

Effect of seasonal variations of organic loading rate and acid phase on methane yield of food waste leachate in South Korea

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Received: 17 November 2016 / Accepted: 8 January 2017 / Published online: 21 January 2017
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Abstract The objective of this study was to determine the effect of seasonal variations of organic loading rate (OLR) and acidogenic phase on methane yield of food waste leachate (FWL) treated in biogas facility in South Korea. A biogas facility operating in G city was selected as the target for this study. Remarkable seasonal fluctuations in methane yield occurred in this facility repeatedly. Methane yield in the summer was significantly lower compared to that in other seasons. In order to determine the operation efficiency, precision investigation (methane yield, OLR, etc) was conducted from March 2014 to April 2015. Characteristic parameters and operating factors of a two-stage anaerobic digestion were analyzed to obtain volatile fatty acids (VFAs), chemical oxygen demand, nutrients, total nitrogen, and so on. Data comparison revealed that the monthly average values of OLR and VFAs tended to increase rapidly in the summer (up to 3.92 kgVS/m³ day and 9263 mg/L, respectively). In contrast, methane yield in the same season was at 0.28 Sm³CH₄/kg VS, which was much lower than the average value (0.42 Sm³CH₄/kg VS) of methane yield in other seasons. The decrease in methane

yield ranged from 69.0 to 57.9% in the summer. These results suggested that methane yield might be influenced by the operating conditions with seasonal organic loading fluctuations. In other words, methane yield might be affected by a shock load of VFAs due to inapposite operation of acidogenic phase with easily degradable FWL, particularly in the summer. The results of this study will provide important information on how an ongoing biogas facility of FWL should be operated in the summer.

Keywords Methane yield · Organic loading rate · Anaerobic digestion · Two-stage digestion · Food waste leachate

Introduction

According to prohibition for direct landfill ('05) and ocean dumping ('13) of organic waste (including food waste), the treatment of organic waste is getting urgent and serious (KME 2012). After ban of landfilling and ocean dumping of organic waste in Korea, the disposal of food waste (FW)/food waste leachate (FWL) which accounts for more than 25% of municipal waste was needed (KME 2013, 2015a). Among the treatment methods, anaerobic digestion is emerging as an alternative to treat the high organic waste such as FW and FWL (Peter 2010). It is used as a useful organic source to bio-gasification facilities with high total solid contents and biodegradability.

According to microorganisms processing steps, bio-gasifications are divided into single-stage and two-stage anaerobic digestion. In laboratory scale, two-stage digestion process in bio-gasification was more efficient at operating parameters such as methane yield compared with single-stage digestion process. Shen et al. (2013) and Ueno

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et al. (2007) measured methane yield in various organic loading rate (OLR), hydraulic retention time (HRT), and type of anaerobic digestion process, single or two-stage. Shen et al. (2013) determined that two-stage digestion presented more settled operation status with 7.0–15.8% increase in methane production comparing to single-stage digestion in condition of more than 2.0 kg VS/m³ day. Ueno et al. (2007) compared the performance of two-stage and single methanogenic process. The methane yields were single-stage 0.22 and two-stage 0.40 Sm³CH₄/kgVS, respectively. Viturtia et al. (1995) examined the biogas production and methane contents (%) in different conditions of OLR and HRT in two-stage digestion. The longer retention time of acidogenic phase had a tendency to increase the methane content 75.0–79.9% and biogas production 1.09–1.65 dm³/dm³ day, individually. Study on difference in bacterial community depending on digestion system type was also reported in the following reference. Zhang and Noike (1991) and Shin et al. (2004) observed the number of bacterial populations so that acetate-utilizing methanogens in methanogenic fermenter phase of two-stage digestion showed a higher about 2–10 times than the number of single-stage digestion.

Few researches on organic loading into digestion system have been conducted in the aspects of contrasting two stage and single stage. Dinsdale et al. (1997) evaluated that 77% of COD removal efficiency in the condition of OLR 16 kg COD/m³ day and HRT 12 h in two-stage digestion for treating instant coffee as organic source. Vinas et al. (1993) derived COD removal efficiency of 90% at OLR 11 kg COD/m³ day and HRT 21 h applied two-stage upflow anaerobic sludge blanket reactor. Ince (1998) achieved 90% reduction in COD in continuous stirred tank reactor at OLR 5 kg COD/m³ day and HRT 2 days. On the other hand, many full-scale plants particularly treating FWL in Korea showed different patterns comparing to laboratory or pilot scale experiments. In full-scale plants, deterioration of operation efficiency is easily affected by seasonal variation of organic loading and immaturity of field operation. Especially in some full-scale plants in summer, methane yield showed a significantly lower value comparing with those of other seasons. However, it was hard to interpret the operation conditions of the domestic full-scale plants because of the lack of field data in South Korea. Korean FWL discharging from home might be likely perishable especially in the summer. Consequently, bio-gasification of FWL in full-scale plants has been shown much difference in awfully sensitive to seasons.

In this study, the effect of seasonal variations of OLR and acidogenic phase on methane yield of FWL treated in biogas facility in South Korea was determined. The results of this study will provide important information on the relationship between methane yield and other operating factors with seasonal variations. It will help biogas facility

to troubleshoot low methane yield problem encountered in the summer to meet full-scale production of methane in Korea.

Materials and methods

Amount of FW/FWL production in G city and input FWL G plant

During the precision investigation period from March 2014 to April 2015, seasonal production of FW/FWL in G city was examined based on annual statistics released by Korea Ministry of Environment to inspect the tendency of seasonal flow (KME 2014). FW/FWL was conducted in a site survey by separating them to household section and non-household section. The amount of FW/FWL was calculated in accordance with sources such as detached houses, apartment, and so on. Physical compositions were determined for samples from various sources.

Target facility description and sampling

A biogas facility in G city (referred to G plant) where FWL has been treated by a two-stage anaerobic digestion was selected for this study. The detail outlines of G plant are described in Table 1. This facility has treated FWL generated in the shredding/selection process of FW. FWL collected from G city was transported to G plant at an average of 211 tons per day (81.8% of design capacity based on FWL) from March 2014 to April 2015. It was pre-treated to fine particles (less than 5 mm) by passing through head and end processes such as screening, crushing, dehydration, and shearing. After the pre-treatment process, FWL was stored in a collection tank for 2–3 days before being fed into the digester system. Anaerobic digestion processes of this facility were configured to a two-phase digestion process (an acid phase and a methanogenic fermenter operated in hermopillic condition at approximately 54.6 °C). The volume of digestion processes was 4830 m³, and the HRT was an average of 24.1 days during the investigation period as mentioned above.

Sampling at acidogenic phase was conducted during all seasons excepting winter. Sampling was performed from the outlet of acidogenic phase and from the two-stage anaerobic digester. It was carried out twice in each season. Each sample was stored in refrigerated condition before being analyzed.

Analysis methods

All six samples from acidogenic phase were experimented twice to determine 18 different items including volatile

Table 1 Outline of G plant in this study

	Treatment material	Digestion method	Volume of digester	Design capacity	HRT
G bio-gasification facility (G plant)	FWL	Wet type, high temperature, two-stages	4830 m ³	259 ton/day	24.1 day

fatty acids (VFAs), chemical oxygen demand (COD_{cr}), nutrients, total nitrogen (TN), total phosphorus (TP), and so on. COD_{cr} was analyzed by closed reflux titration method (5220C) (APHA 1998). The nutrients of FWL (representative nutrients such as carbohydrate, protein, and fat) were determined by Korean Food Standard Codex (MFDS 2015a, b). TN and ammonium nitrogen (NH₃-N) were analyzed according to oxidation method and UV/visible spectrometry of Official Testing Method with respect to water pollution process (KME 2015b, c). TP and phosphate phosphorus (PO₄-P) were analyzed according to UV/visible spectrometry and ascorbic acid method of Official Testing Method with respect to water pollution process (KME 2015d, e). *n*-Hexane was determined by Official Testing Method with respect to water pollution process (KME 2015f). Nine kinds of VFAs (acetic acid, propionic acid, isobutyric acid, butyric acid, isovaleric acid, valeric acid, isocaproic acid, hexanoic acid, and heptanoic acid) were determined according to standard method 5560D gas chromatographic method 4.a. (APHA 1998). Sample extracted with diethyl ether was used to determine VFA contents by gas chromatography. Gas chromatograph (Agilent 6890, USA) was equipped with FID and DB-FFAP column (length, 25 m; inner diameter, 0.32 mm; film thickness, 0.5 μm). Detailed conditions for GC operation are summarized in Table 2.

Precision investigation in anaerobic digestion process

To investigate the operating status of methanogenic fermenter phase of G plant, precision investigation was carried out for a year. Operating data including gas production (Sm³), volatile solids (VS, %), methane contents (%) in biogas, and so on were collected in precision investigation from March 2014 to April 2015. Each parameter was

elicited by calculating its monthly average value. Approximately five daily data in each month were used to calculate the monthly value. Methane yield (Sm³CH₄/kg VS) was calculated based on daily operation data such as biogas production (Sm³/day), methane content (%), treatment amount of FWL (ton/day), and VS_{inlet} in G plant.

Results and discussion

Statistic amount of FWL production in G city

The production amount of FW from G city and the input amount of FWL to G plant are illustrated in Fig. 1. Those two amounts were generated the highest in the summer. Composition of FW was classified to vegetables, fruits, grains, fish/meat, leachate, and so on. Overall, vegetables occupied the most quantity overall. In summer, discharge of fruits was increased approximately 1.3 times comparing to their amounts in other seasons. Total discharge amount of FWL also was augmented simultaneously.

Characteristics of acidogenic phase in G plant

Proliferation of microorganisms depends on the properties of organic substrate. In the two-stage anaerobic digestion system, methanogen that generates biogas in methanogenic fermenter phase could be affected by effluents from the acidogenic phase (Speece 2008; El-Machad et al. 2008). Seasonal properties of internal acidogenic phase in G plant are shown in Tables 3 and 4. In general, nutrient components of food waste are known to be readily degradable in the following order: carbohydrate, fat, and protein. As shown in Table 3, carbohydrate, the most easily biodegradable nutrient, had the highest content (4.78 g/

Table 2 Analysis conditions of volatile fatty acids (GC-FID)

GC condition	
Model	Agilent 6890
Column	DB-FFAP (25 m × 0.32 mm × 0.5 μm)
Oven temp	95 °C (2 min) → 10 °C/min → 140 °C (2 min) → 40 °C/min → 240 °C (5 min)
Injection temp	240 °C
Injection mode	Split (10:1)
Flow rate	1.0 mL/min

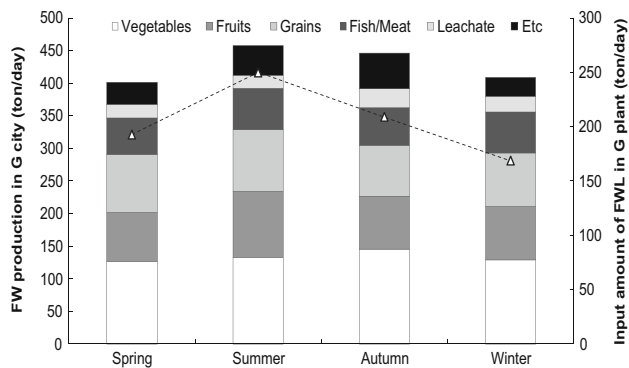


Fig. 1 Food waste (FW) production in G city and input of FWL in G plant

100 g) in the summer comparing to that in other seasons. The average contents of nutrient components such as fat, protein, and carbohydrate were 2.58, 2.11, and 4.30 g/100 g, respectively. *n*-Hexane can cause scum problem. Its content in the anaerobic digester was at an average of 7647 mg/L, with a maximum level of 9680 mg/L in the summer at the acidogenic phase. The average contents of other factors such as COD_{Cr}, TN, NH₃-N, TP, and PO₄-P were 112,425, 4502, 717, 619, and 112 mg/L, respectively.

The increase in VFA concentration is generally associated with microbial growth rate of acidogenic microorganisms. If too high concentration of VFAs flows into the methanogenic fermenter phase, the buffering capacity of methanogen can be deteriorated due to the shock load of VFAs (Speece 2008; Lee et al. 2015). Total VFAs of acidogenic phase in G plant in the spring and autumn were at 6244 and 6120 mg/L, respectively (Table 4). In particular, the value in the summer was the highest (9263 mg/L) among all seasons. Individual VFAs were the highest included acetic acid (98.4%) in the spring, propionic acid (43.6%) in the summer, and propionic acid (52.8%) in the autumn. A reasonable explanation for these findings is that overloading of fruits and vegetables with very high water content in FWL occurs in summer. Especially, the production amount of fruits in FWL in the summer was about 1.3 times higher than other seasons. It has been reported that accumulation of propionic acids can adversely affect the consortia of propionic acid utilizers during the

acidogenesis process due to overloading of FWL in the summer (Burak and Orhan 2002).

Operation status of methanogenic fermenter phase in bio-gasification facility

VFA variations in the methanogenic fermenter phase during the investigation period are shown in Fig. 2. In general, VFAs would have inhibitory effect on methanogen when their levels are above 3000 mg/L based on the guideline provided by the local government of Bayem (Gronauer and Effenberger 2007; Stockl and Oechsner 2012). VFAs/alkalinity ratio is known as a key monitoring parameter in the anaerobic digestion process. The maximum VFAs/alkalinity ratio was 0.51 in August of 2014. After August of 2014, VFAs/alkalinity ratio was gradually decreased. It maintained an optimal range at 0.2–0.3 which was its standard range proposed by NIER 2014 in South Korea (NIER 2014). Lee et al. (2015) have proposed that the maximum allowable concentration of VFAs should be kept lower than 4000 mg/L in full-scale biogas plants treating FWL that is very easily degradable. The contents of VFAs began to increase in June, the beginning of the summer. It reached a maximum peak in September of 2014.

Methane (CH₄) gas and biogas production in G plant is shown in Fig. 3. The biogas production in the methanogenic fermenter phase showed similar tendency to VFAs in the acid phase. From March 2014 to September 2014, biogas production tended to be increased continuously due to the increase amount of input. In contrast, methane gas production seemed to have a slight degree of fluctuation during the same period. More detail results of methane contents are shown in Fig. 4.

With increasing load to the methanogenic fermenter phase, biogas production and methane content ratio in biogas showed contrasting patterns of seasonal variations. Methane content was decreased gradually. It was decreased to 57.9% in July 2014. Then it recovered in November. From July to August, methane contents failed to meet the Korean standard (60%) suggested by the Ministry of Environment of Korea (NIER 2014).

Table 3 Seasonal characteristics of acidogenic phase

	Nutrients (g/100 g)			COD _{Cr} (mg/L)	TN (mg/L)	NH ₃ -N (mg/L)	TP (mg/L)	PO ₄ -P (mg/L)	<i>n</i> -hex (mg/L)
	Fats	Proteins	Carbohydrates						
Spring	4.29 ± 0.17	2.46 ± 0.12	3.62 ± 0.13	–	4493 ± 112	1313 ± 46	–	–	5080 ± 173
Summer	2.41 ± 0.22	2.35 ± 0.16	4.78 ± 0.38	159,680 ± 9581	4260 ± 234	559 ± 36	710 ± 47	88.5 ± 4.07	9680 ± 649
Autumn	1.03 ± 0.06	1.52 ± 0.09	4.49 ± 0.20	85,170 ± 2555	4752 ± 71	280 ± 8	528 ± 13	136 ± 1.77	8180 ± 172
Average	2.58	2.11	4.30	112,425	4502	717	619	112.3	7647

Table 4 Seasonal characteristics of VFAs in acidogenic phase

Volatile fatty acids (mg/L)		Volatile fatty acids (mg/L)										Total
		Acetic acid	Propionic acid	Isobutyric acid	Butyric acid	Isovaleric acid	Valeric acid	Isocaproic acid	Hexanoic acid	Heptanoic acid		
Spring		6146 ± 141 (98.4%)	49 ± 1.67 (0.8%)	N/D	48 ± 1.34 (0.8%)	N/D	N/D	N/D	–	N/D	6244 ± 144	
Summer		2691 ± 172 (29.1%)	4041 ± 287 (43.6%)	958 ± 54 (10.3%)	120 ± 8.28 (1.3%)	1192 ± 70 (12.9%)	165 ± 9.08 (1.8%)	40 ± 1.84 (0.4%)	47 ± 2.54 (0.5%)	11 ± 0.61 (0.1%)	9263 ± 605	
Autumn		1472 ± 49 (24.1%)	3231 ± 116 (52.8%)	702 ± 19 (11.5%)	86 ± 3.61 (1.4%)	453 ± 15 (7.4%)	105 ± 2.10 (1.7%)	37 ± 0.78 (0.6%)	33 ± 0.56 (0.5%)	N/D	6120 ± 206	
Average		3436 (47.7%)	2440 (33.9%)	553 (7.7%)	85 (1.2%)	548 (7.6%)	90 (1.2%)	26 (0.4%)	40 (0.6%)	4 (0.1%)	7209	

N/D not detected

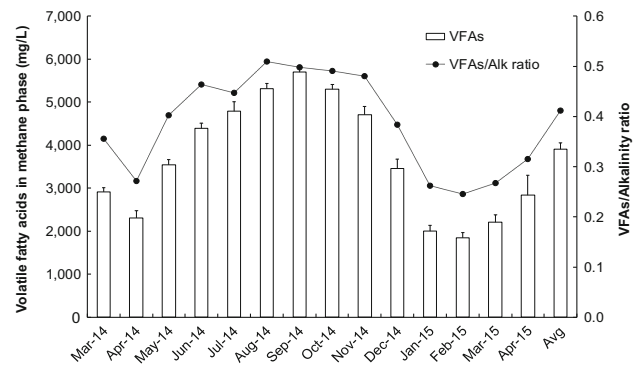


Fig. 2 Volatile fatty acids (VFAs) concentration and VFAs/alkalinity ratio in methanogenic fermenter phase

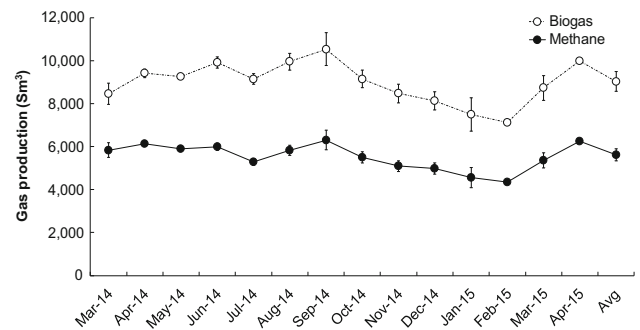


Fig. 3 Biogas and methane gas production in methanogenic fermenter phase

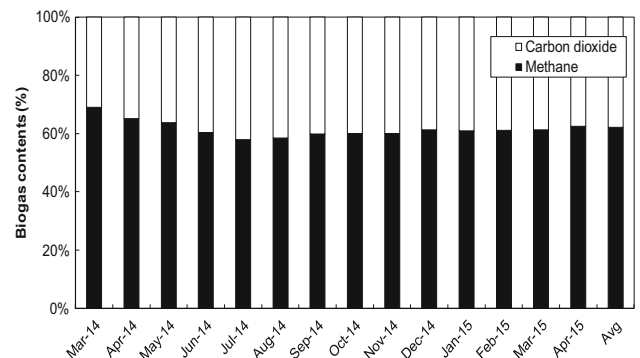


Fig. 4 Biogas composition in methanogenic fermenter phase

Correlation between methane yield and OLR is shown in Fig. 5. The average methane yield and OLR were 0.42 Sm³CH₄/kgVS and 2.95 kgVS/m³ day, respectively. Overall, OLR and methane yield showed reverse inclination. Especially from June 2014 to July 2014, both of them showed distinct correlation. Optimal guideline for FW/FWL biogas facility in Germany and South Korea has suggested an OLR standard of 1.5–4.0 kgVS/m³ day and methane yield of more than 0.48 Sm³CH₄/kgVS in actual field conditions (Gronauer and Effenberger 2007; NIER 2014). As shown in Fig. 5, the maximum average OLR

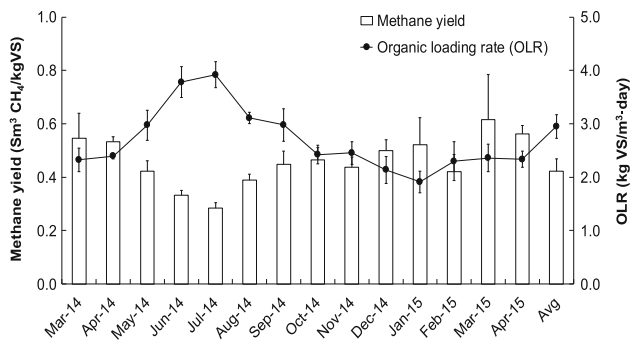


Fig. 5 Methane yield and organic loading rate (OLR) in anaerobic digestion facility

was 3.92 kgVS/m³ day with a maximum level of 4.58 kgVS/m³ day and a minimum methane yield of 0.28 Sm³CH₄/kgVS in July 2014. This could be caused by excessive accumulation of VFAs due to inappropriate operation of the acidogenic phase and multiplication of organic loading into the G plant in the summer.

Previous studies have determined those causal factors of process failure as mentioned above. Hawkes et al. (1994) have concluded that bicarbonate alkalinity should be performed to monitor the instability and the overloading of the anaerobic digester. Anderson and Yang (1992) have determined that the concentration of VFAs and COD could have profound effect on the alkalinity demands for pH control in the anaerobic digestion process.

Most guidelines for the operation of anaerobic digestion system have presented troubleshooting and indications for various anomalies in the anaerobic digestion processes. In case of Germany, all problem situations have been recommended for gradual mitigation. If VFAs/Alk ratio is increased and methane yield is decreased due to abrupt rising of input organic quantities, it can be solved by adjusting OLR or monitoring the operation factors of digester (Gronauer and Effenberger 2007). Reducing the input flow rate and securing the microbial population by reposing the acidogenic phase for a certain period time have also been recommended (EPA 1976; Park et al. 2009a, b; NIER 2014).

However, these resolutions should be deliberated in conjunction with seasonal features in South Korea. South Korea has distinct characteristics with various ranges of temperatures in each season. For example, the average temperature in Korea is 10–15 °C in the whole year. It is 23–26 °C in the summer and –6–3 °C in the winter (KMA 2014). Thus, reducing the retention time of acidogenic phase, bypassing the FW/FWL into the methanogenic fermenter phase, and regulating excessive OLR are suggested to have stable operation of the anaerobic digester when there is abrupt augmentation of VFAs. This could be considered as substantial alternatives in terms of

convenience operation and economic aspects rather than mediating alkalinity by injecting chemicals represented in small-scale experiments.

Therefore, considering the seasonal features such as unexpected VFAs augmentation in the summer, acidogenic phase in two-stage biogas facilities should be operated by reducing the retention time to directly manage the quantities of VFAs in the methanogenic fermenter phase. In addition, proper OLR (4.0 kgVS/m³ day) should be maintained because FW produced by each household has been significantly increased in Korea, especially in the summer.

In conclusion due to large differences in the amount of FW generated in each season and large variations in ambient temperatures in Korea during different seasons, appropriate solutions are needed for biogas facility to have stable methane production.

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