegration. Sludge disintegration and increased biodegradability is due to rapid internal heating of microwave radiation and the floc destruction achieved by ultrasonic treatment.

- FTIR bands for combined microwave-ultrasonic-treated digested sludge confirmed the increased polysaccharide, protein and fatty acid decomposition as compared to the other techniques. Microwave-treated sludge also showed a similar trend. The combination of microwave and ultrasonic pretreatment techniques did not result in direct additive effect. There is rather a complementary synergy between the two pretreatment techniques causing enhanced sludge disintegration, floc destruction, cell wall disruption and release of soluble organics. The floc structure and particle size were smaller in the combined microwave-ultrasonic pretreatment due to the cavitation effects and hydro mechanical shear forces which reduce floc size and enhance release of organics and radicals important to improve the biodegradability.

- The dewaterability slightly deteriorated in case of combined microwave-ultrasonic pretreatment due to greater size reduction and floc disruption higher percentage of fines causes compaction and increase in the amount of bound water trapped within solubilized organics and EPS. EAS also showed better dewaterability compared to other sludge types. Smaller sludge particles resulted in densification and higher resistance to the flow of water. The reduced dewaterability for combined microwave-ultrasonic pretreated sludge with smaller digested sludge particles confirms this. Reduction in particle size may also result in release of extracellular polymeric substances (biopolymers) may trap more bound water hindering the separation of water from the sludge during filtration.

- It can be understood that the anaerobic digestion enhancement is much greater when combined microwave–ultrasonic pretreatment is applied on Excess activated and thickened excess activated sludge.

- Moreover, separate pretreatment of TEAS before mixing with primary sludge resulted in substantial improvement in the biodegradability, solid reduction, methane production kinetics and biogas quality, protein removal, microbial destruction and overall performance of anaerobic digestion process. Besides, higher percentage of the pretreated TEAS increases the digestion kinetics, the methane production capacity and the biogas quality. The significance of the findings of this study in large scale wastewater treatment plants is enormous in terms of reducing the sludge treatment and handling costs. It will also help to enhance anaerobic digestion kinetics and overall performance.

- Organic loading rate for shorter HRT produced higher amount of methane for combined microwave-ultrasonic pretreated sludge compared to the untreated sludge. This confirms the availability of more readily available organics for microbial attach which helps to maintain higher biogas production and higher reduction of COD and VS even at a lower HRT.

- The predictions based on the FIS model show that alkalinity, Sludge feed flow rate, pH volatile fatty acid and solid content of the sludge correlate well to biogas production. The optimum surface responses for alkalinity and VFA, pH and VFA and sludge feed flow and volatile solid content for optimum biogas production and higher methane quality were identified. Sugeno-fuzzy model, hybrid training algorithm, gaussian type membership function and linear output variables were used for better predictions. Biogas production and anaerobic digester performance can be enhanced by monitoring and maintaining the key operational parameters and the ANFIS can be used as an intelligent tool to predict the optimum working conditions for a better and continuous process control.

11.2 Recommendations for further research

The outcome of this research has revealed the impacts of combined microwave-ultrasonic pretreatment on digester performance in comparison to digestion of microwave pretreated, ultrasonic pretreated or untreated sludge. The optimum pretreatment conditions were determined. Effects of pretreatment on anaerobic digestion of Primary, excess activated and mixed sludge on anaerobic digestion were compared. The optimum mixing ratios between untreated primary and untreated thickened excess activated sludge and treated activated sludge were determined. Model based prediction of operational parameters was performed using actual operational data from BWWTP. Based on the findings of this study the following research directions are recommended for further investigations.

11.2.1 Pilot scale experimental research and feasibility study on effect of combined microwave-ultrasonic pretreatment

It is recommended to duplicate the findings of this research on effects of combined microwave-ultrasonic pretreatment and other parameters affecting digester operation at a large pilot scale reactor. It is important to investigate the effects of SRT, OLR, pH and temperature on methane production potential, sludge biodegradability, solid reduction capacity, and process kinetics and sludge dewaterability at large scale. Digester performance analysis and sludge pretreatment study at large scale helps to perform cost-benefit analysis and predict the feasibility of the process at industrial or commercial scale.

Large scale anaerobic digestion study can be performed as a two stage process involving thermophilic acidogenesis and mesophilic methanogenesis to further enhance anaerobic digestion kinetics and performance. The collection of data and control of the process can be automated to enhance the process stability and methane production. A data logger can be attached to the thermophilic and mesophilic digesters to store data and monitor temperature, pH, organic loading rate and biogas quality. Periodic analysis of both liquid and gas samples will be performed after steady state is achieved. When the equilibrium sludge retention time anticipated is reached the characteristics of the digested sludge can be analysed and compared with that of the reactor feed sludge.

11.2.2 Simultaneous combined microwave-ultrasonic pretreatment on sludge disintegration and molecular mechanism.

The synergy between microwave and ultrasonic pretreatment calls for further research on sludge pretreatment where the microwave and ultrasonic pretreatment take place simultaneously and possibly using in-situ pretreatment techniques either on the digester feed pipelines or inside the anaerobic digesters. The combined pretreatment helped to enhance gas quality, process kinetics, solid and COD reduction capacity of the anaerobic digestion process. Simultaneous microwave-ultrasonic pretreatment would result in rapid cell disintegration effect reducing the pretreatment duration and cost.

Further Investigation is required on the opportunities to adapt combined microwave-ultrasonic pretreatment at large scale in technically and economically feasible manner.

Further in-depth analysis of the molecular mechanisms of microwave and ultrasonic pretreatment effects is required to clearly understand the underlying mechanisms of cell disintegration and biodegradation. Model based analysis of the kinetics at molecular level enables better optimization of the process and increases flexibility for application.

The decrease in dewaterability with increasing pretreatment time was one of the drawbacks of microwave-ultrasonic pretreatment. Further investigation to reduce the pretreatment time and density without compromising the sludge disintegration effects to enhance dewaterability is an important research direction. It was found out from this research that dewaterability can be enhanced when pretreatment is applied to a limited extent for a shorter duration.

11.2.3 Rheological and other characteristic investigation during pretreatment and anaerobic digestion

Further research can focus on the effects of pretreatment on sludge characteristics mainly rheology. It has been shown in this research that the rheological properties of sludge change with microwave and ultrasonic pretreatment conditions and during the anaerobic digestion process. Hence, investigating the rheological properties during pretreatment and in the course of the anaerobic digestion process helps to understand the flow and mixing behaviour. It is also interesting to investigate the effects of change in rheological properties of the sludge on digester performance for a pretreated sludge sample.

11.2.4 Application of ANFIS for continuous operational performance assessment of Wastewater treatment plants.

The modelling and sensitivity study based on ANFIS has revealed that optimum working conditions can be determined and the relationships between parameters can be understood using the plant historical data as an input. Further research can focus on development of adaptive model for continuous monitoring and control of operational parameters to optimize process parameters and maximize digester performance. The ANFIS model can also be applied to other unit processes in the wastewater treatment plant before and after the anaerobic digestion unit so that the digester performance can be enhanced by optimizing processes before and after the digester. The ANFIS can also be used to study the interaction between different operational parameters over a long historical period. Generally, the learning ability of ANFIS allows flexible application of the model for broader research in wastewater treatment plants. ANFIS modelling study can be coupled with sludge treatment cost minimization research in wastewater treatment plants.

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