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Effects of mixture ratio on anaerobic co-digestion with fruit and vegetable waste and food waste of China

Jia Lin[1,2], Jiane Zuo[1,2,*], Lili Gan[1,2], Peng Li[1,2], Fenglin Liu[1,2], Kaijun Wang[1,2], Lei Chen[2], Hainan Gan[2]

1. State Key Joint Laboratory of Environment Simulation and Pollution Control (SKLESPC), School of Environment, Tsinghua University, Beijing 100084, China. E-mail: tsinghuaenv@gmail.com
2. Joint R & D Center of Tsinghua University (Department of Environmental Science and Engineering) and Shandong Sifon Environmental Protection & Bio-Energy Co., Ltd., Beijing 100084, China

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Abstract

The biochemical methane potentials for typical fruit and vegetable waste (FVW) and food waste (FW) from a northern China city were investigated, which were 0.30, 0.56 m^3 CH_4/kgVS (volatile solids) with biodegradabilities of 59.3% and 83.6%, respectively. Individual anaerobic digestion testes of FVW and FW were conducted at the organic loading rate (OLR) of 3 kg VS/(m^3·day) using a lab-scale continuous stirred-tank reactor at 35°C. FVW could be digested stably with the biogas production rate of 2.17 m^3/(m^3·day) and methane production yield of 0.42 m^3 CH_4/kg VS. However, anaerobic digestion process for FW was failed due to acids accumulation. The effects of FVW: FW ratio on co-digestion stability and performance were further investigated at the same OLR. At FVW and FW mixing ratios of 2:1 and 1:1, the performance and operation of the digester were maintained stable, with no accumulation of volatile fatty acids (VFA) and ammonia. Changing the feed to a higher FW content in a ratio of FVW to FW 1:2, resulted in an increase in VFAs concentration to 1100–1200 mg/L, and the methanogenesis was slightly inhibited. At the optimum mixture ratio 1:1 for co-digestion of FVW with FW, the methane production yield was 0.49 m^3 CH_4/kg VS, and the volatile solids and soluble chemical oxygen demand (sCOD) removal efficiencies were 74.9% and 96.1%, respectively.

Key words: anaerobic co-digestion; biochemical methane potential; continuous stirred-tank reactor (CSTR); fruit and vegetable waste; food waste

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Introduction

The amount of municipal solid wastes (MSW) in China increased rapidly in recent years. According to China Statistical Yearbook 2009 by China Statistics Press, a total amount of MSW, which were collected and disposed concentratedly in the year of 2008, was about 154 million tons. Currently, the main disposal methods for MSW in China is landfill (90.5%), and small percentage of MSW is disposed through incineration and composting. In some area, the organic content in MSW is higher than 60% due to high percentage of fruit and vegetable waste (FVW) and food waste (FW). In some FVW and FW samples, the volatile solids (VS) content is 80%–90%, and the water content is 75%–95%. The high organic and water contents are the main cause of heavy odor and plenty of leachate during the collection, transportation and landfill of MSW. Anaerobic digestion has been suggested as an alternative method for treatment of high organic content waste to recovery renewable energy–biogas; as well as to make organic matters more stable. Some research groups have developed different anaerobic digestion processes for different organic wastes including FW, FVW, and organic fraction of MSW (OFMSW) (Bouallagui et al., 2005; Forster-Carneiro et al., 2008). Compared with conventional aerobic composting processes, anaerobic processes need a lower operational energy input and a lower initial investment cost (Nguyen et al., 2007), and most important, it produces biogas – a clean and renewable energy. Recently in Europe, there are more and more biogas plants applying anaerobic digestion for energy production from organic wastes, animal manure, and other energy crops (Forster-Carneiro et al., 2008).

However, the operation of anaerobic digester fed with FW was not very effective and stable due to the accumulation of volatile fatty acids (VFAs). When the FVW is anaerobically digested individually, sometimes low stability and low efficiency of operation may happen due

* Corresponding author. E-mail: jiane.zuo@tsinghua.edu.cn

to its low C/N ratio. Therefore, co-digestion of FW or FVW with other organic wastes, such as municipal sludge, animal manure, and agricultural biomass become more popular (Bouallagui et al., 2009; Habiba et al., 2009; Nayono et al., 2009; Neves et al., 2009). Co-digestion is combined digestion in a single digester with some different wastes (solid or liquid organic wastes) with complementary characteristics (Habiba et al., 2009). The main advantages of co-digestion with different bio-wastes are: (1) increasing methane production yield due to mixed supply of additional nutrients from co-substrates; (2) more efficient utilization of equipments; and (3) cost-sharing by processing multiple waste streams in a single facility (Alatriste-Mondragon et al., 2006).

FVW and FW are two major compositions of OFMSW in China, their lower cellulose content and lower C/N ratio may cause rapid acidification and ammonia releasing, and may cause further inhibition to methanogenesis in digester. Co-digestion of FVW with FW may give an alternative solution for possible operational problems, but there is a limited research conducted on the co-digestion with FVW and FW. In this article, the anaerobic co-digestion with raw FVW and FW in a lab-scale continuous stirred-tank reactor (CSTR) was investigated, and the effects of FVW to FW ratio were investigated.

1 Materials and methods

1.1 Fruit and vegetable waste, and food waste

Raw FVW were collected from a fruit and vegetable market in Beijing, China in different seasons of 2009, which mainly contains residues of vegetables such as Chinese cabbage, carrot, lettuce, and fruits, such as apple, banana, pear, and watermelon. Raw FW were collected daily for one week from a dining hall of Tsinghua University, Beijing, China, which mainly contains leftovers of cooked foods, such as meats, fishes, rice, breads, noodles and vegetables. After shredding to small size and homogenizing, the raw FVW and FW were stored at 4°C. The characteristics of FVW and FW are shown in Table 1.

1.2 Inocula

Anaerobic granular sludge with good methanogenesis activity was used as inocula to the digester, which were taken from a full-scale UASB reactor treating starch processing wastewater at 35°C in Qinhuangdao City, Hebei, China. The water content and VS content were 87.5% and 79.63%, respectively.

Table 1 Characteristics of typical fruit and vegetable waste (FVW) and food waste (FW) from a northern China city

	FVW	FW
pH	4.24	3.55
Water content (%)	92.6	77.9
Total solids (TS) (%)	7.4	22.2
Volatile solids (VS) (%)	6.5	20.5
VS/TS (%)	88.1	92.5

1.3 Biochemical methane potential test

Biochemical methane potential (BMP) tests were performed in 500 mL Erlenmeyer flask at 35°C using modified method by Gunaseelan (2004). The initial concentrations of inocula and raw wastes were both 10 gVS/L in each flask. All flasks were purged with nitrogen gas before sealing and operation. Flasks were incubated for 20 days in a incubator at 35°C and continuously shaking at 50 r/min. Biogas produced from each flask was collected, measured, and then the volume was normalized to standard volume (0°C, 1.013×10^5 Pa). All samples were analyzed in duplicate.

1.4 Digester and operating conditions

A lab-scale CSTR with a volume of 4.0 L had been operated for 178 days at (35 ± 1)°C. The digester was equipped with inlet and outlet ports for feeding and effluent discharging, and with a port for gas collection. A mechanical stirrer (120 r/min) was used for mixing in digester. The test mixture ratios of FVW: FW based on VS contents were 2:1, 1:1, and 1:2. The digester was operated with a draw and fill method, the residue was drew out and raw material was fed into digester once a day with a peristaltic pump at the same organic loading rate (OLR) of 3 kg VS/(m^3·day). The gas production was measured with a gas meter (LML-2, Changchun Qichelvqingqi Co., Ltd., China).

1.5 Chemical analysis

The following parameters were analyzed: pH, total alkalinity, soluble chemical oxygen demand (sCOD), ammonium concentration, VFAs, TS, VS and phosphorous. All samples were analyzed in triplicates. The oxidation reduction potential (ORP) was monitored by ORP-meter (PHS-25, Shanghai Precision & Scientific Instrument Co., Ltd., China). The ORP value kept between –350 to –360 mV during the whole experiment. Samples pretreated by filtration with a 0.45-μm cellulose acetate membrane after centrifugation at 15,000 r/min for 20 min used for sCOD, NH_4^+-N, VFA analysis according to standard methods (APHA, 2005). TS, VS, total alkalinity of the samples were determined directly according to APHA Standard Methods (2005). The elemental compositions of raw wastes were analyzed by elemental analyzer (CE-440, EAI Co., USA). Biogas compositions were analyzed using a gas chromatography (N2000, Beijing Beifenyiqichang, China) equipped with a thermal conductivity detector (TCD). VFAs concentrations in effluent supernatant were determined using a gas chromatography (6890N, Agilent, USA) equipped with a flame ionization detector and a capillary column (25 m × 0.32 mm × 0.5 μm; Hewlett Packard-FFAP, USA).

1.6 Anaerobic biodegradability of wastes

Organic matters in raw wastes can be represented with formulation of $C_aH_bO_cN_d$. Assuming all organic constituents in raw wastes can be completely converted into CH_4 and CO_2, the theoretical methane production (TMP)

V_T can be estimated using Eqs. (1) and (2) (Sosnowski et al., 2003). The anaerobic biodegradability could be calculated using Eq. (3) (Penaud et al., 1999), in which V_C is cumulative methane production (CMP) obtained from BMP test.

$$C_aH_bO_cN_d + \frac{(4a-b-2c+3d)}{4}H_2O \rightarrow \frac{(4a+b-2c-3d)}{8}CH_4 + \frac{(4a-b+2c+3d)}{8}CO_2 + dNH_3 \quad (1)$$

$$V_T = \frac{(4a+b-2c-3d)\times 2.8}{12a+b+16c+14d} \quad (2)$$

$$\text{Biodegradability} = \frac{V_C}{V_T} \times 100\% \quad (3)$$

The results of elemental compositions of FVW and FW are summarized in Table 2. Organic matter in raw FVW and FW were represented with formulation of $C_{18.2}H_{26.2}O_{12}N$ and $C_{20.1}H_{34.7}O_{8.6}N$. Based on Eqs. (1) and (2), TMPs for FVW and FW can be estimated as 0.51 and 0.67 m^3 CH_4/kg VS, respectively.

Table 2 Elemental compositions, C/N ratios, and theoretical methane production (TMP) of FVW and FW

Wastes	Elemental compositions (wt.% TS)				C/N	TMP (m^3 CH_4/kg VS)
	C	H	O	N		
FVW	43.3	5.2	38.0	2.8	15.6	0.51
FW	51.0	7.3	29.2	3.0	17.2	0.67

TS: total solid.

2 Results and discussion

2.1 BMPs and biodegradabilities of fruit waste and vegetable waste and food waste

The total methane production during the 20 days BMP test for FVW and FW were 0.30, 0.56 m^3 CH_4/kg VS, thus the biodegradability of FVW and FW could be calculated to be 59.3%, 83.6%, respectively. The organic fractions of FVW include sugar, cellulose, hemicellulose and lignin, while FW were mainly composed of protein, starch, sugar and fat, etc. The higher biodegradable organic matter content in FW resulted in a higher methane production.

The C/N ratios for FVW (15.6) and FW (17.2) both are lower than the numbers (22–35) suggested in literature for stable operation of the digester (Habiba et al., 2009; Kayhanian, 1999; Yen and Brune, 2007). The low C/N of FVW and FW implied that they contain a large quantity of nitrogen, mainly in organic forms, such as proteins.

2.2 pH, NH_4^+-N and VFAs concentrations during digestion

pH value in the digester is an important parameter to imply the process stability. Variation of pH values of the digester is presented in Table 3. In the first four phases, despite the low pH of the feed (3.55–4.24), the pH values in digester maintained within a stable neutral range of 7.20–7.85. pH values increased along with the increasing of influent FW proportion, and reached the highest value at the FVW/FW ratio of 1:2. The resistance to pH-change in digester depends on its buffering capacity, which is, in a well operating digester, mainly bicarbonate/carbon dioxide. If other ions present the solution, they may also contribute to the alkalinity. The simultaneous presence of ammonia and bicarbonate in the digester results in the formation of another buffer system (Cecchi et al., 2003). Therefore, in an anaerobic system, a proper ammonia concentration would be of benefit to enhance the buffering capacity due to formation of NH_4HCO_3. FW are rich in high nitrogen matters such as proteins, when these matters are degraded, ammonia will be released to the solution and form ammonium bicarbonate, results in additional buffering capacity of the digester (Murto et al., 2004). However, pH value declined rapidly when FW was fed alone, this is mainly because of the rate of anaerobic degradation of cellulose-poor wastes would be limited by methanogenesis rather than hydrolysis process, a major inhibition to methanogenesis was caused by the fast pH decreasing due to rapid acidifiation (Bouallagui et al., 2005, 2009; Misi and Forster, 2002). The results showed that co-digestion of FVW and FW with a proper mix ratio can improve pH buffering capacity in digester, which will be of benefit to stable operation of digester. But when FW was fed into digester individually, the fast acidification process resulted in very high VFAs accumulation, finally inhibited the methanogenesis, and caused biogas production stoped.

Ammonia is produced by biological degradation of nitrogenous organic matters in FVW and FW, mostly in form of proteins, phospholipids, nitrogenous lipids and nucleic acids etc. (Kayhanian, 1999). Microorganisms need some ammonia to form celluar protoplasm for growth and reproduction. However, if ammonia concentration is relatively high, the methanogenic process will be inhibited, resulting in a sharp drop of pH value and methane production. Therefore suitable C/N ratio in substrates is important to anaerobic digester. If it is too high, the nutrient deficiency for growth and reproduction of anaerobes may occur; if it is too low, high ammonia (total or free) inhibition to methanogenesis may occur. The impacts of C/N ratio to anaerobic process were well studied, and the optimal C/N ratio was suggested as 20–30 (Mshandete et al., 2004; Yen and Brune, 2007). The free ammonia (FA) was reported that having more significant inhibition effects on methanogenesis than ionized ammonium nitrogen (NH_4^+) (Bhattacharya and Parkin, 1989). A maximum tolerable FA nitrogen concentration of (55 ± 11) mg N/L was suggested (Bhattacharya and Parkin, 1989). However, due to the adaptation ability of microorganisms, the tolerance to 4000 mg/L total ammonia had been reported (Angelidaki and Ahring, 1993). Table 3 shows the variations of effluent NH_4^+-N concentrations of the digesters in different operational phases. The average NH_4^+-N concentration increased from 585.3 to 2329.7 mg/L gradually as influent FW proportion increased. The highest NH_4^+-N concentration was 2518.0 mg/L when the digester fed with only FVW. It could be concluded that the NH_4^+-N concentration in the digester increased with the increasing influent

Table 3 Summary of performance parameters in different operational phases

		Phase I	Phase II	Phase III	Phase IV	Phase V
Operation conditions						
Time (days)		0–30	30–60	60–90	90–131	131–178
OLR (kg VS/(m^3·day))		3	3	3	3	3
Substrates	FVW (g VS)	12	8	6	4	0
	FW (g VS)	0	4	6	8	12
Effluent characteristics						
TS (%)		3.7	3.3	4.1	4.3	6.5
VS (%)		1.8	2.1	2.5	2.8	4.3
sCOD (g/L)		2.2	1.6	2.6	4.1	21.0
pH		7.37 ± 0.03	7.40 ± 0.03	7.56 ± 0.03	7.73 ± 0.02	7.07 ± 0.14
NH_4^+-N (mg/L)		585.3 ± 61.7	608.9 ± 27.0	763.9 ± 25.8	1242.1 ± 60.0	2329.7 ± 143.5
Free ammonia (mg/L)		15.09 ± 2.2	16.90 ± 1.7	32.0 ± 1.0	70.5 ± 5.3	29.8 ± 7.5
VFAs (mg/L)		69.7 ± 11.42	181.7 ± 33.60	170.64 ± 44.00	1216.5 ± 77.15	8887.0 ± 754.05
Digester performances						
sCOD removal (%)		96.3	97.4	96.1	94.7	73.3
GPR (m^3/(m^3·day))		2.17	2.25	2.35	2.45	0.35
Methane content (%)		60.0	60.5	63.8	61.0	53.7
MPY (m^3 CH_4/kg VS)		0.42	0.44	0.49	0.49	0.06

GPR: biogas production rate; MPY: methane production yield.

FW proportion The FA concentration can be calculated through pH value and TAN according to a mathematical equation (Kayhanian, 1999). The calculated FA concentration variations in different operational phases are shown in Table 3. In the first three rational phases, NH_4^+-N and FA concentrations in the digester were estimated to be lower than the reported inhibition concentrations to methanogens (Angelidaki and Ahring, 1993), but during phase IV the FA concentration was increased over 40 mg/L, the maximum FA concentration was 97.9 mg/L. It could be concluded that FA inhibition occurred at 40 mg/L.

Both C/N ratio of FVW and FW were about 16:1, but after anaerobic digested separately, the ammonia concentrations in digester were considerably different, which is 585.3 mg/L for FVW, and 2329.7 mg/L for FW. The final ammonium concentration in the digester fed with FW was considerably higher than fed with FVW. This can be explained by the fact that FW is easy biodegradable and released faster ammonium then the less biodegradable organic matter in FVW. In this case, the addition of FW to the digester increased the releasable nitrogen, consequently, the concentration of ammonia increased, but there was no apparent inhibition to methanogenesis in the digester at the current operational conditions.

VFAs are end products of the fermentation stage, and their concentrations are very good indicators to the metabolic status of an anaerobic degradation process (Habiba et al., 2009). The variation in VFAs concentration at different operational phases is shown in Fig. 1. Total VFAs concentration increased from 70 to 9900 mg/L along with increasing influent FW which was generally due to the high carbohydrates and fat matters content in FW. In the first three operational phases, when the influent ratio of FW was low, the average total VFAs were much lower than other reports (Bouallagui et al., 2009; Habiba et al., 2009). However, when influent FW to FVW ratio increased to 2:1, the VFA production and utilization rates were unbalanced. It was indicated that the methanogenesis was inhibited slightly due to the free ammonia and became the limitation step, resulted in the

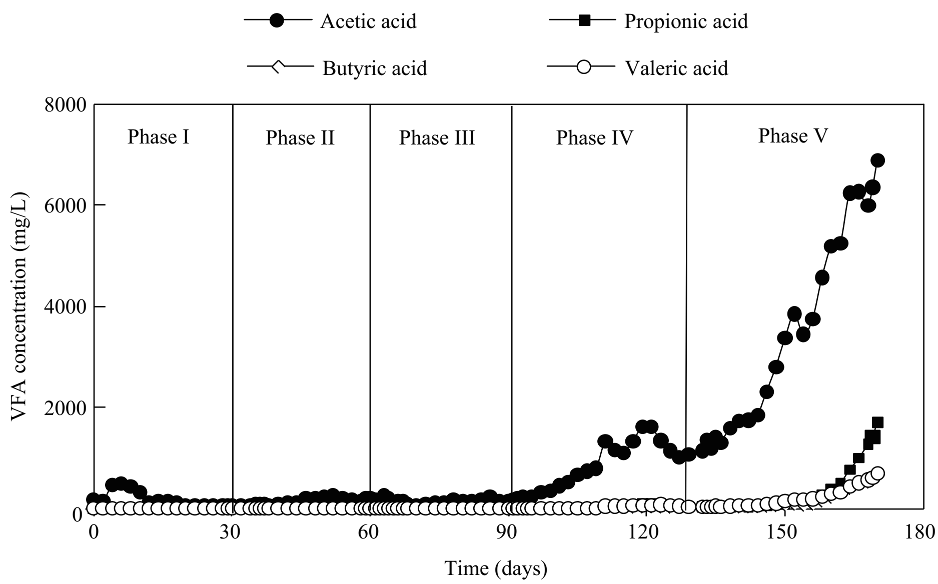

Fig. 1 VFAs variations in different operation phases.

accumulation of VFAs, which concentration increased to 1100–1200 mg/L at the end of the 4th phase. And then, only FW was fed into the digester. As shown in Fig. 1, the TVFAs concentrations in the digester increased rapidly from 1200 to 9900 mg/L. After the acetic acid concentration increased over 2000 mg/L, valeric acid began to accumulate. Methanogenesis can also be inhibited by high non-dissociated VFA concentration that were the major inhibitor and may cause very heavy inhibition (Anderson et al., 1982). Barredo and Evison (1991) reported that the number of methanogens was affected by at least two orders of magnitude at propionic acid concentrations between 1500 and 2200 mg/L. Hajarnis and Ranade (1994) studied the effect of propionate toxicity on methanogens and found that 5000 mg/L of propionate inhibited methane production at neutral pH to 22%–38% of the control value. When the pH was reduced, the inhibition drastically increased. In this study, when acetic acid concentration increased over 5000 mg/L, propionic acid and butyric acid concentration also increased from 169 to 1958 mg/L and from 91 and 934 mg/L, respectively, and at the same time, the biogas production rate reduced dramatically. Under this condition, methanogenes can not remove hydrogen and VFAs as quickly as they were produced by fermentative bacteria. As the operation went on, the more acids accumulated, the more buffering capacity depleted, and the further pH decreased, and the less biogas produced.

2.3 Volatile solids reduction and sCOD removal at different ratios of fruit and vegetable waste to food waste

Volatile solid contents in raw FVW and FW were 6.5% and 20.5%, respectively. Therefore, the influent VS concentration increased with the increase of influent FW ratio. As shown in Fig. 2, when the digester was fed only with FVW, the effluent VS concentration was the lowest. The effluent VS concentration went up with increasing FW proportion in influent. The highest effluent VS concentration was 4.3% when only FW was fed into the digester. It is clear that the VS removal efficiencies were affected as the change of influent FW ratio. The total VS removal was increased from 71.7% to 79.3% with increasing influent FW.

Variations of soluble COD concentration and its removal efficiency in digester are shown in Table 3. The initial sCOD concentrations were 59.4 g/L and 78.8 g/L for FVW and FW, respectively. When FVW was fed to digester alone, the sCOD concentration in effluent was about 2.2 g/L and sCOD removal efficiency was 96.3%. But when digester was fed with only FW, sCOD concentration increased rapidly from 4.10 to 21.0 g/L due to the accumulation of VFAs, the sCOD removal efficiency decreased to less than 75%. When the mixture ratio was 2:1, the effluent sCOD concentration became to the lowest concentration, which was 1.6 g/L, and the highest sCOD removal efficiency of 97.4% was reached. After that, the effluent sCOD concentrations went up with FW proportion increased.

2.4 Biogas production rate and methane production yield at different FVW:FW ratios

Table 3 shows the biogas production rate (GPR), methane content in biogas and methane production yield (MPY) of the digester operated at different mixtures at the same OLR. During the whole process, the methane content in biogas is generally stable, which is about 50%–65%. When the influent FVW to FW ratio was 1:1, the highest methane content in biogas was reached to 63.8%, and when only FW was fed, the lowest methane content of 53.7% was reached.

The GPR improved with the increasing proportion of FW in the mixture. Among five phases, the GPR was the highest at feed mixture of 1:2. It was indicated that the GPR was affected by the biodegradability of the mixture as shown in the BMP test. The MPY increased with a higher proportion of FW in the mixture. The maximum MPY was 0.49 m^3 CH_4/kg VS at the OLR of 3 kg VS/(m^3·day) with the mixture of 1:1. However, the decrease of the FVW:FW ratio to 1 : 2 in the feedstock led to the same MPY due to the incomplete utilization of acetic acid. When FW was

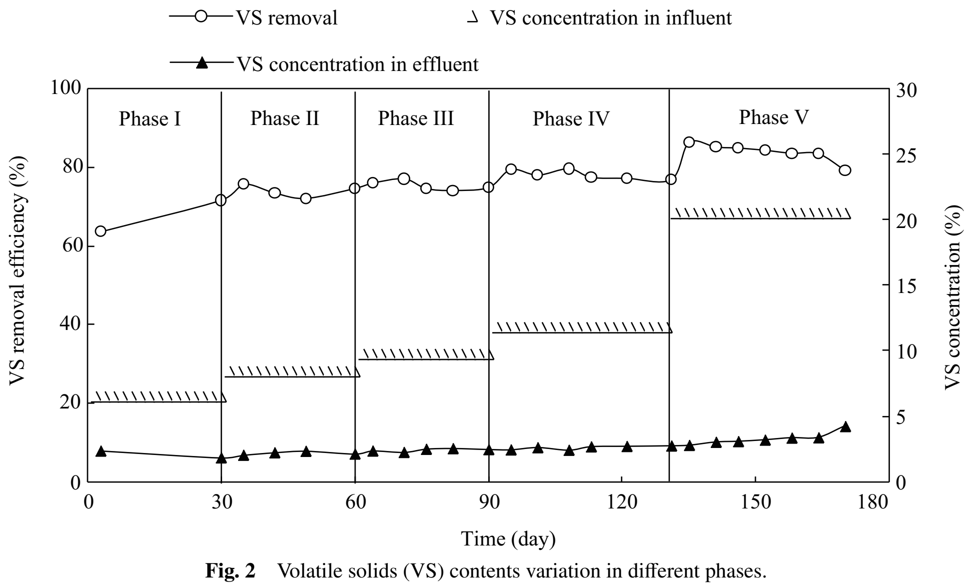

Fig. 2 Volatile solids (VS) contents variation in different phases.

fed as the only substrate, the MPY was the lowest due to the inhibition of acids accumulation.

3 Conclusions

The BMPs for FVW and FW from a northern China city in China were 0.30, 0.56 m^3 CH_4/kg VS, respectively. Both the FVW and FW are easily biodegradable wastes, and their biodegradabilities were 59.3% and 83.6%, respectively.

At OLR of 3 kg VS/(m^3·day), when FVW was fed individually, the CSTR digester can be operated stably with GPR2.17 m^3/(m^3·day) and MPY 0.42 m^3 CH_4/kg VS. However, anaerobic digestion with only FW was failed due to VFAs accumulation.

The optimal mix ratio for co-digestion of FVW with FW was found to be 1:1. Under this condition, the methane production yield, VS and sCOD removal efficiencies achieved to 0.49 m^3 CH_4/kg VS, 74.9% and 96.1%, respectively. The co-digestion with FVW and FW could not only improve the stability of anaerobic process, but also can achieve a higher biogas production and organic matter removal efficiency.

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