High-rate methane fermentation of lipid-rich food wastes by a high-solids co-digestion process

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Abstract This paper presents an experimental study on anaerobic degradation of lipids-rich food wastes by using the high solids co-digestion process. The experiments were conducted under mesophilic (35°C) and thermophilic (55°C) condition, respectively, by using a semi-continuous flow completely mixed reactor. The influent TS level was controlled at around 10%, while the hydraulic retention time (HRT) was changed from 15 days to 7.5 days. The lipids (fats or oil and grease) content in the influent TS was changed from 8% to 40% by adding salad oil (vegetable) and lard (animal) to the food wastes. The result of this study showed that the food wastes containing high lipids content was effectively degraded by the high solids co-digestion process and over 85% of lipid was degraded to biogas with 60–65% of methane. In addition, thermophilic methane fermentation was more effective for reducing lipids and had more higher loading capacity compared with mesophilic condition.

Keywords Anaerobic co-digestion; high-solids; lipids (fats, oil and greases); mesophilic; methane fermentation; thermophilic

Introduction

Lipids, characterized either as fats or oils and greases (Forster, 1992), are one of the major organic matters in food wastes and some industrial wastewaters (Mackie *et al*., 1991). Lipids-rich wastewaters are widely found in the industries of wool, leather, meat, dairy and food processing. These types of wastewaters have received some form of preliminary physical-chemical treatment, e.g. trapping, intercepting, and flotation separation, before passing to the biological stage. As a result, a large volume of sludge with high lipids content is produced. Wastes from meat-packing and slaughterhouses are especially high in lipids, with flotation foams containing 60 to 65% lipid on dry basis. Economic recovery of lipids is often impracticable because of their association with tissue, soil or fecal matter. The treatment of the wastes with high lipids contents is, therefore, a big technical problem.

It has been known since 1950s that complete degradation of lipids to CH_4 and CO_2 can be achieved in a well mixed anaerobic digester (Heukelekian and Mueller, 1958). Modern concepts of anaerobic microbiology indicate that the methane fermentation of lipids is achieved by the concerted action of three groups of bacteria: hydrolytic fermentative, syntrophic acetogenic and methanogenic bacteria. During anaerobic digestion, lipids are first hydrolyzed to long-chain fatty acids (LCFA) and glycerol by hydrolytic fermentative bacteria (Mackie *et al*., 1991; Jarvis *et al*., 1998), and the glycerol is then readily fermented to volatile fatty acids (VFA). The resulting LCFA are further degraded to acetate and hydrogen (Eq. (1)) via beta-oxidation (Weng and Jeris, 1976).

$$CH_3(CH_2)_{2m}COOH + 2m\,H_2O \rightarrow (m+1)CH_3COOH + 2mH_2 \quad (1)$$

These reactions are carried out by "H_2-producing acetogenic bacteria" (Bryant, 1979), a special group of bacteria, which also named as "syntrophic acetogenic bacteria" because they degrade LCFA and some VFA to acetate and hydrogen (Roy *et al*., 1986; Mackie *et al*.,

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1991) and require methanogenic bacteria to remove inhibitory levels of acetate and hydrogen (Lowe *et al*., 1993). Acetate and hydrogen produced by hydrolytic fermentative bacteria and syntrophic acetogenic bacteria are finally converted to biogas by methanogenic bacteria. In these complex biological reactions, the first step proceeds easily and fast and is not usually the rate-limiting step in the anaerobic digestion of lipids (Heukelekian and Mueller, 1958; Novak and Carlson, 1970; Hanaki *et al*., 1981; Broughton *et al*., 1998). The overall conversion rate is considered to be limited either by the physical processes of dissolution and mass transfer of insoluble lipids or by the biological conversion of the LCFA (Novak and Carlson, 1970; Hanaki *et al*., 1981)

Lipids in wastewaters are normally recalcitrant to biological treatment because of lipids propensity to form floating aggregates. The low surface area to volume ratios of this aggregate slows their degradation by microorganism (Gujer and Zehnder, 1983). For an effective degradation of lipids, it is very important to disperse the lipids in the substrate for making uniformity feed. In addition, a sufficient mixing for maintaining a good contact between the bacteria and influent lipids in the digester is also necessary (Heukelekian and Mueller, 1958; Renzema *et al*., 1993). These two points are the most important physical factors affecting the lipids degradation.

$$CH_3(CH_2)_{14}COO^- + 14\,H_2O \rightarrow 8\,CH_3COO^- + 7H^+ + 14H_2\ (G'^o = +345.6\ \text{kJ/mol}) \qquad (2)$$

On the other hand, as shown in eq.(2), the values of standard free energy change G'^o are positive in the reaction of LCFA degradation. These reactions are thermodynamically unfavorable unless the hydrogen partial pressure is maintained at an extremely low level. This fact means that the degradation of LCFA depends largely on the activity of methanogenic bacteria. In addition, LCFA are well known inhibitors of various microorganisms at low concentration and, consequently, cause some serious problems in anaerobic digestion. It has been established that a concentration of 1 $g.L^{-1}$ LCFA in anaerobic digester sludge is sufficient to cause severe inhibition of the digestion process (Hanaki *et al*., 1981; Angelidaki and Ahring, 1992; Rinzema *et al*., 1994; Chu *et al*., 1997). It is reasonable to expect that both methanogenic bacteria and acetogenic bacteria, the two consortia related to LCFA degradation, suffer from LCFA inhibition by protecting the accumulation of LCFA. For this purpose, both increasing the total biomass concentration and promoting methanogenic rate in the methane fermentation reactor are effective measures because the former way is expected to increase the LCFA degrading bacteria and the latter may contribute to improve beta-oxidation reactions, as well as Eq. (2), by removing the hydrogen and acetate.

Based on the above fundamental information, a two-stage process composed of mixing unit and high solids digestion unit was developed to treat lipids-rich wastes in this study. In the mixing unit, food waste was used as a dispersant for lipids to make uniformity high solids slurry. The well dispersed slurry by mixing unit was then used as a substrate of high solids co-digestion to convert lipids to biogas. This paper presents an experimental study on this high solids co-digestion process. Effects of two factors, temperature condition (mesophilic and thermophilic) and lipids content in the mixed slurry, on the removal efficiencies of total solids and lipids, biogas production, loading rate in the high solids co-digestion were investigated in this study.

Methods

Food wastes and lipids

The food waste used in this study was composed of fruits, vegetables, meat & fish, and staple foods, as shown in Table 1, which is selected based on the typical component of the

OFMSW in Japan. They were shredded and mixed with a high-speed blender and then adjusted by adding water and mineral nutrient ($FeCl_2$, 100 $mg.L^{-1}$, $NiCl_2$, 10 $mg.L^{-1}$, $CoCl_2$, 10 $mg.L^{-1}$). Table 2 gives the average characteristic of this shredded food waste. A mixture of salad oil and lard (animal fat) in weight bases of 1:1 was added to the food waste to increase its lipids content. In this study, the lipids percent in the influent TS was changed from 8% to 40%.

Seed sludge

The original seeds used for mesophilic and thermophlic digestion were taken from a full-scale sewage sludge digester operated at mesophilic or thermophilic condition, respectively. These seeds have been sufficiently adapted to the food waste for over two years in another experimental study on high solids digestion, before being used for this study.

Experimental apparatus and conditions

Figure 1 illustrates the scheme of the experimental apparatus. A two-stage system composed of mixing unit and high solids digestion unit was developed to treat lipids-rich wastes in this study. The first stage is a heated mixing tank for receiving the food waste and lipids and then mixing them to make uniformity high solids slurry. The temperature of the mixing tank was kept at 40°C for the liquefaction of lipids. The well dispersed slurry by mixing unit was fed to the high solids digester in the second stage. A semi-continuous flow, completely mixed reactor with a working volume of 5 L was used as the digester to convert lipids to biogas. The digester was water-jacketed and operated at a constant temperature of 36°C or 55°C, respectively, for mesophilic and thermophilic methane fermentation. The

Table 1 The composition of the food waste used in this study (percentage in wet base)

Fruit	Apple	10
	Grapefruit (rind)	5
	Orange (rind)	5
	Banana (rind)	10
Vegetables	Cabbage	12
	Potato	12
	Carrot	12
Meat and fish	Meat	5
	Fish (with born)	5
	Egg	4
Staple foods	Rice	10
	Bread	5
	Noodles	2.5
	Chinese noodle	2.5

Table 2 Characteristics of the food waste

	Unit	Average	Range
TS	($kg.m^{-3}$)	99.9	97.4–108
VS	($kg.m^{-3}$)	89.5	88.0–94.1
T-COD	($kg.m^{-3}$)	151.6	143–159
T-Carbohydrates	($kg.m^{-3}$)	40.6	37.0–42.7
T-Proteins	($kg.m^{-3}$)	23.5	20.7–26.4
T-Lipids	($kg.m^{-3}$)	9.64	9.1–10.0
SS	($kg.m^{-3}$)	61.6	60.4–62.4
VSS	($kg.m^{-3}$)	56.8	53.7–57.8
S-COD	($kg.m^{-3}$)	65.0	62.2–68.8
pH	(–)	4.37	4.28–4.45

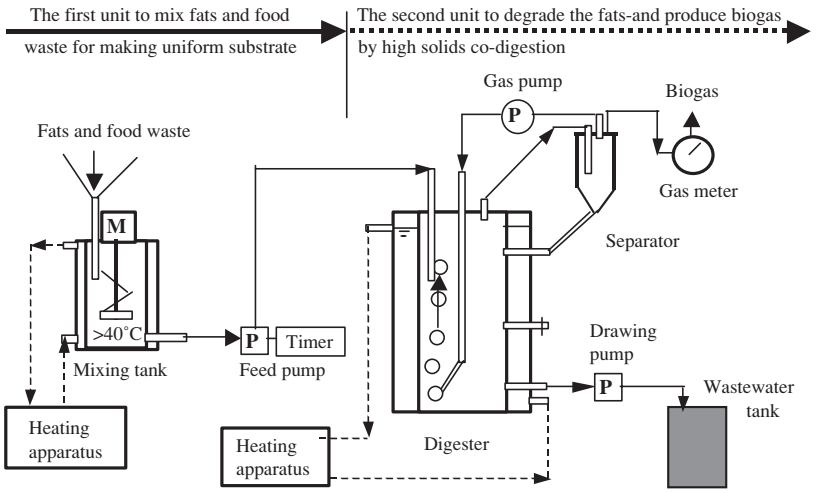

Figure 1 Schematic of the experimental apparatus for the two-stage co-digestion

substrate for digester was fed 6 to 12 times a day according to the retention time by using a timer-controlled feed pump.

As summarized in Table 3, the experiments were carried out in a total of four run by changing the lipids content from low (9.1 $g.L^{-1}$) to high (44.3 $g.L^{-1}$) stepwise. The influent TS concentrations for each run were almost the same at around 100 $g.L^{-1}$, but the influent COD concentrations were increased from 152 $g.L^{-1}$ to 250 $g.L^{-1}$ with the increase in lipids content. For each run, two digesters were operated in parallel, respectively under mesophlic and thermophlic conditions. The HRT for each digester was changed from 15 days to 7.5 days according to the lipids loading rate. For each experimental condition, the reactor was continuously operated for over 50 days (three times of HRT) to get the steady. Both the feed and digester content were analyzed 1 to 2 times a week. The average of each parameter was used as the indicator for each condition.

Analyses

The total gas production was measured with a wet gas meter. The percentage of CH_4 and CO_2 in the biogas was analyzed using a gas chromatograph (Hitachi-163 model) equipped with a thermal conductivity detector and a 3 mm × 3-meter stainless column packed with Unibeads C. Helium was used as the carrier gas at a pressure of 5 $kg.cm^{-2}$. The operational temperatures of the oven and detector were same at 140°C. The concentrations of the VFAs

Table 3 Summary of the experimental conditions

Substrate condition	Lipids content	Influent concentration ($g.L^{-1}$)			Temperature	HRT	COD_{Cr} loading	Lipids loading	Continuous operation period
		TS	COD_{Cr}	Lipids	(°C)	(days)	($g.L^{-1}.d^{-1}$)	($g.L^{-1}.d^{-1}$)	
					Meso.36	15	10.1	0.56	60 days
Run 1	Low 1	108	152	9.1		7.5	20.3	1.12	50 days
					Thermo.55	15	10.1	0.56	60 days
						7.5	20.3	1.12	50 days
Run 2	Low 2	102	166	14.3	Meso.36	7.5	22.1	1.87	60 days
					Thermo.55	7.5	22.1	1.87	60 days
Run 3	Medium	101	188	23	Meso.36	7.5* 15	12.5	1.53	80 days
					Thermo.55	7.5	25.1	3.07	80 days
Run 4	High	110	250	44.3	Meso.36	15	16.7	2.95	80 days
					Thermo.55	7.5	33.3	5.91	80 days

* The HRT was changed from 7.5 days to 15 days, because of overloading

were determined by a second gas chromatograph (HP-6890) equipped with an Innowax polyethylene glycol capillary column and a flame ionization detector (FID), using helium as the carrier gas at a pressure of 1.0 kg/cm^2. Injector and detector temperatures were 200°C and 280°C, respectively. The temperature of column oven was increased stepwise from 50°C to 170°C. *Standard Methods* (APHA, 1995) were used for the measurement of pH, alkalinity, ammonia, total solids (TS), volatile solids (VS), suspended solids (SS), volatile suspended solids (VSS) and COD (using dichromate). Lipids concentration was measured by the Bligh-Dyer method.

Results and discussion

Operation performance of mesophilic and thermophilic co-digestion process

Figure 2 illustrates the operation performance of experiments Run 2, 3 and 4, cope with the low lipids content, medium lipids content and high lipids content, respectively. At the low lipids condition (Run 2), three typical operational parameters: pH, methane gas production rate and total VFA concentration were stable in both the mesophilic and thermophilic digesters, indicating that these two digesters were operated successfully at the COD

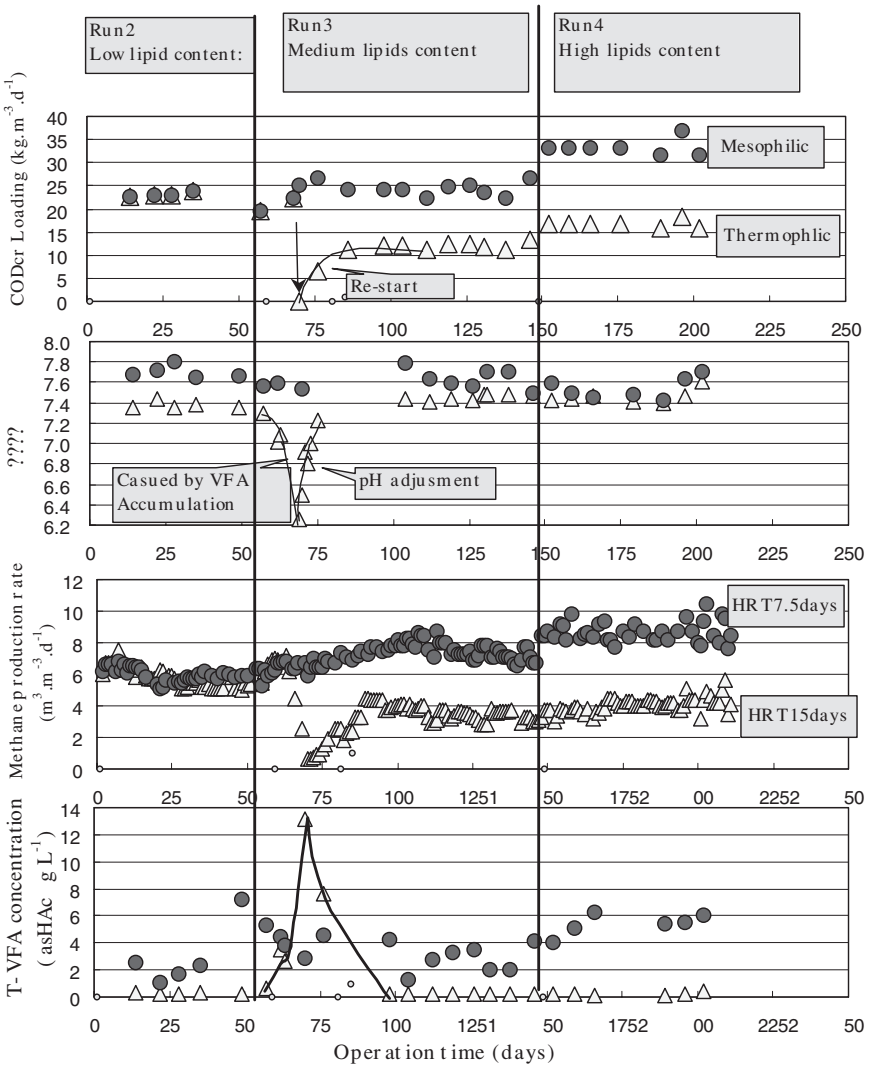

Figure 2 Operational progress of the experiments Run 2, 3 and 4

loading rate up to 22.1 kg.m^{-3}.d^{-1} corresponding to a HRT of 7.5 days. When the influent lipids content was increased to medium (Run 3), operation of the mesophilic digester was unsuccessful at HRT 7.5 days due to overloading. There were rapid drops in pH and methane production rate at the operation time of 70 days, and VFA accumulated up to 13,000 mg.L^{-1} resulting in the process failure. The mesophilic digester was then re-started at a HRT of 15 days for reducing the COD loading. After that, the mesophilic digester worked well even the lipids content was increased to be high as 44 g.L^{-1} (Run 4). On the other hand, the thermophilic digester was operated successfully at the HRT of 7.5 days throughout the three lipids conditions, while the concentration of VFA ranged in 2,000 to 6,000 mg.L^{-1}.

The results shown in Figure 2 indicate that a longer HRT as well as 15 days is necessary for maintaining a mesophilic digester to be stable in treating the food wastes with a high lipids content, compared to the HRT 7.5 days for thermophlic digester.

Loading capacity comparison between mesophilic and thermophlic co-digestion

Figure 3 illustrates the comparison between mesophlic and thermophilic digestion in loading rate and degradation efficiency of VS and COD, based on the results obtained from this study. During mesophilic co-digestion, the removal efficiencies of VS and COD had clear drop tendency when the COD loading rate was higher than 16.7 kg.m^{-3}.d^{-1}, indicating the COD loading capacity for the high solids mesophilic co-digestion is below 20 kg.m^{-3}.d^{-1}. For the thermophilic co-digestion, no significant decrease in the removal efficiencies of VS, COD and lipids was observed at the COD loading rate up to 33 kg.m^{-3}.d^{-1}. These results mean that thermophilic co-digestion has twice the COD loading capacity of

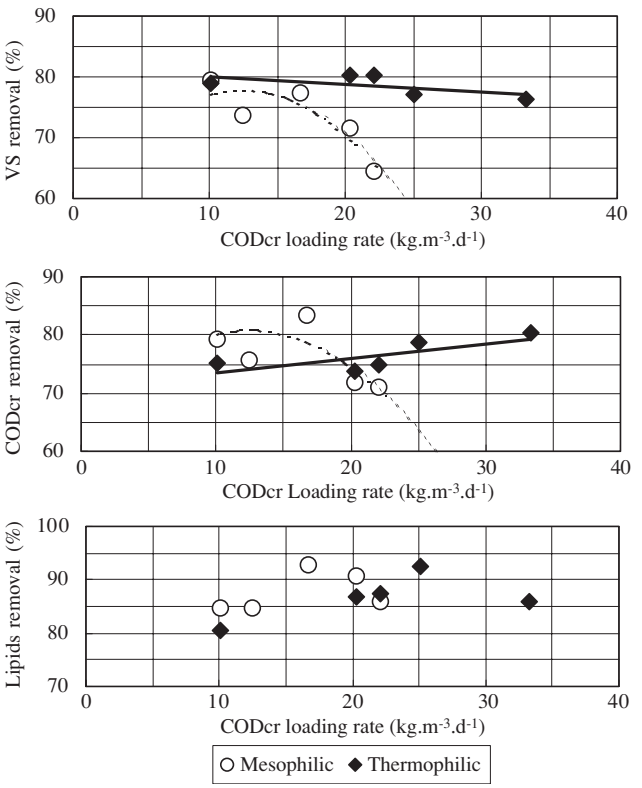

Figure 3 Comparison between mesophlic and thermophilic digestion in the loading capacity

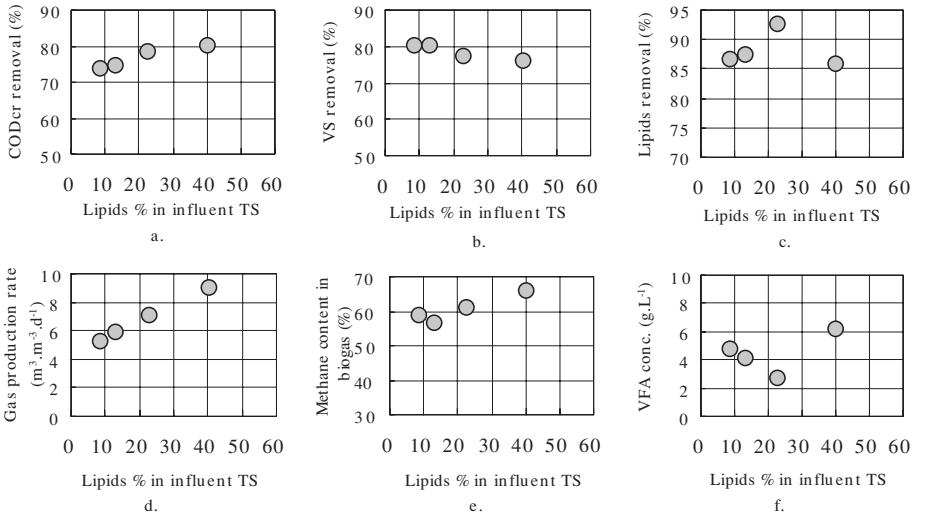

Figure 4 Effect of lipids content in substrate on the performance of thermophilic co-digestion (HRT was kept at 7.5 days, COD loading rate ranged from 20 to 33 kg.m^{-3}.d^{-1})

mesophilic condition. This point is in agreement with the previous reports in literature (Fair and Moore, 1937; Mackie and Bryant, 1995; Li, 1998; Li *et al.*, 1999). Also, it must be noted that to the best of our knowledge, this is the first paper showing successful operation of a high-solid digester at a COD loading rate as high as over 30 kg.m^{-3}.d^{-1}.

Effect of lipids content on the co-digestion performance

Figure 4 illustrates the effect of the lipids content of the substrate on the performance of the thermophilic high-solids co-digestion treating food wastes and lipids at a HRT of 7.5 days. With the increase of lipids content from 8% to 40%, the COD removal was slightly increased from 73% to 80%, while the VS removal was stable around 80%. The lipids removal efficiency was high as over 85% (86–93%) at all experimental conditions. It is clear that a good treatment performance for co-digestion was obtained at the lipids content up to 40%.

Interestingly, the biogas production rate and methane percentage increased linearly with the influent lipids content. This is not only because the degradation efficiency of lipids is high, but also because lipids have a higher gas production potential compared with carbohydrates and proteins, as shown in Table 4. This means that lipids are the most important substances in the anaerobic digestion. It is reasonable to expect that lipids produced from wastewaters and food be converted to biogas by using anaerobic biotechnology. The results of this study demonstrated the feasibility of a high solids co-digestion system in treating lipids-rich wastes.

Conclusions

Based on the results from this study, the following conclusions may be drawn.

1. A high solids co-digestion process was successfully developed to treat the lipids-rich food wastes. Over 85% of influent lipids were degraded to biogas in this process. Thermophilic methane fermentation was more effective for reducing lipids and had more higher loading capacity compared with mesophilic condition.
2. During the mesophilic co-digestion, the VS and COD removal were about 75% at a COD loading rate below 20 kg.m^{-3}.d^{-1}, but the degradation efficiency decreased at the more higher loading rate, indicating that the maximum COD loading rate for mesophilic methane fermentation was below 20 kg.m^{-3}.d^{-1}. The experimental results

Table 4 Comparison of biogas production potential of different component in the food waste

Component	Reaction of methane fermentation	Gas production	CH_4 % in biogas
Lipids	$C_{15}H_{90}O_6 + 24.5H_2O = 34.75CH_4 + 15.25CO_2$	1.425 L/g	69.5
Carbohydrates	$(C_6H_{10}O_5) + nH_2O = 3nCH_4 + 3nCO_2$	0.830 L/g	50.0
Proteins	$C_{11}H_{24}O_5N_4 + 14.5H_2O = 8.25CH_4 + 3.75CO_2 + 4NH_4^+ + 4HCO_3$	0.921 L/g	68.8

demonstrated that the suitable HRT for treating lipids-rich food wastes at mesophilic condition should be 15 days.

3. The COD loading capacity for thermophilic high solids co-digestion was high to be 33 $kg.m^{-3}.d^{-1}$. The thermophilic digester was also operated successfully at a short HRT of 7.5 days, and the removal efficiencies of VS, total COD and lipids were high as, respectively, in the ranges of 76–80%, 73–80%, and 85–93%.

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