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Biogas Production from Brown Grease and the Kinetic Studies

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<http://dx.doi.org/10.5772/intechopen.74083>

Abstract

Food wastes were mostly treated in landfills due to economic reasons, which create rich organic leachate and soil contamination risks. An alternative for food waste treatment is to use the anaerobic digestion technique to reduce the organic content, as well as to recover bioenergy in the form of biogas. In this chapter, a common type of food waste brown grease was selected and investigated using a pilot-scale high-rate completely mixed digester, and the digestibility, biogas production, and the impact of blending of liquid waste streams from a nearby pulp and paper mill were assessed. Using a pseudo-first-order rate law, the observed degradation constant was estimated to be 0.10–0.19 d⁻¹ compared to 0.03–0.40 d⁻¹ for other organic solids. These results demonstrate that brown grease is a readily digestible substrate that has excellent potential for energy recovery through anaerobic digestion.

Keywords: brown grease, biogas, anaerobic digestion, renewable energy

1. Introduction

Biogas as an alternative source of energy is gaining more traction throughout several nations of the world [1]. Researchers have been conducting massive experiments on evaluating the conversion of miscellaneous wastes like animal manure, municipal solid waste, energy crops, municipal biosolids, food waste, and so on to biogas [2–4]. Biogas as an end-product could be produced from either artificially engineering anaerobic digester processing or naturally through the organic waste decaying process. Both the artificial and natural processes will include the main steps in anaerobic digestion; these are: hydrolysis, acidogenesis, and methanogenesis. Optimization of biogas generation from artificially engineering the anaerobic

system is centrally focused on the digester design and operation, although it has been stated that the feedstock is as important as the digester technology.

A type of food waste, brown grease (BG) was selected here for investigation. In the United States, there are 3800 million pounds of trap grease produced every year (per National Renewable Energy Laboratory Report [5]). Considering 100% conversion efficiency, approximately 14 million m³ of methane could be generated from these grease wastes [6, 7]. This substantial amount of energy could be used in different ways. Also, the environmental effect would be minimized compared to the effect of dumping to landfills [8].

BG is a mixture of fat, oil and grease from animal fat, vegetable oil, and other grease typically found in grease interceptors in restaurants and food industries [9]. Most of the time BG will be disposed to landfill, as the landfill treatment cost is only 5 cents per pound of brown grease [5]. However, landfills will generate some side effects such as soil and water pollution; these side effects will make the soil sterile and unusable to support plant life [5] and then garner ever-growing environmental concerns. These problems have a significant negative impact on the industrial cost and environmental effect of the BG treatment processes [10]. Anaerobic digestion technique is a viable option of the BG treatment [11]. The benefits of using anaerobic digestion include that the technique requires less reactor size, eliminates off-gas air pollution, produces less sludge, and generates substantial amounts of biogas as energy recovery; the generated biogas could then support the plant operation after converted to mechanical energy [12–14]. However, nowadays, only a small portion of BG has been treated by anaerobic digestion process, mainly because the biogas energy benefit cannot meet the anaerobic process investment [15, 16].

Kinetic simulation models of the anaerobic digestion process have been used to predict the digestion patterns and help to optimize design parameters of digestion reactors. As we have known that anaerobic digestion is a complex microorganism reaction process, to simplify the model, a pseudo first-order kinetic model can be applied to provide information, such as the relationship between feed rate and reactor size, and so on.

In this chapter, the biogas forming potential of BG as well as its COD and solids removal efficiency during the anaerobic digestion process was illustrated; moreover, system kinetic study has been performed to estimate the effects of input variations and substrate composition to the overall stability of the process. To make the investigation more comprehensive, various substrates from paper mill including foul condensate (FC) and screw press liquor (SPL) have also been introduced. The process parameters including substrate composition, hydraulic retention time (HRT), organic loading rate (OLR), and others have been studied in the process to get the optimal value.

2. Methodology

The anaerobic digestion system employed in this chapter includes three CSTR tanks: balance tank (BAL), facultative tank (FAC), anaerobic digester (AD), and a final batch sedimentation

tank (ST). The raw substrate streams were equalized in BAL and then been fed to FAC for bioaugmentation and predigestion, then the predigested substrates were pumped continuously into AD for digestion. During the treatment process, the mixing condition in AD vessel was checked periodically to confirm the accordance of sample. The BAL and FAC tanks were always operating under an internal mixing condition so that the pH adjusting and grease hydrolysis would be favored.

The whole evaluation process takes 343 days. Due to system maintenance recovery and feeding transition issues during the operation, five periods (S1–S5) of stable system performance were selected for assessment. **Table 1** summarizes the divided evaluation periods and correlated operating parameters. During each operating period, a steady-state operation was selected for further analysis. The settling tank was introduced at the 196th day. In S3 and S5, FC and SPL was fed as a cosubstrate to investigate their impact for the system.

The characteristics of BG, FC, and SPL were listed in **Table 2**. BG used in this study was obtained from a food waste plant in Houston, TX. Before feeding to the BAL, the stream was prescreened and the rest was dehydrated; lime was introduced to increased pH to neutral (see **Table 2**) in order to maintain the minimum microorganism activity; afterward, the BG was shattered by a beater to ensure complete emulsification with tap water.

	System start-up	S1	S2	S3	S4	S5
Date	4/13/11–7/26/11	7/27/11–8/7/11	8/8/11–10/24/11	10/25/11–12/7/11	12/8/11–2/29/12	3/1/12–3/21/12
Days of operation (d)	/	1–12	13–90	91–135	136–217	218–238
Days with consistent data (d)	/	1–12	34–43	107–133	184–217	218–238
Settling tank	No	No	No	Yes	Yes	Yes
Feeding	BG	BG	BG	BG + FC	BG	BG + SPL
Influent COD (mg L ⁻¹)	/	19,208 ± 1579	26,205 ± 2685	26,570 ± 6264	33,881 ± 9176	30,200 ± 1503
Influent VS (mg L ⁻¹)	/	10,367 ± 662	12,802 ± 925	10,139 ± 754	13,224 ± 3236	13,225 ± 1891
OLR*	/	2.0 ± 0.2	2.0 ± 0.2	0.8 ± 0.2	0.6 ± 0.2	0.9 ± 0.3
HRT*	/	7.3 ± 0.6	11.9 ± 1.1	15.2 ± 1.1	15.8 ± 1.9	11.0 ± 0.1
Activity	Seeding and initiating	Establish BG steady state	Reduce flow rate	Establish BG + FC steady state	Maintenance and recovery	Establish BG + SPL steady state

*OLR and HRT in S1 and S2 were calculated based on digester only (considering recycle); OLR and HRT in S3–S5 were calculated based on digester + sedimentation tank (not considering recycle).

Table 1. Feeding schedule and operating periods.

Parameter	Brown grease (BG) ^a ($\mu \pm \sigma$, n = 17)	Foul condensate (FC) ($\mu \pm \sigma$, n = 11)	Screw press liquor (SPL) ($\mu \pm \sigma$, n = 13)
COD (mg L ⁻¹)	910,634 ± 229,993	2973 ± 142	4498 ± 2020
dCOD (mg L ⁻¹)	/	2740 ± 125	609 ± 189
TS (mg L ⁻¹)	437,778 ± 91,348	406 ± 104	8768 ± 7957
VS (mg L ⁻¹)	372,111 ± 77,646	210 ± 14	3742 ± 1666
VS/TS ratio	0.85 ± 0.06	0.53 ± 0.1	0.5 ± 0.1
TSS (mg L ⁻¹)	/	357 ± 577	4048 ± 1750
VSS (mg L ⁻¹)	/	339 ± 461	1997 ± 875
VSS/TSS ratio	/	0.83 ± 0.25	0.49 ± 0.06
Alkalinity (mg L ⁻¹ as CaCO ₃)	/	205 ± 50	/
pH ^b	6.51 ± 0.77	9.28 ± 0.18	8.44 ± 0.83
TN (mg L ⁻¹)	/	52.2 ± 4	2.3 ± 0.1
TP (mg L ⁻¹)	/	0.24 ± 0.09	0.41 ± 0.04
Sulfide (mg L ⁻¹)	/	52.2 ± 20.5	/
Sulfate (mg L ⁻¹)	/	<40	/
Moisture content (wt%)	56 ± 9	/	/

^aIn BG, COD, TS, and VS are measured as mg/kg.

^bpH of BG was measured by suspending 100 g BG in 1 L tap water. Tap water has pH of 8.05 and alkalinity of 55 mg L⁻¹ as CaCO₃.

Table 2. Substrate characteristics.

During S3 and S5, instead of mixing with tap water, FC and SPL were introduced as cosubstrate, respectively. Compared with BG, FC and SPL have a relatively low COD concentration and solids content (see **Table 2**). Also, since BG contains enough amount of total nitrogen (TN) and total phosphorous (TP) for anaerobic digestion, no additional nutrients were added to the batch.

3. Results and discussion

3.1. Performance of anaerobic process

The summary of anaerobic operating parameters and results was listed in **Table 3**. For comparison purpose, typical values for anaerobic digestion processes were also listed. During the entire process, the pH in digester has been controlled at the neutral range (7.01–7.34, optimal pH range for anaerobic digestion 6.9–7.6) and the digestion occurred mostly at mesophilic temperature (34.3–37.9°C). The low DO concentration (< 0.1 mg L⁻¹) and ORP value (< -200 mV) indicate that the system is strictly anaerobic. VFA concentration is lower than

Operating periods	S1	S2	S3	S4	S5	Typical range
pH	7.34 ± 0.05	/	7.12 ± 0.08	7.10 ± 0.07	7.01 ± 0.17	6.5–8.5
T(°C)	36.0 ± 0.7	36.3 ± 0.7	34.3 ± 1.8	34.3 ± 2.1	37.9 ± 1.0	35–40
DO (mg L ⁻¹)	0.01 ± 0.00	/	0.06 ± 0.04	0.15 ± 0.05	0.10 ± 0.03	/
ORP (mV)	-209 ± 14	-228 ± 24	-243 ± 40	-247 ± 37	-263 ± 23	-400 to -150
TN (mg L ⁻¹)	591 ± 83	409 ± 37	237 ± 74	314 ± 50	306 ± 46	60–1000
TP (mg L ⁻¹)	3.4 ± 2.4	1.5 ± 0.4	0.9 ± 0.4	2.3 ± 1.1	2.2 ± 0.4	6–50
Alkalinity (mg L ⁻¹ as CaCO ₃)	3087 ± 282	/	1455 ± 457	2478 ± 291	2204 ± 222	1500–5000
VFA (mg L ⁻¹ as HAc)	274 ± 97	/	199 ± 76	394 ± 84	629 ± 378	<1800
COD removal efficiency (%)	42.1 ± 6.7	50.6 ± 5.8	82.3 ± 11.0	61.7 ± 12.3	53.5 ± 8.7	/
VS removal efficiency (%)	26.8 ± 7.9	37.1 ± 4.3	70.1 ± 8.4	65.6 ± 7.0	62.3 ± 7.2	/
CH ₄ content (%)	74.3 ± 2.0	74.6 ± 1.0	75.9 ± 1.9	74.6 ± 1.8	75.4 ± 1.0	/
CO ₂ content (%)	22.3 ± 1.3	/	23.9 ± 1.9	25.2 ± 1.8	24.2 ± 1.0	/
H ₂ S content (ppm)	38.2 ± 4.1	/	147.2 ± 34.8	185.2 ± 28.1	371.7 ± 127.6	/
CH ₄ yield (m ³ -CH ₄ Kg-VS ⁻¹)	0.99	0.66	0.52	0.48	0.45	0.30–1.00

Table 3. Operating conditions in each period.

400 mg L⁻¹ as HAc except S5 when the VFA level is somewhat elevated up to 630 mg L⁻¹ as HAc. TN and TP concentration in system is 230–600 mg L⁻¹ as N and 1–4 mg L⁻¹ as P respectively, which indicates enough nitrogen but slightly lower in phosphorus concentration.

Figure 1 shows the COD and VS variation and removal efficiency during each operating period. During S1 and S2, the ST has not been introduced to system yet, the effluent from AD was considered as final effluent and some of the sludge from AD was recycled to FAC manually that results for the higher effluent COD concentration (20,000–30,000 mg L⁻¹) compared with other stages (~10,000 mg L⁻¹). The COD removal efficiency in these periods is relatively lower than other periods, about 30–60% (**Figure 1a**). After ST was added (S3–S5), the effluent COD was kept in a relatively stable range (~10,000 mg L⁻¹) even if the influent COD was varied from 15,000 to 80,000 mg L⁻¹ (**Figure 1a**). This implies that sedimentation tank was efficiently in the elimination of a substantial amount of COD and polishing the quality of final effluent. With the stable effluent COD, during S3, FC was added as a cosubstrate and the initial COD loading was increased; thus, the COD removal efficiency was increased (70–95%, **Figure 1a**) to the highest value in the overall process.

During each operating period, VS variation has a similar trend with COD; the VS removal efficiency during S3–S5 did not change too much, in the range of 40–70% (**Figure 1b**), while the effluent VS concentration in S4 seems higher that may be due to the higher influent VS concentration. After added the ST, the VS removal efficiency was also improved from 20–40% to 40–70% (**Figure 1b**).

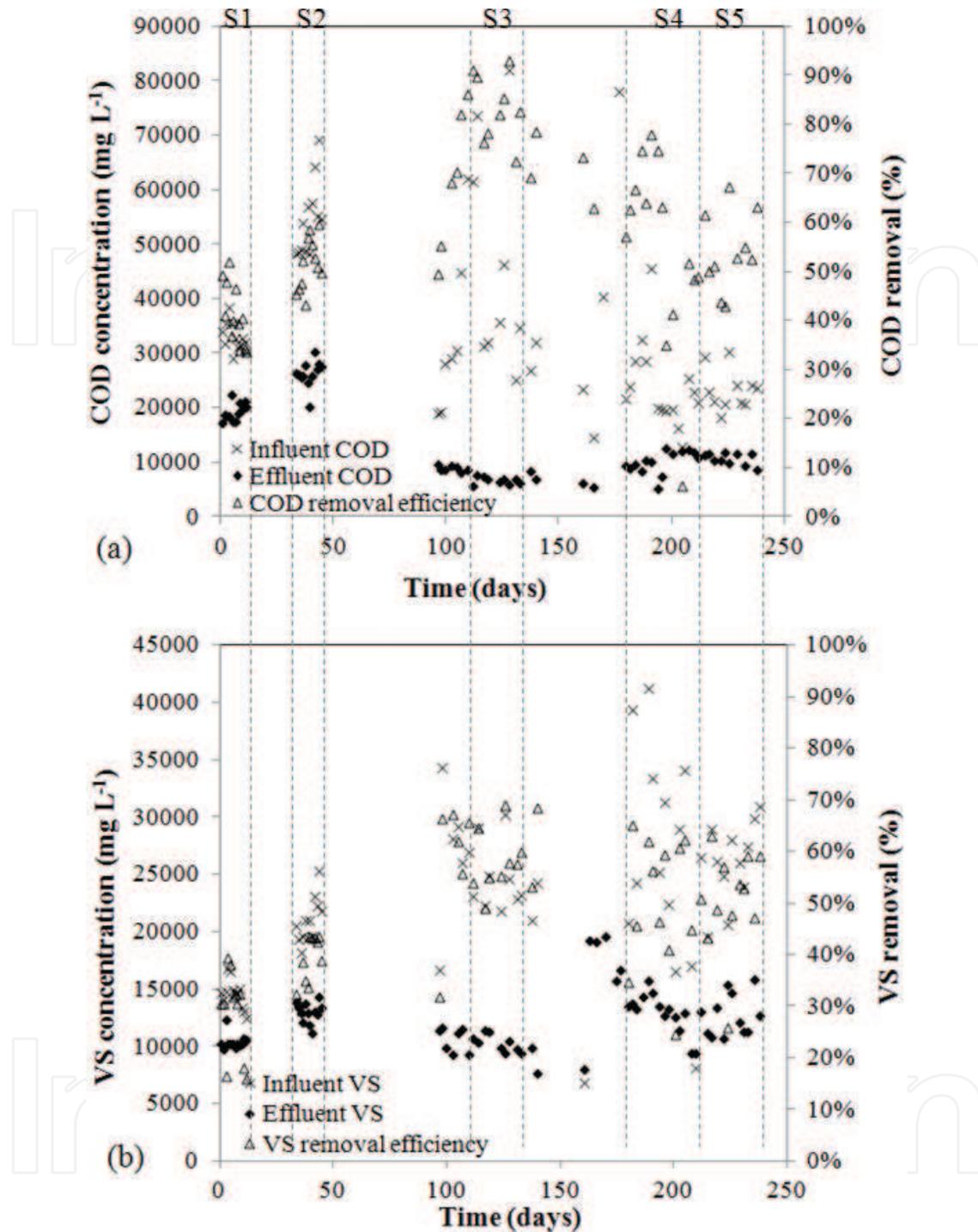


Figure 1. COD (a) and VS (b) concentration variation before and after AD, and their removal efficiency. Five stable operating periods (S1–S5) were marked.

The indicator of system organic removal is the volatile ratio (VS/TS and VSS/TSS) before and after AD. **Figure 2** shows the VS/TS ratio (**Figure 2a**) and VSS/TSS ratio (**Figure 2b**) in FAC and AD, respectively. In FAC, the volatile ratio is 0.84–0.86, and this ratio decreases to 0.66–0.69 in AD. The reduced ratio indicates that there was organic digestion since the inorganic parts should always be consistent in the anaerobic digestion process. Compared with the

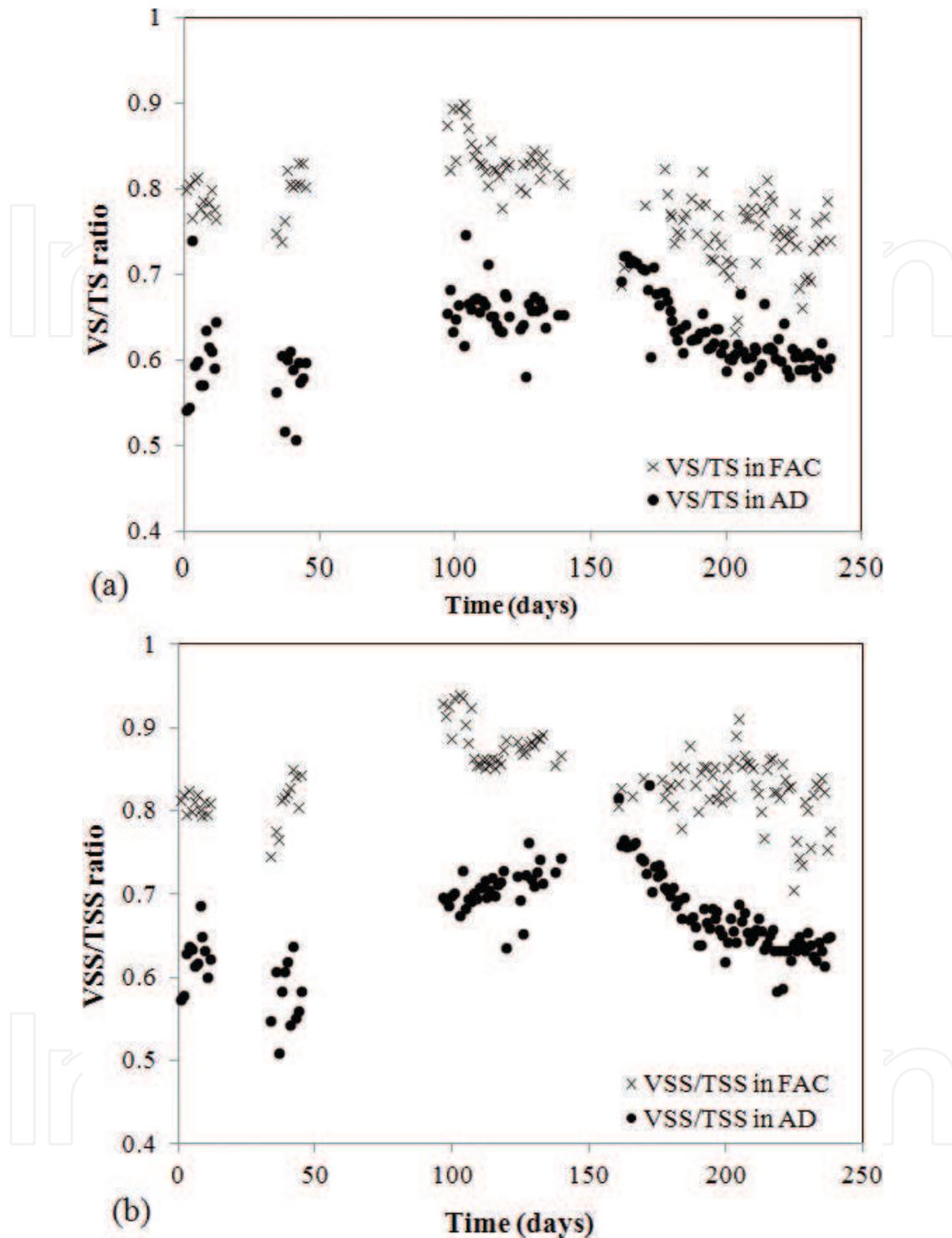


Figure 2. Volatile fraction including VS/TS (a) and VSS/TSS (b) in both FAC and AD.

dCOD reduction from FAC to AD (see Table 3), the decrease of volatile fraction should come of hydrolysis, acidogenesis, and methanogenesis together.

Figure 3 shows the scattered plot between system organic removal and organic loading rate in terms of COD (Figure 3a) and VS (Figure 3b). In S1 and S2, the OLR is higher than S3–S5 since the clarifier was not included and the OLR was calculated with considering the recycling

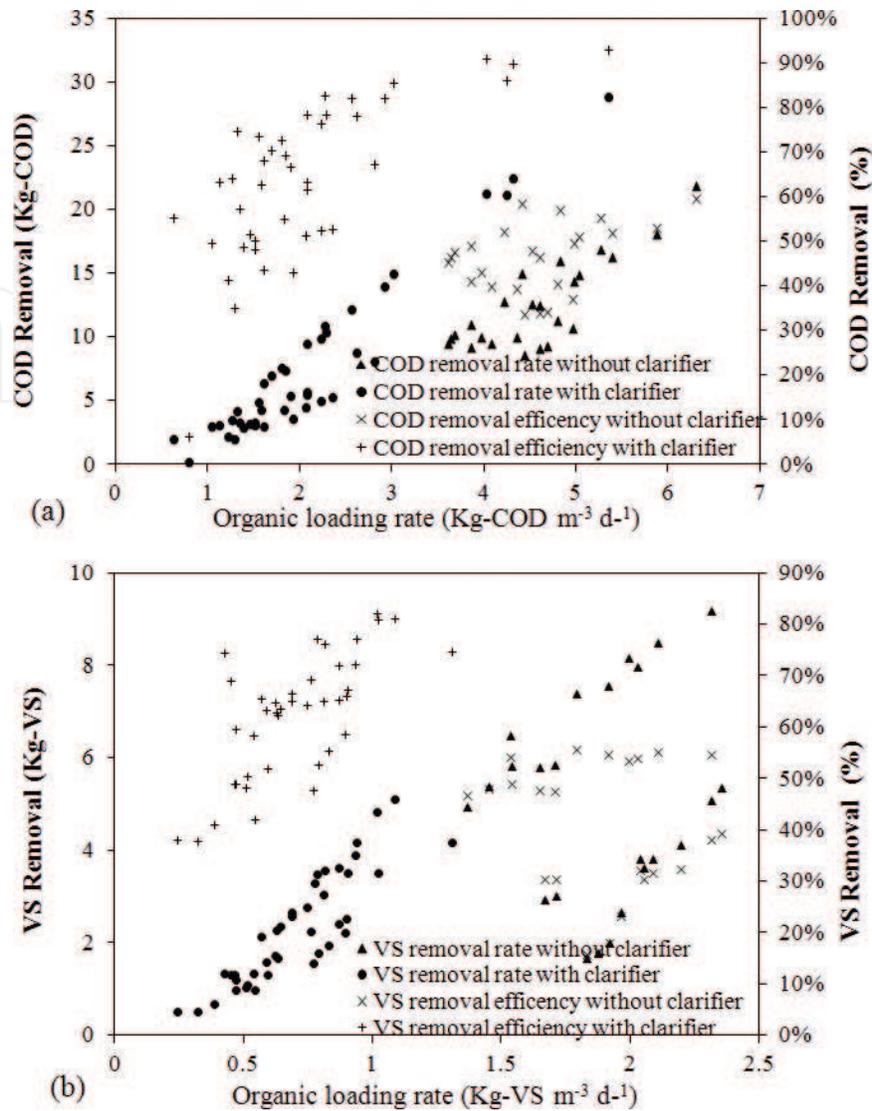


Figure 3. Scatter plots between organic loading rate and organic removal in terms of COD (a) and VS (b).

sludge. Both on COD and VS, the removal efficiency was not significantly affected by OLR variation, resulting in linear increase of COD and VS removal with respect to the applied OLR. This indicates that the system throughput could be improved within the OLR range applied during the evaluation periods to obtain higher organic removal efficiency.

VFA is also an important parameter to investigate the anaerobic process. As the source for methanogenesis, the system needs a certain amount of VFA; while VFA accumulation to greater than 1800 mg L⁻¹ has been shown suggested to significantly decrease pH and *in vitro* toxicity, thus somewhat alkalinity was needed to offset the extra amount of VFA as well. **Figure 4** shows the VFA concentration in FAC and AD as well as the alkalinity in AD. The mean alkalinity during overall process is 2122 mg L⁻¹, which is adequate for extra VFA. As shown in **Figure 4**, when substrate moves from FAC to AD, the average VFA concentration decreased from 800 to 413 mg L⁻¹, which indicates that FAC was efficiently augmenting VFA generation and improving the methanogenesis process in AD.

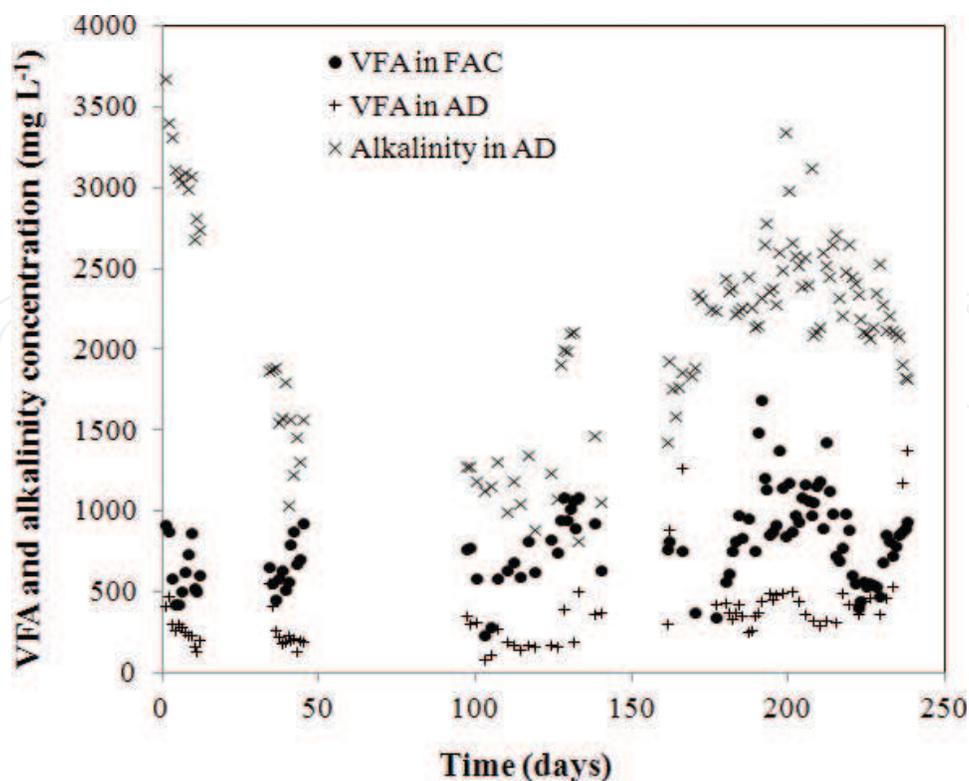


Figure 4. VFA variation before and after AD, and alkalinity level in AD.

3.2. Methane yield and kinetic analysis

The produced biogas has a consistently high CH₄ content (~75%, see **Table 3**). The other major gas (CO₂) consisted of the other ~25% by volume (**Table 3**) and trace gases (e.g., H₂S). From day 160–175, the system was recovered from system maintenance and the methane content built up from 40% to 75% quickly (**Figure 5**). During the entire evaluation, the average H₂S concentration was 189 ppm, significantly lower than that level that may cause H₂S toxicity (~1500 ppm).

The cumulative CH₄ production and digested VS in S3–S5 are shown in **Figure 6**. The methane yields of S3–S5 were calculated directly as the ratio of the two slopes. The value was reported based on VS removal because the organic content of the BG feed was mainly in the suspended solid phase. The methane yield of BG in S3–S5 was consistent in the range of 0.45–0.49 m³-CH₄ Kg-VS⁻¹ (at standard temperature and pressure, STP). All the gas volumes mentioned hereafter have been normalized to STP.

For the first two stages (S1 and S2), the apparent VS removal efficiency (25–40%, **Table 3**) was significantly lower than in S3–S5 (55–75%, **Table 3**) because ST had not been introduced to the system. Based on that, the effluent VS during S1 and S2 contains a large amount of biomass produced from the anaerobic digestion of brown grease. During S3–S5, ST was used to collect and recycle most of the generated biomass back to the AD, resulting in the higher organic removal efficiency. To estimate the BG conversion into biogas during S1 and S2, a mass balance analysis on solids before and after the AD was performed as follows:

$$(1-f)F = (1-\alpha)X + (1-\beta)Y \quad (1)$$

$$\alpha X + \beta Y = M \quad (2)$$

Eq. (1) represents the mass balance of the inorganic (fixed) solids where f is the volatile fraction of influent BG obtained from measurement (0.808 in S1 and 0.816 in S2), F is the mass flow of influent total solid (Kg d^{-1}), α and β are the volatile fraction (VS/TS) of biomass and undigested BG substrate, respectively ($\alpha \approx 0.80$). X and Y are the mass flow of biomass and undigested BG, respectively (Kg d^{-1}). Eq. (2) represents the VS composition in the effluent, where M is the mass flow of VS in the effluent (Kg d^{-1}). Using the solid measurements, we estimated that the generated biomass constitutes 25–50 wt% in the effluent.

Based on the mass balance results, the cumulative CH_4 production and the digested VS during S1–S5 were plotted in **Figure 6**. The methane yield was then calculated as the ratio of the slopes of the two lines in the respective period. The estimated methane yield in S1 ($0.40\text{--}0.49 \text{ m}^3\text{-CH}_4 \text{ Kg-VS}^{-1}$) was comparable with S3–S5 ($0.45\text{--}0.49 \text{ m}^3\text{-CH}_4 \text{ Kg-VS}^{-1}$). S2 has a higher methane yield ($0.58\text{--}0.77 \text{ m}^3\text{-CH}_4 \text{ Kg-VS}^{-1}$) at the cost of lower organic removal at higher organic loading (**Table 3**), which would require greater treatment effort for the digester effluent. During S3–S5, the organic removal was obviously higher with reduced methane yield. Therefore, the mode of process operation will depend on the treatment objective (better organic removal or higher methane yield). Also, the added cosubstrate (FC and SPL) did not adversely affect the methane yield during S3–S5 (**Table 3**).

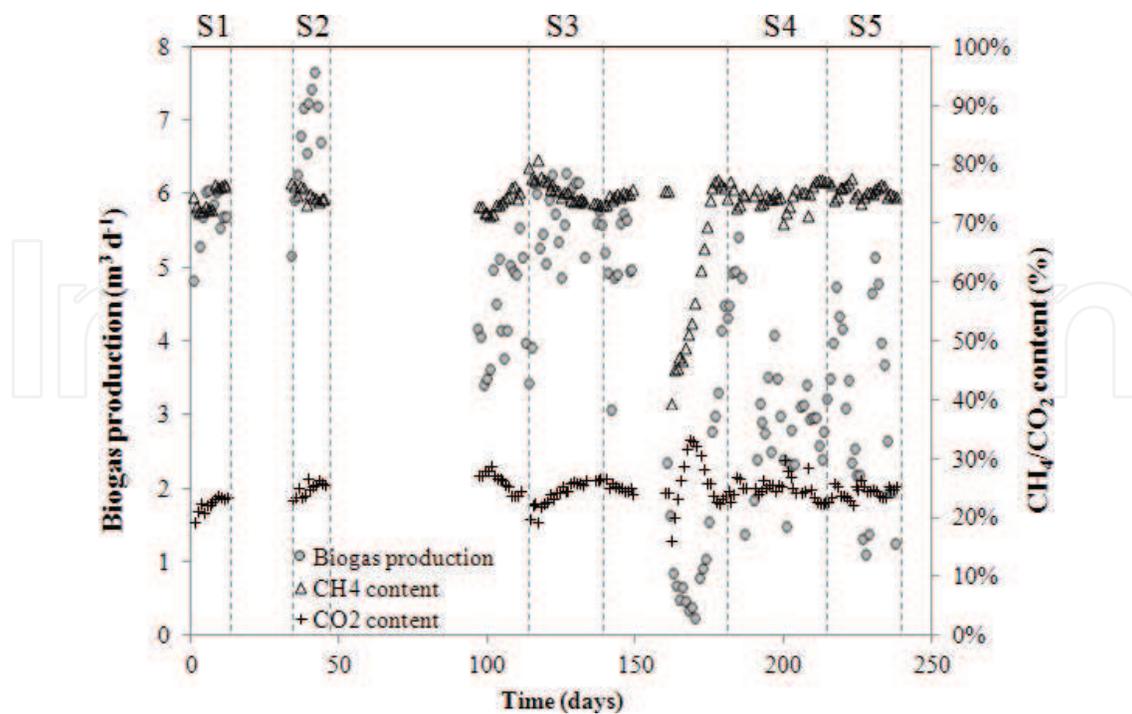


Figure 5. Measured daily biogas production and CH_4/CO_2 content.

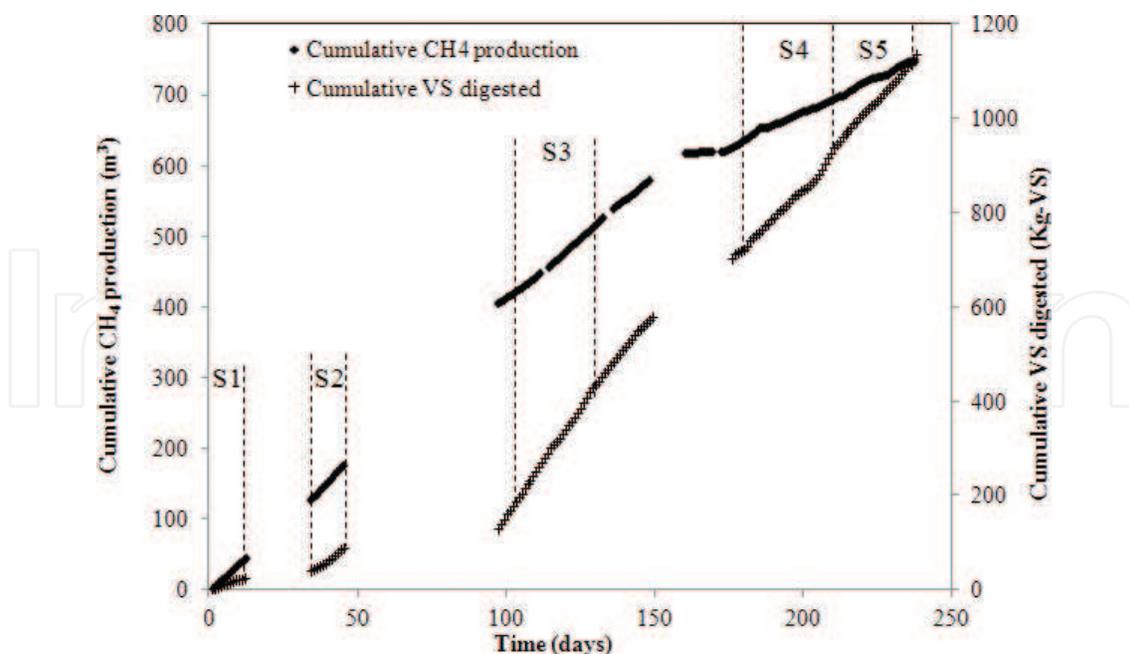


Figure 6. Cumulative CH₄ production at STP and cumulative VS digested during five selected stages (S1–S5). The slopes of each linear stage were used to calculate corresponding CH₄ yield. In S1 and S2, the mass of digested VS was corrected by biomass calculation.

A pseudo-first-order kinetic model was applied to analyze the substrate utilization. Similar approaches have been used earlier [2, 17]. The substrate concentration was calculated based on VS. For a CSTR at a steady state, the effluent concentration (C) can be estimated as:

$$C = \frac{C_0}{1 + k\theta} \quad (3)$$

where C_0 is the influent substrate concentration (mg L^{-1} VS), k is the first-order substrate utilization rate constant (d^{-1}), and θ is the HRT (d). The estimated k value is in a relatively consistent range of $0.10\text{--}0.19 \text{ d}^{-1}$ throughout the evaluation process.

For comparison purposes, the previous reported methane yields of food wastes and their first-order kinetic parameters are shown in **Table 4** [18–25]. Different degradation rate constants were obtained for different substrates and bench-scale reactors. Generally, the rate constants were in the range of $0.03\text{--}0.4 \text{ d}^{-1}$. The rate constant obtained in this study ($0.10\text{--}0.19 \text{ d}^{-1}$) has probably been adversely affected by the greater difficulty of controlling the digestion conditions (temperature and mixing) in a pilot-scale system due to the ambient temperature variation ($>15^\circ\text{C}$ diurnal change). It was slightly lower than that of municipal solid sludge in batch reactors ($0.2\text{--}0.4 \text{ d}^{-1}$), comparable to that of municipal solid sludge in CSTR (0.175 d^{-1}), and higher than that of canary grass in CSTR ($0.03\text{--}0.04 \text{ d}^{-1}$). The methane yields in this study range from 0.45 to $0.85 \text{ m}^3\text{-CH}_4 \text{ Kg-VS}^{-1}$, higher than earlier reported data ($0.11\text{--}0.42 \text{ m}^3\text{-CH}_4 \text{ Kg-VS}^{-1}$, **Table 4** [18–25]) from food wastes in solid digesters. The biogas quality produced by BG is excellent ($\sim 75\%$, **Table 3**), possibly due to the high lipid content of BG. These pilot-plant data suggest that BG can be effectively digested anaerobically for high-quality biogas production.

Substrate	Reactor type	Pseudo-first-order rate constant (d ⁻¹)	Methane yield (m ³ -CH ₄ Kg-VS ⁻¹)
Brown grease	CSTR	0.10–0.19	0.40–0.77
Municipal sludge	CSTR	0.175	0.309
Municipal sludge	Batch	0.2–0.4	/
Corn stover	Batch	/	0.239
Rice straw	/	/	0.225
Canary grass	CSTR	0.03–0.04	0.19–0.33
Sunflower oil cake	Batch	/	0.107–0.227
Winter wheat	Batch	/	0.311–0.360
Waste activated sludge	Batch	/	0.116
Waste activated sludge + fatty wastewater	Batch	/	0.362
Synthetic kitchen waste	Batch	/	0.117
Synthetic kitchen waste + municipal grease waste	Batch	/	0.324–0.418

Table 4. Comparison of reported and calculated first-order degradation rate constants and methane yields [18–25].

4. Benefits of anaerobic digestion process of BG

In this chapter, the anaerobic digestion process of BG as well as its kinetic study was investigated. An average COD removal of 58% and VS removal of 55% was achieved. The organic removal efficiency was consistent and comparable with earlier studies. Kinetic analysis indicated that the pseudo-first-order degradation rate constant of BG was in the range of 0.10–0.19 d⁻¹.

The organic removal rate and kinetic coefficient listed above indicate that anaerobic digestion process could be introduced as an effective pretreatment process of BG for initial COD removal and energy recovery. After anaerobic treatment process, the effluent had a consistent effluent organic strength (COD ~ 10,000 mg L⁻¹) that can be treated aerobically. Based on the analysis, BG was found to be a readily digestible substrate. The recovered biogas could be considered as a readily in-plant-usable energy source; therefore, the energy utilization efficiency will be increased, and also the capital of organic treatment will be decreased, which is an industrial win-win situation. Once the AD process was linked as a gate-to-gate life cycle to the BG treatment chain, the efficiency of the complete cradle-to-gate evaluation will be improved and the total cost will be reduced.

The pilot-scale system produced excellent quality biogas (75% CH₄ content), with a methane yield in the range of 0.40–0.77 m³-CH₄ Kg-VS⁻¹. The addition of paper mill waste streams (FC and SPL) as cosubstrate did not adversely affect the methane yield. Currently, anaerobic digestion technique has not been widely used in the pulp and paper industry due to the recalcitrant nature of the paper process wastes. However, the combining treatment process of paper process and food process wastes does not show negative affections and could be considered as an alternative treatment method in the future.

The conclusion of this chapter is that BG has the industrial potential to be anaerobically treated as an energy feedstock and there has been ongoing commercial effort to build large-scale digesters using BG as the primary substrate. Using BG for biogas production could serve as a profitable model for converting waste to renewable energy.

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