Experimental study on the mechanisms involved in the synergetic effect of anaerobic co-digestion of mature leachate with food waste

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ABSTRACT

In this paper, a leachate anaerobic digestion experiment was carried out in a sequencing batch anaerobic digestion reactor. The anaerobic co-digestion characteristics of leachate and food waste in landfills were studied. To conduct the experiment, 10, 20, 30 and 40 g of TS/L food waste were added to leachate to observe the effect of food waste on the degradation of dissolved organic matter (DOM) in the leachate. The results of this experiment showed that adding food waste into the anaerobic digestion system could significantly increase the gas and methane production per unit organic load, and promote the degradation of DOM in leachate. When the initial load rate of food waste was 30 g TS/L, the gas production was increased by 117.69%, the dissolved organic carbon (DOC) of biogas slurry after anaerobic digestion of leachate and food waste is about 50% of the DOC in the anaerobic mono-digestion leachate. The results show that the combination of leachate and food waste can not only promote the anaerobic digestion of food waste, but also increase the gas and methane production rates; this combination can also promote the degradation of DOM in leachate. The combined anaerobic digestion of leachate and food waste had a synergistic effect.

Keywords: Food waste; Landfill leachate; Anaerobic digestion; Humus substance

1. Introduction

Sanitary landfills are the most important treatment method for municipal solid waste [1,2]. For example, in China, according to the 2017 and 2018 Statistical Yearbooks, more than 130 million tons of domestic waste were disposed of in the country's sanitary landfills. The total resulting leachate generated by domestic waste landfills exceeded 60 million tons. Landfill leachate contains a large number of organic pollutants [3] and other toxic and harmful substances [4]. In landfill leachate, dissolved organic matter (DOM) accounts for approximately 85% of the total organic matter [5,6]. Due to the complex composition of DOM, which is composed of compounds with different chemical properties, detailed information on its molecular structure and composition is not clear [7]. However, researchers have found that the main component of organic matter in leachate species is humus (HS) [8]. The humus content in leachate was found to increase with an increase in landfill years [9], which leads to a decrease in the biodegradability of leachate. Therefore, improving biodegradability is the key factor to effectively degrade mature leachate.

Food waste has become a serious problem because of the eating habits in China and the country's generally

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booming food service industry. It is estimated that the economic value of wasted food exceeds 200 billion yuan every year, most of which is produced in public locations, such as restaurants and other commercial service places [10]. The moisture content in food waste is often high (70%–90%) and the nutrient content is usually sufficient, which makes it an ideal substrate for anaerobic digestion [11]. When food waste is anaerobically digested on its own, the hydrolysis and acidification processes are easily controlled by lactic acid fermentation [12]. The accumulation of propionic acid in volatile fatty acids (VFAs) is not conducive to the utilization of methanogens [13], leading to acid inhibition and ammonia inhibition in the anaerobic digestion system. Some scholars have reviewed the research progress of anaerobic digestion of food waste and believe that the combined anaerobic digestion of food waste and other biomass wastes could improve the efficiency of anaerobic digestion [14].

Landfill leachate treatment and food waste resource utilization are two main problems in domestic waste management in China. When the locations of municipal public utilities are planned, many anaerobic digestion and food waste treatment facilities and treatment facilities of food waste are proposed to be built nearby and end up in close proximity to landfill sites. The co-digestion of leachate and food waste proposed in this paper can be realized through the integrated construction and management of leachate treatment facilities and food waste treatment facilities, which is conducive to improving the operating efficiency of these treatment facilities.

Anaerobic digestion of food waste mainly uses microorganisms to degrade the organic components of biomass. The microorganisms in the system primarily come from the inoculum [15]. In the process of anaerobic digestion, the quantity and quality of the inoculum are very important for the operation efficacy and stability of the methane production stage in anaerobic digestion [16]. At present, in the study of anaerobic digestion of food waste, researchers usually use domesticated anaerobic sludge as the inoculum [17-19], and some researchers use materials such as cow dung as inoculum [20-22]. A sanitary landfill is actually a giant anaerobic digestion bioreactor [23-25], and it is rich in various anaerobic microorganisms that have been domesticated for a long time [26-28]. The landfill leachate must be rich in these microorganisms, and it can also be one of the sources of microorganisms in the anaerobic digestion system [29,30]. Therefore, landfill leachate may be a good substitute for inoculum as the digestion solution for anaerobic digestion of food waste.

In this paper, a leachate anaerobic digestion experiment was carried out in a sequencing batch anaerobic digestion reactor, food waste was added to leachate to observe the effect of food waste on the degradation of DOM in the leachate. Three-dimensional fluorescence spectrum and fluorescence regional integration (FRI) analysis methods were used to reveal the conversion mechanisms of soluble organic matter in the combined digestion of leachate and food waste, to clarify the synergistic effect of leachate involved in combined anaerobic digestion. The discovery of these co-digestion and transformation mechanisms will help improve the DOM biodegradation problem in leachate. The knowledge acquired from exploratory this kind of research can be applied to practical solutions for future landfill leachate treatment. This kind of synergistic effect can be realized in engineering applications, thus resulting in large-scale economic and environmental benefits.

2. Materials and methods

2.1. Experimental material

Raw material leachate was taken from Chenjiachong landfill in Wuhan, China. The Chenjiachong landfill has been in operation since 2007. Table 1 summarizes the biochemical parameters of raw leachate and food waste. Food waste was collected from the canteen of Huazhong University of Science and Technology. The main components of the food waste were rice, vegetables and meat. The inoculation sludge was obtained from the anaerobic digestion system of Cofco Meat Food Co. Ltd., (Wuhan, China). The sludge moisture content was approximately 87.30%, the suspended solid concentration (MLSS) was approximately 112.82 g TS/L, the volatile suspended solid concentration (MLVSS) was approximately 79.68 g TS/L, the MLVSS/MLSS was approximately 0.706, and the particle size was approximately 1.5 mm.

2.2. Experimental methods

Six identical batch reactors were used for the anaerobic co-digestion experiments. The total amount of raw material and inoculation sludge in each reactor was 1,500 g, and the sludge inoculation rate was 20% (300 g). To prevent the inhibitory effect of ammonia nitrogen, water was added to the leachate to adjust the concentration of ammonia nitrogen before combined anaerobic digestion. The concentration of anaerobic digestion stock was 2,000 mg/L. There were four different initial organic loading rates of 10-40 g TS/L, and their experimental numbers were Load21-Load24. In addition, three sets of experimental reference systems, Load30, Leach34 and Water04, were set up. In Load30, only leachate was added to the inoculated sludge; in Leach34, the concentration of ammonia nitrogen in the digestive stock was adjusted to 3,000 mg/L, and Water04 is a separate anaerobic digestion system for food waste without that lacks leachate in the digestive stock. Table 2 shows the experiment numbers and conceptual design of the 7 groups of experiments.

The anaerobic digestion experiment was conducted at mesophilic conditions (35°C), and the temperature deviated less than 1°C above or below this value. When the anaerobic digestion experiment started, the gas produced during the digestion process was recorded on a daily basis. A Shimazu GC-2014c gas chromatograph (Japan) was used to measure carbon dioxide and methane in biogas. The measurement interval was determined according to the conditions of the reactor. Parameters such as the pH and VFA of the biogas slurry were measured on an irregular basis. The VFA was measured jointly of acetate, propionate, butyrate and others. Chemical oxygen demand (COD) in leachate was determined by the combination of COD intelligent digestion instrument (6B-12, Shanghai ShengAoHua Environment Protection Technology Co., Ltd., Shanghai) and visible light spectrophotometer (UV1800, Shanghai



Fig. 1. Curve of relative parameters of anaerobic digestive system with time: (a and b) change of gas production rate per unit mass with time, (c) Variation of methane concentration with time under different initial loading conditions, (d) VFA changes with time under different initial load conditions, (e) pH changes with time under different initial load conditions, and (f) pH changes with time under different reference conditions.

Chromatographic Instrument Co., Ltd., Shanghai), referred to the Standard Method HJ/T 399-2007. pH value was determined by pH meter (PHS-3C, Shanghai ShengAoHua Environment Protection Technology Co., Ltd., Shanghai). NH₄–N concentration was determined by Micro-Kjeldahl Method. VFAs value was determined by titration analysis, referred to the Standard Method Q/YZJ10-03-02-20001.

2.3. Three-dimensional excitation-emission matrix fluorescence spectrum analysis

Various kinds of fluorescent substances contained in DOM have different reactions when excited at certain wavelengths, which show as fluorescence peaks at different positions in the three-dimensional fluorescence spectrum [31]. The three-dimensional excitation-emission matrix and fluorescence region integral methods were adopted [32,33]. These methods can be used for quantitative analysis of various organic compounds in DOM, and even though they have been applied to analyze DOM in landfill leachate [34–38], they are seldom used to analyze DOM in biogas slurry [39].

In this study, the dissolved organic carbon (DOC; mg/L) of leachate and biogas slurry was determined by the direct determination method (NPOC) using a German multi N/C 2100 DOC tester. An FP-6500 fluorescence spectrometer was used to determine the three-dimensional excitationemission matrix and perform fluorescence spectroscopy. The excitation light source was a xenon lamp, the PMT voltage was 700V, the excitation and emission slit widths were

Table 1		
Parameters	of digestion	materials

Parameters	Raw leachate	Parameters	Food waste
COD (mg/L)	2,500 ± 25	Protein (%)	3.37 ± 0.12
DOC (mg/L)	$1,298 \pm 11$	Fat (%)	4.43 ± 0.14
NH_3^+-N (mg/L)	$3,625 \pm 25$	TOC (g/kg TS)	450 ± 5
рН	8.54 ± 0.12	Carbohydrate (%)	21.37 ± 0.28
Salinity (g/L)	8.18 ± 0.12	Moisture (%)	69.52 ± 0.42
		C/N	25.39 ± 0.22
		Salinity (g/L)	8.40 ± 0.12

COD: chemical oxygen demand; DOC: dissolved organic carbon; TOC: total organic carbon; C/N: carbon to nitrogen.

Table 2
Combinations of co-digestion materials

Experimental group	Sample number	Food waste load (g TS/L)	Food waste (g)	Landfill leachate (mL)	Water (mL)	NH ₃ –N (mg/L)
Load30	L30	0	0	1,200	0	3,625
Load21	L21	10	39.2	639.2	521.6	2,000
Load22	L22	20	79.2	617.6	503.2	2,000
Load23	L23	30	116.0	597.6	486.4	2,000
Load24	L24	40	157.6	574.4	468.0	2,000
Load34	L34	40	157.6	861.6	180.8	3,000
Water04	W04	40	157.6	0	1,042.4	0

5 nm, the excitation wavelength range was 200–450 nm (wavelength was 5 nm), the emission wavelength range was 250–550 nm (wavelength was 2 nm), and the scanning speed was 1,200 nm/min. Before analysis, deionized water was used as a blank sample to calibrate Raman scattering, and the water samples were adjusted to pH = 7. The dilution ratio of DOC in the biogas slurry in each group was the same. The fluorescence spectrum of the sample was subtracted from that of deionized water to remove the effects of Raman scattering and Rayleigh scattering.

3. Results and discussion

3.1. Cumulative gas production per unit dry material

Fig. 1a shows that the cumulative gas production per unit dry material increases with time. The first 6 d after the start of anaerobic digestion comprise phase 1, which is typically the peak production period of the anaerobic digestion system. The second phase is the continuous gas generation phase. As seen in Fig. 1a, the unit dry material gas production of Load24 in stage 1 is greater than that of Load23; and the unit dry material gas yield of Load23 is much higher than that of Load21 and Load22.

It can be seen from Table 3 that in the first 6 d after the experiment began, the gas production per unit dry material accounted for approximately half of the total gas production per unit dry material for systems Load21-Load23, and for Load24 it accounted for 73.15% of it. Thus, it can be seen that for the experimental anaerobic digestion system

Load24, the effect of leachate on the anaerobic digestion of kitchen waste is mainly reflected in the start-up stage of the anaerobic digestion system.

3.2. Gas rate and methane concentration

Fig. 1b shows the gas production rate of the anaerobic co-digestion reactor when the ammonia nitrogen concentration of the biogas slurry is 2,000 mg/L. It can be seen from the figure that the gas production rate of the other three tanks is relatively stable, except that the gas production rate of the Load23 reactor fluctuates greatly many times. Among the four groups of experimental data, the Load24 reactor with an initial load of 40 g/L TS had the highest peak gas production rate of Load23 with an initial load of 30 g/L TS fluctuated at a larger amplitude, which indicates an obvious unstable state.

Fig. 1c shows the change in methane concentration over time. It also shows that the methane concentrations of Load21 and Load22 typically remained at a higher level with a small fluctuation range. Additionally, it shows that the Load23 reactor produces the largest amount of methane, has the lowest methane concentration and has the most fluctuation in its gas production rate curve. When the initial load is 40 g TS/L, the stability of the anaerobic digestion system is the best, and the methane concentration is the most stable.

3.3. pH and VFA

Methanogens require a meta-alkalescent environment. The optimal pH range is 7.3-8.6. During the anaerobic digestion of food waste, the hydrolysis and acidification rates are too fast, and acid inhibition easily occurs. Leachate is generally more alkaline when it is mature, so leachate has a certain pH buffer capacity. The results of this experiment show that the acid buffer capacity of leachate is important for the stability of methane production. As seen in Figs. 1d-f the combined anaerobic digestive system with leachate added has a strong acid buffer capacity. When the initial load is less than or equal to 40 g TS/L, the combined anaerobic digestion experimental system enters the stage of stable gas production. The combined digestive system can recover from the acidic state to the alkaline state even at high rates of hydrolysis and acidification, with VFAS up to 40,000 mg/L (as shown in Fig. 1d). However, when water is used as the anaerobic digestive stock (Water04), the anaerobic digestive system of food waste is rapidly acidified and acid inhibition occurs.

3.4. Three-dimensional fluorescence spectrum

Fig. 2 shows the fluorescence spectrum of the leachate sample and anaerobic digester. As shown in Fig. 2, the FRI method was adopted to divide the fluorescence spectrum into 5 regions [32]: tyrosine protein regions (Ex: 200–260; Em: 250–340), tryptophan protein regions (Ex: 200–260; Em: 340–380), fulvic acid regions (Ex: 200–260; Em: 380–550), soluble microbial byproducts regions (Ex: 260–450; Em: 200–380), and humic acid regions (Ex: 260–450; Em: 380–550).

As seen from Fig. 2, the leachate sample raw leachate (LR) had three fluorescence peaks, which were located in the protein fluorescence region, the fulvic acid fluorescence region and the humic acid fluorescence region. In the samples form anaerobic digesters with leachate and food waste, the fluorescence peak in the fluorescent region of humic acid disappeared. The three-dimensional fluorescence spectra of 5 samples (L21, L22, L23, L24, and L34) showed a high degree of consistency. The spectrum of sample W04 with water as the digestion solution was completely different. The three fluorescence peaks were located in the protein-like region and microbial by-product region. The fluorescence range was found to be significantly smaller than that of other samples. This demonstrates that the combination of leachate and food waste for anaerobic digestion can promote the degradation of protein soluble organic matter and microbial by-products.

3.5. Fluorescence spectral region integral

The relative intensities $P_{i,n}$ and the standard volume of the total fluorescence region $\Phi_{T,n}$ of each region were calculated by the continuous integral formula. The components of DOM were analyzed quantitatively [32,33,40]. Table 4 shows the DOC of leachate and biogas slurry, the relative fluorescence intensity $P_{i,n}$ and the total fluorescence volume $\Phi_{T,n}$ in each region. $\Phi_{T,n}$ can indirectly and quantitatively reflect the degradation of organic matter. Generally, the higher the $\Phi_{T,n}$ is, the lower the content of non-fluorescent organic matter in the biogas slurry is, which indicates that the organic matter in the digested raw material has been fully degraded. Table 4 shows that the $\Phi_{T,n}$ value of sample L22 is the highest, while that of W04 is the lowest, indicating that anaerobic co-digestion is helpful to the degradation of organic matter.

As seen from Table 4 and Fig. 3, the content of humus (humic acid + fulvic acid) in the 5 samples combined with anaerobic digestion was not significantly different, with percentages of 69%, 66%, 70%, 68%, and 64%, but the DOC of L21, L22 and L23 was approximately 50% of LR in the leachate sample. After combining leachate with food waste for anaerobic digestion, DOM was reduced by approximately half. When the leachate was digested by anaerobic mono-digestion, the content of DOC was 72% of the leachate, which emphasizes how anaerobic co-digestion was more conducive to the reduction of DOC. It can be concluded that food waste can promote the degradation of leachate DOM when the initial load is 30 g/L TS and that the percentage of humus in biogas slurry can be increased. Different results were observed when the content of DOC in biogas slurry of food waste containing water is as high as 8,630 mg/L. However, when the digestion stock is leachate, the content of DOC in the biogas slurry is greatly reduced. Thus, leachate can promote DOM degradation in food waste.

Literature reports [41–43] suggest that, compared with fulvic acid organic matter and humic acid organic matter, tyrosine organic matter, tryptophan organic matter and microbial by-products are more easily degraded, which increases the humus proportion in DOM after anaerobic digestion. On the other hand, studies have shown [32] that humic acid has better solubility, higher quinone content, and is more easily reduced by microorganisms than fulvic acid humus. Cui et al. [44] believe that there are microorganisms in leachate and that some microorganisms will synthesize carbon sources into humus substances under sufficient carbon sources. However, when carbon source is poor, other microorganisms will degrade humus,

Table 3

Comparison of gas production in stage 1 and stage 2 of Load1-Load4 reactors (mL/g)

Experimental number	Load21	Load22	Load23	Load24
Total gas production per unit dry material	588	756	1,280	1,036
Phase 1 gas production (1–6 d)	304	370	672	758
Phase 2 gas production (7–30 d)	284	386	608	278
Phase 1 gas production/total gas production (%)	51.70	49.00	52.50	73.15
Phase 2 gas production/total gas production (%)	48.30	51.00	47.50	26.85







L21: 10g TS/Linitial lord biogas slurry



L23: 30g TS/Linitial lord biogas slurry





initial load is 40g TS/L



L30: Anaerobic mono-digestionleachate



L22: 20g TS/Linitial lord biogas slurry



L24: 40g TS/Linitial lord biogas slurry



W04: biogas slurry which initial NH₃-N is 0mg/L



DOC of leachate and digestate, relative fluorescence intensity of each region ($P_{i,n}$), and total fluorescence volume ($\Phi_{T,n}$)								
Sample	Food waste	ood waste DOC	P _{i,n} (%)					$\sum \Phi_{T_n}$ (×10 ⁻⁶)
number	load (g TS/L)	(mg/L)	Humic acid	Microbial by-products	Fulvic acid	Tryptophan	Tyrosine	(AU-nm ² -[mg/L] ⁻¹)
LR	_	1,298	14	17	47	14	8	242.3
L30	0	935	15	11	54	16	4	277.3
L21	10	500	16	14	50	15	5	313.3
L22	20	472	15	10	55	15	5	304.6
L23	30	574	15	11	53	16	5	276.5
L24	40	1,113	16	17	48	17	2	159.1
L34	40	1,604	4	16	53	12	15	146.6
W04	40	8,630	17	23	27	21	12	29.5



Fig. 3. Relative fluorescence intensity of DOM in leachate and biogas slurry.

which can be used as nutrients for their own growth. The experimental results of this paper are consistent with the above conclusions.

4. Conclusions

Table 4

Anaerobic co-digestion of leachate and food waste can not only promote the anaerobic digestion process of food waste, but also improve the gas production and methane production rates. It can also promote the degradation of DOM in leachate. The anaerobic co-digestion of leachate and food waste has a synergistic effect. The anaerobic co-digestion of landfill leachate and food waste can maintain the buffer balance between ammonia and VFAs, overcome the inhibitory effect of ammonia and VFAs, and improve the stability of the system. When the initial load rate of food waste is 20 g TS/L, the DOC content in biogas slurry can be reduced to 36% of raw leachate and 50% of leachate from anaerobic mono-digestion. The results of this study showed that anaerobic co-digestion of leachate and food waste had synergistic effect. This kind of synergistic effect can be realized in engineering applications, thus resulting in large scale economic and environmental benefits.

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