Advancing High-Solids Anaerobic Digestion of the Organic Fraction of Municipal Solid Wastes

by

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Abstract

In the first phase, the major focus was on high-solids anaerobic digestion (HSAD) of the organic fraction of municipal solid waste (OFMSW) with percolate recirculation supported by conductive materials for enhancing biomethane recovery to address the kinetic limitation identified in previous studies. In this study, two identical lab-scale mesophilic (36^{o}C) HSAD systems were operated with OFMSW under similar operating conditions, including feedstock to inoculum ratio and daily percolate recirculation rate. The percolate tank of the test HSAD system was amended with 15 g/L of PAC. After 30 days of operation, the cumulative biomethane yield for the test reactor was 17% higher than the control reactor (109 vs. 93 L CH_4/kg VS). Furthermore, PAC addition showed additional benefits by lowering free ammonia nitrogen (FAN) and volatile fatty acids (VFAs) concentrations that can inhibit anaerobic digestion.

In the second phase, the effects of feedstock to inoculum (F:I) ratio and percolate recirculation time (PRT) were studied for the HSAD of OFMSW. Six mesophilic HSAD systems were operated at different F:I ratios (1 to 3 kg VS/kg VS; PRT=2.5 h/d) and PRTs (1.5 to 3.5 h/d; F:I=2). The F:I ratio of 1 provided up to 86% of the theoretical methane potential of OFMSW. In contrast, F:I ratio of 3 provided only 34% methane recovery due to VFAs accumulation and pH drop. Despite F:I ratio of 2 could provide 70% methane recovery, it could enable almost 45% higher organics processing capacity (VS basis) and lower solids washout during percolate recirculation, as compared to the F:I ratio of 1. However, different examined PRTs showed marginal impacts on methane yields with comparable changes in profiles of percolate characteristics.

Keywords: High-solids anaerobic digestion; Organic fraction of municipal solid waste; Powdered activated carbon; Feedstock to inoculum ratio; Percolate recirculation time; Methane

Preface

Chapter 1 and 2 include research background and literature review (i.e., on Biochemical Digestion of Organic Waste), in which fundamental concepts, technical terms, different technologies and operational concepts, as well as the relevant cutting-edge technologies and the state-of-the-art breakthroughs have been discussed and/or evaluated with respect to optimum operating conditions and high-quality/yield biomethane. The present review has also summarized and discussed the recent observations and results about the impact of operating parameters and feedstock characteristics on biochemical conversion rate, possible pretreatment/post-treatment options for higher biofuel quality and quantity, and their safe applications.

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Dedication

To my undergraduate's supervisor, Dr. Teimour Amani (rest in peace), who taught me abundant precious lessons and skills on how to live and work ethically and professionally...

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Table of Contents

List of Tables

List of Figures

Abbreviations and Units

AD	Anarobic digestion
Alk	Total alkalinity
C	Carbon
°C	Celsius
CH_4	Methane
CO_2	Carbon dioxide
cm	Centimeters
cm^3	Cubic centimeters
d	Day
DIET	Direct interspecies electron transfer
FAN	Free ammonium nitrogen, NH_3
F:I	Feedstock to Inoculum
g	Gram
GAC	Granular activated carobn
GHGs	Greenhouse gases
GJ/y^{-1}	Gigajoules per year
kg	Kilogram
h	Hour
H_2	Hydrogen
H_2S	Sulfric acid
HCl	Hydrocloric acid
HSAD	High-solids anaerobic digestion

L	Liter
LCFA	Long-chain fatty acids
IIET	Indirect interspecies electron transfer
LSAD	Low-solid anaerobic digestion; so-called wet-type AD
MC	Moisture content
MIET	Mediated interspecies electron transfer
min	Minute
mg/kg	Miligram per kilogeram
MJ/kg	Megajoules per kilogeram
mg/L	Milligrams per liter
mm	Milimeter
MSW	Municipal solid wastes
μm	Micrometers
N	Nitrogen
NaOH	Sodium hydroxide
NH_3	Free ammonia-nitrogen
NH_4-N	Ammonium-nitrogen
O_2	Oxygen
OFMSW	Organic fraction of municipal solid wastes
OLR	Organic loading rate
PAC	Powdered activated carbon
ppm	Milligrams per liter
s	Second
S	Sulfur

SCOD	Dissolved chemical oxygen demand
SCFA	Short-chain fatty acids
T	Temperature
TAN	Total ammonium-nitrogen, NH_4-N
ton	Tonne
TS	Total solids
TSS	Total suspended solids
VFA	Volatile fatty acids
VS	Valotile solids
VSS	Total volatile suspended solids
wt.%	Weight percentage
y	Year

Chapter 1

Background

1.1. Introduction

Nowadays, establishing a widespread sustainable food waste management is accentuated as an urgent global act to mitigate the rising environmental pollution and excessive financial costs due to the high-load disposal of food waste (Pagliaccia et al., 2019). Nowadays, the proper management of municipal solid waste (MSW) is still one of the most demanding tasks in various countries (such India, China, and other developing countries), which is due to rapid urbanization and population growth and excessive production of organic wastes such as food waste, lignocellulosic residuals and sewage sludge (Kondusamy and Kalamdhad, 2014; Dasgupta and Chandel, 2019). One of the major portions of solid waste is food waste, which is more than paper or plastic. To be specific, up to one-third of food is wasted globally, so that the annual amount of food waste accounted for 1.3 billion tons in 2014 (Kondusamy and Kalamdhad, 2014; FAO, 2015). The rising trend of word's population and urbanization increases the annual MSW generation from ~2.0 billion tons (in 2016) to 3.4 billion tons (in 2050) (Dasgupta and Chandel, 2019; Hoornweg and Bhada-Tata 2012; WorldBank 2017, 2018), thereby requiring further processing or transfer of MSW into landfills. According to literature, dumping or landfilling of MSW is the widely practiced and cheapest approach throughout the globe (Hodge et al., 2016). However, owing to the perpetual menace of environmental pollution and posing risks to public health (WHO, 2018), various rules pertaining to efficient MSW management have been legislated to urges the authorities to only landfill the non-usable inert waste (Joshi and Ahmed, 2016; The-European-Union-European-Union-Landfill-Directive, 1999). Apart from the recycling and reuse of source separated MSW, biochemical and thermochemical processes have been recommended as promising technologies to valorize the organic fraction of municipal solid waste (OFMSW) into various biofuels and value-added chemicals (Kumar and Samadder, 2020; Zhang et al., 2019), and to reduce the mass of waste (Guilford et al., 2019; Rezaee et al., 2020). Nevertheless, the definition and classification of OFMSW alter regionally and nationally. In the European Union, OFMSW (the combination of food waste, garden and park wastes) (Al Seadi et al., 2013) is distinguished from the source selected

food waste, which is specifically based on the time elapsed between the production and the collection of each classification (Alibardi and Cossu, 2015).

Generally, MSW with high contents of biodegradable substance and moisture have an invaluable potential of being considered as a viable feedstock for anaerobic digestion (AD) process and sustainable renewable recovery (Kalyani and Pandey, 2014; Rozzi and Remigi, 2004). In fact, AD has been enumerated as one of the best environmentally-friendly alternatives for the valorization and management of organic wastes (Pagliaccia et al., 2019; Rozzi and Remigi, 2004). The AD process employs a diverse set of microorganisms for the decomposition of organic compounds and converting them into renewable biomethane (Rozzi and Remigi, 2004). Notably, the AD of organic compounds comprises a series of biochemical reactions, including hydrolysis, acidification, acetogenesis, and methanogenesis (Ai et al., 2018; Elyasi et al., 2015). Among the aforementioned steps, hydrolysis is always considered as the rate-limiting stage in the AD process for complex substrates, while methanogenesis can be the rate-limiting step for easily biodegradable substances (Rozzi and Remigi, 2004; Gunaseelan, 2004). Thermophilic condition possesses some advantages, including higher solubilization of the organic compounds, higher rates of biological and chemical reaction, and further death rate of pathogens, however the mesophilic temperature regime is dominant in most large-scale AD plants owing to higher process stability and lower energy consumption (Appels et al., 2008; Khalid et al., 2011; Nielsen and Petersen, 2000). Reportedly, a combination of AD and landfill (with the potential of the recovery and reuse of both biomethane and digestate as an alternative for chemical fertilizer) demonstrated a better performance in terms of mitigating global warming, as it was compared to other approaches such as landfilling alone, composting and combustion with energy recovery (Hodge et al., 2016). However, unlike landfills, the higher cost of AD used from the processing solid organic waste has been restricted its widespread application. In fact, the higher cost of conventional low-solid anaerobic digestion (LSAD) process is attributed to the complex, heterogeneous and variable characteristics of the feedstock requiring expensive pretreatment, including sorting, shredding, decontaminating and adding water, before undergoing conventional AD system (De Baere and Mattheeuws, 2013; Guilford, 2009). These disadvantages of LSAD have led to its bare application in North America, with only 58 digesters in the U.S. and around 15 digesters in Canada without any national database (Pennington, 2018).

Various low- and high-solid AD (HSAD) processes have been studied for the biomethanation of OFMSW, which despite the advantages and disadvantages of each one, they generally require a lengthy operational retention time ranging from 20 to 40 days (Shahriari et al., 2012; Pennington, 2018; Fagbohungbe et al., 2015). Additionally, mesophilic (30 °C–40 °C) and thermophilic (45 °C–60 °C) conditions are the common operating temperature of AD process, which influences the following factors: (i) the growth rate, metabolism and population dynamics of microorganisms in the anaerobic digesters, as well as (ii) the physicochemical properties of the substrate components (Appels et al., 2008). Apart from the abovementioned influential operating parameters, implementing AD process have encountered a number of hurdles and high operating costs, which is linked to heterogeneous physical characteristics and complex chemical composition of OFMSW, as well as requiring different pretreatment approaches prior to operating AD process (Zheng et al., 2014; De Baere and Mattheeuws, 2013; Guilford, 2009). Mostly, it is rather tough to report a specific composition analysis for representing a taken sample due to the heterogeneous nature of OFMSW. However, the composition of the biodegradable fraction of MSW could be considered, in order to provide a reliable baseline for comparison between experiments and make clear conclusions (Dasgupta and Chandel, 2019).

In order to overcome these shortages associated with HSAD (i.e., lower biomethane yield and longer digestion time), the feedstock is processed under different operational conditions, such as organic loading rate (OLR), feedstock to inoculum ratio (F:I), percolate recirculation time (PRT), addition of conductive materials, and implementing different pretreatment methods, like mechanical, chemical, biochemical and thermal approaches (Atelge et al., 2020; Rocamora et al., 2020; Phuttaro et al., 2019; Elalami et al., 2020; Zhao et al., 2019). These approaches and operating conditions are to effectively improve the characteristics of substrate and amplify hydrolysis rate during AD processes. For example, some recent studies have investigated the impact of different conductive materials, such as powdered activated carbon (PAC), granular activated carbon (GAC), biochar, carbon cloth, etc., on the performance of conventional LSAD systems operating under mesophilic and/or thermophilic AD of food waste under batch, semi-continuous, and continuous operations (Zhang et al., 2017; Ryue et al., 2019; Xu et al. 2015). Conductive materials have demonstrated a positive impact on escalating the performance of the AD processes, which in fact, originates from improving the microbial communities' structure and abundance within the AD reactor, as well as accelerating its kinetic rates by improving syntrophic interactions between

bacterial and archaeal communities (Pan et al., 2020; Park et al., 2018). These facts promote the AD mechanisms towards further degradation of organic fractions and conversion of intermediate products, thereby further biomethane production and yield (Chowdhury et al., 2019; Wu et al., 2019; Yan et al., 2018).

1.2. Scope and Objective

The scope of this thesis was focused on high-solids anaerobic digestion (HSAD) of the organic fraction of municipal solid waste (OFMSW). The overall research goal was to investigate the impacts of influential operating conditions on mesophilic AD of OFMSW in promoting HSAD reaction rate and biomethane yield, which is intrinsically a slow and low-yield process due to the nature of operation and feedstock characteristics. Two specific objectives of this study were:

a) To investigate the effects of conductive material (powdered activated carbon; PAC) added to percolate on mesophilic HSAD of OFMSW.

b) To study the influence of feedstock to inoculum (F:I) ratio and percolate recirculation time (PRT) on the degradation of OFMSW and biomethane yields.

Two bench-scale research were conducted to figure out the effects of PAC, F:I ratio and PRT on (i) kinetics and biomethane production yield and, (ii) accumulation of volatile fatty acids (VFAs), (iii) variation of other influential parameters, such as pH, alkalinity, total and free ammonia-nitrogen, and VFA/Alkalinity ratio.

1.3. Thesis Outline

This thesis is divided into five chapters. In Chapter 1 (present chapter) the background of the investigated topic is highlighted and specific objectives of the proposed research is summarized. In Chapter 2, a holistic literature review related to the proposed research is scrutinized. Afterward, the experimental findings from this thesis are presented in Chapter 3 and Chapter 4 in research article format. Finally, in Chapter 5, the summary of significant results, their scientific and engineering implications, along with future outlook (recommendations) are discussed.

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Chapter 2

Literature Review

2.1. Introduction

2.1.1. Feedstock Characteristics

Lignocellulose is commonly present in agricultural residues, forest residues, energy crops, OFMSW, and food waste (Romero-Cedillo et al., 2017). Lignocellulosic fraction of OFMSW, such as garden waste, contains three polymeric components with the following general ratios: 30%–50% of cellulose, 20%–40% of hemicellulose, and 25%–35% of lignin (dry-wt.), so that the first two, i.e., cellulose and hemicellulose, are enumerated as a source of carbohydrates in the feedstock (Kabir and Hameed, 2017; Bhange et al., 2015). Lignin, which is known as a non-biodegradable or un-fermentable fraction of the biomasses, rigidly encompasses hemicellulose and cellulose by its randomly cross-linked phenylpropanoid units. These sophisticated arrangements and strong bonds have made cellulose to unreachable by biochemical microorganisms and thermochemical reaction media (Huang et al., 2012; Hadar, 2013; Geng, 2013). Therefore, unlike starch containing wastes, which can easily be hydrolyzed into its carbohydrate components, other organic wastes containing higher contents of cellulose, hemicellulose and lignin require further pretreatment in order to reduce the lignin matrix and enhance subsequent microbial breakdown (Zheng et al., 2014).

Over recent decades, various biomass pretreatment approaches have been implemented, in order to tackle the challenges associated with the complex and rigid structure of lignocellulosic biomasses resulted from their high lignin contents, thereby (i) facilitating the enzymatic hydrolysis rate during the AD process (Carlsson et al., 2012; Loow et al., 2016), and (ii) promoting the production of suitable and useful products and value-added chemicals (Neyens et al., 2003; Loow et al., 2016; Braguglia et al., 2018). To date, various pretreatment methods such as physical (Nuruddin et al., 2016), chemical (Wang et al., 2017), biochemical (Rouches, et al., 2016) and hydrothermal pretreatment (Kim et al., 2016), have been studied to break down the different types of organic and/or lignocellulosic substrates. Moreover, green ionic liquids and eutectic solvents have been suggested as novel complementary pretreatment used for the fractionation of

lignocellulosic structure and/or the removal/stabilization of distracting contaminants (like heavy metals). Consequently, these methods streamline hydrolysis/saccharification and thermochemical conversions, thereby producing biofuels with higher yield and quality (Dastyar et al., 2019; Xu et al., 2016; Zhang et al., 2016; Elgharbawy et al., 2016). However, this arena of pretreatment investigations has been developed over the past decade to figure out their impact on the yield and quality of various biofuel products. In this research, feedstock is introduced directly to digester after a mechanical screening and grinding to particle size lower than 3 inches without ant further focus on the impact of pretreatment of OFMSW.

2.2. Operating Conditions

As previously indicated in Chapter 1, various LSAD and HSAD processes have been investigated for the AD of OFMSW (Shahriari et al., 2012; Pennington, 2018; Fagbohungbe et al., 2015). Each approach possesses its specific operational conditions, along with its advantages and disadvantages, which have been listed in Table 2.1 (Shahriari et al., 2012; Pennington, 2018; Fagbohungbe et al., 2015).

As can be seen in Table 2.1, unlike LSAD, the main advantages of HSAD are as follows: (i) higher OLR, (ii) lower water consumption, (iii) less energy demand for heating smaller reactor, (iv) high-quality digestate production (due to its lower loss of nutrients content), and (v) no need of technologies for dewatering digestate. However, this approach has some shortages and limitations that are required to be addressed in the future investigation as a major breakthrough to promote the performance of HSAD. As compared to LSAD, some of the drawbacks are (i) lower biogas production (Table 2.2), (ii) lower VS and TS removals, (iii) slower kinetic rate, (vi) tougher and demanding operational conditions like loading and unloading the solid, (v) demanding procedure for recycling biogas and leachate due to clogging issues (Fagbohungbe et al., 2015, Bi et al., 2020; Di Maria, et al., 2017).

As can be seen in Table 2.2, despite a lower methane yield obtained from HSAD of a feedstock, the methane yield per unit volume of the digester ranges 2 to 10 times higher than LSAD (Rocamora et al., 2020). HSAD processes operate with different organic solid wastes at higher TS contents than typical LSAD. Unlike LSAD, the HSAD provides treatment of higher quantities of organic waste per unit volume of digester. However, HSAD produces a lower biomethane

production yield per unit mass of VS, as compared to LSAD (Table 2.2). Furthermore, other problems associated with HSAD are the accumulation of inhibitors (VFA, TAN, FAN, pH, and so on), which are directly associated with the higher solids content and higher F:I ratios in the system without proper mixing causing the formation of hot-spot within digester (Rocamora et al., 2020).

Table 2.1. LSAD and HSAD processes: A short comparison (Fagbohungbe et al., 2015).

Factor	LSAD	HSAD
Total solid content	<10%	10% - 40%
Operational type	Single and multi-stage AD	Single and multi-stage AD
Feeding method	Semi and continuous	Batch, sequential batch, semi and continuous
Biogas feature	High biogas production with high moisture content	Low biogas production, but low moisture
Volatile solid removal	50% - 70%	< 40%
Feedstock loading rate	<7 kg VS/m^3.d	7-15 kg VS/m^3.d
Inhibitory factors	More dispersion and diffusion	Less dispersion and high adsorption into substrate
Mixing regime	Internal mixing device, or liquor and biogas recirculation	Mixing by recirculating percolate and biogas, or by partial mixing
Heating condition	Requiring high heating due to large digester volume	Requiring low heating due to smaller digester volume
Operational issues	Requiring less sophisticated pumping device, due to higher water or moisture	Requiring complex pumping equipment due to dry nature of operation
Feedstock requirement	Not appropriate for hydrophobic feedstock like lignocellulosic biomasses	Most proper for hydrophobic organic biomasses
Digestate management	Requiring considerable dewatering of digestate	Minimal dewatering is needed
Digestate characteristic	Less stable but with higher nutrient content	More stable but with lower nutrient content

Moreover, the lower water content in HSAD is considered as a key cause for less gas and liquid diffusion within the system, thus reducing feedstock availability to the inoculum microorganisms, lessening their metabolism, and causing inhibitors accumulation (Ge et al., 2016; Visvanathan, 2010). As reported in the literature, an increased water content or percolate recirculation can enhance biomethane yields, which is attributed to the provision of a better homogenized substrate, consequently enhancing mass transfer (i.e., diffusion) of intermediate

compounds, improving microorganisms and nutrients interactions, as well as diluting potential inhibitors (Fagbohungbe et al., 2015, Rocamora et al., 2020; Forster-Carneiro et al., 2008; Kusch et al., 2008).

Table 2.2. Biomethane yields obtained from feedstocks processed by LSAD and HSAD.

Feedstock Type	Digestion mode	OLR (kg VS/m^3.d)	Biomethane yield (m^3/kg $VS_{Feedstock}$)	Biomethane yield (m^3/m^3 Digester)	Reference
OFMSW	LSAD	30.0	0.20	6.0	(Di Maria et al., 2017)
	HSAD	90.0	0.14	12.2	
Wheat straw	LSAD	14.5	0.14	2.0	(Brown et al., 2012)
	HSAD	106.1	0.12	12.7	
Chicken manure	LSAD	1.8	0.35	0.1	(Bi et al., 2020)
	HSAD	5.3	0.18	1.0	

In regard to operating batch HSAD of organic solid waste, some critical process parameters have been reported, including F:I ratio, inoculum fraction, percolate recirculation, temperature, and bed characteristics (such as porosity, permeability and field capacity of substrate within digester). Briefly, F:I ratio and temperature are considered as a defining factor for commencing operation and provide the essential microbial community to the new fresh feedstock (Chen et al., 2008; Di Maria et al., 2013). However, percolate recirculation and bed structure have been reported as other influential parameters enhancing waste content and homogeneity (Fagbohungbe et al., 2015; Rocamora et al., 2020).

There are different sources that can be employed as the inoculum, including sewage sludge from wastewater treatment plants, digested manure or previous digestate from AD reactors. However, the characteristics of digestate vary with the source, time, and operating conditions (Karthikeyan and Visvanathan, 2013). As discussed elsewhere, a lower F:I ratio (i.e., higher inoculum content) could potentially lead to decreasing retention time and increasing biogas yield (Forster-Carneiro et al., 2008; Lopes et al., 2004; Liu et al., 2009). In the contrary, a higher F:I ratio (i.e., reducing inoculum content), despite increasing waste treatment capacity, causes longer

retention times and lower biomethane production yields owing to lower availability of microorganisms to interact with feedstock, the accumulation of VFA, TAN, FAN, as well as increase of VFA/alkalinity and drop of pH (Fagbohungbe et al., 2015, Rocamora et al., 2020). Despite the vital role of the F:I ratio in HSAD systems, there is neither a specific nor a general optimum ratio. This is attributed to the differences in the composition of each feedstock, solid and liquid inoculum, as well as operating conditions and the system type. Nonetheless, some authors and technology suppliers have suggested some F:I ratio values used for operating their designed systems. For example, the Bekon company (Harsewinkel, Germany) has recommended using F:I ratio of 1 (total weight) while reusing 50% of the recycled digestate as inoculum in their garage-type digester to start a new cycle (Karthikeyan and Visvanathan, 2013). Similarly, Di Maria et al. (2012) recommended an F:I ratio between 1.5 to 2.5 (total weight) for a large-scale HSAD of food waste equipped with percolate recirculation. Kusch et al. (2008) and Hashimoto (1989) reported 20%, 50% and 34% inoculum for other substrates, inoculum, and operating conditions, respectively.

In previous literature, percolate recirculation has been reported as a common solution to reduce the use of solid inoculum and enhance moisture content within the system (Rocamora et al., 2020; Pezzolla et al., 2017). Reportedly, percolate recirculation can enhance the performance of HSAD and biomethane production yield, which results from greater reactor homogeneity, further contact between the methanogenic archaea and nutrients, as well as washing adverse impact of the inhibitors formed in the HSAD, including VFA, TAN, FAN, etc. (Rocamora et al., 2020; Pezzolla et al., 2017). For example, by employing mature leachate as liquid inoculum, the solid inoculum content in a HSAD system could be decreased from 40% to 10% or completely avoided, without methane loss (Wilson et al., 2016; Kusch et al., 2008). Having said that, operating HSAD with higher solid inoculum and percolate recirculation shortens operating times required to attain the ultimate biomethane yield and elevate final products (Chan et al., 2002; Benbelkacem et al., 2010). According to these literatures, an increase of ~400% and 60% in biomethane production have been observed with a shorter time to obtain the maximum gas production rate with percolate was recirculated during different feedstock such as MSW, sewage sludge and marine dredging, etc. (Chan et al., 2002; Benbelkacem et al., 2010). Different strategies, i.e., continuous and intermittent schemes, have been reported for percolate recirculation, which can be found elsewhere (Rocamora et al., 2020), so that all of them census on that percolate

recirculation with higher frequencies improves biomethane production yield, as compared to no recirculation. Yet, some negative impacts have been reported when percolate recirculation time or frequency exceeded an excessive value (Chen et al., 2008; Sponza and Ağdağ, 2004), especially when dealing with feedstock with low C/N ratios, i.e., nitrogen-rich feedstock like OFMSW or food waste.

Therefore, the major goal of future studies should be focused on the following items: 1) enhancing the process performance in terms of kinetic rate towards increasing biomethane production yield, 2) effective pretreatment and further degradation of feedstock, 3) combing different thermochemical and biochemical processes to address other shortages, thereby promoting overall outcome, and 4) enhancing process performance by tailoring liquid inoculum (percolate) by its effective pretreatment, conditioning and recirculation (Rocamora et al., 2020; Guilford et al., 2019; Fagbohungbe et al., 2015).

2.3. HSAD Technologies

To address the shortages associated with HSAD, various single- and multi-stage technologies have been operated. A list of reported technologies and set-ups has been illustrated in Figure 2.1. As can be seen and is discussed in the following sections, in some of these processes, various techniques have been employed to tackle the lack of proper mixing by recirculation of biogas and/or percolate obtained from the digester to the bottom and top of the feedstock-inoculum blended medium, respectively, to enhance substrate homogenization and biogas mobilization (Rocamora et al., 2020; Guilford et al., 2019; Fagbohungbe et al., 2015; Wilson et al., 2016). As can be observed in Figure 2.1, Table 2.3 and Table 2.4, these tabulated single- and multi-stage systems are operated with dissimilar TS contents (20% to 60%), mixing/recirculation regimes, OLR (up to 15 kg VS/m^3.d), operation conditions, set-up arrangements depending on feedstock type and operating temperature.

According to the literature, currently, the garage-type reactor (i.e., German rectangular digester with percolate recirculation) is one of the commercially available batch systems available in the market (Rocamora et al., 2020; Fagbohungbe et al., 2015). In these digesters equipped with percolate recirculation, the percolate channels through the mixed dense substrate and is collected in a drainage tank placed at the bottom of the AD systems, then is recirculated and sprayed through

the top of the substrate within the system based on a defined schedule as needed. The description of a single-stage HSAD systems, along with their pros and cons, have been listed in Table 2.3.

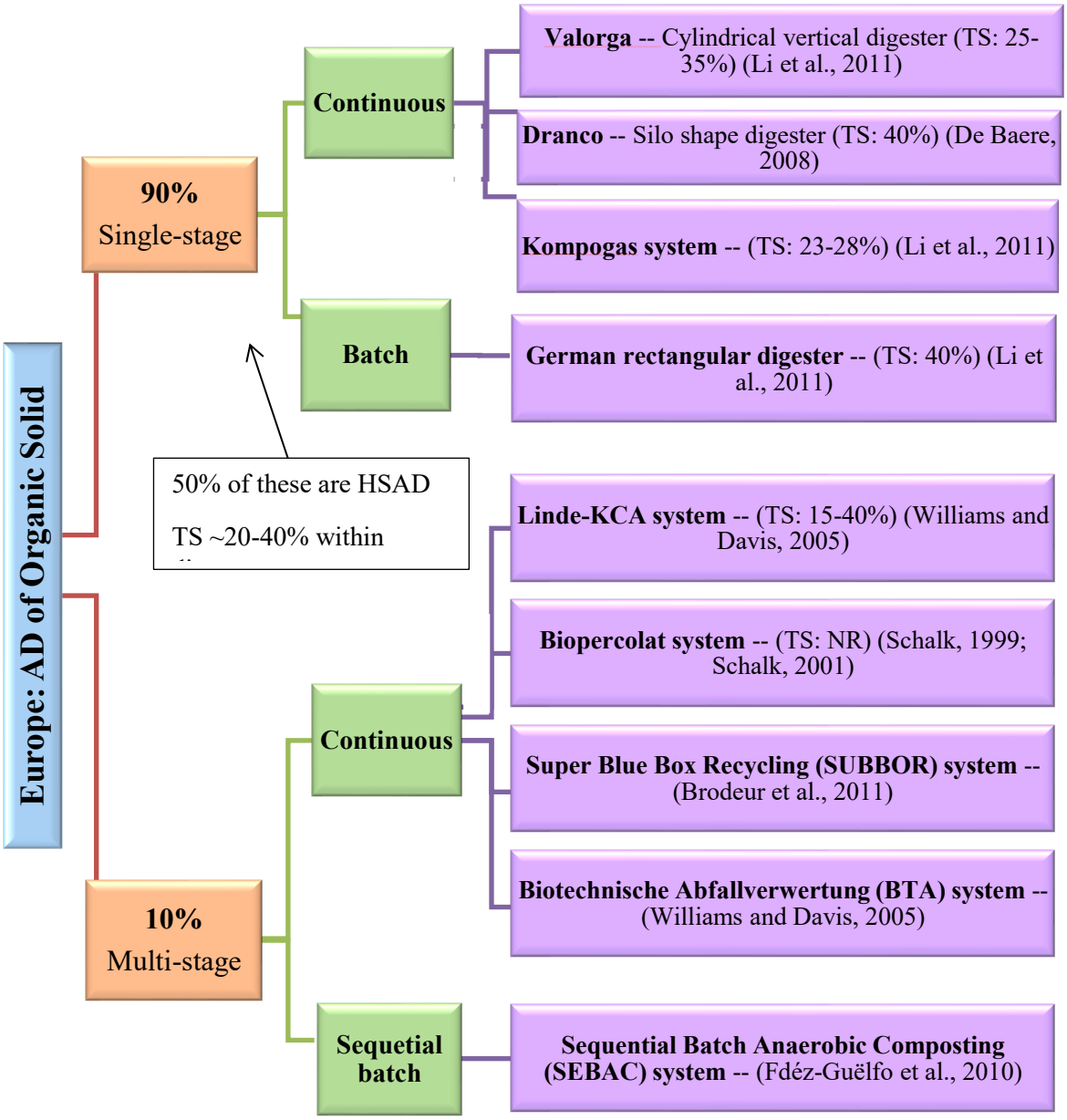

Figure 2.1. Different HSAD technologies (Fagbohungbe et al., 2015).

Table 2.3. Conventional single stage HSAD systems along with their pros and cons.

Technology	Description	Pros	Cons
Valorga system (Li et al., 2011)	- A cylindrical vertical digester - Recirculation high pressure biogas through nozzle	- Employing vertical wall for enhancing circular flow of feedstock - Biogas recirculation prevents separation of phases within the digester and improves the distribution of intermediates	- High energy required for pressurizing the biogas - High tendency for the blockage of biogas nozzles by organic particles
Dranco system (De Baere, 2008)	- Vertical silo shaped reactor without recirculation - Employs a mixing unit to combine fresh feedstock and recycled digestate (ratio of 1:6)	- Avoiding excessive energy input and clogging of nozzles - Lack of intermittent mixing using biogas/leachate	- Limited quantity of fresh substrate added per cycle - Lower efficiency resulted from the high volume of recycled digestate
Kompogas system (Li et al., 2011)	- Modified design of low solid horizontal plug flow digester with a slowly rotating internal axial mixer	- Keeping dense solids in suspension - Increasing feedstock and microorganisms contact	- Limited quantity of fresh substrate added per cycle
German rectangular digester (Li et al., 2011; Berge et al., 2009)	- A batch process with intermittent leachate recirculation	- Recirculation of leachate to improve performance - Possibility of mixing fresh feedstock and recycled digestate - Limited value for commercial usage	- Improper distribution mixing feedstock and solid/liquid inoculum - Uneven biodegradation of feedstock - Interrupted methane production due to unloading/reloading the digester

As for Table 2.3, the recirculation approach in the above-mentioned single-stage HSAD systems can be summarized as follows:

- ❖ The Valorga recirculates pressurized biogas (but not leachate and digestate)
- ❖ The Dranco system reuses digestate (but not leachate and biogas)
- ❖ The Kompogas system reuses digestate (but not the leachate or digester)
- ❖ The German rectangular digester recirculates leachate and reuses digestate (but not biogas).

However, typically mechanical mixing is not a substantial operating characteristic of HSAD, in some of the above-mentioned HSAD systems (Figure 2.1) different ways of mixing have been employed, either mechanical or using biogas/biomethane recirculation. As a conclusion on the mixing approach used in these systems, it can be seen due to the hydrodynamic and rheological properties of the solid, liquid and gas within the digesters, there is a correlation between the type of internal mixing equipment and the mixing approach:

- ❖ Typical axial mixer → TS up to 28% (e.g., Kompogas system)
- ❖ Pressurized recirculating biogas → TS up to 35% (e.g., Valorga system)
- ❖ Without internal mixers → TS ~ 40% (e.g., Dranco and German rectangular systems)
- ❖ With leachate recirculation → almost no limit for TS loading (e.g., German rectangular system)

To address some of the limitation and operating shortages of single stage HSAD systems (see in Figure 2.1 and Table 2.3), various successful multi-stage AD systems have been operated over the last couple of decades (in Germany, Japan, and Canada), which have been summarized in Table 2.4 (Vandevivere et al., 2003; Fagbohungbe et al., 2015). However, owing to the high expenses required for construction, operation, and maintenance of these multi-stage HSAD systems, few of them have been commercially developed.

Table 2.4. Novel multi-stages HSAD systems along with their pros and cons.

Technology	Description	Pros	Cons
Sequential Batch Anaerobic Composting (SEBAC) system (Chynoweth et al., 1991; Fdéz-Guëlfo et al. 2010) (Figure 2.2)			
- Two stages: AD + AD - Sequential addition of feedstocks and continuous spraying mature leachate until biomethane production stabilizes in the new batch reactor.		- Minimizing the constraint of mixing and handling high solid feedstocks	- Long period of startup and stabilization (at least 110 days)
Linde-KCA system (Williams and Davis, 2005; Curtis, 2010) (Figure 2.3)			
- Two stages: Aerobic + AD - The anaerobic system performs like a plug flow digester with axial mixers		- Continuous treatment of feedstock with 15% - 40% TS content	- The aerobic pre-treatment stage can adversely lower the energy content of the substrate
Biopercolat system (Garcia and Schalk, 1999; Schalk, 2001) (Figure 2.4)			
- Two stages: Aerobic + AD - Partial aeration of feedstock in the aerobic stage		- Convenient operation and processing of the separated liquid fraction derived from homogenized feedstock	- The aerobic pretreatment adversely affects the energy value of the feedstock, thereby reducing biomethane output.
Super Blue Box Recycling (SUBBOR) (Brodeur et al., 2011; Vogt et al., 2002) (Figure 2.5)			
- Three stages: Hydrolysis + Steam + AD - Steam explosion with high pressure (55–63 bar) breaks down feedstock's complex structure, thereby enhancing microbial fermentation		- Shorter retention time resulted from steam explosion reaction - Lack of adverse impact on the energy value of feedstock	- Additional energy required for the steam explosion - Potentially less sustainable for small-scale AD operators
Biotechnische Abfallverwertung (BTA) system (Chavez-Vazquez and Bagley, 2002; Williams and Davis, 2005) (Figure 2.6)			
- Multi stages: Physical + Physical + Hydrolysis + AD - Solid feedstock is blended with pre-treated percolate. Afterwards, it is pumped into a hydrolysis container, followed by AD process		- Data from a Canadian installation showed that VS reduction of between 70% and 75%.	- Poor degradation of the OFMSW.

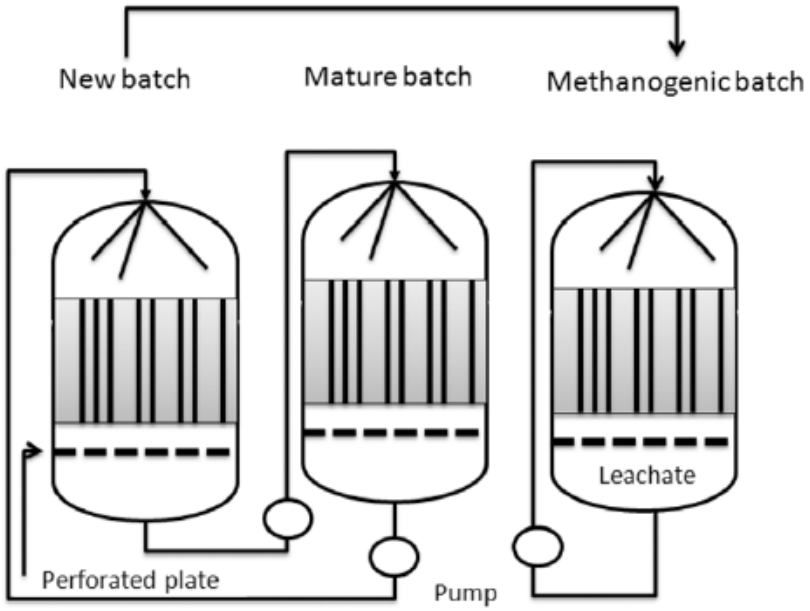

Figure 2.2. Sequential Batch Anaerobic Composting (SEBAC) process diagram.

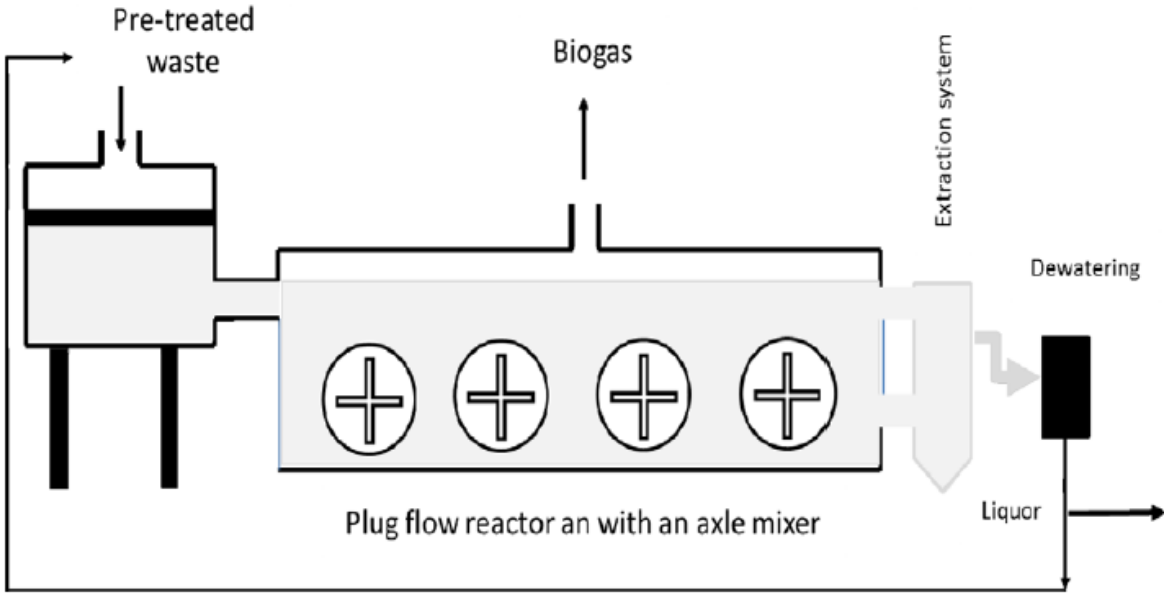

Figure 2.3. Linde-KCA two-stage HSAD.

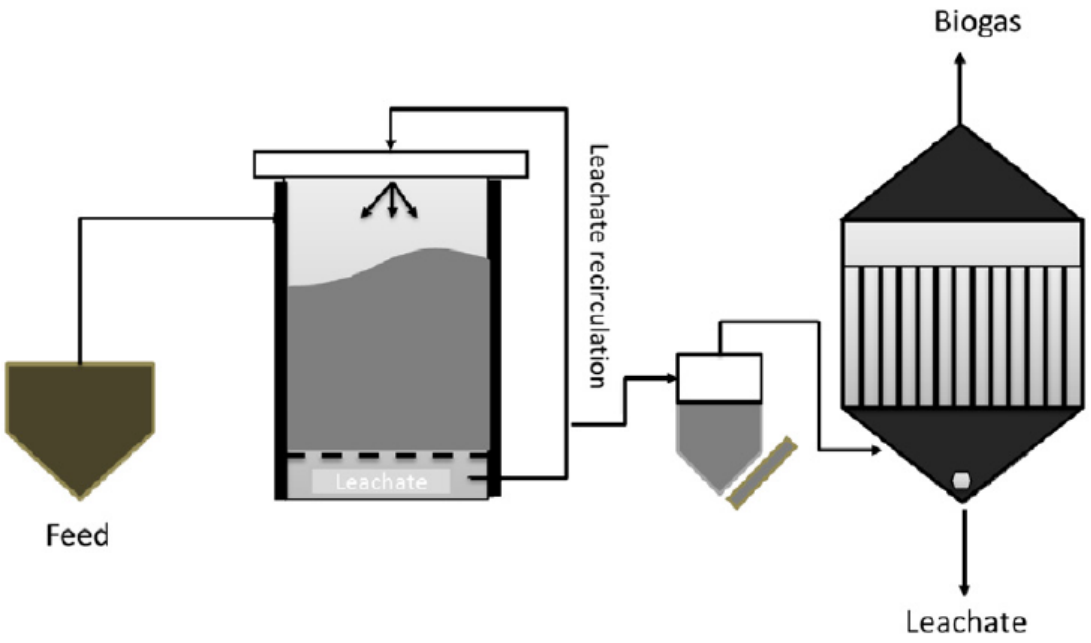

Figure 2.4. Two-stage Biopercolat process diagram.

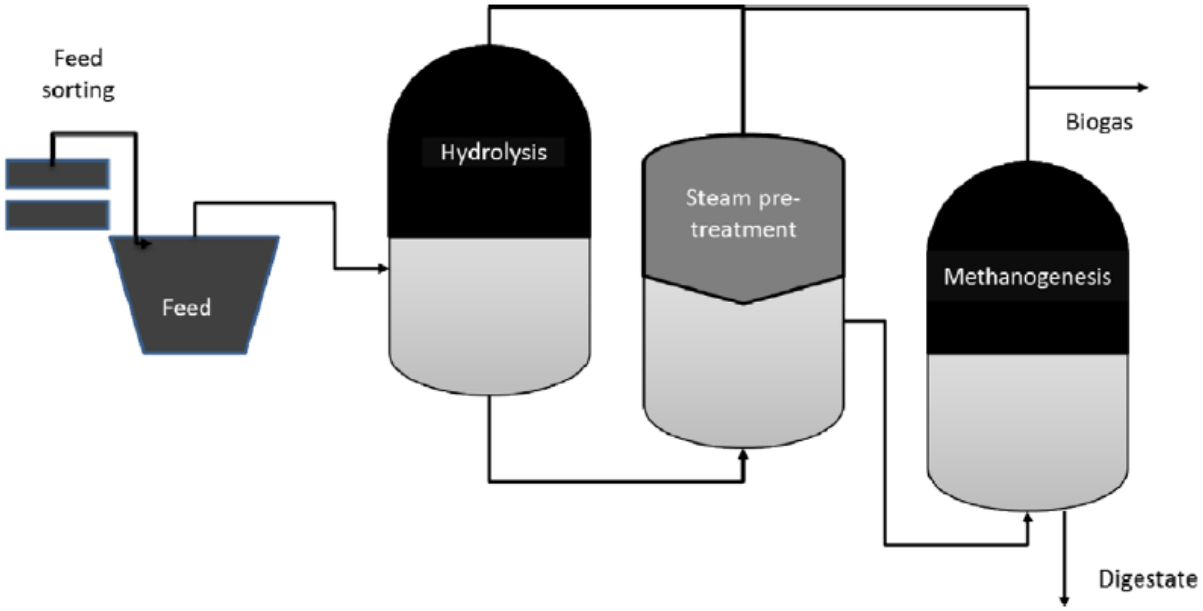

Figure 2.5. A schematic draw of two-stage Super Blue Box Recycling (SUBBOR) processes.

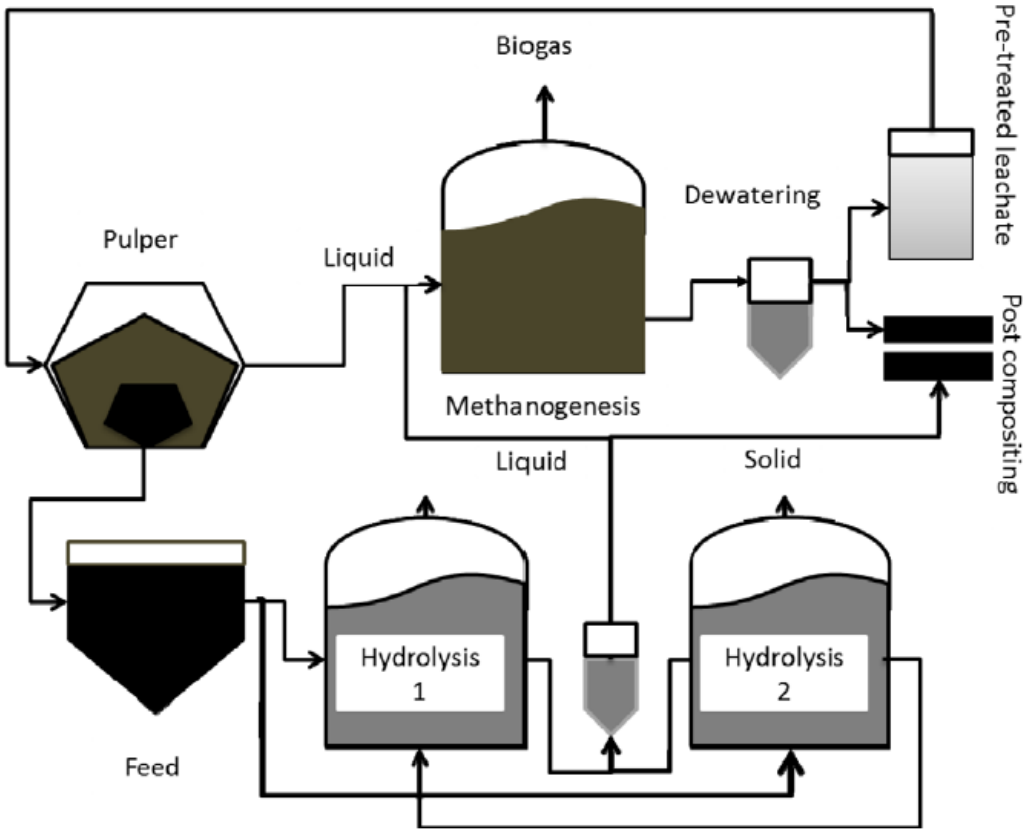

Figure 2.6. Diagram of Biotechnische Abfallverwertung (BTA) system: multi-digestion.

2.4. Inhibitory Factors

Physical and chemical characteristics of OFMSW largely affect the process stability, biogas quality and yields (Pagliaccia et al., 2019). The constituents of OFMSW could be classified into three major categories, as follows: carbohydrates, proteins and lipids, which each originate from different materials. For example, as one of the major constituents of OFMSW, lipids play the most significant contribution to higher biomethane yields, as compared to carbohydrates and proteins. However, lipids exhibit the lowest hydrolysis rate due to their adsorption on the surfaces of microorganisms and causing acidification, thereby reducing pH, lessening susceptibility to enzymatic reactions, and reducing biomethane production (Neves et al., 2008). Nonetheless, it is impossible to standardize the AD if complex matrices like OFMSW and food waste. For these complex substrates, a case-by-case approach is needed to obtain the quantity and quality of the biogas produced in terms of yields and composition (Curry and Pillay, 2012). Thus, an accurate characterization of OFMSW or food waste, in terms of protein, carbohydrate, lipid and

lignocellulosic matrix (so-called macromolecular compounds analysis), can play an essential role in studying their conversion in AD processes (Alibardi and Cossu, 2016; Fisgativa et al., 2017).

Previous literature has reported various challenges and inhibitory factors affecting the performance of HSAD processes, shown in Figure 2.7 and discussed in the following section. Some of these influential factors include but not limited to F:I ratio, percolate recirculation, operating temperature, pH, as well as toxicity resulting from high concentrations of some compounds, including VFAs, TAN, FAN, D-limonene and furfurals, which could be inhibitory to methanogenic bacteria depending on their concentrations in the digester (Fagbohungbe et al., 2015; Rocamora et al., 2020). Although HSAD has the potential for higher biomethane yield per unit volume of digester, some operational challenges have limited this technology towards its proliferation as a viable and sustainable AD process, particularly because of the role of VFAs and putative inhibitors in influencing biomethane production during HSAD (Nagao et al., 2012). In addition to operating temperature (discusses later), the low water or moisture content of the feedstock in HSAD is the major cause for the poor distribution of fermentative intermediates, including VFAs and acetic acid, thereby producing low biomethane yield (Nagao et al., 2012). In fact, water enhances the diffusion of VFAs and acetic acid to the structure of microbial cells, which causes further biomethane production.

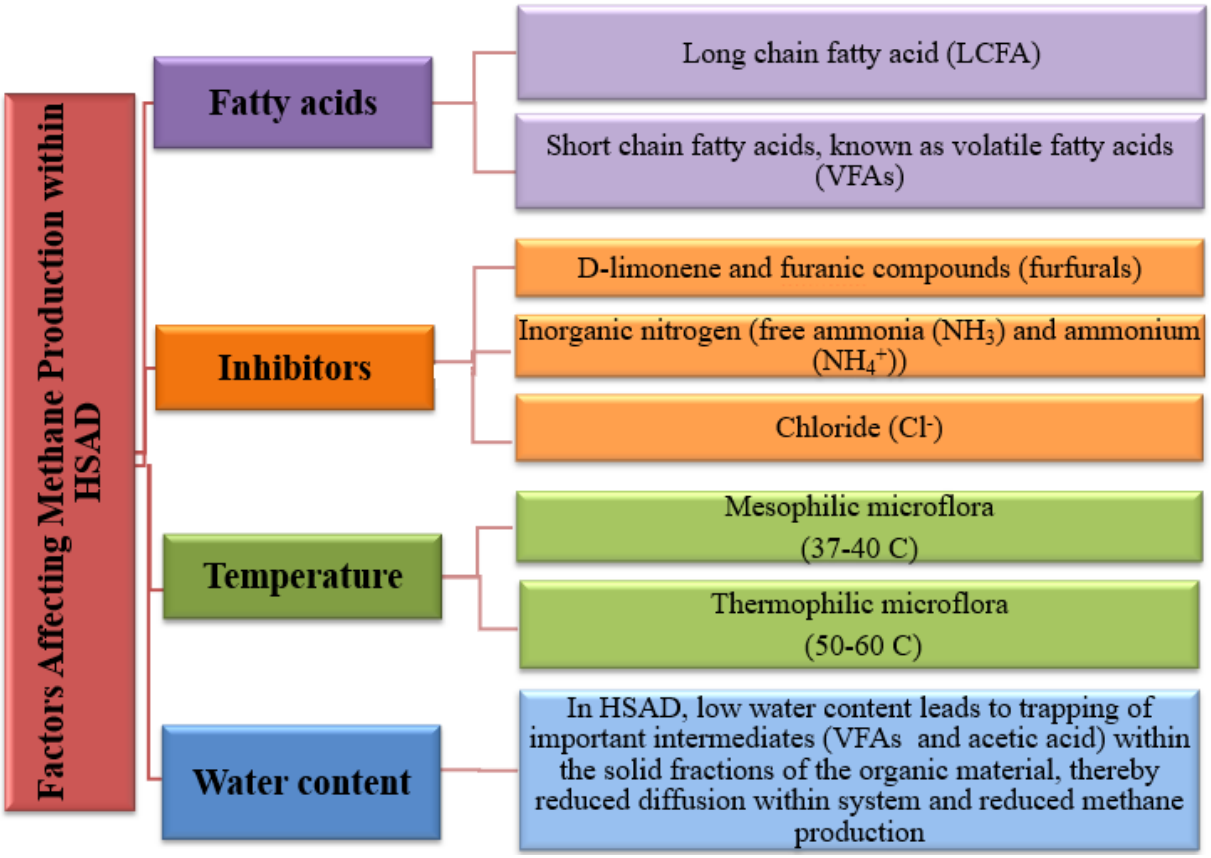

Figure 2.7. HSAD: affecting factors (Fagbohungbe et al., 2015).

2.4.1. Volatile Fatty Acids

The fatty acid category includes long-chain fatty acids (LCFAs) and short-chain fatty acids (SCFAs), which both are enumerated as fermentative intermediate products that are formed by the acidogenesis and acetogenesis reactions during the AD of organic feedstock (Madsen et al., 2011). The LCFAs originate from lipid-rich substrates (Rinzema et al., 1994), and SCFAs (shortly known as volatile fatty acids (VFAs)) emanate from acetic acid, propionic acid, butyric acid, and so on (Chen et al., 2008). Generally, two-third of the biomethane produced from the AD of organic substrates strongly depends on the availability of important intermediate formed compounds such as VFAs (Buyukkamaci and Filibeli, 2004). However, both SCFAs and LCFAs constituents could have inhibitory impacts on the methanogens. Reportedly, the maximum methanogens tolerance is 1 mM of LCFAs (Sousa et al., 2013), whereas, a very low content of VFAs released in an AD digester with an OLR of 7 to 15 kg VS/m^3.d can significantly inhibit methanogens, thereby causing

dramatic lower biomethane production (Dong et al., 2010; Nagao et al., 2012). High concentrations of VFAs will escalate the acidity of the AD systems causing methanogenesis inhibition (Fagbohungbe et al., 2015).

Noteworthy, HSAD systems could be more stable at high VFAs concentrations, as compared to LSAD. This outcome is because of the trapping of the VFAs intermediates within the solid medium of the organic substrate, the poor dissolution of the organic compounds and limited accessibility to methanogenic microorganisms resulted from low water content, thereby reducing their diffusion within the system (Dong et al. 2010; Nagao et al. 2012). However, the recirculation of leachate into HSAD systems is considered as the most economical mixing approach to enhance the accessibility of VFAs to the methanogenic communities. In fact, percolate recirculation provided a more homogeneous distribution of fermentative intermediate products, thereby enhancing biomethane production, reducing hydraulic retention time, and improving biomethane production (Sponza and Ağdağ, 2004). In addition, leachate recirculation lessens the possibility of hot-spots formation within the HSAD, where the inhibitory compounds accumulate exceedingly (Vavilin et al., 2003).

The F:I ratio also influences the consumption of VFAs and biomethane yield (Ali Shah et al., 2014), and represents the presence of the favorable microorganisms to accomplish the AD process (Eskicioglu et al., 2011). In other words, at a very high ratio of F:I, the accumulation of VFA methanogenesis could be inhibitory. Each feedstock has an optimum F:I ratio, which depends on (i) total VFAs produced and (ii) the medium capability to buffer the ammonium produced from the hydrolysis of protein substrate (Lesteur et al., 2010). For example, Ta and Bable (2019) demonstrated for the AD of green waste, the optimum F:I ratio was 2 in terms of higher biogas yield, at which the VFA concentration and VFA/alkalinity ratio (4,937 mg/L and 1.87) were lower than that at F:I ratio of 1 (VFA 7,994 mg/L and VFA/alkalinity 1.89). These facts signified further conversion of VFA to biogas at the acetotrophic phase when F:I ratio was 2, thereby decreasing the amount of VFAs. Rouches et al. (2019) reported that although increasing F:I ratio enhanced biomethane productivity in an HSAD of fungal pretreated wheat straw, it also raised the risk of reactor failure owing to VFAs accumulation and increasing VFA/alkalinity ratio, thereby causing acidification. According to their results, HSAD reactors could recover from acidification when

VFA/alkalinity ratio was lower than 2 gHAc_eq/g$CaCO_3$ (with total VFA content < 10,000 mg/L and pH ~ 5.5).

2.4.2. Ammonium-Nitrogen

In regard to ammonia-nitrogen inhibition, it can be indicated that, protein is a source of inorganic nitrogen, namely free ammonia (NH_3) and ammonium (NH_4^+). According to literature, low concentrations of ammonia are necessary for microbial growth; although its higher concentrations (2,000–10,000 mg/L) can cause significant inhibitions in AD process (Koster and Lettinga, 1988). It has been reported that NH_3 is more toxic than NH_4^+ because of NH_3 ability to penetrate the cell membranes causing proton imbalances and sometimes interfering with the metabolic enzymes; thus, inhibiting degradation of VFAs (Gallert and Winter, 1997; Sung and Liu, 2003). These fermentative intermediate products (i.e., NH_3 and NH_4^+) originate from protein-rich substrates that emerge during AD, so that their concentrations are associated with operating temperature and pH within digester (Anthonisen et al., 1976) or imbalanced C/N ratio of feedstock (Yangin-Gomec and Ozturk, 2013). There are two possible ways to reduce the inhibition of NH_3:

- Co-digestion with organic feedstocks containing high carbon content, in order to adjust the C/N ratio, which is well-known as the most economical option to prevent NH_3 inhibition (Yangin-Gomec and Ozturk, 2013; Fagbohungbe et al., 2015).
- Taking time to give ability of methanogens to acclimate to NH_3 toxicity (Koster and Lettinga, 1988).

2.4.3. Operating Temperature

Under AD conditions, the interactions between microorganisms and organic substrates are dependent on temperature. Microorganisms can be classified as two main categories in terms of their viability within different operating temperature regime (Fernández-Rodríguez et al., 2013):

- Mesophilic microflora (37–40 °C)
- Thermophilic microflora (50–60 °C)

Generally, the use of mesophilic temperature is broader, as compared to systems running at thermophilic condition, which is mainly due to less energy consumption and more robust methanogens at mesophilic conditions (Biey et al., 2003). However, thermophilic condition result

in a higher biomethane production rate with lower HRT (Fernández-Rodríguez et al., 2013; Hidaka et al., 2013). As compared with thermophilic conditions, the advantages of mesophilic temperatures for running AD processes are (i) enhancing the diversity of methanogenic microorganisms and (ii) lower energy demand (Biey et al., 2003). However, it has two drawbacks, including a lower biomethane production rate and longer HRT (Biey et al., 2003). Nevertheless, a higher temperature regime (i.e., thermophilic condition) can increase metabolic activities of microorganisms, solubilization and viscosity of organic substrates, thereby further biomethane yield at shorter HRTs (Fernández-Rodríguez et al., 2013; Hidaka et al., 2013; Bollon et al., 2013). However, operating AD under thermophilic conditions requires higher energy demand and lessens the diversity of robust methanogens required for consistent biomethane production (Biey et al., 2003).

2.5. Conductive Materials

The mechanism of the AD process includes a chain of biochemical reactions, such as hydrolysis, acidification, acetogenesis and methanogenesis (Ai et al., 2018; Elyasi et al., 2015). Over initial steps, named hydrolysis and fermentation, acetate and electron carriers (such as formate or hydrogen) are formed by degradation of organic substrate, which can perform as the precursors for methanogenesis (Stams and Plugge, 2009). These electron carriers (like formate or hydrogen) are converted into biomethane by the interaction between bacteria and methanogens via indirect or mediated interspecies electron transfer (IIET). Of note, this natural mechanism or microorganisms' interaction with formed compounds is a very slow process. To boost this stage and biochemical process kinetics, recently researchers have employed different types of conductive additives (including graphite, carbon fibers, carbon cloth, biochar, magnetite, granular activated carbon (GAC), powder activated carbon (PAC), etc.) to amend AD systems, thereby enhancing biomethane production yield (Guo et al., 2018; Tsui et al., 2020). In fact, conductive materials streamline direct interspecies electron transfer (DIET), which is a syntrophic relationship that permits faster and direct transfer of electrons from the bacterium to the methanogen (Baek et al., 2018; Barua and Dhar, 2017; Ren et al., 2020; Sharma et al., 2019).

Despite recent applications of different conductive additives for improving the AD processes by other researchers (Barua et al., 2019; Cheng et al., 2019; Indren et al., 2020; Lei et al., 2018), there have been limited investigations on their use to boost the performance of HSAD systems used

for OFMSW. For example, Zhang et al. (2017) studied the addition of 15 g PAC/L (with the particle size of 100–400 mesh) to the mesophilic AD of food waste under semi-continuous operation (i.e., LSAD) with a gradual increase in the OLR from 1.5 to 4.4 g $VS_{food\ waste}$/L.d and mixing at 80 rpm. Their results revealed that the PAC-amended LSAD sustained stable operation with 0.35 L CH_4/L g VS at high OLR of 4.38 g $VS_{food\ waste}$/L.d, while the control bioreactor displayed less stability resulted from further acidification. In addition to the enrichment of some specific dominant bacteria in PAC-amended LSAD, the PAC addition improved the performance of the AD process by lessening the comparative abundance of bacterial pathogens by 18%, as compared to the control digester (Zhang et al., 2017). Furthermore, Ryue et al. (2019) conducted a comparative study on the addition of conductive materials (15 g GAC/L) to the AD of food waste under mesophilic and thermophilic conditions. Accordingly, the lag phases for biomethane production significantly declined by GAC addition, as it was compared with the unamended control reactor at both mesophilic and thermophilic. Moreover, their results illustrated that the addition of GAC exhibited its significant beneficial impacts on the AD process in terms of biomethane productivity and kinetics, particularly in mesophilic conditions. These could result from the enhancement of the microbial community of the AD in terms of the diversity, abundance and richness of each bacterial and archaeal community (Ryue et al. 2019). In another study, the comparative results of employing PAC (with 80–100 mesh) and GAC (with 10–20 mesh) have been presented to show their different influence on biomethane recovery from a lab-scale UASB reactor used for AD of brewery wastewater (Xu et al. 2015). According to their results, the introduction of GAC and PAC to the system enhanced biomethane yield and substrate degradation, which were associated with the enrichment of methanogens and accelerating the start-up of methanogenesis. Interestingly, under heavy OLR, adding PAC outcompeted GAC in terms of enhancing biomethane recovery. This was due to a more abundant micropore–mesopore structure of the PAC surface, so that it can colonize further numbers of DIET-active *Methanosarcina sp*. sites, thereby promoting syntrophic associations and boosting biochemical kinetic rate (Xu et al., 2015).

2.6. References

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Chapter 3

Powdered activated carbon amendment in percolate tank enhances high-solids anaerobic digestion of organic fraction of municipal solid waste

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3.1. Introduction

The ongoing increase of the world's population growth and urbanization would cause a 70% increment of annual solid waste generation from ~2.0 billion tons in 2016 to 3.4 billion tons in 2050 (Hoornweg and Bhada-Tata, 2012; WorldBank, 2017, 2018). Moreover, the rising trend of the population would have led to the further global food demand and duplication or even triplication of current universal demand for biomass-based renewable energy by 2050 (Johansson et al., 2012). Moreover, growing population and urbanization would also instigate the landfilling of municipal solid waste (MSW) due to a lack of sustainable waste management practices and policies. Reportedly, the extensive open-air processing and storage of wastes in landfills cause emission of greenhouse gases (GHGs), which poses serious risks to human well-being and global warming (WHO, 2018). So far, various efforts have been attempted to study and implement different practical and economic pathways to mitigate the menaces mentioned above. One of the most practical ways of diverting MSW from landfills is its valorization using thermochemical and biochemical technologies into various value-added products, such as compost, chemicals, and biofuels (Kumar and Samadder, 2020; Lim et al., 2016; Olatunji et al., 2019; Zhang et al., 2019).

Among various biochemical technologies, high-solids anaerobic digestion (HSAD) has been accepted as a practical and eco-friendly approach in addressing both environmental challenges and energy crisis through the waste diversion from landfills and the recovery of bioenergy (Dastyar et al., 2015; Guilford, 2009; Rezaee et al., 2020). Various advantages of HSAD include: (i) lower water consumption and energy demand, (ii) capability of handling higher organic loading rates (OLRs), and (iii) alleviating the requirement of dewatering of residuals (Fagbohungbe et al., 2015).

However, compared to wet-type digesters (also known as low-solids anaerobic digestion, LSAD), heterogeneous and variable characteristics of the feedstock often lead to inferior process kinetics, lower energy recovery and solids removal from HSAD process (Fagbohungbe et al., 2015). The timeline of the HSAD process's evolution reveals that, to date, numerous investigations have explored various reactor configurations coupled with different feedstock pretreatment techniques. Previous studies have aimed to investigate the significance of various operating parameters for HSAD systems, including temperature, F:I ratio, external and/or internal mixing, recirculation of percolate and biogas, etc. (Chen et al., 2018; Guilford et al., 2019; Guo et al., 2020; Wang et al., 2018; Westerholm et al., 2020). Notably, designs of several commercial HSAD systems adopted percolate/leachate recirculation to eliminate mass transfer limitations in the process via homogenization of organics and nutrients (Fagbohungbe et al., 2015).

The process of AD consists of a series of biochemical reactions: hydrolysis, acidification, acetogenesis, and methanogenesis (Ai et al., 2018; Elyasi et al., 2015). The initial hydrolysis and fermentation of organics materialize acetate and other electron carriers that can function as the precursors for methanogenesis (Stams and Plugge, 2009). The electron carriers like formate or hydrogen are transformed into biomethane via the interaction between bacteria and methanogens via indirect or mediated interspecies electron transfer (MIET), which is a very slow process indeed. Recently, the amendment of anaerobic digesters with conductive additives has shown significant potential to boost the biochemical process kinetics, enhance organics degradation, and improve biomethane recovery (Guo et al., 2018; Tsui et al., 2020). Conductive materials could promote direct interspecies electron transfer (DIET), a syntrophic partnership that streamlines the direct transfer of electrons from the bacterium to the methanogen (Baek et al., 2018; Barua and Dhar, 2017; Ren et al., 2020; Sharma et al., 2019). Various conductive additives, such as graphite, granular activated carbon (GAC), powder activated carbon (PAC), biochar, magnetite, carbon fibers, carbon cloth, etc., have been explored to enhance the AD process (Barua et al., 2019; Cheng et al., 2019; Indren et al., 2020; Lei et al., 2018). However, there have been limited studies on the application of conductive materials to improve the performance of HSAD processing the organic fraction of municipal solid waste (OFMSW). Moreover, to the best of the authors' knowledge, no studies investigated the amendment of conductive additives in HSAD systems with percolate recirculation.

The present work investigates the effects of PAC on biomethane recovery from OFMSW in an HSAD system with percolate recirculation. The first objective of this research is to explore how the addition of PAC material influences HSAD process kinetics and degradation of OFMSW, compared to an unamended control reactor. The second objective is to examine the impact of PAC on the potentially inhibitory factors, including pH, accumulation of volatile fatty acids (VFAs), and free ammonia nitrogen (FAN) levels. In this study, PAC was added to the percolate tank of an HSAD system, while in previous studies, conductive additives were directly added into the digester tank along with feedstock and inoculum (Chowdhury et al., 2019a; Cuetos et al., 2017; Pan et al., 2020b; Ryue et al., 2019b; Xu et al., 2015b; Zhang et al., 2017c). Thus, this present study investigates whether recirculation of percolate amended with conductive materials can enhance the HSAD system.

3.2. Material and Methods

3.2.1. Organic Fraction of Municipal Solid Waste (OFMSW)

The organic fraction of municipal solid waste (OFMSW) was obtained from the Edmonton Waste Management Centre (EWMC) located in Edmonton, Alberta, Canada. The collected sample was stored in plastic buckets in a cold room (at 4°C) prior to use. The sample collected from the facility was mainly composed of a mixture of grass clippings, food waste, fruit and vegetable wastes, wood, paper wastes, as well as some minor particles of plastic, styrofoam, glass, and metals (see Supplementary Information). The large particles of plastic, styrofoam and metals were separated manually before the experiment. The remaining was labeled as OFMSW and blended completely to make it more homogeneous prior to characterization and introduction to high-solids anaerobic digesters. The average characteristics of OFMSW were as follows: moisture content (MC): 47.7 ± 1.1%; total solids (TS): 52.3 ± 1.1%; volatile solids (VS): 27.5 ± 1.0%.

3.2.2. Inoculum

The required solid and liquid inoculum for the start-up of these HSAD reactors were also collected from the Edmonton Waste Management Centre (Edmonton, Alberta, Canada). Dewatered anaerobic digester sludge (i.e., biosolids) was used as the solid inoculum in this study. The samples were stored in plastic buckets in a cold room (at 4 °C) before use. The solid inoculum (i.e., biosolids) was acclimated at 36 °C for a couple of weeks before the initial characterization and the

start-up of reactors. The average characteristics of solid inoculum were as follows: MC: 77.0 ± 0.5%; TS: 23.0 ± 0.5%; VS: 13.1 ± 0.3%. The percolate was also acclimated at 36 °C for a couple of weeks before conducting initial analyses and start-up of systems. The percolate had the following average characteristics: MC: 98.9 ± 0.0%; TSS: 1.1 ± 0.0%; VSS: 0.7 ± 0.0%; TCOD: 22,809 ± 272 mg/L; SCOD: 20,238 ± 403 mg/L; TAN: 4471 ± 84.9 mg/L; alkalinity: 11,650 ± 33 (mg/L, as $CaCO_3$); and pH: 8.13.

3.2.3. Reactor Configuration and Operation

Two custom-built lab-scale HSAD systems consist of the digester and percolate tank were used in this study (see Supplementary Information). The cylindrical digester tanks (d×h: 20 cm×50cm) were built with polycarbonate. As shown in Figure 3.1, the body of each digester tank consisted of two major sections: (i) the bottom part with a volume of ~1.5 L was for the collection of the percolate, and (ii) the upper part with a working volume of ~14.5 L to encompass the solid waste. These two sections were separated using a stainless mesh plate (1 mm thickness and ~2 mm hole diameter). The exterior parts of the upper section in both digesters were harmonically covered by heating tubes connected to a re-circulating water bath to uniformly maintain an operating temperature of 36±1 °C in both reactors. In addition, the heating tubes were completely wrapped with bubble-wrap insulators to minimize heat loss.

During start-up, the acclimated inoculum (at 36 °C) was completely blended with OFMSW at an F:I ratio of 2:1 (kg VS_{OFMSW}/kg $VS_{Inoculum}$) and then loaded into the digester tanks. Thus, both digesters initially contained the same total VS content (~812 ± 26 g). Therefore, the biomethane yield and final VS removal were calculated based on the aggregate initial VS. Consequently, reactors were properly sealed, and ultra-pure nitrogen gas was purged through the reactors to provide anaerobic conditions. Each digester tank was connected to a specific glass percolate tank containing 2 L of liquid inoculum. 15 grams of PAC per liter of percolate was added to one of the percolate tanks (referred to as PAC-amended HSAD system). The other system served as the unamended control (no PAC). It should be noted that the loading of 15 g PAC/$L_{percolate}$ used here was within the optimum range of PAC loadings reported in the literature for wet-type digesters (dos Santos et al., 2018; Xu et al., 2015a; Zhang et al., 2017a; Zhang et al., 2017b). More specifically, the dosage of PAC used in this study was in the typical range of PAC or GAC used in other works, either wet- or dry-type anaerobic digestion of various feedstock (Chowdhury et al.,

2019a; Cuetos et al., 2017; Pan et al., 2020b; Ryue et al., 2019b; Xu et al., 2015b; Zhang et al., 2017c). However, compared to these studies, the dosage was lower in terms of the mass of PAC per dry mass of feedstock. The percolate tanks were wrapped with heating tapes (110v×200w) connected to controllers to maintain a constant temperature of 36±1 °C. In addition, each percolate tank was equipped with a mechanical mixer coupled with electric motors to provide a continuous mixing of 300 rpm using an agitator. The percolate recirculation commenced from day 5 with a flow rate of 0.15 L/min for 4 h/d using a peristaltic pump.

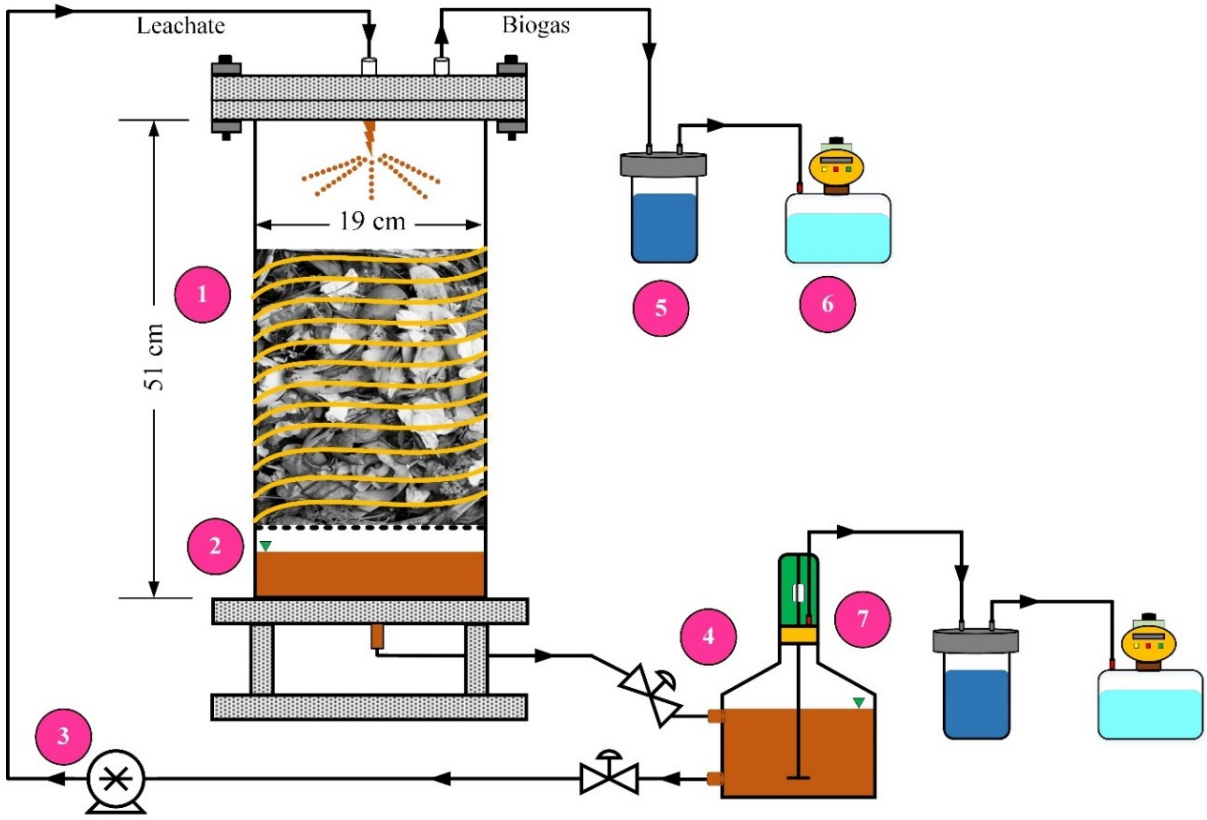

Figure 3.1. Schematic diagram of high-solid anaerobic digester: 1) reactor body, 2) percolate storage, 3) peristaltic pump, 4) percolate tank equipped with mixer, 5) CO_2 capture bottle 6) gas-flow meter, and 7) percolate sampling ports.

3.2.4. Analytical Methods

The moisture and solids (TS and VS) contents were measured using the standard methods (APHA, 1999). The pH was measured with a pH meter, and alkalinity was measured using the standard titration method by sulfuric acid (APHA, 1999). Moreover, chemical oxygen demand

(COD) and total ammonia nitrogen (TAN) concentrations were measured using HACH reagent kits (HACH, Loveland, CO, USA). The concentrations of volatile fatty acids (VFAs) (acetate, propionate, butyrate, and iso-butyrate) were measured using an ion chromatograph equipped with an electro-chemical detector and microbore AS19 column. Considering the COD equivalent of various VFAs, the concentrations were expressed as mg COD/L (Ryue et al., 2019b). Moisture content, TS, VS, VFAs concentrations were measured in triplicate for each sample, while pH, alkalinity, TAN and COD were measured in duplicate. At the end of the operation, for each digester tank, six digestate samples were taken from different zones of the tank for TS and VS analyses.

The volumes of the biomethane produced from both digester and percolate tanks were monitored daily using gas-flow meters coupled to CO_2 sequestration bottles (ISES-Canada, Vaughan, ON, Canada). The bottles contained 3 M of NaOH solution possessing thymolphthalein indicator applied for adsorbing CO_2 from biogas; a detailed description could be found in previous literature (Barua et al., 2018). Furthermore, the concentrations of free ammonia nitrogen (FAN) in both digesters were calculated according to the literature (Koster, 1986). Furthermore, to confirm whether the VFA results obtained had significant differences in characteristics, statistical analyses were performed using Minitab 19 software. The P-values were calculated using Tukey Pairwise comparisons with a significance level of 0.05.

3.3. Results and Discussion

3.3.1. Effect of PAC Addition on Biomethane Productivity

The daily biomethane production and cumulative methane yield from both configurations are shown in Figure 3.2. As shown in Figure 3.2a, during the initial lag phase of approximately 7 days, both reactors exhibit almost similar daily biomethane production rates. Over this commencing period, minimal biomethane production was observed from both digesters, which resulted in roughly the same amounts of cumulative biomethane yields (L CH_4/kg VS) (Figure 3.2b). However, in both reactors, an appreciable biomethane production commenced after 7 days of operation. The PAC addition showed a distinct influence on the daily biomethane production between day 8 to day 16. Subsequently, both reactors sharply reach a peak biomethane production rate at the same time (day 12), albeit the peak for the digester supplemented with PAC is comparable. The peak point of the biomethane production rate in the PAC-amended digester is

~8.9 L/d, which is 50% higher than the control digester possessing a peak point of ~5.9 L/d. This result is in complete accordance with previous studies, reporting that the addition of different types of activated carbon can accelerate the start-up of methanogenesis (Kindzierski et al., 1992; Xu et al., 2015a). Afterward, in both reactors, the daily biomethane production curves dramatically plunged on day 16. From day 17 to day 30, further gradual declines with almost similar trends can be observed for both digesters. Overall, PAC addition ultimately causes a considerable increment in biomethane productivity. To be exact, the total cumulative biomethane yield from the PAC-amended HSAD reactor is 108.7 L CH_4/kg VS, corresponding to a 17% increase as compared to the control (93.0 L CH_4/kg VS).

The effects of different conductive materials have been extensively investigated for conventional wet-type anaerobic digesters operated with food waste under mesophilic conditions (Ryue et al., 2019a; Xu et al., 2015a; Zhang et al., 2017b). According to Zhang et al. (2017b), adding 15 g PAC/L into a wet-type AD ensured a stable operation and higher methane production than a control reactor at a high OLR of 4.38 g $VS_{food\ waste}$/L-d. Furthermore, Ryue et al. (2019a) demonstrated that adding 15 g GAC/L into a wet-type AD operated with food waste significantly declined the lag phases for biomethane production at both mesophilic and thermophilic conditions. Moreover, the GAC addition enhanced biomethane productivity and kinetics, particularly at mesophilic conditions, attributed to improving the diversity, abundance, and richness of bacterial and archaeal communities (Ryue et al., 2019a). Another study suggested that a more abundant micropore–mesopore structure of the PAC surface could promote substantial colonization of DIET-active *Methanosarcina* sp. and syntrophic bacteria and improve AD of brewery wastewater revealed that under high OLRs (Xu et al., 2015a).

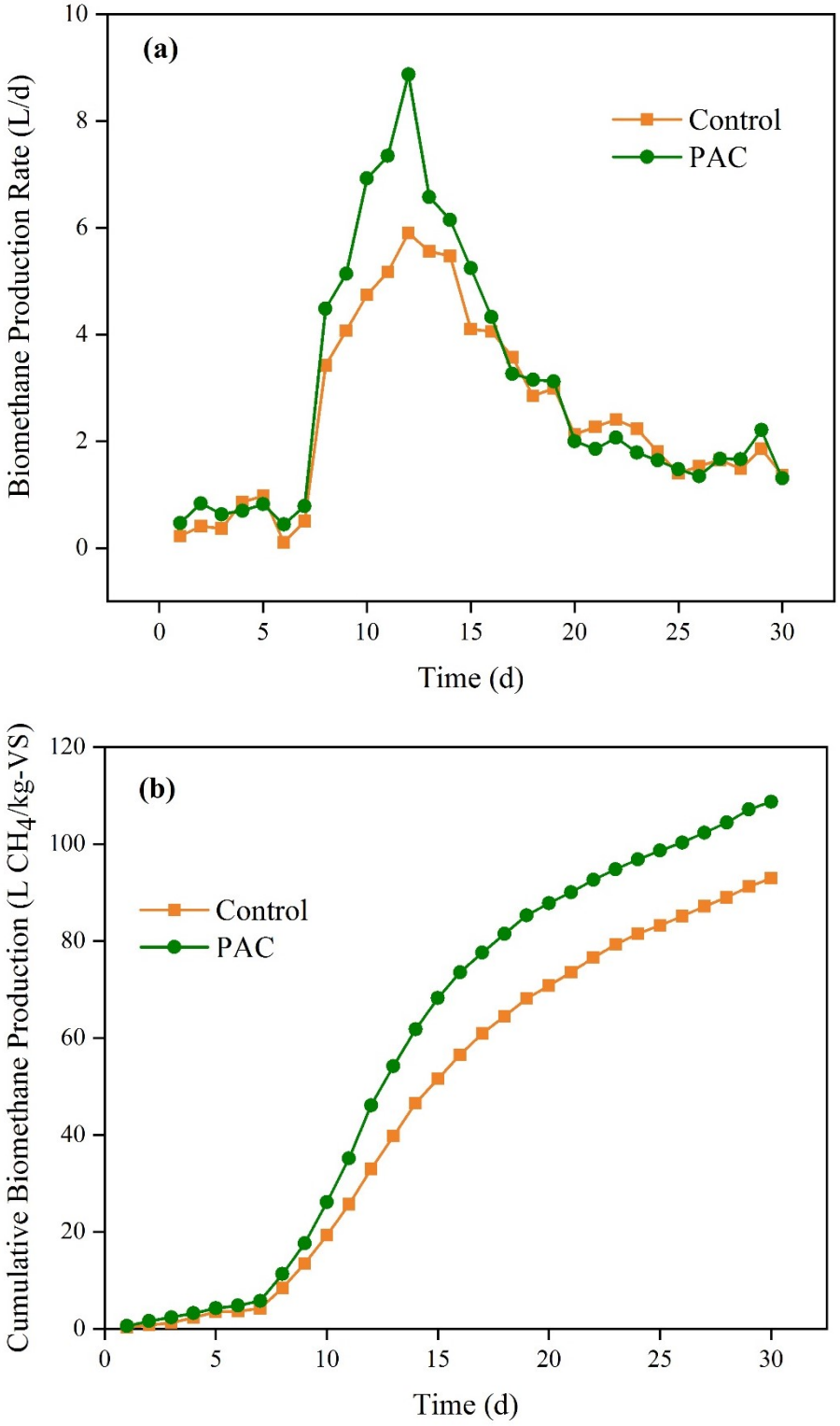

Figure 3.2. Effect of PAC addition on a) daily biomethane production rate, and (b) cumulative biomethane yield.

Conductive materials have demonstrated a positive impact on escalating the digester performance, which originates from shaping the microbial community structures of the digester, accelerating its kinetic rates (Pan et al., 2020a; Park et al., 2018). Notably, kinetically efficient microbial communities can promote faster degradation of organics and conversion of intermediate products, thereby reduce lag phases (Chowdhury et al., 2019b; Wu et al., 2019; Yan et al., 2018). However, the results of this study emphasize that the addition of PAC may not necessarily reduce the lag phases during mesophilic HSAD of OFMSW. The possible reasons are differences in (I) type of feedstock (e.g., biodegradability), (II) operating conditions (e.g., F:I ratios), and (III) features of conductive additives (such as type, specific surface area, and loading of additives) (Ryue et al., 2019a; Xu et al., 2015a; Zhang et al., 2017b). Hence, for further development, future research should focus on different F:I ratios, other types of conductive additives, and microbial analysis towards improving biomethane productivity from HSAD of OFMSW.

The VS content of the final digestate collected from both reactors was comparable (13.4%), corresponding to apparent VS removal efficiency of 34%. It should be noted that several factors may influence the apparent VS removal efficiencies for HSAD systems with percolate recirculation. Depending on the field capacity (i.e., moisture-retaining capacity) of the solid matrix (i.e., the mixture of OFMSW and biosolids), a portion of percolate can be retained by the solids. It is expected that the degradation of organics during operation will also generate leachate/percolate. Due to the natural compaction and gravity, a portion of moisture can be squeezed out of the solid matrix. Moreover, evaporation of the liquid may occur due to the operation under mesophilic conditions. Similar aspects have previously been highlighted for leachate generation and percolation within municipal solid waste landfills by (Yang et al., 2015). Considering the initial moisture content of OFMSW (47.7%) and biosolids (77.1%) in this study, the initial moisture content of their mixture in the digester tank was around 62.4%. After 30 days of operation, the moisture content of the initial solid matrix increased from 62.4% to 66.9% in the final digestate from both digesters. While 2 L of liquid inoculum was introduced to each percolate tank during start-up, only ~1.35 L of percolate was recovered from each system at the end of the operation. Thus, it was evident that a portion of the percolate was trapped within the solid matrix in the digester tank over 30 days.

3.3.2. Impact of PAC Addition on Accumulation/Degradation of VFAs

The VFAs variations during the operation of the control and PAC-amended digesters are illustrated in Figure 3.3. As can be seen, during the lag phase on day 4, the initial concentrations of the total VFAs are 3,069 mg COD/L and 3,144 mg COD/L in the control and PAC-amended digesters, respectively. Subsequently, on day 9, the total VFAs concentrations sharply increase to the highest points of 14,506 mg COD/L and 13,901 mg COD/L in the control and PAC-amended digesters, respectively. Evidently, it can be observed that on day 9, the total and individual VFAs in the PAC-amended reactor are harmonically lower than that of the control. Afterward, by day 29, the concentrations of total VFAs gradually decrease from 14,506 to 5,922 mg COD/L and from 13,901 to 6,346 mg COD/L in the control and PAC-amended digesters, respectively (see Figure 3.3).

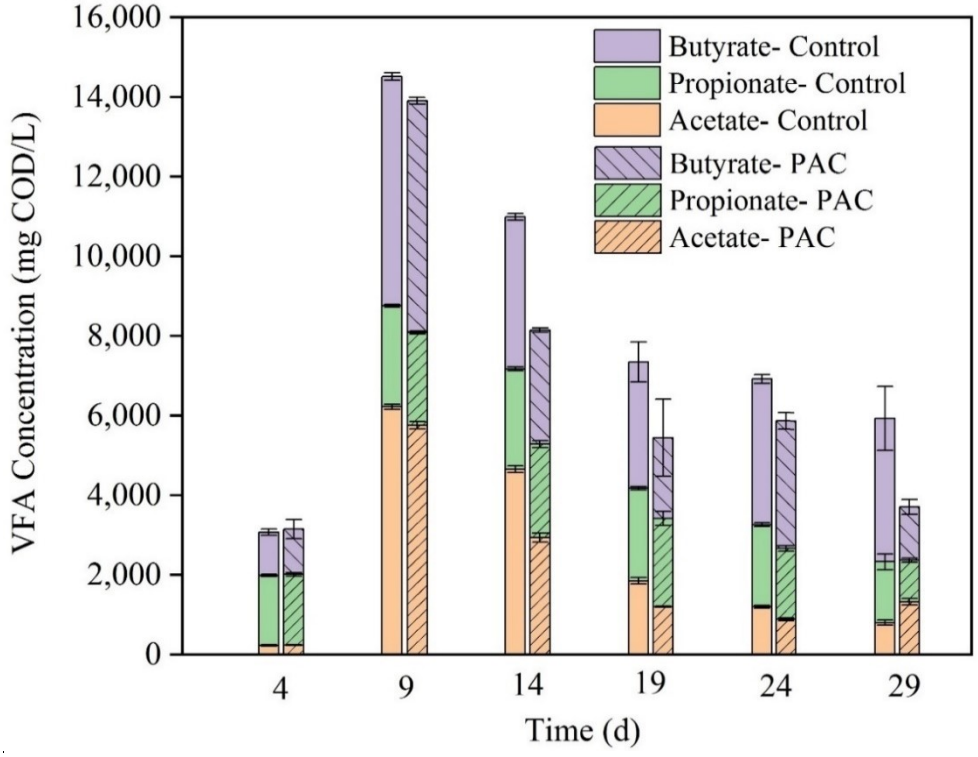

Figure 3.3. Impact of PAC addition on VFAs concentrations.

Throughout the experiment, the concentration of total VFAs, as well as individual VFAs concentrations in PAC-amended reactor, are always lower than that in the control digester. This excludes the acetate concentration with a slightly higher level on day 29 (Figure 3.3). Over the lag

phase (until day 7), acetate concentrations in the control and PAC-amended digesters are considerably lower than propionate and butyrate. Yet, during the peak period of methane production between day 9 and day 14, acetate concentrations have higher levels than butyrate and propionate in both digesters. For the rest of the period, despite reducing the total VFAs in both digesters, the changes in individual VFAs display different trends. Overall, both control and PAC-amended digesters demonstrate active degradation of various VFAs (as depicted in Figure 3.3), while the final VFAs concentrations in both digesters are less than 6,000 mg COD/L. From the relation between methane production patterns and VFAs profiles, it can be inferred that one of the reasons for a lower biomethane production rate in the control digester can be attributed to the higher VFAs accumulation throughout the 30 days of operation, as compared to PAC-amended HSAD.

The accumulation of VFAs can reduce the buffering capacity (alkalinity) of anaerobic digesters and increase VFA/alkalinity ratios (Guwy et al., 1997). However, methanogens can consume VFAs if a suitable buffer capacity in the process is maintained (Ward et al., 2008). The VFAs to total alkalinity ratios varied in the range of 0.28 – 0.87 and 0.21 – 1.10 (with the highest values on day 9) in the control and PAC-amended digester, respectively (Figure 3.4). Notably, maintaining a VFAs/alkalinity ratio below 0.4 could ensure the stability of the digester (Zickefoose and Hayes, 1976), while VFAs/alkalinity ratios higher than 0.8 can cause process instability (Callaghan et al., 2002). Consequently, the further accumulation of the total VFAs (with higher levels of individual VFAs) in the control reactor leads to the further consumption of alkalinity and subsequent reduction of the biomethane production rate than the PAC-amended reactor.

The VFAs degradation trends are consistent with previous studies that have reported the effects of the PAC and GAC addition in mesophilic anaerobic digestion of poultry blood and food waste, respectively (Cuetos et al., 2017; Ryue et al., 2019a). According to Cuetos et al. (2017), the concentration of total and individual VFAs similarly increased to a peak, and again decreased to their minimum values in the following operating days. Furthermore, Ryue et al. (2019a) have reported a comparatively lower amount of the total VFAs accumulation in the GAC-amended digester, than the unamended control reactor. This resulted from the active degradation of various VFAs by microbial communities (Capson-Tojo et al., 2018; Dang et al., 2016; Ryue et al., 2019a). In this study, the enhanced acetate and butyrate consumptions can be observed from the peak point

on, while propionate concentration shows a relatively small variation in the range of 1,529 to 2,536 mg COD/L and 1,040 to 2,349 mg COD/L in the control and PAC-amended digester, respectively.

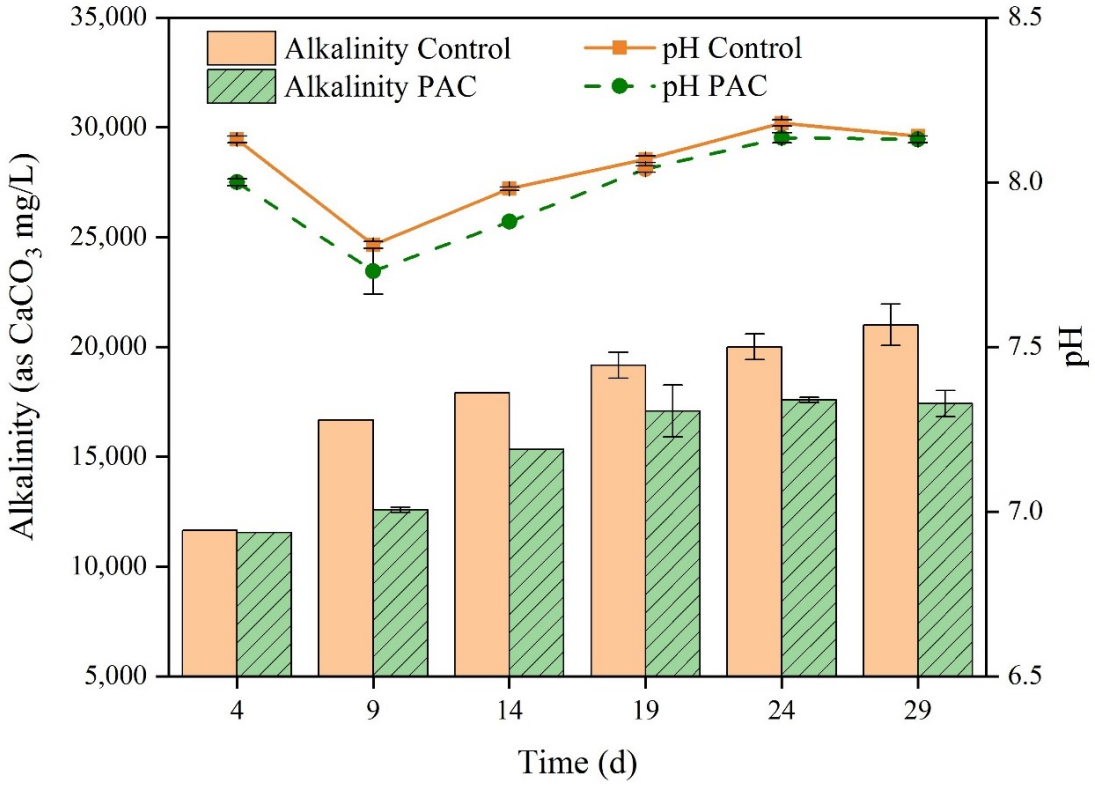

Figure 3.4. Influence of PAC addition on the variations of alkalinity and pH in the PAC-amended and control digesters.

However, the roughly constant propionate levels in this study contrast with Ryue et al. (2019a), reporting enhanced propionate degradation by GAC addition in wet-type anaerobic digestion of food waste. This contradictory behavior could be due to the differences in characteristics of conductive materials, the type of feedstocks, and the F:I ratio between these studies.

A total VFA/alkalinity ratio of < 0.4 has been recommended for the stable operation of anaerobic digesters. In a study conducted by Rouches et al. (2019), the maximum threshold was found out to be different from the threshold of 1 reported in previous literature, which is due to different experimental conditions (Kim and Kafle, 2010; Rouches et al., 2019). The accumulation of VFAs was attributed to a slower growth rate of methanogens than acidogenic bacteria (Vavilin and Angelidaki, 2005). Previous studies have recommended dynamic monitoring of

VFA/alkalinity ratio during the AD process because the dramatic increase of this parameter can reflect the potential instability of the process (Kim and Kafle, 2010; Voß and Weichgrebe, 2009).

A statistical test on the significance of the similarities/differences of VFA data between the control and PAC might not be likely represented with using the same data set in the same group. However, any differences in the replication of measurements of VFAs caused in the same group between the two digesters can be examined. The P-values results obtained from Tukey Pairwise comparisons show that the control and PAC samples are generally different at various levels of total VFAs and individual VFAs (see Supplementary Information). Based on the Tukey simultaneous tests for differences, two distinct groups (Control, in group A versus PAC, in group B) were noticed. In addition, the confidence interval does not contain zero (–3374, –1071) (see Figure A-5). Similar observations were noticed while examining the acetate level (see Table A-4).

3.3.3. Influence of PAC Addition on Free Ammonia-Nitrogen

In anaerobic digestion, inorganic ammonia nitrogen is produced through the hydrolysis of protein-rich substrates (Chen et al., 2008; Yenigün and Demirel, 2013). Two major forms of ammonia nitrogen are free ammonia (NH_3) and ammonium (NH_4^+). The free ammonia nitrogen (FAN) is more toxic than NH_4^+ because free NH_3 can penetrate the cell membranes causing proton imbalances and sometimes interfering with the metabolic enzymes, thus inhibiting the degradation of VFAs (Sung and Liu, 2003). Although a low concentration of ammonia (as a base) is essential for neutralizing the organic acids and maintaining neutral pH conditions required for cell growth microbial growth, the higher concentrations of TAN and FAN pose inhibitory effects (Gallert et al., 1998; McCarty, 1964). To be specific, wide ranges of TAN and FAN concentrations have been reported as critical thresholds under the mesophilic anaerobic digestion of municipal organic waste starting from 1,200 to over 8,000 mg/L and from 45 to over 680 mg/L, respectively (El Hadj et al., 2009; Gallert and Winter, 1997; Kayhanian, 1999).

The changes in TAN and calculated FAN concentrations in the control and PAC-amended digesters have been depicted in Figure 3.5. As illustrated, throughout the operating period in both digesters, the concentrations of TAN and FAN initially decrease and then again increase, which signifies the same trends for the variation of ammonium in the reactors. The TAN contents vary in the ranges of 3,799 mg/L – 4,487 mg/L and 3,210 mg/L – 4,608 mg/L in control and PAC-amended

reactors, respectively. In addition, FAN concentrations in both digesters were lower than 825 mg/L. According to above-mentioned inhibitory ranges in previous literature, both TAN and FAN were within their inhibitory ranges (El Hadj et al., 2009; Gallert and Winter, 1997; Kayhanian, 1999). Moreover, in each day of analysis, FAN concentration in the control digester was higher than the PAC-amended reactor. To be exact, on day 4, the concentrations of FAN in the control and PAC-amended reactors are 766.9 ± 14.6 and 607.6 ± 6.3 mg/L, respectively. Subsequently, FAN concentrations decline to the minimum levels of 399.9 ± 4.8 mg/L and 291.7 ± 2.6 mg/L in the control and PAC-amended digesters, respectively. At the minimum points, the concentrations of FAN in the control and PAC-amended HSAD are reduced by ~48% and ~52%, respectively, compared to their initial concentrations. In the following days, FAN concentrations gradually increase in each digester until reaching the maximum peaks of 785.0 ± 0.3 mg/L and 680.6 ± 6.0 mg/L, respectively. The final FAN contents in both digesters are higher than their initial levels.

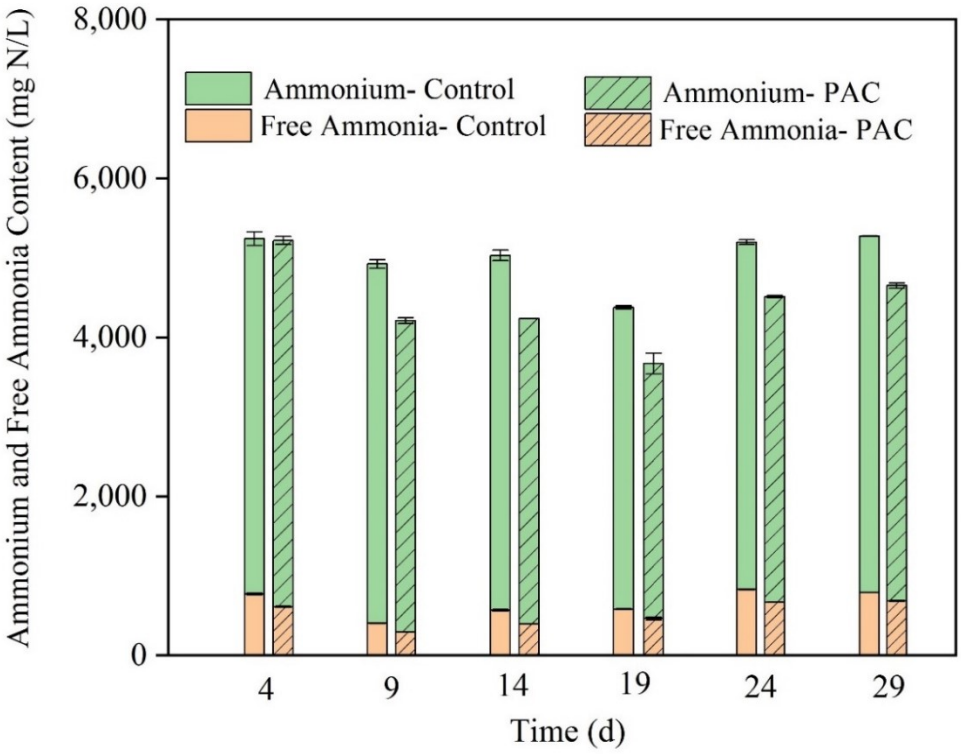

Figure 3.5. Effect of PAC addition on TAN and FAN concentrations.

According to Ali and Yue (2020), the decline of ammonia concentration in an alternate layer bioreactor landfill caused early onset of methanogenesis and considerable increment of biomethane content (from 11% to 40%–50% (v/v)), due to the coexistence of *Methanosarcinales* (3%–50%)

and *Methanomicrobiales* (26%–28%) archaea. According to previous research, NH_4-N has been found as the most critical inhibitory factor that often affects the functional activities of archaea and bacteria in digesters (Ali and Yue, 2020). The higher FAN concentrations can decelerate the degradation of VFAs by methanogens (Chen et al., 2008; Siegert and Banks, 2005). According to previous studies, ammonia content could become toxic when it exceeds 1,500–3,000 mg/l of TAN at pH > 7.4 (Monson et al., 2007). The enhanced biomethane production from the PAC-amended reactor could have resulted from the lower FAN levels in the current work. Overall, despite the potential inhibitory effects of FAN (at high pH) on methanogenesis at some points in both bioreactors, the digester performance was stable. Still, a higher accumulative biomethane yield was observed for PAC-amended HSAD.

According to previous literature, an increase in pH from 7 to 8 will cause an eight-fold increment of the FAN concentration in mesophilic conditions (Hansen et al., 1998). Therefore, in this study, the higher FAN in the control reactor can be attributed to the relatively higher alkaline pH in the control reactor, as compares to the PAC-amended reactor. This slightly higher concentration of TAN with a higher pH level in the control reactor has caused the further conversion of TAN to FAN, thereby occurring higher ratio of FAN/TAN in the control reactor (ranging from 0.09 to 0.19) as compares with PAC-amended digester (0.07 to 0.17) at each specific point. Apart from this, despite the increase of VFAs on day 9, pH does not decrease in any of the reactors. This behavior can be linked to the role of ammonia to balance the alkalinity level in HSAD systems.

3.3.4. Significance of Results and Economic Feasibility

The results of this study first demonstrated that the amendment of PAC in the percolate tank could considerably increase the performance of the HSAD system. The positive impact of PAC addition could be related to the enhancement of methanogenesis kinetics resulted from (i) the formation of DIET-active syntrophic communities, (ii) improvement of MIET kinetics owing to the declined interspecies distance required for hydrogen transfer, and (iii) adsorption of intermediate products (such as ammonia and VFAs) (Cuetos et al., 2017; Pan et al., 2020a; Park et al., 2018; Xu et al., 2015a).

According to previous studies, the lower accumulation of VFAs and higher biomethane yield in PAC-amended HSAD could be attributed to the abundant surface area of PAC structure available for further colonization of syntrophic bacteria and methanogenic archaea, as well as its better assimilation of C3–C5 organic acids. Thus, these alterations in the microbial network and metabolic pathways resulted from the presence of PAC can enhance syntrophic metabolism (Cuetos et al., 2017; Xu et al., 2015a; Zhang et al., 2017a). Moreover, the powdered structure of PAC might have adsorbed a portion of inhibitory compounds (e.g., VFAs, ammonia, and other toxic substances), given the appreciable lower final butyrate and propionate concentrations and the slightly higher acetate in the PAC-amended digester. For instance, a previous study reported the ammonia adsorption capacity of PAC within HSAD has positively influenced biomethane production (Xue et al., 2018). Thus, the enhancement in biomethane productivity could be due to the aforementioned reasons resulted from PAC addition to the system. In fact, PAC addition could have a favorable holistic influence on the reaction mechanisms and kinetics compared to the control digester.

Nonetheless, the economic feasibility of the application of conductive additives should be considered. The current market price of PAC is mostly in the range of $400–2,000/ton PAC (Alibaba.com, September 2020), depending on the nature and characteristics of the product. Thus, a loading rate of 30 g PAC/digester (15 g/L of percolate) in this study will result in a cost of 6–30 $/ton OFMSW. Reportedly, the price of produced biomethane from AD processes worth 0.28 $/m^3 (Dhar et al., 2012). Thus, the excess biomethane recovered from OFMSW in PAC-amended HSAD possesses a potential for extra revenue of ~4.48 $/ton VS. Although this preliminary assessment indicates that the addition of PAC is not economically attractive, reusing PAC for successive batch cycles can possibly make this approach economically feasible. Noteworthy, the transfer of PAC from the percolate tank to the digester tank (resulted from periodic percolate recirculation) is confirmed by the alterations of total suspended solids (TSS) analysis (see Figure A-4). Obviously, it is challenging to separate PAC from digestate, purify, and then reuse it. As the percolate inlet port is located at the top of the digester tank, it is possible that a major portion of the transferred PAC would be trapped in the upper section of the digester tank due to the dense nature of the substrate. Thus, reusing digestate from the top section of the tank in the following batch HSAD cycles can be considered to retain and reutilize the trapped PAC material in digestate and present a promising way to propel this approach more economically feasible. Of note, as the full-scale

HSAD process is typically operated in a batch mode, a portion of the digestate in most HSAD systems is recycled as an inoculum for the following batch cycle and mixed with fresh feedstock (Fagbohungbe et al., 2015). Nonetheless, further studies are required to conduct a multi-cycle HSAD experiment followed by a comprehensive economic assessment.

3.4. Conclusions

The PAC addition had a significant impact on enhancing mesophilic HSAD of OFMSW and resulted in a 17% higher cumulative biomethane yield than an identical unamended control reactor. The daily biomethane production patterns demonstrated similar trends over the initial 7 days and final 13 days; whereas, during day 8 to day 16, the PAC-amended digester exhibited significantly higher daily biomethane production rates over the control. Thus, these results suggested that the amendment of conductive additives in the percolate tank could be a technically feasible option for enhancing the HSAD process with percolate recirculation. However, a preliminary economic assessment indicated that the application of HSAD amended with PAC would be economically challenging. It will only be economically feasible if the PAC can be recycled and reused for long-term operation or several cycles. Therefore, further investigation would be required to ensure long-term recyclability and reusability of PAC or other conductive additives in HSAD systems.

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Chapter 4

High-solids anaerobic digestion of organic fraction of municipal solid waste: effects of feedstock to inoculum ratio and percolate recirculation time

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4.1. Introduction

It has been projected that the annual worldwide municipal solid waste (MSW) generation will be escalated to a critical level of ~3.4 billion tons in 2050, which is owing to the rising world's population and rapid urbanization (Hoornweg & Bhada-Tata, 2012; WorldBank, 2018; WorldBank, 2017). Reportedly, OFMSW represents a significant portion of MSW generated worldwide (Moula et al., 2017). In most countries, the majority of the OFMSW is managed through landfilling, contributing to climate change due to greenhouse gas emissions, pose a human health risk due to seepage of leachate to groundwater and imbalance of natural ecosystem (WHO, 2018). In order to divert MSW from landfills and alleviate the aforementioned menaces, there have been growing interests in the valorization of none-recyclable materials and OFMSW into various valuable products (such as biofuels, bio-fertilizer, and various value-added platform chemicals) using different thermochemical and biochemical processes (Kumar & Samadder, 2020; Lim et al., 2016; Olatunji et al., 2019; Zhang et al., 2019). Among these technologies, anaerobic digestion (AD) has been recognized as a reliable, eco-friendly, and attractive approach to convert OFMSW into renewable biomethane (Chowdhury et al., 2020; Dastyar et al., 2015; Guilford, 2009; Panigrahi & Dubey, 2019). The process of AD primarily involves multiple interconnected biochemical steps: hydrolysis, fermentation, and methanogenesis (Rocamora et al., 2020).

Recently, high-solids anaerobic digestion (HSAD) (also known as dry-type or solid-state anaerobic digestion) has attracted particular attention due to its distinct advantages for processing OFMSW (de Albuquerque, 2021; Fagbohungbe et al., 2015c; Guilford et al., 2019; Ting et al., 2020). Compared to wet-type anaerobic digesters used for aqueous slurry feedstock (typically with <10% total solids (TS) content), HSAD systems are operated with organic solid wastes with higher

TS content (typically 20-40% TS) (Fagbohungbe et al., 2015). Nonetheless, underlying biochemical steps remained the same. Heterogeneity and high-solids content in organic solid wastes can create clogging issues in conventional wet-type AD systems (Chanakya et al., 1997). Therefore, HSAD systems are considered most appropriate for organic solid wastes. Most HSAD systems are operated with active mixing; however, percolate and/or biogas is recirculated to minimize mass transfer limitations (Abid et al., 2021; Dastyar et al., 2021; Fagbohungbe et al., 2015b).

Some of the key benefits of HSAD systems are: (i) less water consumption, (ii) less heating cost, (iii) handling higher organic loading rates (OLRs), and (iv) producing high-quality digestate without requiring sophisticated dewatering technologies (Fagbohungbe et al., 2015a). However, the heterogeneous and variable physicochemical characteristics of OFMSW can still lead to some limitations and/or challenges during HSAD systems operation, including slower biochemical reaction kinetics and lower biomethane recovery, as compared to wet-type digesters (Fagbohungbe et al., 2015a). Typical HSAD systems are operated without active mixing and often suffer from gas/liquid mass transfer limitations, such as inadequate interactions between substrate and microorganisms, localized inhibition by high VFAs and ammonia nitrogen (Capson-Tojo et al., 2017; Riya et al., 2018). To date, numerous investigations have been dedicated to enhanc biomethane recovery from HSAD of different organic wastes (such as OFMSW, sewage sludge, agricultural residues, animal manure) by manipulating reactor configuration, optimizing operational parameters, implementing different pretreatment techniques, exploring co-digestion, and supplementing additives (Ma et al., 2018; Panigrahi & Dubey, 2019; Riya et al., 2018). Notably, incorporating recirculation of percolate (often called leachate) has been demonstrated to be an effective strategy to minimize mass transfer limitations, as HSAD systems are usually operated without mixing (Di Maria et al., 2017; Fagbohungbe et al., 2015c). Moreover, washing inhibitory compounds from the solid matrix to percolate can minimize potential inhibitions by higher VFAs, TAN, etc. levels (Guilford et al., 2019; Qian et al., 2017).

The influential operating parameters of HSAD systems include temperature, F:I ratio, total solids (TS) content, PRT, etc. (Capson-Tojo et al., 2017; Ma et al., 2018; Qian et al., 2017; Rajagopal et al., 2017). As HSAD systems are mostly operated in batch mode, increasing F:I ratio (ratio of feedstock and inoculum) is always desirable to attain process intensification (i.e., high

organic processing capacity). However, operating digesters at high F:I ratios often lead to unfavorable process conditions and inhibition of digester microbial communities due to higher accumulation of VFAs and ammonia nitrogen (Capson-Tojo et al., 2017; Di Maria et al., 2017). Various F:I ratios have been extensively investigated for conventional wet-type digesters and HSAD without percolate recirculation. Despite most studies reported process disturbance with increasing F:I ratios, values of optimum F:I ratios widely depend on feedstock characteristics (Abid et al., 2021; Rocamora et al., 2020). For example, Capson-Tojo et al. (2017) found that maintaining F: I ratio <0.25 would be vital to avoid digester failure via acidification during HSAD (without percolate recirculation) of food waste and cardboard. However, there has been a lack of information on optimum F:I ratios for HSAD with percolate recirculation, especially for OFMSW.

Despite percolate recirculation has been demonstrated to be an effective strategy to alleviate gas/liquid mass transfer limitations, previous studies highlighted the significance of optimizing PRT. It has been suggested that higher PRTs may often lead to excessive leaching of inhibitory compounds (VFAs and ammonia) in percolate, completely suppress the activity of liquid inoculum (Qian et al., 2017). For instance, increasing PRT from 3 h/d to 8 h/d led to a 48% reduction in biomethane yield during HSAD of ensiled straw (Li et al., 2020). Likewise, Qian et al. (2017) also reported a negative impact of increasing PRTs in the co-digestion of OFMSW and corn straw. Nonetheless, the impact of different PRTs on HSAD is yet understudied. To date, limited information is available on the optimum PRTs in HSAD systems processing OFMSW. In contrast, the feedstock used for HSAD systems in centralized waste management facilities in urban areas is often limited to either OFMSW or source-separated organics (SSO).

The present work investigates the optimization of different F:I ratios and PRTs for HSAD of OFMSW. First, the effects of different F:I ratios and PRTs on process performance in terms of biomethane generation rates and yields were evaluated. Second, the temporal changes in physicochemical characteristics of percolate were monitored to intensify the understanding of how process performance changes with different process conditions. Moreover, elemental analysis followed by estimation of theoretical methane potentials was performed to evaluate process efficiencies further and propose potential process intensification opportunities through a trade-off between organic processing capacity and biomethane recovery efficiencies.

4.2. Material and Methods

4.2.1. Feedstock and Inoculum

The feedstock (OFMSW) and solid inoculum (biosolids or dewatered anaerobic digester sludge) were collected from the Edmonton Waste Management Centre (EWMC) located in Edmonton, Alberta, Canada. The collected samples were kept in plastic buckets and stored in a cold room (at 4°C) prior to use. The OFMSW sample was a mixture of food waste, grass clippings, fruit and vegetable residue, lignocellulosic materials, as well as some particles of paper, plastic, styrofoam debris, and tiny glass pieces (see Supplementary Information). Before the experiment, the OFMSW sample was manually inspected to remove any undesired items (e.g., large particles of plastics, glass pieces, etc.). Then, the remaining was manually mixed to provide a homogeneous sample before characterization and use. The average characteristics of OFMSW were as follows: moisture content (MC): 67.1 ± 0.3%; total solids (TS): 32.9 ± 0.3%; volatile solids (VS): 22.2 ± 0.2%; and carbon to nitrogen ratio (C/N): 13.90 ± 0.6. Before the experiment, the solid inoculum (biosolids) was acclimated at 37°C for fourteen days. The acclimatized solid inoculum was characterized before being blended with OFMSW. The average characteristics were as follows: MC: 77.1 ± 1.6%; TS: 23.0 ± 1.6%; VS: 13.1 ± 0.8%; and C/N: 6.66 ± 0.03.

The dewatering centrate from mesophilic anaerobic digester sludge collected from the Gold Bar Wastewater Treatment Plant (Edmonton, Alberta, Canada) was used as the liquid inoculum (referred to as percolate). The centrate was collected by centrifuging anaerobic digester sludge at 3600 rpm for 5 min at room temperature. The average characteristics of the percolate were as follows: MC: 97.7 ± 0.1%; total suspended solid (TSS): 2.3 ± 0.0%; volatile suspended solid (VSS): 2.1 ± 0.2%; TCOD: 33,100 ± 1,278 mg/L; SCOD: 31,843 ± 4 mg/L; total ammonia nitrogen (TAN): 1,896 ± 44 mg N/L; FAN: 7.95 ± 0.2 mg N/L; alkalinity: 8,655 ± 1 (mg/L, as $CaCO_3$); and pH: 6.46 ± 0.0.

4.2.2. Reactor Configuration and Operation

In this work, six cylindrical HSAD systems were used. The detailed description of the system could be found in the literature (Dastyar et al., 2021). Briefly, each system consisted of a cylindrical digester tank connected to a glass percolate tank with a working volume of 2.5 L. The body of the digester tank was manufactured with polycarbonate material and divided into two sections: (i) the

upper section was for encompassing feedstock (volume ~14.5 L), and (ii) the bottom section was designated for collecting percolate (volume ~1.5 L). These two sections were partitioned by a 1-mm thick stainless mesh plate with well-distributed holes with a diameter size of ~2 mm. The upper sections of all six digester tanks were covered with heating tubes connected to a re-circulating water bath to maintain an operating temperature of 37±2 °C. In addition, the exterior part of the heating tubes was completely wrapped with bubble-wrap insulators to minimize heat loss.

Table 4.1 summarizes the different experimental conditions used to study the effects of F:I ratios and PRTs. To study the effect of different F:I ratios, the first three systems (labeled as R1, R2, and R3) were operated at F:I ratios of 1, 2 and 3 (kg VS_{OFMSW}/kg $VS_{Biosolids}$), while PRT was the same (2.5 h/d) (see Table 4.1). To investigate the impact of PRTs, the other three systems (labeled as R4, R5 and R6) were operated at a constant F:I ratio of 2, while PRTs were varied from 1.5 to 3.5 h/d (R4: 1.5 h/d, R5: 2.5 h/d, and R6: 3.5 h/d). Thus, R2 and R5 had identical operating conditions, which was considered to monitor the probable variation of the systems. The statistical analysis (t-test) was performed for monitored parameters from these two systems to evaluate if any statistically significant differences exist (results not shown). The results confirmed that different monitored parameters from these two reactors were statistically insignificant, ensuring the reliability of the data.

Table 4.1. Summary of experimental conditions.

Digester	F:I ratio (kg VS_{OFMSW}/kg $VS_{Solid\ Inoculum}$)	PRT* (h/d)	OFMSW added (kg VS)	Solid inoculum (biosolids) added (kg VS)	C/N ratio**
R1	1	2.5	0.337	0.325	9.12
R2	2	2.5	0.488	0.236	10.63
R3	3	2.5	0.575	0.185	10.86
R4	2	1.5	0.488	0.236	10.63
R5	2	2.5	0.488	0.236	10.63
R6	2	3.5	0.488	0.236	10.63

*Pumps were set at a constant flow rate of 100 mL/min

**Based on elemental analysis (CHNO)

Before the start-up of reactors, acclimatized solid inoculum (biosolids) was thoroughly blended with OFMSW at different F:I ratios (see Table 4.1) and loaded to the digester. After loading pre-mixed OFMSW and biosolids, all digester tanks were completely sealed. The percolate

tanks were filled with 2.5 L of percolate (liquid inoculum). Then, the reactors were purged with ultra-pure nitrogen gas to provide anaerobic conditions. Percolate tanks were placed in water baths (at 37±1°C). The liquid in the percolate tank was continuously agitated at 300 rpm using a mechanical agitator connected to electric motors to provide a homogeneous liquid medium during percolate recirculation and prevent clogging of valves by settled suspended solids. Each system was connected to a peristaltic pump with double heads (YZ1515x, LongerPump®, Longer Precision Pump Co., Ltd., Tech Industrial Development Zone Baoding, Hebei, China) for daily percolate recirculation. Depending on the experimental conditions (Table 4.1), PRTs were varied, while pumps were set at a constant flow rate of 100 mL/min for all conditions.

Depending on F:I ratios, the amount of feedstock (OFMSW) and biosolids varied among reactors. Therefore, the net cumulative biomethane yield and final VS removal were calculated based on the initial mass of OFMSW (VS basis) in each digester. The biomethane generated from biosolids (solid inoculum) was subtracted to measure the net methane production from OFMSW. The biomethane potential of biosolids was assessed with batch anaerobic digestion tests (at 37°C for 30 days) with inoculum only (see the Supplementary Information). Apart from this, the final VS removal in each reactor was calculated based on the aggregate initial VS of solids (OFMSW + Biosolids) and final corresponding digestates.

4.2.3. Analytical Methods

The solids (TS, VS, TSS, and VSS) and moisture content were measured using the standard methods (APHA, 1999). TCOD, SCOD, alkalinity, and TAN concentrations were measured using HACH reagent kits (HACH, Loveland, CO, USA). The pH was measured using a pH meter (AR15 pH Meter, Thermo Fisher Scientific Inc., Pittsburgh, Pennsylvania, USA). The concentrations of VFAs (acetate, propionate, and butyrate) were measured by an ion chromatograph equipped with an electrochemical detector and microbore AS19 column. For each sample, the analyses of moisture content, TS, VS, VFAs concentrations were triplicated, while pH, alkalinity, TAN, TSS, VSS, TCOD, and SCOD analyses were duplicated. The concentrations of FAN were calculated using an equation reported in the literature (Koster, 1986). The volume of biomethane produced from each system (digester tank + percolate tank) was daily monitored using wettip gas-flow meters (ISES-Canada, Vaughan, ON, Canada). These gas-flow meters were connected to bottles filled with 3 M NaOH solution containing thymolphthalein indicator for capturing acidic gases (e.g.,

CO_2, H_2S, etc.) from biogas, as described in previous literature (Barua et al., 2018). At the end of the operation, three digestate samples were taken from three different zones (i.e., top, middle, and bottom) of each digester for analyses of TS, VS, and elemental composition. The elemental analysis (CHNO, dry-ash free basis, wt.%) of initial OFMSW, biosolids, and final digestate samples were conducted for each digester (Table 4.2). The elemental composition of OFMSW and biosolids were measured using a 2400 Series II CHNS analyzer (PerkinElmer, USA), and oxygen content was measured with the vario MICRO cube (Elementar, HANAU, Germany). The development of empirical molecular formula and theoretical biomethane potential of feedstock was calculated, as previously described in the literature (Chae et al., 2008).

Table 4.2. Molecular formula of OFMSW and biomethane recovery efficiencies obtained based on elemental analysis results.

Digester	VS of OFMSW added (g)	Empirical molecular formula of OFMSW	Methane potential (L) at 37°C		Methane recovery efficiency (%)
			Theoretical	Experimental	
R1	337		179	154	86
R2	489	$C_{16}H_{26}O_{10}N$	260	182	70
R3	575		305	105	34

4.3. Results and Discussion

4.3.1. Biomethane Production

4.3.1.1. Impact of Different F:I Ratios

The effects of different F:I ratios (at a constant PRT of 2.5 h/d) on daily methane production rates and net cumulative methane yields from the R1 (F:I=1), R2 (F:I=2), and R3 (F:I=3) are illustrated in Figure 4.1. As can be seen in a glance, different F:I ratios demonstrated distinct impacts on biomethane recovery and process kinetics in terms of lag phases and daily methane production rates (see Figure 4.1a). As shown in Figure 4.1b, R1 showed the highest net cumulative biomethane yield of 531 L CH_4/kg VS_{OFMSW}, which was 30% and 156% higher than R2 (408 L CH_4/kg VS_{OFMSW}) and R3 (207 L CH_4/kg VS_{OFMSW}), respectively. Moreover, the daily biomethane production profile of R1 (F:I=1) plunged quicker (Figure 4.1a), indicating faster substrate depletion

than reactors operated at higher F:I ratios (R2 and R3). The lowest net biomethane yield (408 L CH_4/kg VS_{OFMSW}) was observed from R3 (F:I=3, PRT= 2.5 h/d) with a lag phase of ~15 days. The peak methane production in R3 reached on day 24, which was about 2-3 times longer than the reactors operated at lower F:I ratios (R1 and R2). Thus, an increase in F:I ratios evidently decreased biomethane yields.

Although limited information is available in the literature on the optimum F:I ratios for HSAD systems with percolate recirculation, the trends observed in this study are consistent with previous HSAD studies conducted with or without percolate recirculation (Capson-Tojo et al., 2017; Di Maria et al., 2017). For instance, Capson-Tojo et al. (2017) explored the influence of F:I ratios (0.25, 1, and 4 g VS/g VS) on co-digestion of food waste and cardboard in mesophilic HSAD systems without percolate recirculation. Their results showed adverse impacts on biomethane production for F:I ratios >0.25. Notably, bioreactors operated at F:I ratios of 1 and 4 failed due to VFAs accumulation, leading to a severe pH drop <5.5 (Capson-Tojo et al., 2017). In this study, minimal biomethane yield from R3 was also attributed to considerable VFAs accumulation, leading to a pH drop below 6.7 (discussed later). Although biomethane yield in R1 is 30% higher than R2, R1 showed a significant washout of solids from the digester tank to the percolate (see Supporting Information). As compared to R2 and R3, clogging of percolate recirculation tubes was also more apparent in R1. Thus, lower F:I ratios caused lower resistance to washout of biosolids during percolation, which could be attributed to the rheological characteristics of the solid matrix (OFMSW + Biosolids) in the digester tank. According to previous literature, the anaerobic digestion process can increase the flowability of sludge (Dai et al., 2014). It is expected that at lower F:I ratios (i.e., a higher proportion of inoculum), the flowability of solids (OFMSW +Biosolids) would be greater. Consequently, a lower F:I ratio can lead to a higher washout of solids. Thus, F:I ratio of 2 (R2) can be considered an optimum from operational perspectives (e.g., clogging-related issues and process intensification).

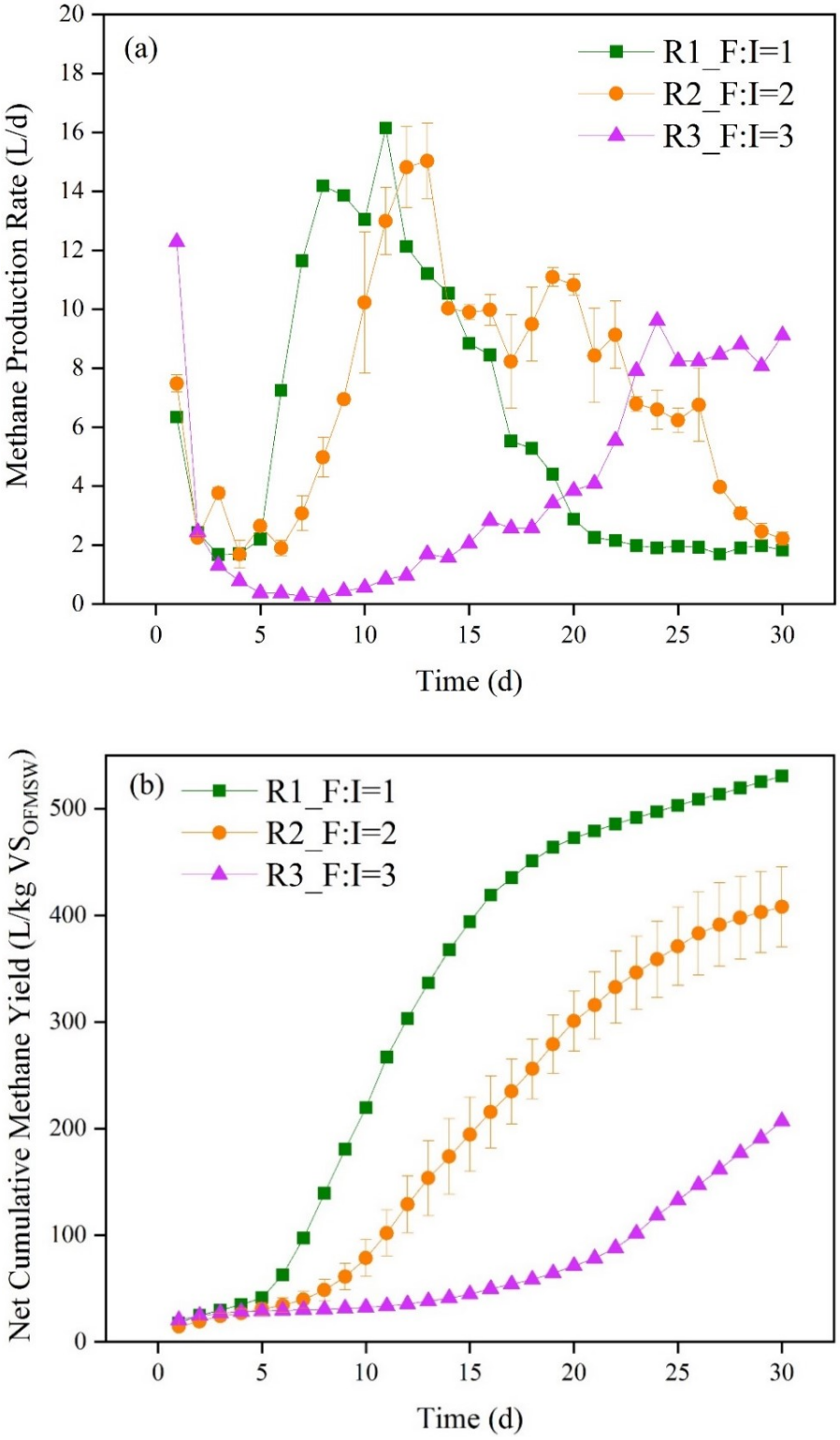

Figure 4.1. a) daily methane production rates, and (b) net cumulative methane production yields at different F:I ratios in R1, R2, R3 digesters (all at constant PRT=2.5 h/d).

4.3.1.2. Impact of Different PRTs

Figure 4.2 depicts the effects of different PRTs on R4, R5, and R6 digesters (1.5, 2.5, and 3.5 h/d, respectively, at a constant F:I ratio of 2). Interestingly, reactors operated under different PRTs showed marginal differences in the time-course profiles of daily methane production and net cumulative methane yields. All three reactors (R4, R5, and R6) also had similar lag phases (~5 days) and reached the same peak daily methane production of ~14 L, albeit maximum methane production rates reached on different days of operation (see Figure 4.2a). After 30 days, all reactors provided almost the same total cumulative biomethane yield of ~420 ± 14 L CH_4/kg VS_{OFMSW} (see Figure 4.2b).

Considering PRTs of 1.5-3.5 h/d at a constant flow rate of 100 mL/min, the frequency of percolate recirculation (FPR, daily percolate recirculation volume to total percolate volume) were ranged from 2.25 to 5.25. Previous studies used FPRs ranged from 0.3 to 4.8 (Guilford et al., 2019; Murto et al., 2013; Pezzolla et al., 2017; Qian et al., 2017); however, none of them explored the effects of different FPRs on HSAD of OFMSW as a sole substrate. Qian et al. found that FPRs ≥2.4 could negatively impact co-digestion of OFMSW and corn straw with a C/N ratio of 24.1 due to considerable VFAs accumulation in percolate (Qian et al., 2017). In contrast, another study reported an increase in methane yield with the increase in PRTs (from 0.75 to 1.5 and 3.0 hr/day) in mesophilic HSAD of combined animal and agriculture residue (initial C/N= 41.3) (Pezzolla et al., 2017).

However, the results of the present study showed marginal impacts of FPRs on HSAD operated with OFMSW only. Such differences can be attributed to differences in the C/N ratios in original feedstock, as well as the decline in C/N ratio of initial substrate resulted from the daily percolate recirculation (Pezzolla et al., 2017). The C/N ratio of OFMSW (13.90) in the present study was lower than that, i.e., 24.1, reported for the co-substrate (OFMSW + corn straw) in previous literature (Qian et al., 2017). It is possible that at a C/N ratio of OFMSW in this study, the balance between hydrolysis/fermentation and subsequent methanogenesis was well maintained at different PRTs. This notion is also supported by comparable COD, VFAs, and ammonia nitrogen levels in percolate samples from different PRTs (discussed later). Thus, based on these results, the lowest PRT of 1.5 h/d can be considered optimum for operating cost minimization (i.e., pumping cost) for processing OFMSW.

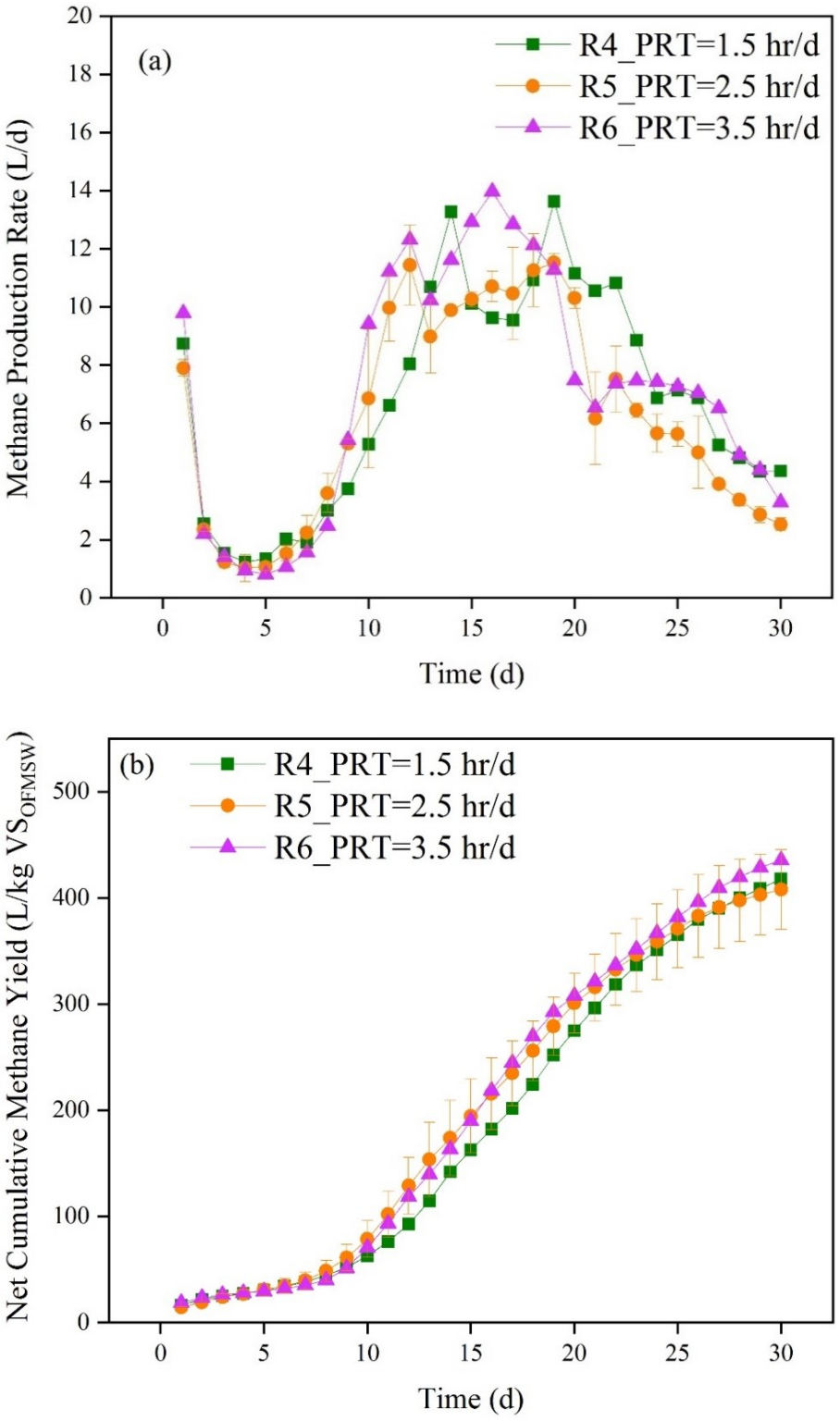

Figure 4.2. a) daily methane production rates, and (b) net cumulative methane production yields for different PRTs in R4, R5, R6 digesters (all at constant F:I=2).

4.3.2. Percolate Characteristics

4.3.2.1. COD and VFAs

Figure 4.3a depicts the variation of TCOD and SCOD concentrations in the corresponding percolate tanks, which are affected by different F:I ratios at constant PRT of 2.5 h/d. The TCOD and SCOD concentrations these digesters (R1, R2, and R3) generally varied in a wide range of 19,076 mg/L – 61,866 mg/L and 8,725 mg/L – 57,475 mg/L, respectively. As can be observed, R1 and R2 digesters with relatively higher biomethane yields had lower SCOD levels than R3 throughout the operation period. Thus, higher F:I ratios ended with a higher accumulation of COD in percolate. As shown in Figure 4.3b, the profiles of TCOD and SCOD concentrations among R4, R5, and R6 were comparable, consistent with comparable methane yields observed from these reactors.

The variation of total VFAs concentrations under different experimental conditions is illustrated in Figure 4.4. On day 5, the total VFAs concentrations in R1, R2, and R3 digesters increased to 19,248 mg COD/L, 23,606 mg COD/L, and 28,048 mg COD/L, respectively, as compared to the initial total VFAs concentration (15,034 mg COD/L) in percolate during start-up (see Figure 4.4a). This increasing trend continued until day 10 before occurring a decrease between day 10 and day 15 followed by a further increase on day 20, specifically for R3 digester. Such rise in VFAs in the later stage of batch anaerobic digestion could be linked to slow hydrolysis/fermentation of slowly biodegradable organics and/or slow degradation of long-chain fatty acids reported elsewhere (Battimelli et al., 2010; Cirne et al., 2007). Afterward, by day 30, VFA levels gradually decreased in R1 and R2 reactors, while VFAs considerably accumulated in R3. The total VFAs concentrations in the final percolate samples from R1 and R2 were substantially lower than R3 (4,712 and 5,590 vs. 25,229 mg COD/L).

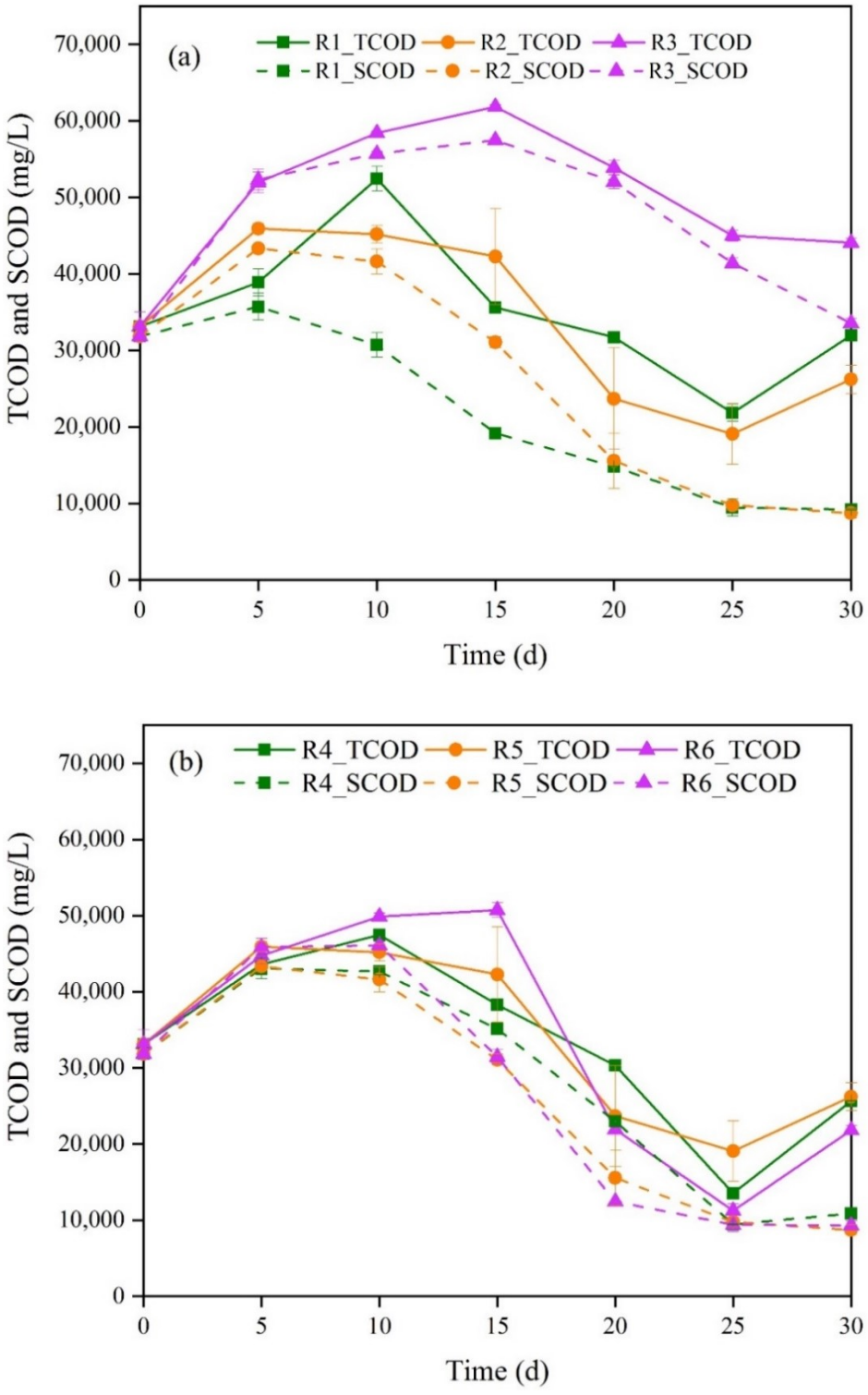

Figure 4.3. The changes in TCOD and SCOD concentrations at a) different F:I ratios (PRT=2.5 h/d), and b) various PRTs (F:I= 2).

Initially, acetate and butyrate were the major VFAs in R1, R2, and R3 reactors (see Figure 4.4a). However, they were efficiently utilized in all reactors, while eventually, propionate started to accumulate in these three reactors. Notably, propionate concentration in R3 was as high as 11,715 mg COD/L on day 25. This observation is consistent with previous reports on the accumulation of propionate when digesters are overloaded with organics; a lower proportion of inoculum (i.e., high F:I ratio) may cause a lower abundance of propionate-utilizing or VFA-degrading microbial population (Acharya et al., 2015; Li et al., 2017; Tale et al., 2015). At higher F:I ratios, the accumulation of VFAs can subsequently reduce digester buffering capacity, leading to pH drop and followed by inhibition of methanogenic activities (Di Maria et al., 2017; Qian et al., 2017). Throughout the operating period, pH in percolate samples from R3 remained lower than R1 and R2 reactors (see Figure 4.5). Notably, acidic pH (<7) until day 15 coincided with the lag phase of methane production in R3 (Figure 4.1a).

Of note, in addition to pH, VFAs to alkalinity ratio has often been considered a critical parameter for monitoring digester stability (Calabro et al., 2018; El Gnaoui et al., 2020; Kim & Kafle, 2010). In previous studies, a VFA/alkalinity ratio of <0.8 has mostly been recommended for efficient operation of HSAD systems, as a ratio >0.6 is an indication of overfeeding (Calabro et al., 2018; Callaghan et al., 2002; Lossie & Pütz, 2008), while a few reports suggested thresholds of ~1 or larger depending on digester operating conditions (Kim & Kafle, 2010; Rouches et al., 2019). The VFAs to alkalinity ratios in the R1, R2, and R3 varied in the range of 0.22–1.74, 0.31–2.66, and 1.64–3.33, respectively (see Figure 4.5). Thus, during batch operation, VFAs/Alkalinity ratios in all these three reactors exceeded the threshold values reported in the literature. Nonetheless, on each sampling day, VFAs/Alkalinity ratios in R3 were consistently higher than R1 and R2. Thus, even though all reactors might face unfavorable VFAs/Alkalinity ratios at some points of operation, the degree of adverse impacts would be higher in R3. Nonetheless, for most operating periods, differences in alkalinity values were marginal in these reactors (see Supporting Information). After day 10, alkalinity remained >10,000 mg/L as $CaCO_3$ in all reactors.

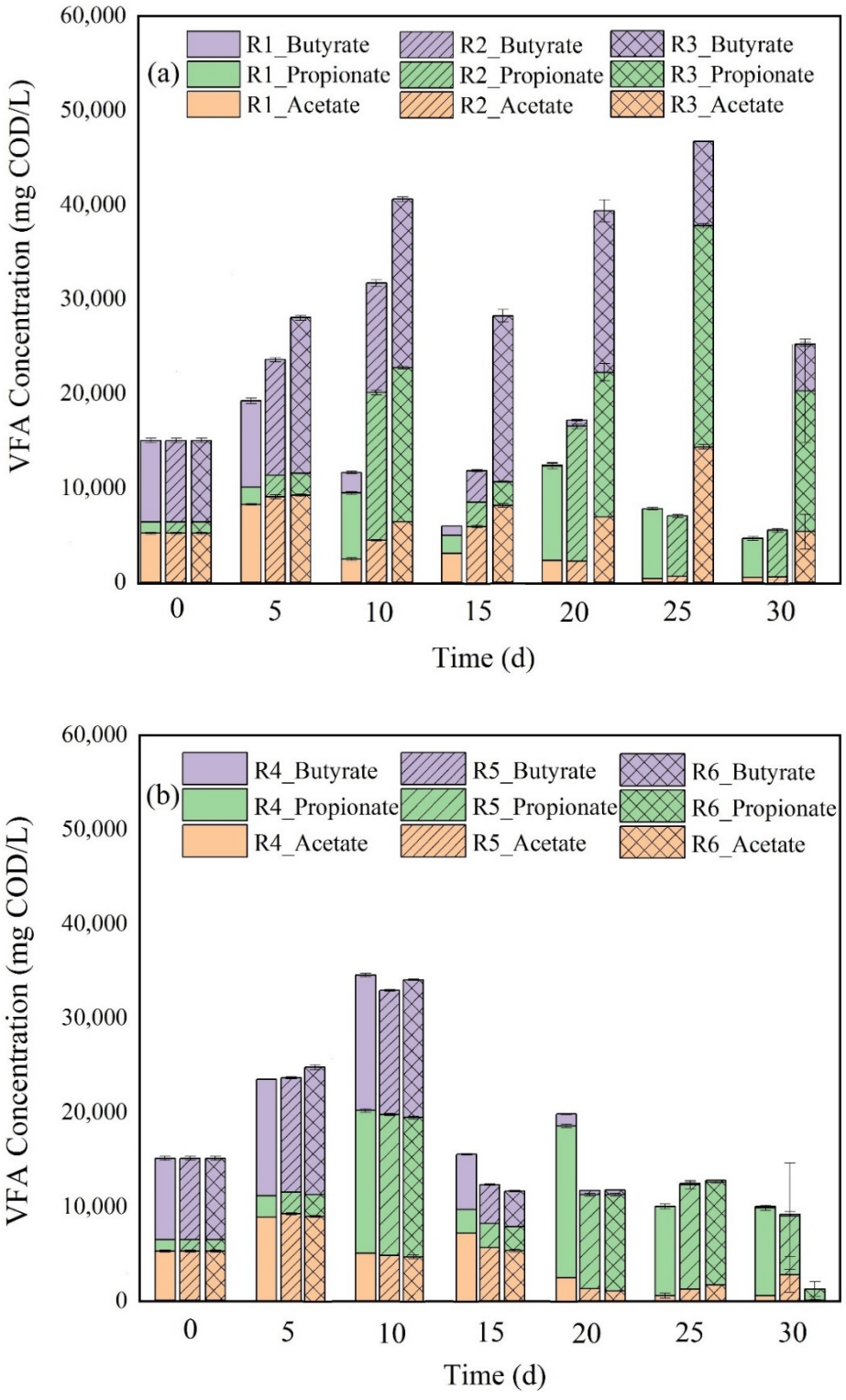

Figure 4.4. The VFA concentrations at a) different F:I ratios (PRT= 2.5 h/d), and b) various PRTs (F:I= 2).

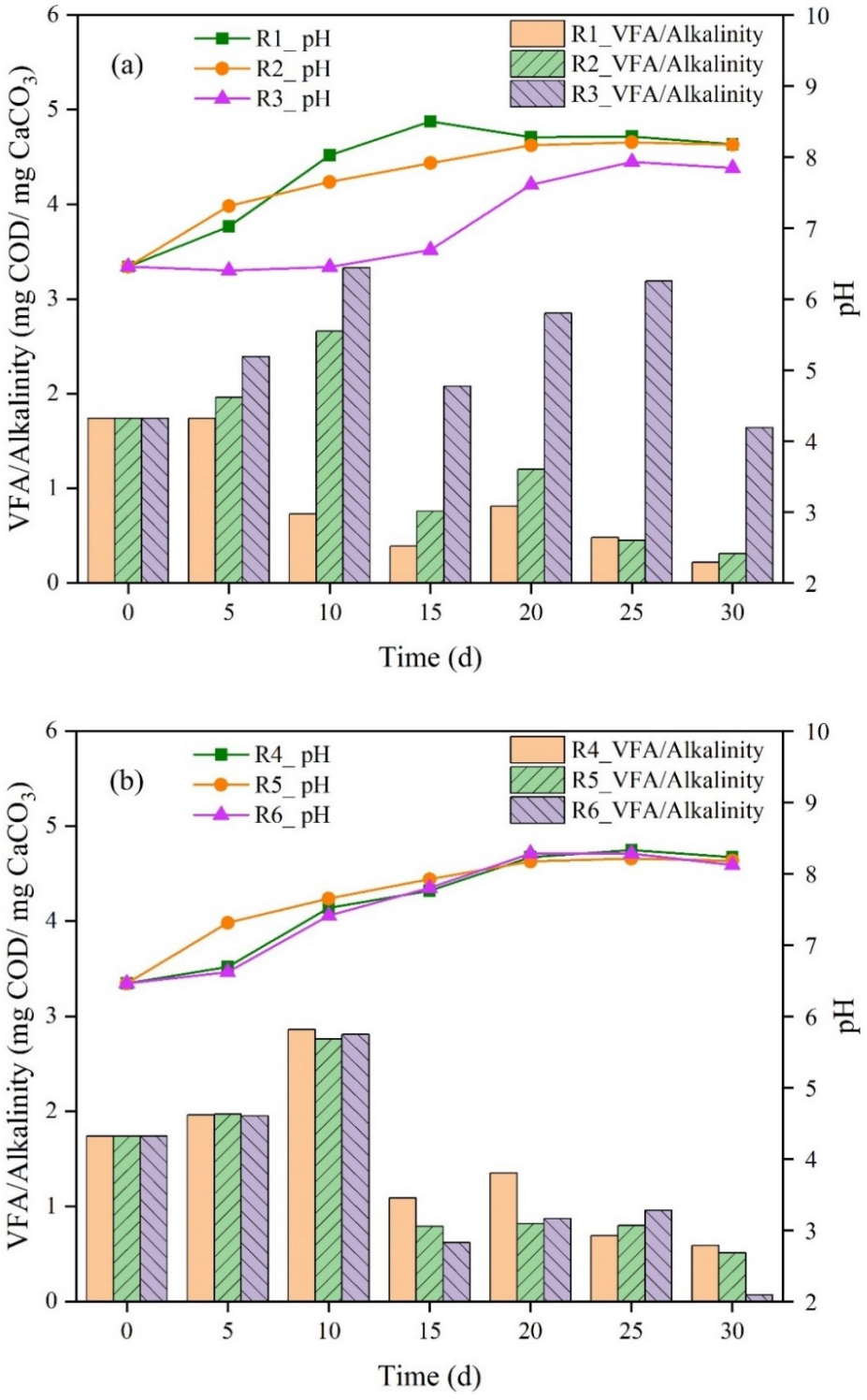

Figure 4.5. The VFA/Alkalinity ratios and pH variations in different (a) R1 to R3, and (b) R4 to R6 digesters.

The VFAs and pH profiles of R4, R5, and R6 were comparable for most of the operating periods (see Figure 4.4b), suggesting marginal effects of different PRTs at a constant F:I ratio of 2. Thus, it can be inferred that the digesters operated under different PRTs showed similar methane production profiles due to analogous trends of changes in VFAs and pH. Despite limited information available on the impact of different PRTs, a study by (Qian et al., 2017) reported a negative impact of higher PRTs due to VFAs accumulation. As discussed earlier, the trivial effect of PRTs observed in this study could be because of the relatively lower C/N ratio in current OFMSW (13.90), compared to the C/N of 24.1 of the feedstock (OFMSW + corn straw) used in a previous study (Qian et al., 2017). Therefore, a thorough investigation of the relationships between different C/N ratios in feedstock and optimum PRTs should be investigated in future studies.

4.3.2.2. Ammonia Nitrogen

The changes in measured TAN and estimated FAN concentrations over time are shown in Figure 4.6. The initial TAN concentration in all reactors was 1,896 mg N/L. For the digesters operated under different F:I ratios (R1, R2, and R3), these TAN levels gradually increased until day 15 due to the ammonia nitrogen release via hydrolysis of organics and subsequent leaching to percolate (see Figure 4.6a). However, on day 10 and onward, TAN concentrations in R3 remained higher than the levels observed in R1 and R2. The reactor operated with the highest F:I ratio of 3 (R3) showed the highest level of TAN (4,136 mg N/L) on day 15, while the highest TAN levels in R1 and R2 were comparable (3,130 and 3,330 mg N/L, respectively). A temporal decline in TAN levels was observed in all reactors between day 15 and day 20. As discussed earlier, during this period, slight increases in VFAs concentrations were also observed in these three reactors (Figure 4.4a). Thus, TAN and alkalinity levels decline between day 15 and day 20 could be attributed to ammonium bicarbonate formation (via the reaction between ammonium and CO_2) that can provide buffer maintain pH (Balaguer et al., 1992). Nonetheless, for R1 and R2, TAN levels further increased from 2,584 mg N/L to 3,094 mg N/L and from 2,679 mg N/L to 3,428 mg N/L, respectively, between day 20 and day 30. In contrast, TAN levels in R3 were relatively stable between day 20 and day 30. Despite different degrees of hydrolysis (as evident from VFAs and TAN profiles) at different F:I ratios, the final TAN concentrations in these three reactors (R1, R2, and R3) were comparable. Thus, these results suggest that a major portion of released ammonia nitrogen in R3 might be utilized for buffering the system.

According to previous studies, a wide range of TAN (1,200 to >8,000 mg N/L) and FAN (45 to > 680 mg N/L) concentrations have been reported to be inhibitory to the mesophilic anaerobic digestion of OFMSW (El Hadj et al., 2009; Gallert & Winter, 1997). Notably, FAN is considered more toxic than ammonium due to its capability to penetrate the cell membranes and causing proton imbalances, which eventually leads to the inhibition of VFAs utilization by methanogenic communities (Sung & Liu, 2003). The equilibrium concentrations of ammonium ions and FAN depend on pH and temperature (Koster, 1986). Thus, TAN would be more toxic at higher pH (pK_a=9.23). As all reactors operated under mesophilic conditions (35^{o}C), it is expected that changes in pH would play a key role in FAN levels. As shown in Figure 4.6a, FAN concentrations in R1, R2, and R3 gradually increased during operation, corroborating the general trends of increased percolate pH in these reactors. However, between day 15 and day 20, a sharp decline in FAN in R1 was observed due to decreased TAN from 3,130 to 2,584 mg N/L and pH from 8.50 to 8.25 during this period (Figure 4.5). According to these results, in all reactors, FAN concentrations were within the inhibitory levels reported in the literature (El Hadj et al., 2009; Gallert & Winter, 1997). Notably, the highest FAN concentration of 1,061 mg N/L was observed in R1 on day 15, while final FAN concentrations in R1 and R2 were comparable. In contrast, FAN concentrations in R3 remained relatively lower than R1 and R2 throughout the operating period due to lower pH. Thus, it was evident that VFAs accumulation followed by pH drop in R3 at a higher F:I ratio of 3 was the primary reason behind its inferior performance.

On the other hand, TAN and FAN in the R4, R5, and R6 operated under different PRTs at a constant F:I ratio of 2 showed marginal differences (Figure 4.6b). Thus, similar ammonia nitrogen profiles under different PRTs suggest that the degree of hydrolysis and leaching of released ammonia from the solid matrix to the percolate were unaffected by different PRTs tested in this study. As discussed earlier, VFAs and pH profiles were also comparable for these three factors. Although FPRs used in this study were within the range used in previous studies with other feedstocks, a future investigation on PRT or frequency should be considered to further minimize operating cost (i.e., pumping cost).

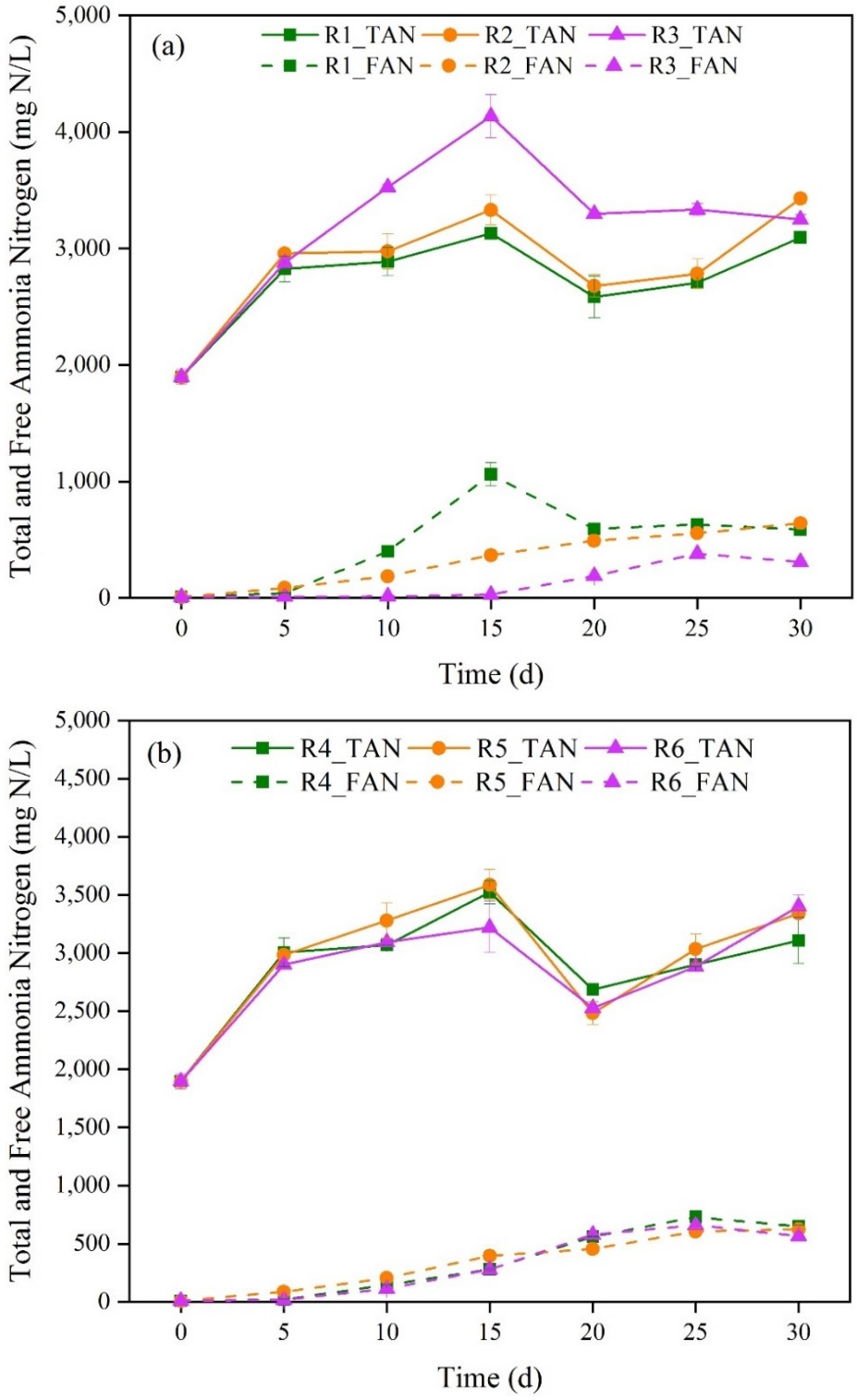

Figure 4.6. The variations of TAN and FAN concentrations at a) different F:I ratios (PRT= 2.5 h/d), and b) various PRTs (F:I= 2).

4.3.3. Elemental Analysis and Process Efficiencies

The elemental composition (C, H, N, O) of OFMSW, biosolids, and final digestate was analyzed to evaluate process efficiencies under different conditions (F:I ratios and PRTs). Detailed results of the elemental analysis results for all reactors are provided in the Supplementary Information. Based on the elemental composition, the empirical molecular formula of OFMSW was estimated at $C_{16}H_{26}O_{10}N$. At different F:I ratios, the estimated theoretical methane potentials of OFMSW added (VS basis) in reactors R1, R2, and R3 were estimated at 179, 260, and 305 L CH_4, respectively. Considering experimentally observed biomethane production, biomethane recovery efficiencies were 86%, 70%, and 34% of the theoretical methane yields, respectively. Thus, biomethane recovery efficiencies considerably decreased with increasing F:I ratios. However, a trade-off between methane recovery efficiencies and F:I ratios should be considered for potential process intensification in terms of organic processing capacity. Despite F:I ratio of 2 provided 16% lower methane recovery efficiency than that achieved at F:I ratio of 1, adopting F:I ratio of 2 can enable the processing of ~45% higher solids (VS basis). Thus, selecting F:I ratio of 2 can provide a reasonable trade-off between organic processing capacity and energy recovery.

4.4. Conclusions

Under the conditions tested in this study, the results identified the F:I ratio as a more critical factor than PRT for HSAD of OFMSW. Notably, an F:I ratio of 3 kg VS/kg VS led to appreciably lower methane yields than other conditions due to VFAs accumulation. In contrast, no noticeable changes were observed for systems operating under different PRTs. Although the F:I ratio of 1 provided 16% higher methane recovery than the F:I ratio of 2, selecting an F:I ratio of 2 can provide ~45% higher organics processing capacity with minimized solids washout and clogging issues during percolate recirculation.

4.5. References

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Chapter 5

5.1. Conclusion

The PAC addition showed a positive impact on enhancing the kinetic and productivity of mesophilic HSAD of OFMSW with ~17% higher cumulative biomethane yield, as compared to the control reactor. These results supported that PAC material added to the percolate tank could be a technically feasible option for improving the efficiency of the HSAD systems. According to results obtained from this feedstock, solid and liquid inoculum, as well as experimental design, it was concluded that the F:I ratio was a more influential factor than PRT for mesophilic HSAD of OFMSW. The reason was attributed to the high risk of acidification formation by increasing F:I ratio ≥3, thereby dramatic decline of methane yields. Unlike the appreciable influence of F:I ratio parameter, no noticeable changes were observed for systems operating under different PRTs ranging from 1.5 -3.5 h/d. Despite 16% lower methane recovery from digester with F:I ratio of 2 (compared with the F:I ratio of 2), it was recommended as the optimum operating condition due to processing ~45% higher organics processing capacity (VS basis) per cycle, with minimized solids washout and clogging issues.

5.2. Recommendation

Although a PAC-amended system could be technically feasible towards increasing the process kinetic and product recovery efficiency, the process could be economically sustainable feasible only when the PAC can be recycled and reused for long-term operation or several cycles. Therefore, further techno-economic studies are required to figure out the best possible way for recovery and regeneration of PAC materials in order to ensure the sustainability of the operation by effective, long-term recyclability and reusability of PAC or other conductive additives in HSAD of OFMSW. Furthermore, operating pilot-scale reactors with frequent recirculation of percolate and convenient loading/unloading substrate is recommended. Determining proper ways to monitor inhibitory factors and prospective troubleshooting procedures during operation is essential, which could be solved by adding chemicals through percolate media to adjust pH, nutrients content, C/N ratio, and tackle biosolid washout and clogging issue.

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Appendix A

Supplementary Information for Chapter 3

Figure A-1. Organic fraction of municipal solid waste.

Figure A-2. Inoculum: anaerobically digested sludge.

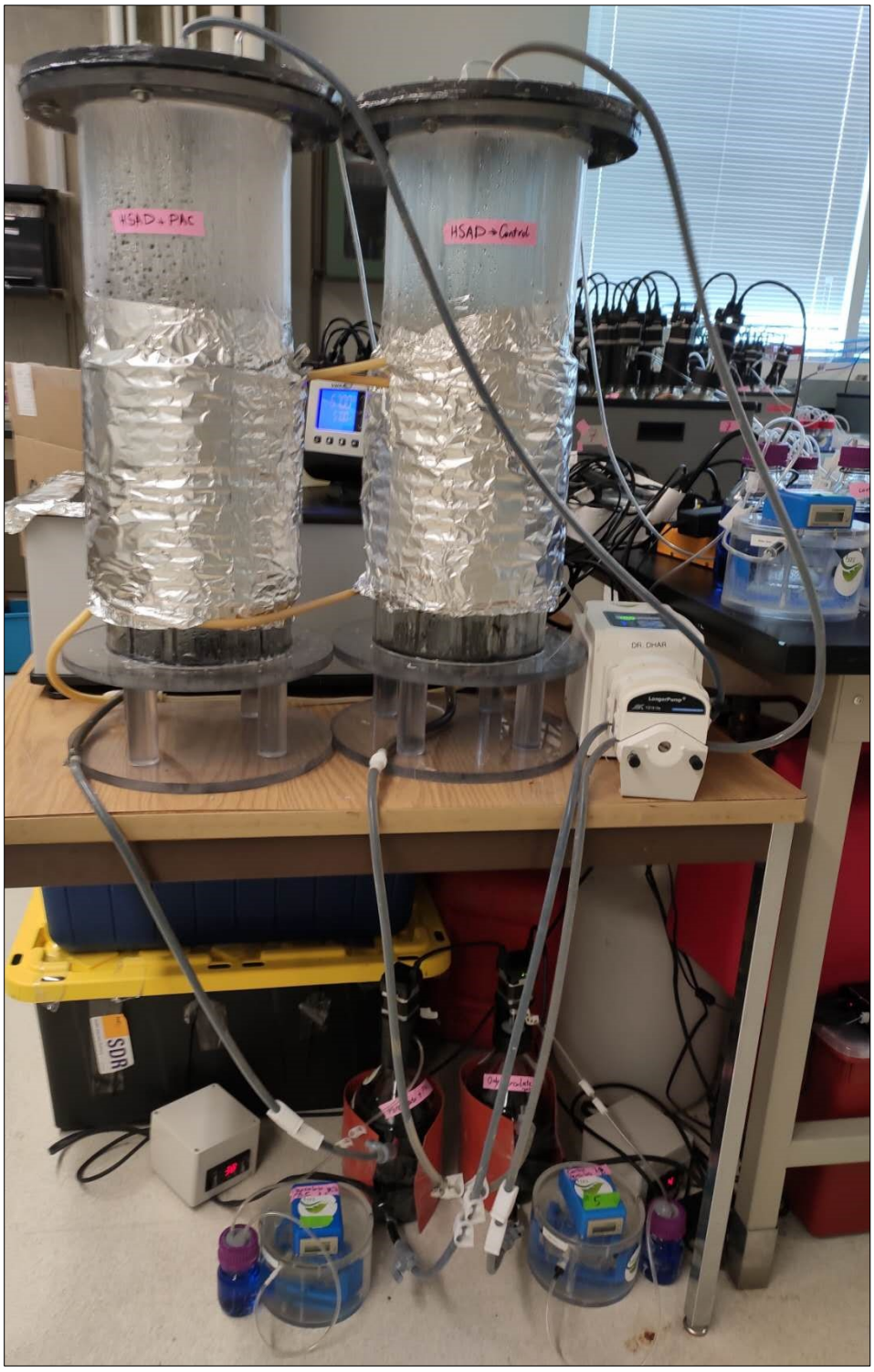


Figure A-3. HSAD set-up (control and PAC-enriched bioreactors).

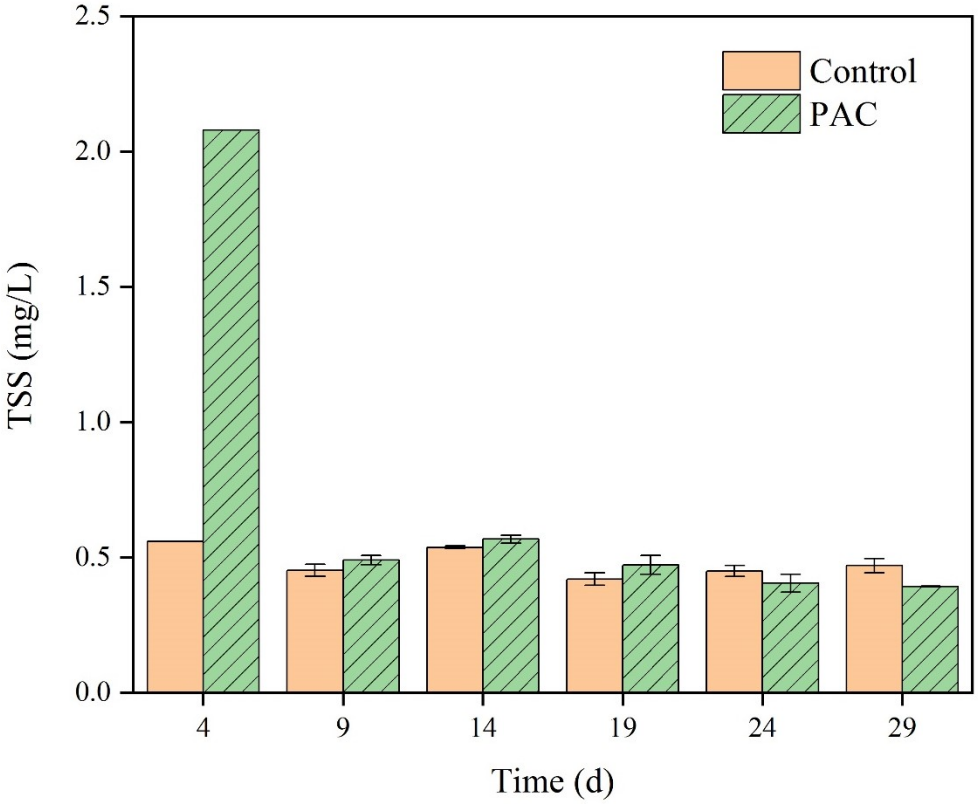


Figure A-4. The variation of TSS content in the PAC-enriched percolate and control tanks.

Table A-1. The ratio of VFA/Alkalinity in the PAC-amended and control digester.

Day	Control	PAC-Amended
4	0.26	0.27
9	0.87	1.10
14	0.55	0.53
19	0.43	0.28
24	0.40	0.33
29	0.28	0.21

Equation A-1

According to Koster (1986):

$$NH_3\ (FAN) = TAN\ \times (1 + \frac{10^{-pH}}{10^{-\left(0.09018+\frac{2729.92}{T(K)}\right)}})^{-1}$$

Where NH_3: Free ammonia-nitrogen (FAN) (mg/L)

TAN: Total ammonia-nitrogen (mg/L)

T: Temperature (K)

Table A-2. Analysis of variance.

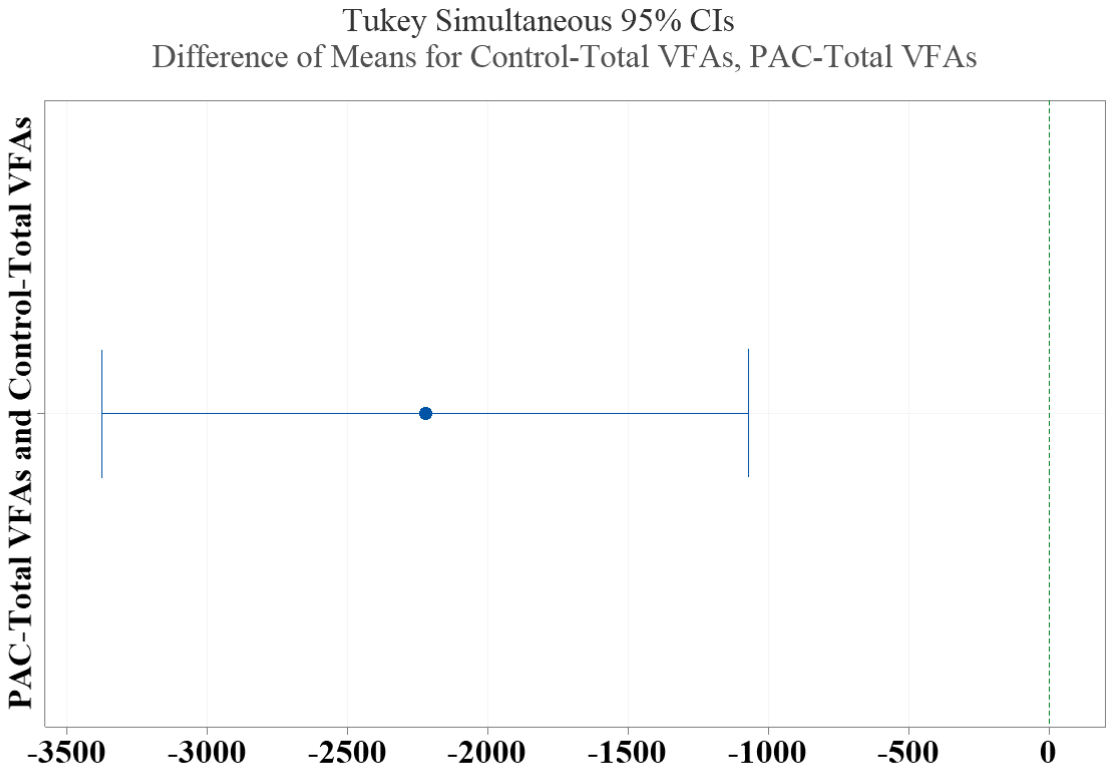
Source	DF	Seq SS	Contribution	Adj SS	Adj MS	F-Value	P-Value
Factor	1	7408683	87.77%	7408683	7408683	28.70	0.006
Error	4	1032730	12.23%	1032730	258183		
Total	5	8441413	100.00%				

Tukey Pairwise Comparisons

Table A-3. Grouping information using the Tukey method and 95% confidence.

Factor	N	Mean	Grouping
Control, Total, Day 29	3	5922	A
PAC, Total, Day 29	3	3700	B

Means that do not share a letter are significantly different.



If an interval does not contain zero, the corresponding means are significantly different.

Figure A-5. Difference of means for the control-Total VFAs and PAC-Total VFAs based on the Tukey simultaneous comparison at 95% confidence interval.

Table A-4. Tukey simultaneous tests for differences of means.

Difference of Levels	Difference of Means	SE of Difference	95% CI	T-Value	Adj P-Value
PAC, Acetate – Control, Acetate	529.0	58.7	(366.0, 692.1)	9.01	0.001

Individual confidence level = 95.00%

Minitab output:

WORKSHEET 5

One-way ANOVA: Control, Total, Day 29, PAC, Total, Day 29

Method

Null hypothesis	All means are equal
Alternative hypothesis	Not all means are equal
Significance level	$\alpha = 0.05$

Equal variances were assumed for the analysis.

Factor information

Factor	Levels	Values
Factor	2	Control, Total, Day 29, PAC, Total, Day 29

Analysis of variance

Source	DF	Seq SS	Contribution	Adj SS	Adj MS	F-Value	P-Value
Factor	1	7408683	87.77%	7408683	7408683	28.70	0.006
Error	4	1032730	12.23%	1032730	258183		
Total	5	8441413	100.00%				

Model summary

S	R-sq	R-sq(adj)	PRESS	R-sq(pred)
508.117	87.77%	84.71%	2323643	72.47%

Means

Factor	N	Mean	StDev	95% CI
Control, Total, Day 29	3	5922	652	(5108, 6737)
PAC, Total, Day 29	3	3700	303	(2885, 4514)

Pooled StDev = 508.117

Tukey Pairwise Comparisons

Grouping information using the Tukey method and 95% confidence.

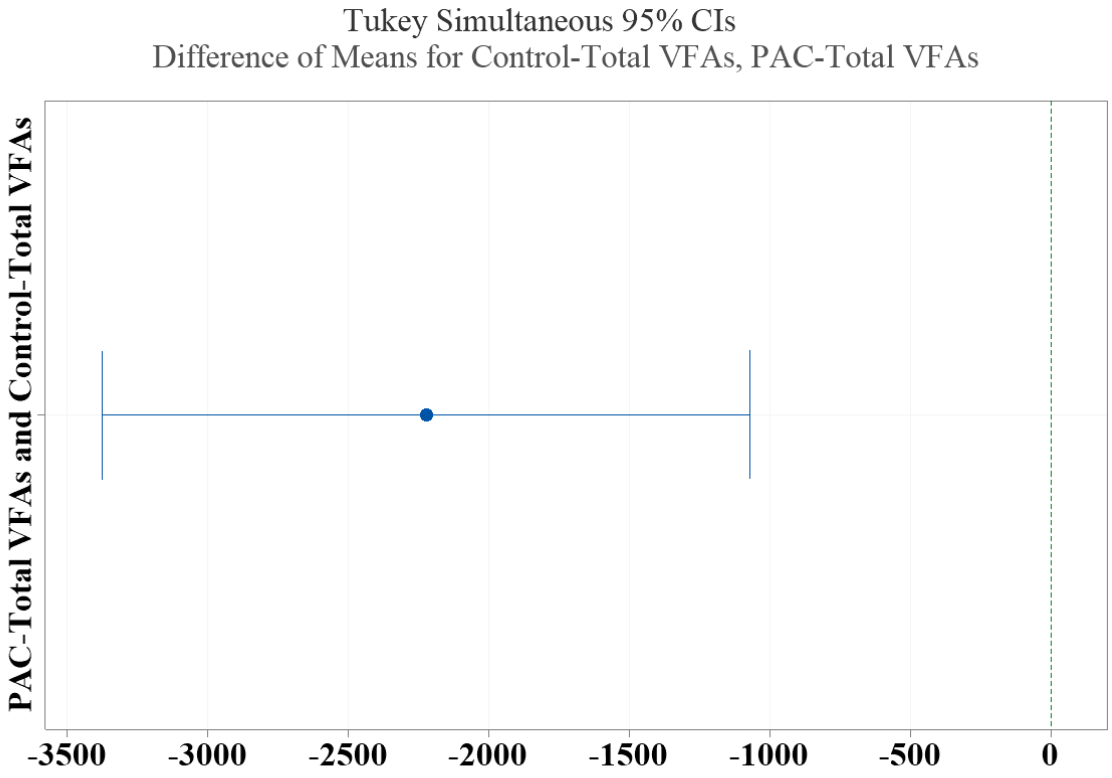
Factor	N	Mean	Grouping
Control, Total, Day 29	3	5922	A
PAC, Total, Day 29	3	3700	B

Means that do not share a letter are significantly different.

Tukey simultaneous tests for differences of means

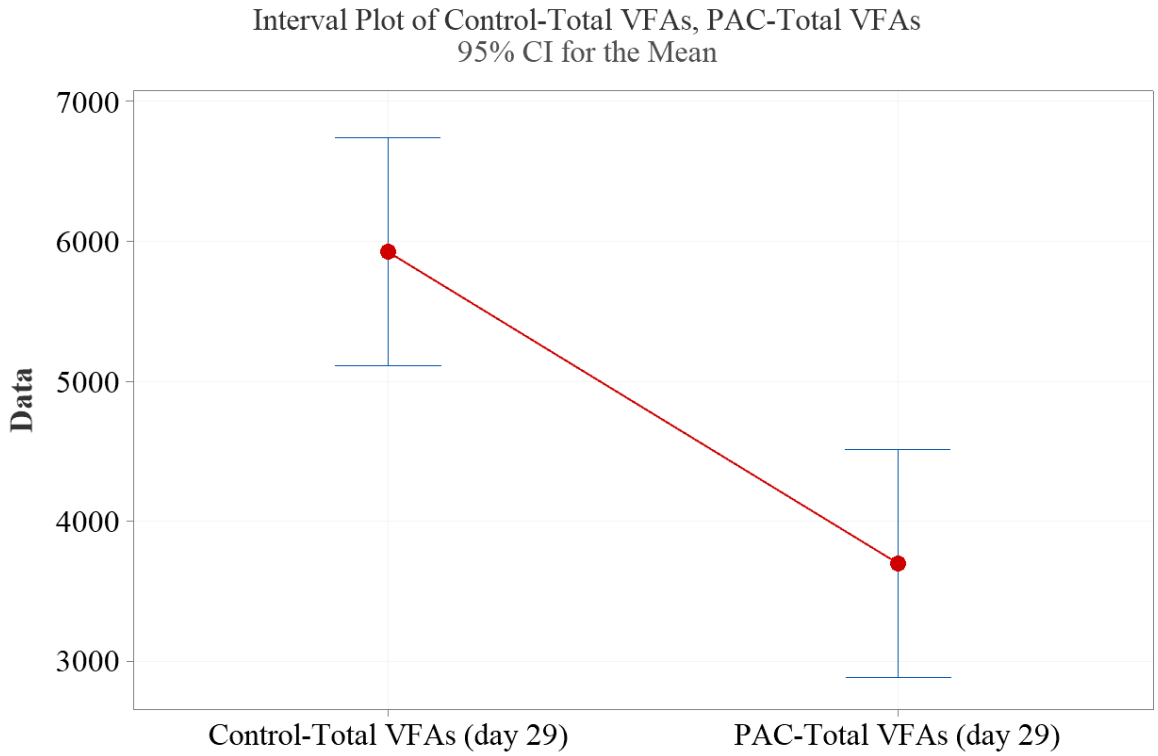
Difference of Levels	Difference of Means	SE of Difference	95% CI	T-Value	Adjusted P-Value
PAC, Total, - Control, Total	-2222	415	(-3374, -1071)	-5.36	0.006

Individual confidence level = 95.00%



If an interval does not contain zero, the corresponding means are significantly different.

Figure A-6. Difference of means for the control-Total VFAs and PAC-Total VFAs based on the Tukey simultaneous comparison at 95% confidence interval.



The pooled standard deviation is used to calculate the intervals.

Figure A-7. Interval plot for the control-Total VFAs and PAC-Total VFAs on day 29 based on the Tukey simultaneous comparison at 95% confidence interval.

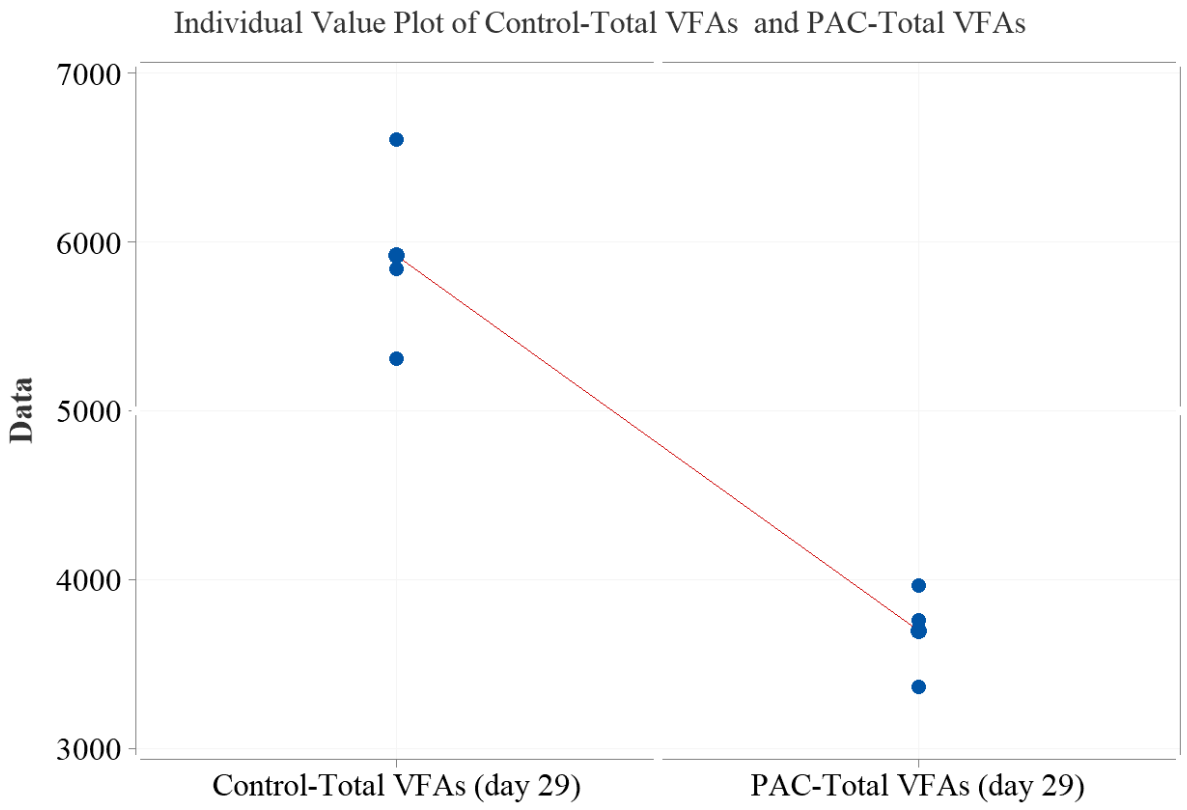


Figure A-8. Differences in the individual values for the control-Total VFAs and PAC-Total VFAs on day 29 based on the Tukey simultaneous comparison at 95% confidence interval.

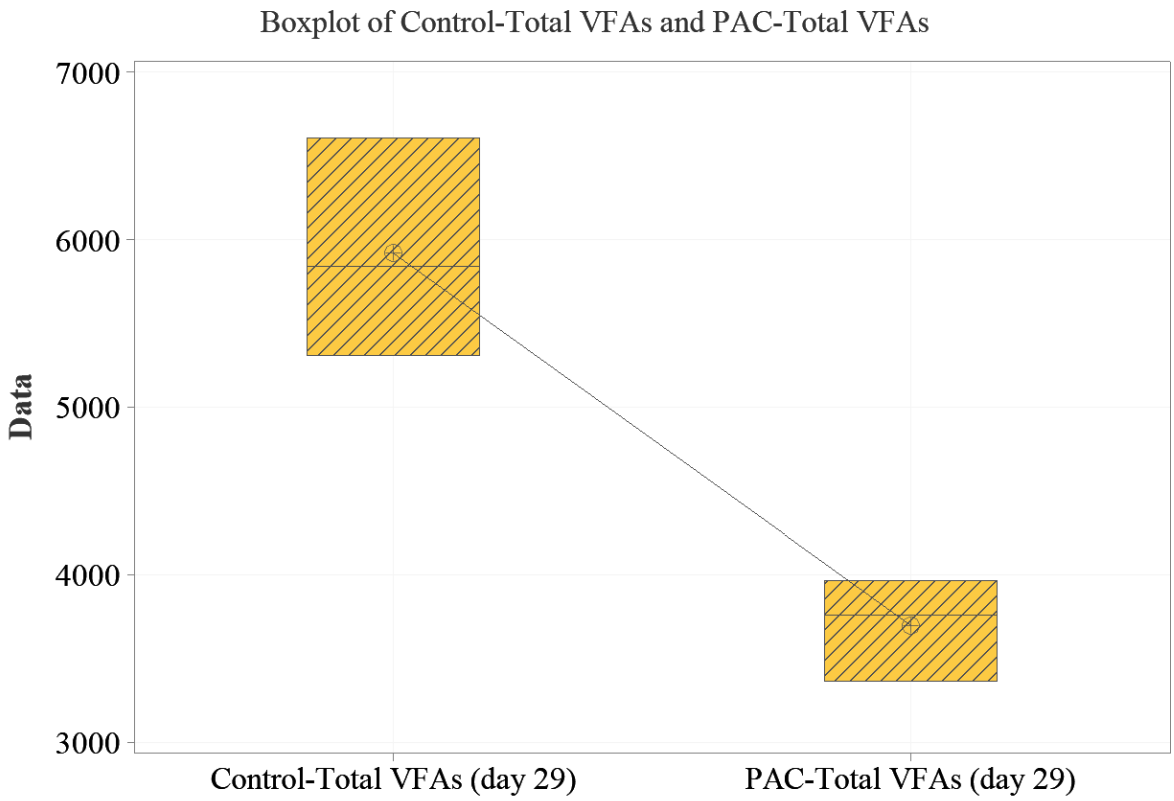


Figure A-9. Box plot for the control-Total VFAs and PAC-Total VFAs on day 29 based on the Tukey simultaneous comparison at 95% confidence interval (n =3, α = 0.05)

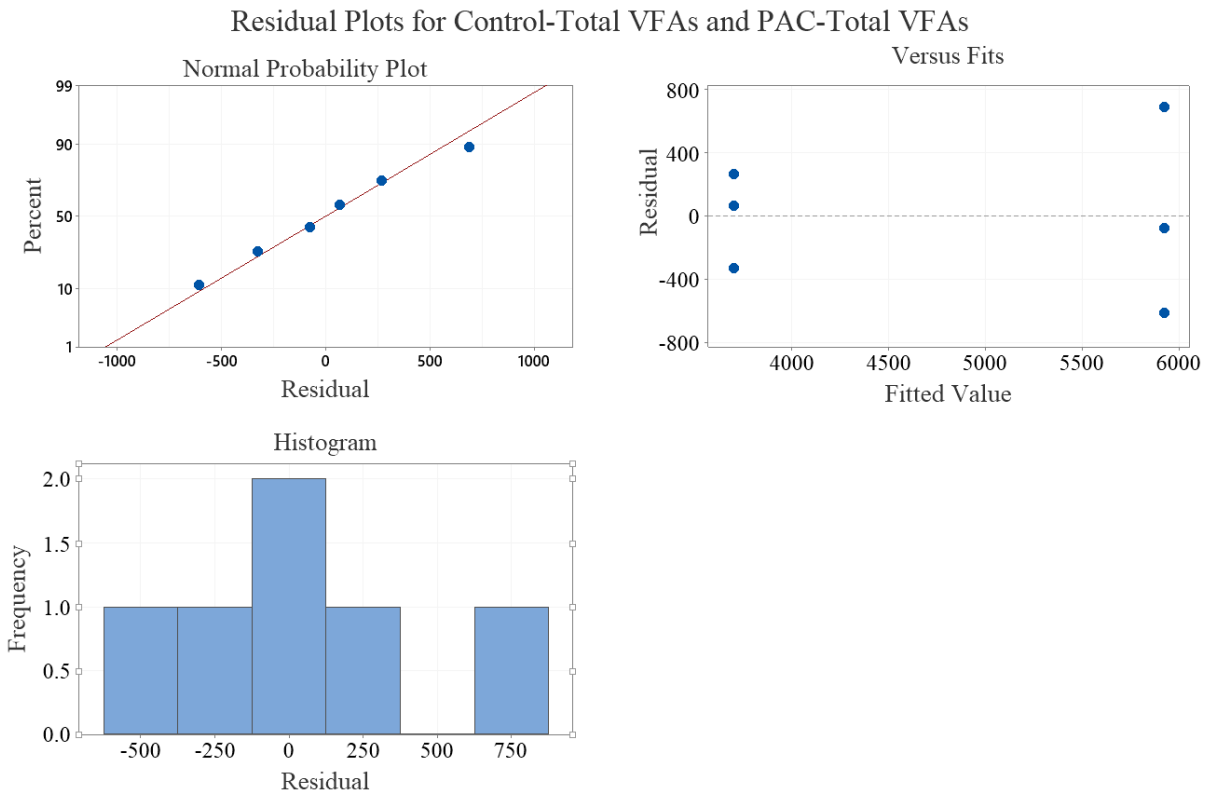


Figure A-10. Residual plots; normal probability linear ship plot, histogram plot and versus fits); for the control-Total VFAs and PAC-Total VFAs on day 29 based on the Tukey simultaneous comparison at 95% confidence interval (n =3, α = 0.05)

Appendix B

Supplementary Information for Chapter 4

Figure B-1. Photograph of experimental setup (Six-HSAD).

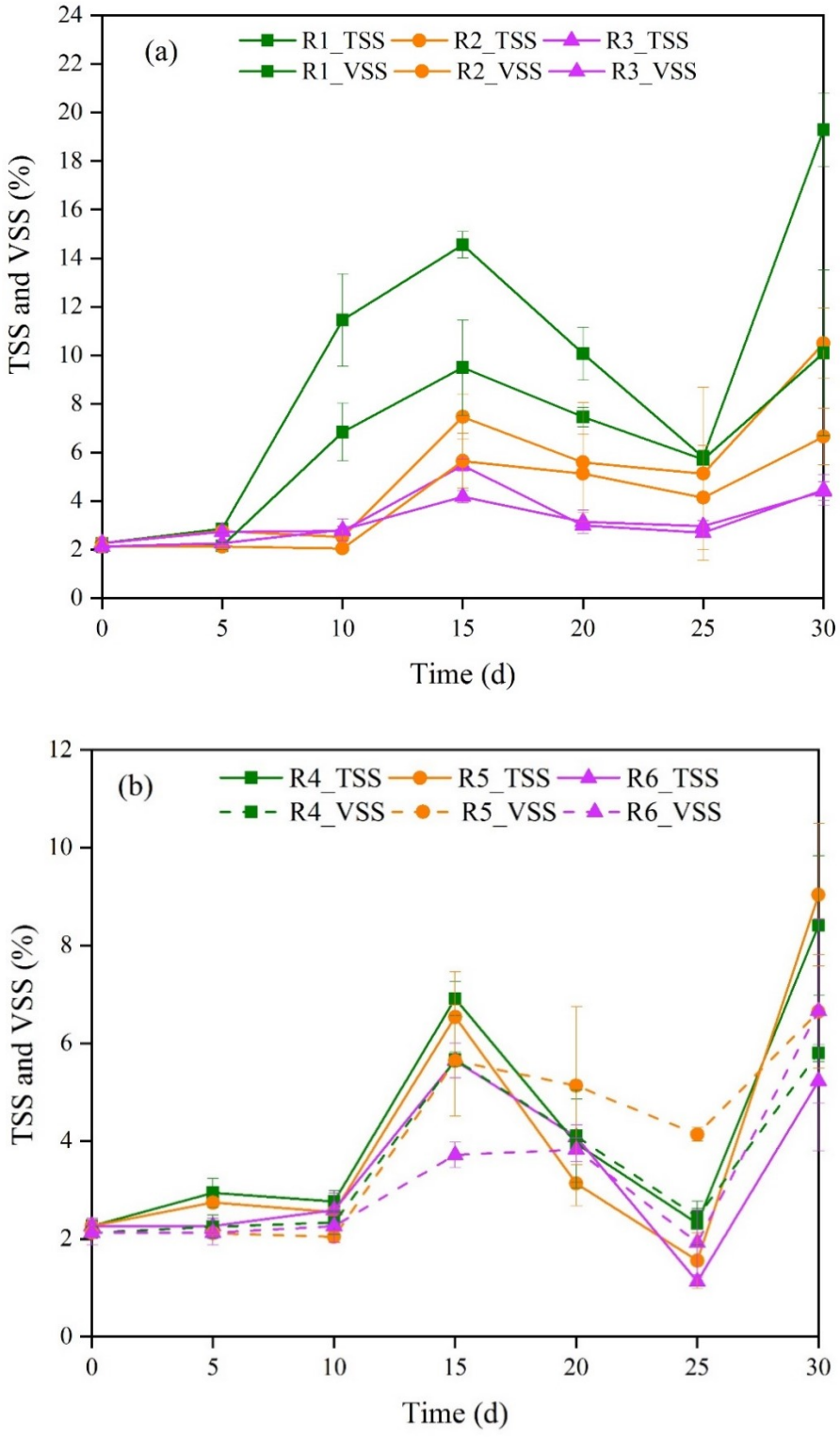


Figure B-2. The variations of TSS and VSS at a) different F:I ratios (constant PRT of 2.5 h/d), and b) different PRTs (constant F:I ratio of 2).

Table B-1. Results from batch biomethane potential (BMP) tests for estimation of methane production from the inoculum. Note. Tests were conducted with two 0.5-L anaerobic glass bioreactors at an F:I ratio of 2. Each reactor was loaded with 180 grams of inoculum and 220 grams of deionized water.

	BMP Tests (resembling F:I ratio of 2)			
	Methane Production Yield (mL)		Cumulative Methane Production (mL)	
Day	Reactor 1	Reactor 2	Reactor 1	Reactor 2
1	64	79	64	79
2	56	70	120	149
3	52	69	172	218
4	48	56	220	274
5	47	75	267	349
6	49	65	316	414
7	49	65	365	479
8	46	64	411	543
9	46	64	457	607
10	44	51	501	658
11	37	56	538	714
12	32	50	570	764
13	38	58	608	822
14	20	46	628	868
15	35	63	663	931
16	22	48	685	979
17	22	42	707	1021
18	26	48	733	1069
19	28	37	761	1106
20	42	57	803	1163
21	46	50	849	1213
22	62	32	911	1245
23	108	58	1019	1303
24	178	51	1197	1354
25	182	52	1379	1406
26	178	55	1557	1461
27	137	56	1694	1517
28	136	42	1830	1559
29	58	30	1888	1589
30	45	28	1933	1617

Table B-2. The total alkalinity in the R1 to R6 digesters at different F:I ratios and PRTs.

Day	Total alkalinity (mg/L as $CaCO_3$)					
	R1	**R2**	**R3**	**R4**	**R5**	**R6**
0	8,655	8,655	8,655	8,655	8,655	8,655
5	11,066	12,037	11,726	11,964	12,037	12,735
10	15,957	11,919	12,200	12,007	11,919	12,139
15	15,446	15,612	13,541	14,280	15,612	18,744
20	15,429	14,385	13,813	14,719	14,385	13,527
25	16,111	15,531	14,629	14,631	15,531	13,363
30	21,370	18,030	15,398	17,005	18,030	18,562

Table B-3. Initial elemental analysis of OFMSW and biosolid; CHNO (wt.%), and their molecular formula.

Item	**Elemental composition (%)**				**C/N ratio**	**Empirical molecular formula**
	C	**H**	**N**	**O**		
OFMSW	37.14	4.93	2.68	31.72	13.90	$C_{16}H_{26}O_{10}N$
Biosolids	30.43	4.28	4.57	20.96	6.66	$C_8H_{13}O_4N$

Table B-4. The elemental analysis of initial substrate and final digestate in the six high-solids digesters, and their molecular formula.

	Initial Substrate (wt.%)				Final Digestate (wt.%)			
	C	**H**	**N**	**O**	**C**	**H**	**N**	**O**
R1	33.56	4.58	3.68	25.99	27.26	3.69	2.88	20.16
	$C_{11}H_{17}O_6N$				$C_{11}H_{18}O_6N$			
R2	34.70	4.69	3.36	27.81	29.52	3.77	2.58	21.35
	$C_{12}H_{20}O_7N$				$C_{13}H_{20}O_7N$			
R3	35.29	4.75	3.20	28.75	30.98	4.02	2.30	22.16
	$C_{13}H_{21}O_8N$				$C_{16}H_{25}O_8N$			
R4	34.70	4.69	3.36	27.81	24.96	3.2	2.44	20.43
	$C_{12}H_{20}O_7N$				$C_{12}H_{18}O_7N$			
R5	34.70	4.69	3.36	27.81	29.52	3.77	2.58	21.35
	$C_{12}H_{20}O_7N$				$C_{13}H_{20}O_7N$			
R6	34.70	4.69	3.36	27.81	28.15	3.61	2.63	20.52
	$C_{12}H_{20}O_7N$				$C_{13}H_{19}O_7N$			

Appendix C

Materials and Equipment

- Agitator and Controller (Model: for GL45 flasks, Bioprocess Control Co., Lund, Sweden).
- Total Alkalinity Kit (25-400 mg/L $CaCO_3$, HACH reagent kits, Hach Company, Loveland, Colorado, USA).
- Total Ammonia-Nitrogen Kit (High range digestion solution for COD 20-1500 mg/L, HACH reagent kits, Hach Company, Loveland, Colorado, USA).
- Balance (Model: AB204-S/FACT, Mettler Toledo Co., Greifensee, Siwtzerland)
- Bioreactor Tank (Working volume: 2 L, fabricated by Integrated Sustainable Environmental Services (ISES), Vaughan, Ontario, Canada).
- Total COD Kit (High range digestion solution for COD 20-1500 mg/L, HACH reagent kits, Hach Company, Loveland, Colorado, USA).
- CO_2 Sequestration Chambers (Integrated Sustainable Environmental Services (ISES), Vaughan, Ontario, Canada).
- Cylindrical Polycarbonate Bioreactors (Total dimension r×h: 21.5 cm × 67 cm, fabricated by Integrated Sustainable Environmental Services (ISES), Vaughan, Ontario, Canada).
- Elemental Analysis (CHNS – dry-ash free basis, wt.%): measured using 2400 Series II CHNS analyzer (PerkinElmer, USA), and oxygen content was measured using vario MICRO cube (Elementar, HANAU, Germany).
- Flexible Latex Tubing (Amber latex rubber tubing, VWR, Avantor Company, Radnor, Pennsylvania, USA).
- Flexible Plastic Tubing (Model: 1481281, Tygon® tubing E3603, Buch & Holm A/S, Herlev, Denmark).
- Furnace (Thermolyne small benchtop muffle furnace, Thermo Fisher Scientific Inc., USA).
- Gas Flow Meter (Benchtop lab-scale ISES-GasMeter, fabricated by Integrated Sustainable Environmental Services (ISES), Vaughan, Ontario, Canada).
- Glass Microfiber Filter (Grade: 934-AH®, Whatman® Schleicher & Schuell®, Sigma-Aldrich, Inc., St. Louis, Missouri, USA).

- ❖ Heated Circulating Water Bath (Model: AD 28L HT, VWR, Avantor Company, Radnor, Pennsylvania, USA).
- ❖ Heating Tape (Dimension 40cm*170cm, 110v and 200w, SunRobot, China).
- ❖ High-speed rotary grinder (COSUAI Company, model CS-700).
- ❖ Hydrochloric Acid (HCl, purity 36.5-38.0%, concentrated reagent 12 M, Certified ACS, VWR, Avantor Company, Radnor, Pennsylvania, USA).
- ❖ Ion Chromatography Auto-sampler Solutions (Dionex AS-AP, Thermo Fisher Scientific Inc., Sunnyvale, California, USA).
- ❖ Ion Chromatography System (Dionex ICS-2100, Thermo Fisher Scientific Inc., Sunnyvale, California, USA).
- ❖ Lab Autosampler Vial (Model: 2 mL Clear Target DP ID™ Vial, National Scientific, Thermo Fisher Scientific Inc., Rockwood, Tennessee, USA).
- ❖ Manual Pipette (Variable adjustable volume pipettes, 10-100 µL and 1-10 mL, PhysioCare Concept®, Eppendorf™ Research plus™, Fisher Scientific Company, Ottawa, Ontario, Canada).
- ❖ Oven (Isotemp Oven, Fisher Scientific Company, Ottawa, Ontario, Canada).
- ❖ Peristaltic Pump (Model: YZ1515x, LongerPump®, Longer Precision Pump Co., Ltd., Tech Industrial Development Zone Baoding, Hebei, China).
- ❖ pH Meter (AR15 pH Meter, Thermo Fisher Scientific Inc., Pittsburgh, Pennsylvania, USA).
- ❖ Powdered Activated Carbon (Alfa Aesar, Thermo Fisher Scientific Inc., Ottawa, Ontario, Canada).
- ❖ Pump Tubing (Model: RK-96400-24, Peroxide-Cured Silicone, Masterflex L/S®, Cole-Parmer Co., Vernon Hills, Illinois, USA).
- ❖ Pump Tubing (Model: RK-96400-24, Peroxide-Cured Silicone, Masterflex L/S®, Cole-Parmer Canada Co., Montreal, Quebec, Canada).
- ❖ Recirculating Water Bath (Model: AD 28L HT [AD28LH200-V11B], VWR® Heated Circulating Baths, USA).
- ❖ Sodium Hydroxide (NaOH, purity 99.1%, Certified ACS, Fisher Chemical, Fisher Scientific Company, Reagent Lane Fair Lawn, New Jersey, USA).
- ❖ Syringe (BD 5 ml Disposable Syringe with Luer-Lok Tip, Fisher Scientific, Thermo Fisher Scientific Inc., Pittsburgh, Pennsylvania, USA).

- ❖ Syringe Filters (Nylone 0.2 µm and 0.45 µm, Fisher Scientific, Thermo Fisher Scientific Inc., Pittsburgh, Pennsylvania, USA).
- ❖ Ultrapure Water (HiPerSolv CHROMANORM® for HPLC, VWR Chemicals BDH®, VWR, Avantor Company, Radnor, Pennsylvania, USA).
- ❖ Ultrasonic Cleaner (Model: B3200R-4, Bransonic®, Branson Ultrasonic Corporation, Danbury, Connecticut, USA).
- ❖ Vacuum Filter Flask Setup (Motor Model: 0211-V45N-G8CX, GAST Manufacturing, MFG Corporation, Benton Harbor, Michigan, USA).
- ❖ Vortex Mixer (Model: 945404, Fisherbrand™ STD vortex mixer 120v, Fisher Scientific, Thermo Fisher Scientific Inc., Pittsburgh, Pennsylvania, USA).
- ❖ Water Bath (Model: WB20, PolyScience®, Niles, Illinois 60714-4516 USA).
- ❖ Weighing Dish (20 mL Aluminum Weighing Dish, Fisher Scientific, Thermo Fisher Scientific Inc., Pittsburgh, Pennsylvania, USA).