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Leachate pretreatment for enhancing organic matter conversion in landfill bioreactor

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Abstract

Direct recycling of leachate from refuse of high food waste content was shown to ineffectively stabilize the refuse. This work aims at evaluating the effects of three pretreatments of leachate on the refuse stabilization efficiency were investigated. Pretreatment of leachate using an anaerobic upflow filtration bioreactor (UFB) or a well-decomposed waste layer could reduce the COD and provide methanogens, both were beneficial to establish early methanogenesis status. Using an aerobic sequential batch reactor (SBR) to pretreat the leachate could reduce its COD to 1000 mg l^{-1}, but the fully developed methanogenesis phase would be built up in a later stage. The organic matters in the effluent leachate inhibited both the hydrolysis/acidogenesis and the methanogenesis steps in the refuse. With the dilution and acid neutralization effects by the recycled leachate, a favorable methanogenetic environment could be produced from the column's top, which moved downward along, and finally made the breakthrough of the column.

Keywords: Landfill bioreactor; Leachate pretreatment; Organic matter conversion

1. Introduction

Landfill bioreactor could provide an accelerated stabilization of the landfilled municipal solid waste (MSW) as well as an enhancement of methane production [1]. Full-scale tests reveal the merit of this application as a potential alternative to the conventional landfill [2–4], with the leachate recycling as the key step to its success in operation. However, some tests conducted using MSW collected at very different composition could note no expected, enhanced effects on landfill stabilization [5]. On the contrary, as we demonstrated in this study, the simple leachate recycling through the landfill unit would inhibit metabolism of organic matter degradation [6]. Veeken et al. [7] and Vavilin et al. [8] noted a similar failure for anaerobic degradation of waste at a high volatile fatty acid (VFA) level.

Kim [9] constructed three simulated landfill reactors, one with leachate recycling without pretreatment (CTR), one with pretreated leachate recycling (CNT), and the third as the dedicated leachate pretreatment zone (DTZ) at active methanogenesis stage linked with the CNT mentioned above. The comparison between landfill bioreactor with and without a dedicated treatment zone showed that the former could accelerate waste stabilization. However, Kim's results lack generality since the waste in the DTZ was of a special nature. Nopharatana et al. [10] operated a process including one reactor containing fresh waste and one reactor containing anaerobically stabilized waste with sequencing leachate recirculating procedure for the degradation of municipal solid waste. Results showed that such process could shorten start-up periods in the fresh waste reactor. Christ et al. [11] showed that the two-stage process involving both thermophilic and mesophilic conditions had higher degradation rates of dissolved carbon-containing compounds than the one-stage process. Veeken and Hamelers [12] demonstrated that both the substrate-seed mixing degree and leachate recirculation rate may determine how fast VFA could transfer in the biowaste-seed bed. Price et al. [13] measured the denitrification potential of landfill bioreactors and demonstrated that a landfill bioreactor is technically viable for conversing nitrate to nitrogen gas. Ex

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situ nitrification process was noted to be effective for removing high concentrations of ammonia in landfill leachate. In addition, the ex situ treatment processes could also prevent the failure of landfill bioreactor as noted by Bae et al. [14], who directly recycled the leachate without any pretreatment back to their landfill bioreactor.

Although different acidification process has been compared by [15] with two simulated sanitary landfill columns compacted with different percentage of biodegradable waste, little works has been done to show the different waste degradation processes in the bioreactor landfills with different percentage of biodegradable waste. Furthermore, ex situ anaerobic pretreatment have been widely applied for leachate recycling in bioreactor landfills, little works have been done on comparing the effects of different types of pretreatment leachate processes for leachate recycling on waste degradation, i.e. aerobic or anaerobic leachate pretreatment.

The objectives of this research are two folds. Firstly, we demonstrate that direct recycling of leachate generated from landfilled MSW of Shanghai, China, did inhibit methanogenesis owing to acid stuck, because of the existence of excess biodegradable food waste. A test with low fraction of biodegradable food waste was also conducted as a comparison. Then, we evaluated how the three pretreatment processes, such as anaerobic upflow filtration bioreactor (UFB) or a mature landfill layer as the anaerobic reactor, or a sequential batch reactor (SBR) as the aerobic reactor, could modify the leachate characteristics, in order to enhance MSW stabilization in landfill bioreactor.

2. Materials and methods

2.1. Materials

The synthetic refuse used in this study was based on the composition noted for municipal solid waste (MSW) in Shanghai, China [16] (Fig. 1). Food waste was collected in a refectory in Tongji University, Shanghai, China. Plastic, paper, glass, metal and textile were from a waste recycle station in Shanghai. The relatively high fraction of food waste (53%, w/w on dry basis) led to high water content of fresh refuse (74%, w/w). The prepared waste sample represented typical municipal solid waste in Shanghai, China. This synthetic waste was placed in column nos. 1, 2, and 4–6 for test. Another synthetic refuse was prepared the same as mentioned above except that the food waste was low, 3.5% (w/w) on dry basis as proposed by Kim [9]. This refuse was placed in column no. 3 for further test.

2.2. Anaerobic degradation

Fig. 2 depicts the alignment of the six refuse degradation systems under investigation. The effluent leachate from column nos. 4–6 had been pretreated before recycling. The pretreated leachate was termed as “treated leachate” in this work.

Six simulative landfill columns (inner diameter of 37 cm, height of 230 cm) were packed with synthetic refuse (Fig. 3). A drainage port, 2.5 cm in diameter, equipped with a ball valve was placed at the column bottom. A gas vent and a liquid addition port, 2.5 cm in diameter each, were installed

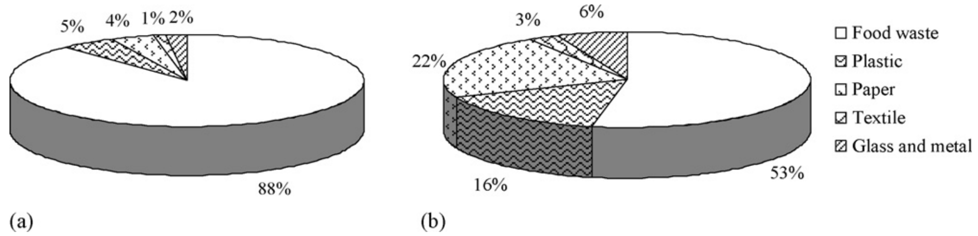

Fig. 1. The composition of the refuse used in column nos. 1 and 2, and 4–6 in this study: (a) moisture content basis and (b) dry basis.

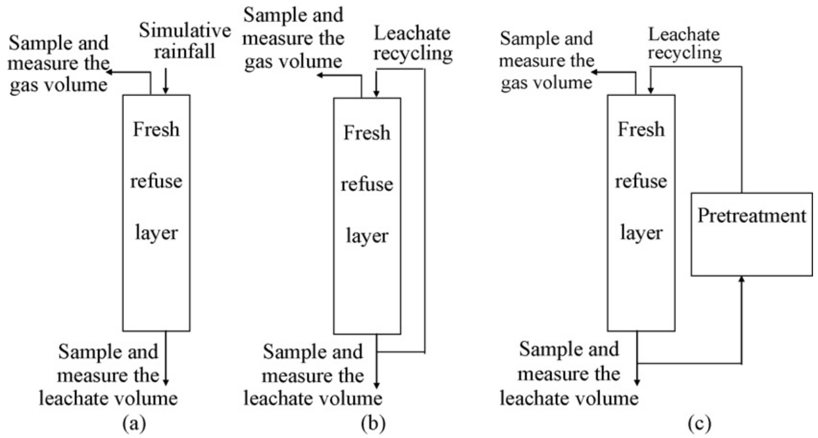

Fig. 2. Schematics of the reactor configuration—(a) column no 1: with simulated rainfall; (b) column nos. 2 and 3 direct recycling of leachate; (c) column nos. 4–6: with leachate pretreatments.

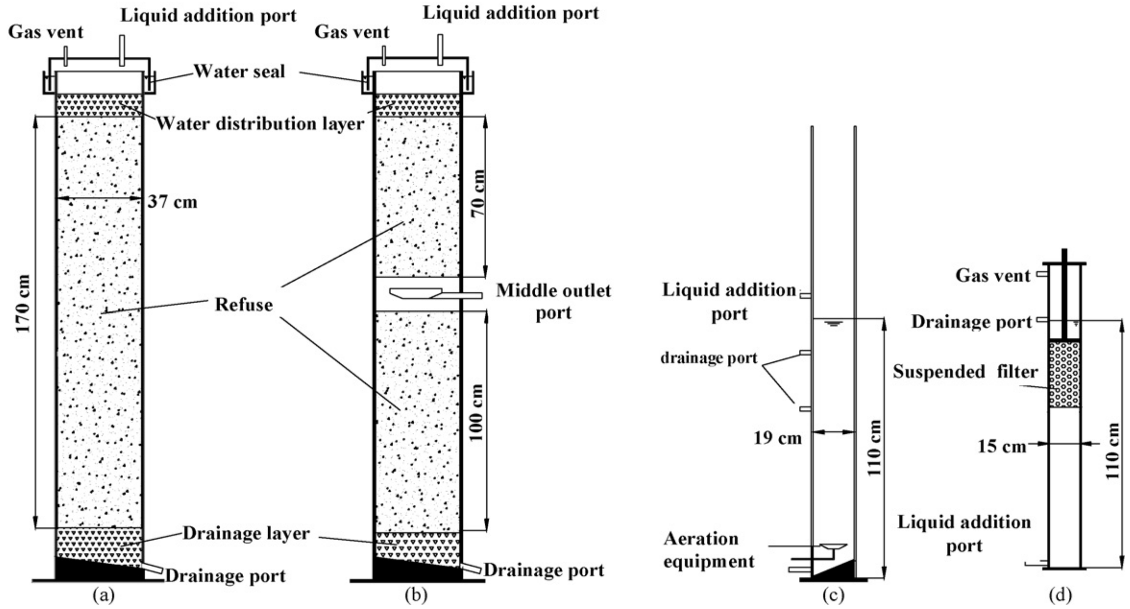

Fig. 3. Schematics of the reactors: (a) simulated column nos. 1–4 and 6; (b) column no. 5; (c) aerobic SBR; (d) anaerobic UFB.

at the column top. Free drainage conditions at the bottom of the column were simulated by placing below the refuse a 10-cm layer consisting of pottery grains (diameter: 1 cm) made from kilned argil. The refuse was then placed in lifts of 15 cm in thickness. Each lift was compacted using a hand tamper. The total thickness of the refuse layer before testing was 170 cm. For column no. 5, two layers of waste of thickness 100 and 70 cm, respectively, were packed to a thickness of 170 cm. The average density of solid waste in the columns was 0.8 tonnes $\mathrm{m^{-3}}$. A layer of pottery grains with thickness of 10 cm was placed over the refuse for even distribution of liquid added to the columns. All the columns were placed in the thermostat, and the temperature of the columns were kept at an average temperature of 35 °C. After being packed, the columns were gas-tight.

Tap water was added on a weekly basis to column no. 1, which was served as the control. The added quantity of the tap water (Q) ranged 0.2–0.6 $\mathrm{mm\,m^{-2}\,d^{-1}}$, and this simulating the monthly rainfall data in Shanghai area (Table 1). (*Note*: we assumed herein that the final cover would be effective for allowing only 10% of the rainfall to flow into the landfill.) The effluent leachate was directly recycled into column nos. 2 and 3 without pretreatment. Note that the fraction in food waste was low in column 3. The effluent leachate from column no. 4 was treated externally in an anaerobic UFB prior to recycling during the first 10 weeks. After the methane content in landfill gas had reached 50% (v/v) at the 8th week, the effluent leachate was directly recycled. The anaerobic UFB (inner diameter 13 cm, height 150 cm) had an effective volume of 17.6 l. Activated sludge (suspended solids of 0.3%) from a wastewater treatment plant in Shanghai had been inoculated in the UFB for several days with high strength leachate. Effluent leachate from column no. 4 was continuously pumped into the bottom of the UFB reactor with the retention time of 8 d. Then, the treated leachate was pumped back to the top of column no. 4 at a rate of $2.2 \pm 0.5\,\mathrm{l\,d^{-1}}$. The schematic of the reactor was showed in Fig. 3. Part of the effluent leachate from column no. 5 was fed through a well-decomposed waste layer, excavated from a section at the Laogang Landfill Works, Shanghai, closed for 5 years at the time of sampling (Table 2). The treated leachate was mixed with part of the effluent leachate from the column to make the recycle stream back to column no. 5. The effluent leachate from column no. 6 was treated in an aerobic SBR prior to recycling during the first 11 weeks. Then, the effluent leachate was recycled without pretreatment. The SBR (inner diameter of 19 cm and height of 200 cm) had an effective volume of 28.4 l and a volume loading of 1–5 $\mathrm{kg\,COD\,m^{-3}\,d^{-1}}$. Intensive aeration was adopted in the first 4 h, and the dissolved oxygen (DO) remained at the level of 0.1–0.3 $\mathrm{mg\,l^{-1}}$. The reactor was operated at low aeration rate of 0.6–1.2 $\mathrm{m^3}$ air $\mathrm{h^{-1}}$ over the remaining 20 h. At the end of the operation period, the mixed liquor (water and excess sludge) was partially drained. The supernatant of the mixed liquor was recycled back to column no. 6. The schematic of the SBR reactor was also shown in Fig. 3.

Table 1
Monthly rainfall data in Shanghai area

Month	Rainfall (mm)
1	44.1
2	62.3
3	79.3
4	103
5	126
6	153
7	93.2
8	115
9	174
10	56.7
11	50.8
12	42.5

Table 2
Characteristics of the well-decomposed waste

	Mass content (%)
Food waste[a]	–
Plastic	13.5
Paper	1.5
Textile	0.5
Glass and metal	5.5
Residue	79.0
Total	100
Element	
C	12.3
H	1.3
N	0.8
S	0.5
O	2.5
Ash	82.6

[a] Food waste could not be picked out from the residue.

Table 3 summarizes the volume and the characteristics of the recycled liquids for each column. The pretreatment was stopped and the effluent leachate was directly recycled in tests using column nos. 4–6 once the COD level of leachate from refuse column had declined to about 20,000 mg l^{-1}. For instance, the anaerobic UFB for column no. 4 stopped operation since week 9, from then the effluent leachate was directly recycled. Similarly, the leachate stopped flowing through the well-decomposed waste layer since week 19, and the aerobic SBR stopped running since week 12.

2.3. Sample analysis

Quality of the influent and effluent of each column was analyzed daily, including COD, total organic carbon (TOC), BOD_5, VFA, ammonia nitrogen (NH_4^+-N), Kjeldahl nitrogen (KN), pH, and the leachate flow rate. The volume and the composition of the produced landfill gas were measured by the wet flow meters and by gas chromatography.

The COD was analyzed by dichromate reactor digestion method and BOD_5 by the dilution and seeding method according to the standard methods (State Environmental Protection Administration of PR China, 1989). The pH value was measured by a PHS-25 digital pH meter (Shanghai Precision & Scientific Instrument Co. Ltd.). Ammonia was analyzed by alkali method and KN was analyzed according to the standard methods [17]. TOC was analyzed with the TN_b/TC multi N/C 3000 Analyzer (Analytik Jena AG). The furnace temperature was 850 °C, and the sample volume was 0.6 ml. The concentrations of the VFA were determined by a gas chromatograph (GC-122) equipped with a flame ionization detector and a 30-m Dikma Capillary Column. The operational temperatures for the injection port, the oven, and the flame ionization detector were 200, 120 and 200 °C, respectively. Hydrogen gas was used as the carrier gas at a flow rate of 50 ml min^{-1}. Before the analysis of the VFA, phosphoric acid was added to control the pH of the samples. Only acetate, propionate and butyrate are tested. The

Table 3
The volume and characteristics of the water or leachate added to the columns

Column (1)	Pretreatment (2)[a]	Week (3)	Experimental start month (4)	Characteristics of the liquid added to the columns					Volume of recycled leachate (mm m^{-2} d^{-1}) (10)	Frequency of recycling (11)
				pH (5)	BOD_5 (mg l^{-1}) (6)	COD (mg l^{-1}) (7)	VFA (mg l^{-1}) (8)	NH_3-N (mg l^{-1}) (9)		
No. 1	Tap water	1–30	June 2002	6.9	0	0	0	0	0.2–0.6	Weekly
No. 2	[b]	1–2	July 2002	5.6–5.8	30000–45000	55000–65000	25000–38000	1200–1600	[c]	Daily
No. 3	[b]	1–19	April 2003	5.3–7.6	400–37000	2650–53500	500–16000	95–670	[c]	Daily
No. 4	Anaerobic UFB	1–8	December 2002	7.8–8.5	2500–3500	5000–7000	2000–3400	1000–1500	20 ± 5 mm	Daily
	[b]	9–27	–	Same as those of the leachate					[c]	Daily
No. 5	[d]	1–18	October 2002	6.4–6.7	6000–8000	ca. 20000	8000–10000	1500–1700	6 ± 2 mm	Daily
	[b]	19–24	–	Same as those of the leachate					[c]	Daily
No. 6	Aerobic SBR	1–11	August 2002	8.0–8.4	60–230	400–1000	<100	0–200	20 ± 5 mm	Daily
	[b]	12–27	–	Same as those of the leachate					[c]	Daily

[a] Except for column no. 1, which was added with tap water throughout the experiment period, pretreatment of leachate was stopped once the COD level of leachate from refuse column had declined to less than 20,000 mg l^{-1}.

[b] Leachate was recycled without pretreatment.

[c] All the leachate produced was recycled.

[d] Recycled liquor is a mixture of leachate with and without pretreatment.

concentrations of methane and carbon dioxide in biogas were analyzed using a gas chromatograph (GC-102) equipped with a thermal conductivity detector. The operational temperatures of the injection port, the oven, and the thermal conductivity detector were 20, 90, and 90 °C, respectively. Hydrogen gas was used as the carrier gas at a flow rate of 60 ml min^{-1}.

3. Results and discussion

3.1. Leachate quality

The residual air in the sealed column would be exhausted during the first several weeks of testing. This initial aerobic period was disregarded in all following discussions for providing a common basis for performance comparison.

All columns produced strong effluent leachate in the initial stage: COD ≈ 60,000 mg l^{-1} (TOC ≈ 20,000 mg l^{-1}), VFA ≈ 30,000 mg COD l^{-1}, and pH 5.5–6.0. This observation revealed that the present refuse was easily hydrolyzed in the landfill columns. Over the testing period the COD concentrations of effluent leachate from column nos.1 and 2 remained at a high level (Fig. 4a). The corresponding pH values were all around 5.5 (Fig. 4b). A mass amount of VFA was produced (Fig. 4c), contributing almost half of the whole COD (Fig. 4a). Hence, adding tap water (column no. 1) or simply recycling the effluent leachate (column no. 2) could not establish favorable landfill stabilization within the testing period. The effluent leachate from column no. 1 reached a level of 70,000 mg COD l^{-1}, which was higher than that from column no. 2. This occurrence may be attributable to the inhibition of organic matters in leachate on waste hydrolysis.

The leachate COD of column no. 3 was reduced from 53,500 to 11,000 mg l^{-1}, 80% reduction achieved in 10 weeks by directly recycling leachate without pretreatment (Fig. 4a). Correspondingly, the leachate pH was raised from 5.6 to 7.6 since week 9 (Fig. 4b). Direct leachate recycling to a landfilled MSW with low food waste fraction (3.5%, w/w) could thereby effectively degrade organic matters in leachate.

For column no. 4, the recycled leachate was pretreated by anaerobic UFB to reduce the COD from 62,600 (Fig. 4a) to 6500 mg l^{-1} (Fig. 4d). Afterward, the effluent leachate's COD declined continuously to a low level of around 800 mg l^{-1} at the end of week 30. On week 9 the UFB had been turned off and the COD of the recycled leachate jumped up to 20,000 mg l^{-1} on week 9 (Fig. 4d). This occurrence had not affected significantly to the effluent leachate quality (Fig. 4a). The pH of leachate started to increase on week 11 from around 5.5 to 7.5 (Fig. 4b).

Over the first 15 weeks of testing, the leachate from column no. 5 was recycled with COD concentration drop from around 65,000 to 20,000 mg l^{-1} after passing through the waste layer. The COD concentration of the leachate from column no. 5 started to drop on week 16, reached 22,000 mg l^{-1} on week 19, and finally a low level of 4000 mg l^{-1} on week 30. In addition, the pH value of the leachate began to rise on week 20 (Fig. 4b). The leachate was totally recycled back to column no. 5 since week 19.

The oxidation reaction in SBR could reduce the COD concentration of the effluent of column no. 6 to a low value of 1000 mg l^{-1} (Fig. 4c), and the treated leachate recycling yielded the continuous decline of the effluent from column no. 6 (Fig. 4a). This pretreatment led to the fully developed methanogenesis over the column on as late as week 23 (Fig. 4b).

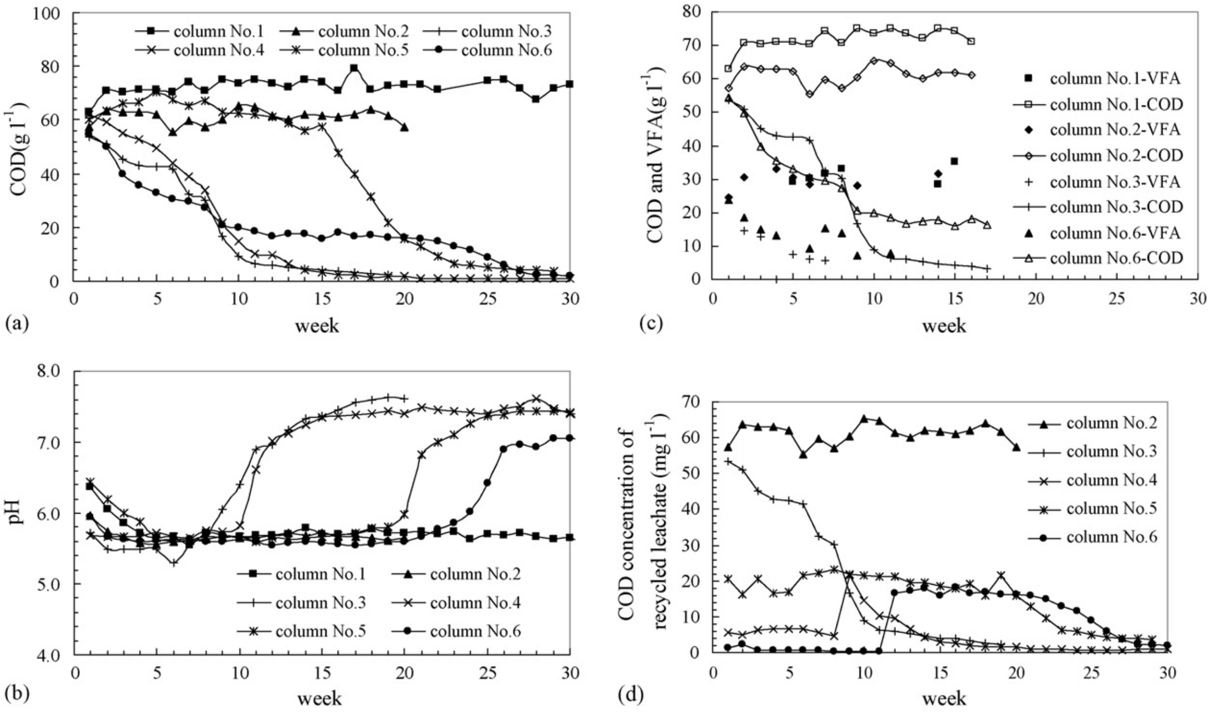

Fig. 4. Evolutions of leachate characteristics from the five columns: (a) COD, (b) pH, (c) VFA, and (d) COD of recycled leachate.

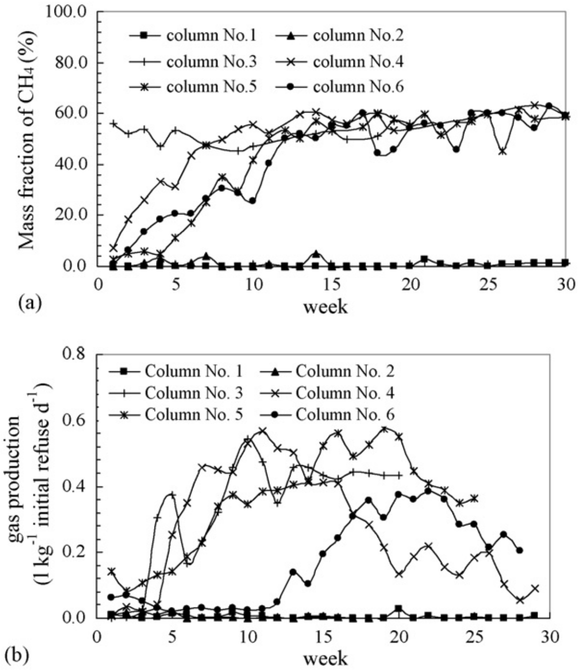

Fig. 5. Evolution of characteristics of landfill gas: (a) mass fraction of methane and (b) gas production rate.

3.2. *Landfill gas*

The methane appeared in the gas phase of column no. 1 and no. 2 since week 21 and week 3, respectively (Fig. 5a). However, their landfill gas production rates were lower than $0.07\,\mathrm{l\,kg^{-1}}$ initial refuse per day (Fig. 5b), and the volume fraction of methane had never exceeded 5%.

Not as those noted in column nos. 1 and 2 in which methanogenesis was largely inhibited, the test of waste with low food waste fraction produced methane since week 1, and the methane concentration remained 45–55% onward (Fig. 5a).

Methane started to appear since the first or the second week from tests with leachate pretreatment (column nos. 4–6). Their volume fraction of methane gradually increased, and reached 50% (v/v), respectively, on weeks 9, 11, and 12, respectively. Except for the test for column no. 4, the rise in leachate pH (Fig. 4b) did not correspond to the trend in methane production (Fig. 5b). This occurrence indicated that the characteristic of the effluent leachate was not an appropriate performance index for the landfill stabilization. More discussion was provided in the last section of this paper.

During the first 4 weeks of testing, the gas production rate from column no. 4 was less than $0.04\,\mathrm{l\,kg^{-1}}$ initial refuse $\mathrm{d^{-1}}$. The production rate considerably increased since week 5. When the effluent leachate was directly recycled since week 9, the gas production rate became even higher, and reached a maximum rate of $0.56\,\mathrm{l\,kg^{-1}}$ initial refuse $\mathrm{d^{-1}}$ on week 11. For column no. 6, the gas production rate remained at a low level, 0.023–$0.067\,\mathrm{l\,kg^{-1}}$ initial refuse $\mathrm{d^{-1}}$ when the leachate was pretreated. More gas was yielded when the SBR stopped running. The peak rate occurred on week 22 at a rate of $0.41\,\mathrm{l\,kg^{-1}}$ initial refuse $\mathrm{d^{-1}}$.

3.3. *Conversion of organic carbon*

The accumulative conversion of carbon from refuse to the gas phase ($\sum$orgCG) or that remained in the liquid phase ($\sum$orgCL) in n weeks could be estimated using Eqs. (1) and (2), respectively.

$$\sum \mathrm{orgCG} = \sum_{i=1}^{n} \left\{ \left[(C_{\mathrm{CH_4},i} + C_{\mathrm{CO_2},i}) \times \frac{V_{\mathrm{G},i}}{22.4} \right] \times 12 \right\} \tag{1}$$

$$\sum \mathrm{orgCL} = \sum_{i=1}^{n} \left\{ \left[(C_{\mathrm{COD,out},i} \times Q_{\mathrm{L,out},i}) - (C_{\mathrm{COD,in},i} \times Q_{\mathrm{L,in},i}) \right] \times \frac{3}{8} \right\} \tag{2}$$

where $C_{\mathrm{CH_4},i}$ and $C_{\mathrm{CO_2},i}$ are volume fraction of CH_4 and CO_2 in the gas phase on week i, $V_{\mathrm{G},i}$ gas production rate on week i ($\mathrm{l\,week^{-1}}$) (in STD), $C_{\mathrm{COD,out},i}$ and $C_{\mathrm{COD,in},i}$ COD concentration of leachate effluent and liquid added into the columns on week i ($\mathrm{mg\,l^{-1}}$), $Q_{\mathrm{L,out},i}$ and $Q_{\mathrm{L,in},i}$ are flow rate of leachate effluent and liquid added into the columns on week i ($\mathrm{l\,week^{-1}}$), respectively. Note that the accumulative amount of organic matter in leachate is calculated based on Eq. (2), assuming that the organic matter comprised most of the reductive materials in the leachate, or 1 g COD = (3/8) g organic carbon.

The total accumulative conversion of organic matter from the refuse ($\sum$orgC) equals ($\sum$orgCG + $\sum$orgCL). Up to the week 8, the total conversions of carbon ($\sum$orgC) were 20.7, 2.3, 4.4, 18.5, 10.9, and 15.2 $\mathrm{g\,kg^{-1}}$ initial dry refuse for column nos. 1–6, respectively (Table 4). Hence, except for column nos. 2 and 3, the organic matters in the refuse were effectively hydrolyzed.

On week 19, at which all pretreatments were ceased to operate, the fractions of hydrolyzed carbon from solid phase in column nos. 1–6 were 34.3%, 3.6%, 16.0%, 52.1%, 50.8%, and 33.8%, respectively. The hydrolysis efficiency in column nos. 4–6 with leachate pretreatment remained at a high level, while 33.2–67.2% of hydrolyzed carbon was converted to CH_4 or CO_2 (Table 4). Adding tap water (no. 1) could only hydrolyze organic carbon from refuse, but could not assist in further methanogenesis. Direct recycling in column no. 2 had no much improvement compared with that on week 8. Nonetheless, direct recycling to waste with low food waste fraction did enhance the stabilization process, as suggested by Pohland and Kim [18]. Carbon conversion in column 3 was 16.0%, slightly more than that presented in food waste fraction in the sample (12.5%). Although food waste seemed almost completely consumed since weeks 10–15 in column no. 3, the gas production rate and methane content remained high up to week 19, the cease of the test. This observation suggested that the food waste fraction, presented as the easily biodegradable materials in MSW, did play an essential role on the success of landfill bioreactor.

3.4. *Effects of leachate pretreatment*

The tests with column no. 4 (anaerobic UFB) could considerably enhance refuse stabilization and yield early methano-

Table 4
Organic carbon conversion in the columns

Column (1)	Initial organic carbon content in the column ($g\,kg^{-1}$) initial dry refuse (2)	Time (week) (3)	$\sum$orgCL ($g\,kg^{-1}$) initial dry refuse (4)	$\sum$orgCG ($g\,kg^{-1}$) initial dry refuse			$\sum$orgC ($g\,kg^{-1}$) initial dry refuse (8)	$\sum$orgCG/ $\sum$orgCL (9)
				CH_4 (5)	CO_2 (6)	Total (7)		
No. 1	69	8th	20.5	0.0	0.2	0.2	20.7	0.01
	69	19th	23.5	0.0	0.4	0.4	23.9	0.02
	69	30th	26.4	0.0	0.5	0.5	26.9	0.02
No. 2[a]	69	8th	2.0	0.0	0.3	0.3	2.3	0.15
	69	19th	2.1	0.0	0.4	0.4	2.5	0.19
No. 3	140	8th	0.2	2.1	2.1	4.2	4.4	21
	140	19th	0.2	11.2	11	22.2	22.4	111
No. 4	69	8th	12.4	2.9	3.2	6.1	18.5	0.49
	69	19th	12.9	12.9	10.5	23.4	36.3	1.81
	69	30th	12.9	16.7	12.4	29.1	42	2.26
No. 5	69	8th	5.8	1.2	3.9	5.1	10.9	0.88
	69	19th	11.6	11.6	12.2	23.8	35.4	2.05
	69	30th	11.7	19.8	17.8	37.6	49.3	3.21
No. 6	69	8th	14.0	0.2	1.0	1.2	15.2	0.09
	69	11th	15.2	0.3	1.2	1.5	16.7	0.10
	69	19th	15.7	3.9	3.9	7.8	23.5	1.50
	69	30th	15.8	10.6	8.6	19.2	35.0	1.22

[a] The operation on column no. 2 stopped on the 20th for the hydrolysis inhibition of organic matters in column no. 2.

genesis. This occurrence might be attributable to the excess methanogenetic bacteria and/or the higher leachate pH recycled back with the pretreated leachate. Recycled leachate to column no. 5 (mature landfill layer) would carry with it the excess methanogenetic bacteria, but may need time to propagate downward with the leachate flow to stimulate methanogenesis [8,19], yielding a delay on the methane production to week 15.

The SBR could effectively reduce the COD of leachate to less than 1000 $mg\,l^{-1}$. With SBR-pretreated leachate recycled back to the column top, the COD of effluent from column bottom dropped from 55,000 to 20,000 $mg\,l^{-1}$ on week 10 (Fig. 4a). The SBR stopped aeration since week 12 but leachate was recycled continuously. Then, the COD of effluent leachate collected at column bottom kept unchanged at around 18,000 $mg\,l^{-1}$ over weeks 12–21 (Fig. 4d), then started to decline again since week 22, dropping to 2000 $mg\,l^{-1}$ on week 30. Meanwhile, the pH of leachate rose to 7.1, indicating the occurrence of the methanogenetic reaction in the landfilled layer.

The mass amount of food waste for the Shanghai's MSW (53%) presented the source for easy hydrolysis (the municipal solid waste generated in US had a fraction of 11.4% (by weight) for food waste [20]). The VFA concentrations (acetate + propionate + butyrate) in column nos. 1–3 and 5 presented in Fig. 4c since the first week. Veeken et al. [7] revealed that high levels of VFA could inhibit both the hydrolysis/acidogenesis stage and the methanogenesis stage. Our results revealed that only if the organic matters of leachate could be considerably removed and the pH increased to neutral or slightly alkaline, waste methanogenesis could be effectively stimulated and enhanced. The methanogenesis in no. 6 was established earlier than no. 4 or 5, indicating that since the SBR could produce a leachate of reduced COD level (1000 $mg\,l^{-1}$), the dilution of local VFA concentration with recycled leachate may also considerably contribute to the enhanced stabilization of MSW.

The liquor inside the refuse column no. 5 was collected at the mid-way of the column height. Fig. 6 shows the COD and pH collected at the mid-way (upper semi-column) and the bottom of column no. 5 (lower semi-column). The decline in COD and

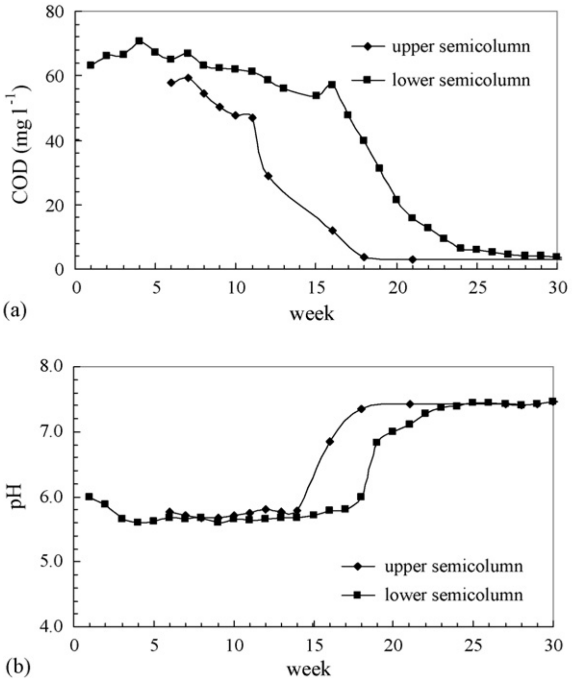

Fig. 6. Characteristics of liquid phase collected at column no. 5: (a) COD and (b) pH.

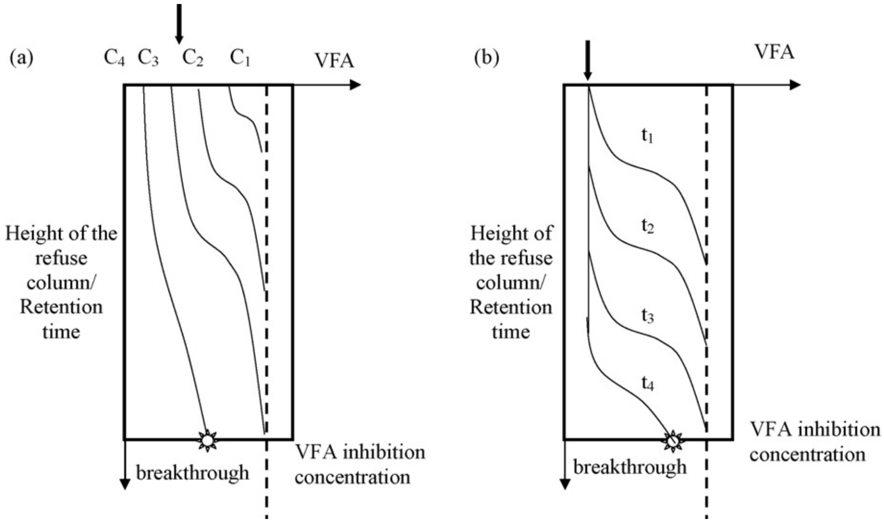

Fig. 7. The conceptual model for VFA penetration through the refuse bed at different loading rates—(a) influent leachate VFA: $C_1 > C_2 > C_3 > C_4$ and (b) time: $t_1 < t_2 < t_3 < t_4$.

the rise in pH occurred earlier at the upper semi-column than at the bottom. This occurrence correlates with the findings of Martin [21].

Fig. 7 depicts a dilution model with recycled leachate. When the recycled VFA was at $C > C_{\text{inhibition}}$, the entire column was inhibited and the leachate COD would not decline, as the case in column no. 2. When the leachate was pretreated to a level at C_1 ($<C_{\text{inhibition}}$), a top section of the column would be diluted. If the organic matters have been diluted to be lower than the inhibition concentration for methanogenesis to occur, methane would be produced if methanogenetic bacteria were present locally. In this regard, the dilution effect would become more profound when the COD of the recycled leachate had been reduced further ($C_4 < C_3 < C_2 < C_1$ in Fig. 7a), which could be the explanation for the earlier methane production for column no. 4 than the column no. 5. When the COD of the recycled leachate was kept at a low level, the leachate COD was consumed and washed downward as time elapsed (Fig. 7b). Breakthrough occurred at time $(t) = t_3$, while before that methane could be produced at the top zone of the column, which interpreted the time lag observed for methane production and the jump in effluent pH in all tests (Fig. 4a and b). This simplified scheme could not be used to explain the long time delay for column no. 6.

A complete scheme for column dynamics should incorporate the factors besides the dilution effect, such as the refuse characteristics, rates of hydrolysis/adsorption of organic matters, local methanogenetic activity, recycled leachate characteristics, and other bed features such as permeability and porosity, and others. However, the basic characteristic of the column dynamics could still be schematically demonstrated in Fig. 7.

Direct leachate recycling could accelerate stabilization of refuse collected at USA in landfill bioreactor [1]. This occurrence might be attributable to its low fraction of food waste in the refuse, which yields lower concentration of organic matters in effluent leachate for possible inhibition. As this work demonstrated, if the food waste (or other easily biodegradable waste) presented in a high level, for example, 53% (w/w) in the present sample, direct recycling of leachate would fail the operation of landfill bioreactor owing to rapid acid stuck. Also, as revealed in Fig. 7a, if the COD in effluent leachate could be effectively removed before recycling, landfill bioreactor could be successfully operated even when MSW of high food waste fraction was handled.

4. Conclusions

Effects of leachate recycling on the stabilization of refuse of the same compositions of that collected at Shanghai City, China, were experimentally explored in six simulated landfill columns. Adding tap water or direct recycling of column leachate could not enhance refuse stabilization. Meanwhile, direct recycling of the waste of low food waste fraction could yield successful operation of landfill bioreactor.

The leachate pretreated with an anaerobic UFB to reduce its COD by 90%, then recycled with methanogenetic bacteria could produce methane from refuse since week 1, and had the whole refuse column entering methanogenetic phase since week 11. Meanwhile, the recycling of leachate pretreated with a well-decomposed waste layer to reduce COD of leachate to about 20,000 mg l^{-1} could have the entire column entering methanogenetic phase on week 20. If based on the amount of leachate pretreated and recycled, the well-decomposed waste layer could initiate earlier methanogenesis with higher methane yield when compared with that with UFB reactor. For column no. 6 with an aerobic SBR the COD of the leachate could be reduced to 1000 mg l^{-1}. However, this pretreatment would lead to a late, fully developed methanogenesis phase.

The high COD level presented in the effluent leachate was speculated to correspond to the noted inhibition effects to both the hydrolysis/acidogenesis and the methanogenesis steps in the landfilled refuse. When the organic matters in the leachate were considerably removed by pretreatments, leachate recycling could accelerate refuse stabilization by producing a diluted, more favorable methanogenetic environment. A methanogenesis "front" would propagate downward from the column's top, producing methane according to the hydrolysis/methanogenesis balance. A conceptual model was used for interpreting the noted inhibition effects.

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