Effects of feed solutions on refuse hydrolysis and landfill leachate characteristics

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Abstract

Tap water, aerobically pre-treated leachate, and anaerobically pre-treated leachate, were each fed into the top of a series of three simulated landfills columns, filled with municipal solid waste collected in Shanghai, China. Changes in leachate, including pH, total organic carbon (TOC), and volatile fatty acids (VFAs), and the produced biogas were monitored over time. The tap-water-fed columns had a low hydrolysis rate that yielded an acidic environment (pH 4.8–5.4) in the leachate that inhibited methanogenesis reaction in the refuse. When aerobically pre-treated leachate was fed into the columns, the hydrolysis rate of total organic carbon fluctuated between 200 and 400 mg d^{-1} and methanogenesis in the refuse column was only partly activated. The hydrolysis rate of refuse fed with anaerobically pre-treated leachate was the highest among the three solutions. The high alkaline levels of the anaerobically pre-treated leachate and its methanogenic bacteria led to an early activation of methanogenesis in the refuse columns. The VFAs contributed approximately 40–60% of TOC in tap-water-fed columns, 60–80% of TOC in the columns fed aerobically pre-treated leachate, and up to 70–90% of TOC in columns fed with anaerobically pre-treated leachate. The feed solution had considerably affected leachate characteristics, and then the build-up of the methanogenesis in the refuse column and the composition of fermentation products in the leachate. The success of a bioreactor landfill depends on whether the recycled leachate could yield a favorable methanogenic environment in the top refuse layer, or whether an appropriate pre-treatment is adopted to modify the leachate characteristics.
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1. Introduction

A bioreactor landfill accelerates stabilization of the landfilled municipal solid waste (MSW), enhances methane production, and costs less than conventional landfill with leachate treatment. Theses factors make it an attractive process for urban waste management (Pohland, 1995). Leachate recycling is a key step in successful bioreactor landfill (Mehta et al., 2002; Reinhart et al., 2002). However, independent tests revealed that bioreactor landfill could fail when MSW of different composition from that by Reinhart et al. (2002) was treated (Bae et al., 1998). Restated, direct recycling leachate

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through the landfill inhibits metabolism of organic matter degradation when the volatile acid levels in leachate is high (Veeken and Sergey, 2000; Zhang et al., 2004). Leachate treated using ex situ processes resolves the inhibition problem carried by the hydrolyzed volatile acids (Kim, 2001).

The characteristics of leachate after ex situ treatment are very different from the original leachate, and the characteristics of the changed leachate play an essential role in refuse stabilization. Moreover, the leachate production rate in practice is typically unstable, particularly during the initial stages of stabilization. Other feed solutions, including leachate from other refuse units could be utilized as feed solutions (Barlaz and Reinhart, 2003). However, the understanding of feed solution characteristics on landfill refuse stabilization is lacking. The leachate flows layer by layer in landfill. Therefore, the characteristics of leachate flowing out from an upper layer would affect the stabilization of its lower layers. The spatial evolution of leachate collected in landfill is not fully understood.

The objective of this research is to explore the effects of different feed solutions, including tap water, refuse leachate pre-treated using an aerobic sequential batch reactor (SBR), and refuse leachate pre-treated using an anaerobic upflow filtration bioreactor (UFB), on the stabilization rates of typical refuse collected in Shanghai, China. This refuse contains a high proportion of easily degradable food waste (He, 2002). The goal of this study is also to demonstrate the spatial interconnection of different refuse layers in landfill site, by utilizing three simulated landfill columns linked in a series that allow the leachate to flow from the upper to the lower columns.

2. Experimental

2.1. Fresh refuse

The refuse tested in this study was prepared based on Shanghai's data for MSW (He, 2002). Its moisture content was 74.5%, mainly due to food waste: its total composition (w/w) was: food waste (85%), plastic (5%), paper (7%), glass (1.6%), textiles (1%), and metals (0.4%). The relatively high proportion of food waste is a characteristic of MSW in China.

2.2. Feed solution

The three feed solutions to be prepared for this study were tap water dechlorinated with aeration, aerobically pre-treated leachate with a SBR, and anaerobically pre-treated leachate with an UFB. The leachate was collected from a real refuse landfill. The characteristics of the feed solutions are shown in Table 1. To compensate for the lack of inorganic nutrients, the following chemicals were added to the feed solutions: $CaCl_2$ 27.5 mg l^{-1}, $FeCl_3 \cdot 6H_2O$ 0.25 mg l^{-1}, $MgSO_4 \cdot 7H_2O$ 22.5 mg l^{-1}, KH_2PO_4 8.5 mg l^{-1}, K_2HPO_4 21.75 mg l^{-1}, $Na_2HPO_4 \cdot 7H_2O$ 33.4 mg l^{-1}, and NH_4Cl 1.7 mg l^{-1}.

As show in Table 1, the main differences between the three feed solutions were their alkalinity, DO level, and the levels of methanogenic bacteria. Tap water tests provided a basis for further comparison with low alkalinity and high DO. The alkalinity of effluent from SBR was close to 3000 mg l^{-1} (mg l^{-1} as $CaCO_3$). The effluent from UFB had a high alkalinity and contained methanogenic bacteria.

2.3. Experiment procedure

The simulated landfill columns were made of PVC plastic with an inner diameter of 9 cm and height of 30 cm. A 2-cm height leachate drainage layer made of crockery balls of diameter 5–6 mm was placed at the bottom of each column. Above this layer there was a fresh 15-cm-high MSW layer that was packed at a density of 800 kg m^{-3}. Above this refuse layer was a layer of textiles that evenly distributed the feed solution. The textile was covered with a 1.5-cm gravel layer to prevent the refuse from floating.

The three identical columns were connected in a series. The feed solution was fed into the top of Column 1. All leachate collected at the column's bottom was fed to the top of Column 2. Then the leachate collected at the bottom of Column 2 was fed into the top of Column 3. Gas meters to measure landfill gas and collecting ports to collect leachate were installed on each column. The leachate collected from the top of each of the three columns was named Q1 to Q3 for tap water, H1 to H3 for pre-treated effluent from SBR, and Y1 to Y3 for pre-treated effluent from UFB, respectively.

All tests were conducted at 35 °C. The leachate was fed into Column 1 at the same time once a day, at a surface loading of 23.6 mm d^{-1}. The surface loading was reduced to 11.8 mm d^{-1} on day 80 for the UFB solution, on day 90 for the SBR solution, and day 100 for tap water. The permeability of leachate flowing through the refuse was estimated at 1.2×10^{-3} cm s^{-1}, leading to a retention time of leachate in each column of 3–4 h.

A leachate sample was collected from the bottom of each column twice a week. The pH of leachate was measured using a PHS-25 pH meter. The leachate's total carbon (TC), total organic carbon (TOC) and inorganic carbon (IC) were measured with an Analytik jena Multi N/C 3000 meter (Germany). Acetic acid, propionic acid, and butyric acid in leachate were measured with a gas chromatograph (GC122M) equipped with a flame ionization detector (FID) and a 30-m Dikma Capillary Column. The temperatures for injection port, the oven, and the FID were at 120 °C, 200 °C, and 200 °C, respectively. Total amount of acetic acid, propionic acid, and butyric

Table 1
Characteristics of the feed solutions

Index	TOC (mgCl^{-1})	TN (mgl^{-1})	pH	TP (mgl^{-1})	BOD$_5$/COD	Alkalinity (mgl^{-1} as CaCO$_3$)	DO (mgl^{-1})	MB[a]
Tap water	0	0	6.6	0	NA	Low	≈5.0	No
Effluent from SBR	50–70	≈0	8.5–8.7	≈0	0.2	2900–3050	0.1–0.2	No
Effluent from UFB	170–190	240–280	8.6–8.8	140–170	0.3	6300–6800	≈0	Yes

[a] Methanogenic bacteria.

acid were calculated and summed as the VFAs. The values of VFAs were expressed as carbon. Alkalinity was analyzed according to Standard Methods (State Environmental Protection Administration of PR China, 1989). To further illustrate the possible role of leachate alkalinity, the so-called ''methane alkalinity'', determined by titrating the leachate at pH 6 since methanogenesis is generally inhibited at a pH level below 6 (Lay et al., 1997), was measured.

The contents of methane and of carbon dioxide in biogas samples were analyzed using a gas chromatograph (GC102M) equipped with a thermal conductivity detector and a 2-m stainless column packed with activated carbon.

3. Results and discussion

3.1. Tap water tests: Q1–Q3

The time evolutions of TOC in the three leachate streams Q1 to Q3 are similar. The TOC concentrations in leachate first dropped from a high concentration, and then followed an increasing-decreasing curve. TOC concentrations peaked following the sequence of Q3, Q2 and Q1, with their peak TOC values being 6800 mgCl^{-1}, 4600 mgCl^{-1} and 2800 mgCl^{-1}, respectively (Fig. 1a). The TOC peak for Q1, Q2 and Q3 occurred on days 38, 42, and 48, respectively. Since the retention time of the fed leachate was about 4h for each column, this time lag between peak times was not attributable to hydrodynamic dispersion. On the contrary, the hydrolysis of refuse was affected by the leachate TOC levels, with a lower hydrolysis rate corresponding to a higher TOC level. If the hydrolyzed TOC could not be effectively degraded in the refuse layer, the leachate TOC could reach a very high level after passing through several refuse columns. This observation interprets the rather strong leachate noted in practical landfill site.

Before day 30, the VFA levels in Q1–Q3 were close and all were lower than 1750 mgCl^{-1}. The VFAs in Q2 and Q3 then increased considerably over time, but those VFAs in Q1 remained unchanged (Fig. 2a). The VFA concentrations in Q2 and Q3 peaked on day 40,
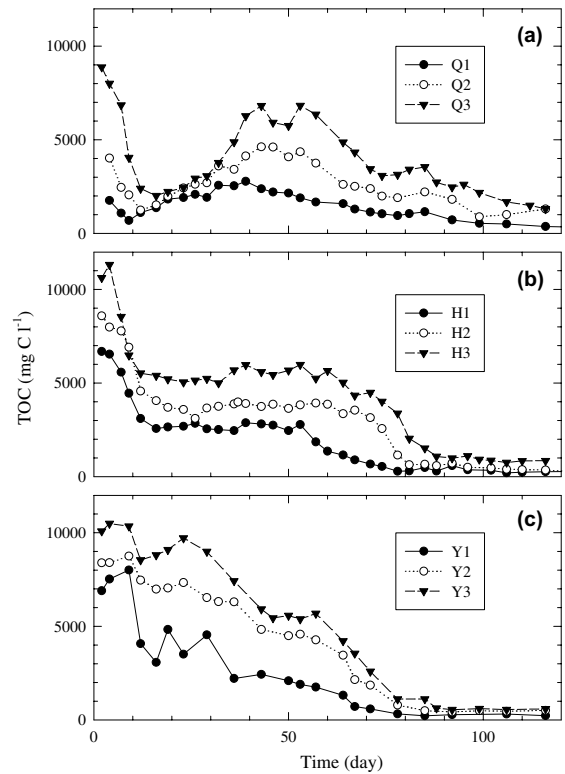

Fig. 1. Time evolutions of the testing leachate's TOC concentrations.

and then dropped gradually over time. A comparison of Figs. 1a and 2a showed that the level of VFAs changed before TOC.

The pH value of Q1 ranged from 5.0 to 5.8, while that of Q2 and Q3 fluctuated between 4.7 and 5.0 (Fig. 3a). This occurrence is a natural result of hydrolysis and acidification of refuse (Kim, 2001). The early release of VFAs suggested that the pH value of leachate would be affected once the hydrolysis reaction occurred in the refuse layer. Since buffer capability of tap water is minimal, a small amount of hydrolyzed VFAs considerably reduced the pH of the leachate (Fig. 3a). The corresponding methane production rate was low (Fig. 4a).
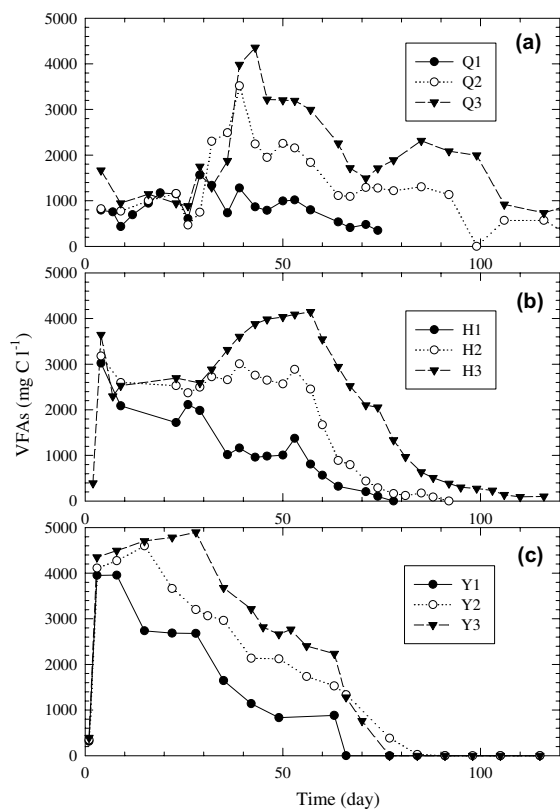
Fig. 2. Time evolutions of the testing leachate's VFA concentrations.

Fig. 3. Time evolutions of the testing leachate's pH concentrations.

3.2. SBR solution tests: H1–H3

Tests of effluent from SBR showed that TOC concentrations in H1–H3 first increased dramatically and then decreased. During day 16–54 of test, the TOC concentrations in the three columns increased sequentially and then remained at their respective plateau values (Fig. 1b). After day 60, the TOC concentrations for all the three columns gradually decreased.

The change in VFAs for H1–H3 approached a plateau or slightly increased over the period day 10–60. The VFA levels then dropped gradually over time. Most VFAs in H1, H2, and H3 were consumed after days 78, 92, and 105, respectively.

Prior to day 40, the pH value of H1 (5.1–5.8) was slightly higher than those of H2 and H3 (4.7–5.5) (Fig. 4b). Hence, the hydrolysis in columns H2 and H3 was also intensive and not inhibited by the leachate that flowed from H1. The increasing methane production rate showed that methanogenic reactions were established in the three columns (Fig. 4b).

From day 40 the pH value in all the three columns started to increase, and reached a value of 8 for H1,
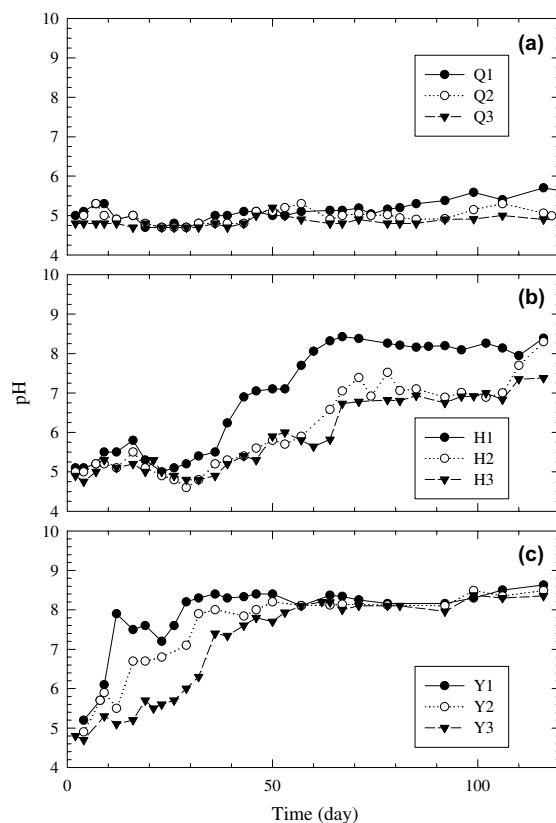
and 7 for H2 and H3 on day 65 and onward. The methane production rate ranged from 5 to $40 \, mgC d^{-1}$ (Fig. 4b). After day 80, all three columns had established their methanogenic phases.

3.3. UFB solution tests: Y1–Y3

Tests of effluent from UFB showed TOC concentrations in Y1–Y3 decreased over time, following Y3 > Y2 > Y1 (Fig. 1c). The changes in VFAs (Fig. 2c) closely followed that the changes in TOC. The VFAs concentrations of Y1, Y2 and Y3 peaked at 3960, 4600, and $4900 \, mgCl^{-1}$, respectively. However, the influence of the TOC levels on the hydrolysis of the refuse was not as clear as with Q1–Q3. This occurrence could be attributable to the high alkalinity and pH of the UFB feed solution.

The pH values for Y1, Y2 and Y3 increased over time, reaching a pH value of 8 on day 28, 32, and 57, respectively (Fig. 3c). The methane production reached a plateau regime with Y3 > Y2 > Y1 (Fig. 4c) over days 75–92, whose magnitudes were higher than H1–H3.
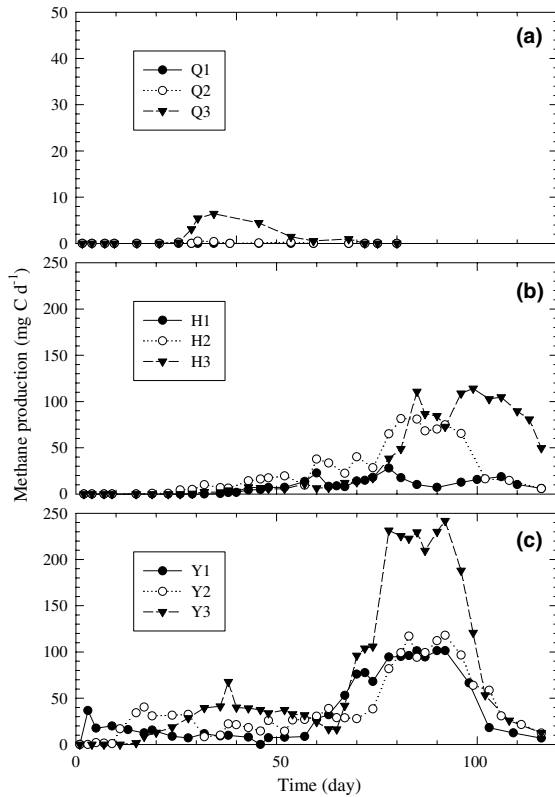
Fig. 4. Time evolutions of methane productions of different simulated landfills.

Fig. 5. Time evolutions of hydrolyzed TOC in different simulated landfills.

4. Discussion

When the feed solution seeped through the three columns in the series, the reactions that occurred in the first column affected reactions in the subsequent columns. This observation also revealed that the situation noted in the field: the leachate produced from a refuse layer or feed solution from the top layers influenced the dynamic behavior of its lower layers. Restated, the dynamics noted in the current three-column test can be regarded as a schematic version of Shanghai's real landfill site.

Over a refuse column balance of leachate TOC is stated as follows:

$$(\text{TOC out}) - (\text{TOC in}) + (\text{TOC consumed})$$
$$- (\text{hydrolyzed TOC}) = 0 \qquad (1)$$

where the term TOC consumed includes the organic carbon assimilated by microbes, transformed into gas phase (i.e., CH_4 and CO_2) and those lost in sampling. Hence, the amount of TOC hydrolysis was estimated as all the other terms in Eq. (1) were experimentally measured.

Columns fed tap water showed slow hydrolysis rates that peaked after 60 days of testing (arrow in Fig. 5a).
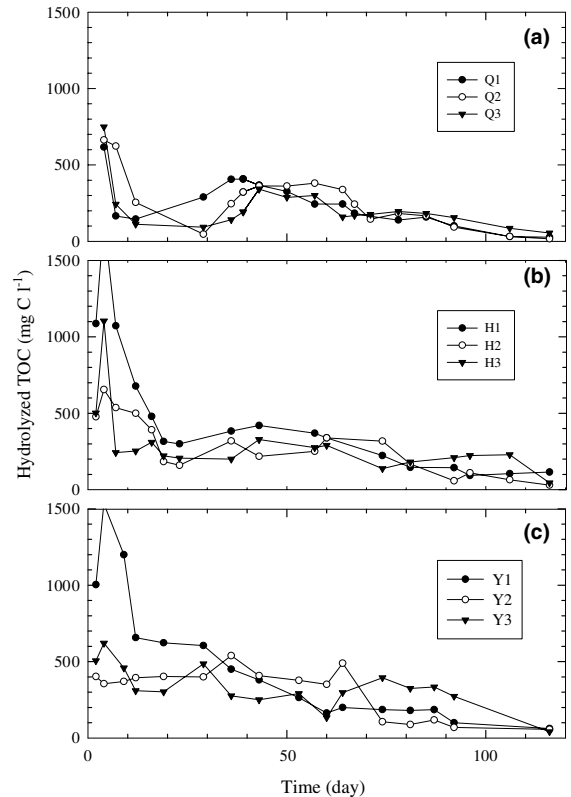
Moreover, during the initial stage, hydrolysis in column Q1 was much faster than that in columns Q2 and Q3. The acidic environment (pH 4.8–5.4 in Fig. 3a) therefore delayed refuse hydrolysis. The acidic environment inhibited methanogenesis in the refuse. The high DO concentration ($5 \, mg \, l^{-1}$) in the fed tap water also inhibited methane production. Both factors led to a low rate of methane production (Fig. 4a).

When the leachate pre-treated by SBR was fed into the columns, the TOC hydrolysis rates declined rapidly over the first ten days, then fluctuated in a range from 200 to $400 \, mg \, d^{-1}$ from day 10 to day 75 (Fig. 5b). The difference between hydrolysis rates in various columns was minor. With an intermediate alkalinity and low DO level, the fed SBR solution stimulated the methanogenesis in the refuse column. This occurrence was attributable to the buffer capability of feed solution to neutralize the H^+ produced during refuse hydrolysis. The H1 started to produce methane later than H2 and H3, and also at a lower rate, most likely because the DO level at 0.1–$0.2 \, mg \, l^{-1}$ was sufficient to depress activity of methanogenic bacteria.

After the leachate pre-treated by UFB was fed into the column, the hydrolysis rate for the column Y1

ranged from 250 to 600 mg d^{-1} up to day 60, was faster than the rates in Y2 and Y3. Although the VFAs were rapidly produced (Fig. 2c), the high alkalinity still effectively neutralized the pH of leachate (Fig. 3c). The high alkalinity and methanogenic bacteria in the feed solution also led to early methanogenesis in column Y1, and stimulated methane production in column Y2 and Y3, following the sequence Y1 to Y2 to Y3. The UFB effluent enhanced hydrolysis and methanogenesis in the refuse layer.

Table 2 lists the alkalinity data and the ''methane alkalinity'' of collected samples. The changes of both alkalinities corresponded to each other. The methane alkalinity of tap water was low, following Q3 > Q2 > Q1, and was gradually consumed during testing. With SBR effluent as the feed solution, the methane alkalinity for H1–H3 was 3 to 5 times higher than that for Q1–Q3 and remained unchanged over the duration of the test. The methane alkalinity of Y1–Y3 was 2 to 3 times higher than that of H1–H3, and dropped when the methanogenesis entering an intensive state (days 70–90 for Y1–Y3). The levels of both alkalinities were continuously recovered as a consequence of effective methanogenesis.

Fig. 6 shows the time evolution of the accumulation of hydrolyzed TOC in Q1, H1 and Y1. Up to day 116 the accumulation of the hydrolyzed TOC reached 24.6, 38.8, and 40.0 g in Q1, H1, and Y1, respectively. Differ-
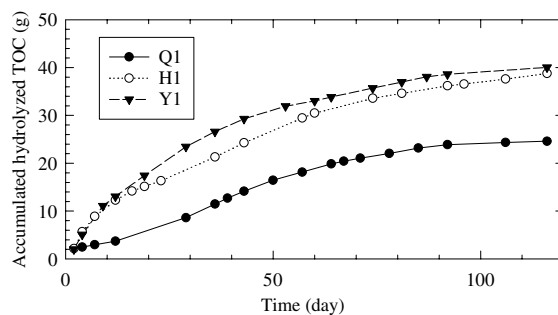

Fig. 6. Time evolutions of the accumulated hydrolyzed TOC.

ent feed solutions affected the hydrolysis rate of refuse (also in Fig. 5). Comparing with the tap-water fed column, the capability to neutralize pH using SBR solution with high alkalinity could increase the accumulated, hydrolyzed TOC by 14.2 g (from 24.6 g to 38.8 g). An even higher alkalinity in the UFB solution could only increase another 1.2 g in accumulated hydrolyzed TOC (from 38.8 g to 40 g).

Also noticeable in Table 3, and Figs. 1 and 2, the VFAs contributed 40% to 60% of the TOC in Q1–Q3, while the VFAs contributed 60–80% for H1–H3, and up to 70–90% for Y1–Y3. The VFA production rate followed both acetic acid > butyric acid > propionic acid, and Y1 > H1 > Q1. Therefore, different feed solutions

Table 2
Time evolution of alkalinity and methane alkalinity of the leachate[a]

Time (d)	Q1		Q2		Q3	
	Methane alkalinity	Alkalinity	Methane alkalinity	Alkalinity	Methane alkalinity	Alkalinity
12	210	930	430	2030	870	3650
29	110	900	400	1870	720	3360
56	170	970	570	2720	670	3760
78	260	750	730	1920	930	2560
102	140	410	370	1400	560	2080
120	220	450	400	1680	580	2030
	H1		H2		H3	
12	1870	3600	2680	4350	2860	4580
29	2430	3330	3070	4020	3200	5010
56	1700	3950	3020	4320	3900	4800
78	2230	3570	3560	3600	3750	5210
102	2420	3430	2365	4030	3660	4400
120	3010	3330	2650	4430	4000	4560
	Y1		Y2		Y3	
17	7370	18 520	8250	20 550	10 200	25 640
51	5600	7980	6890	9130	7210	11 020
72	6210	10 070	6850	12 200	7020	12 370
91	6990	7560	7210	8890	7230	9690
102	7360	8940	7560	9030	8020	10 002
120	10 200	12 250	11 270	12 690	10 230	11 690

[a] Unit: mg l^{-1} as CaCO$_3$.

Table 3
Maximum ratios of VFAs to TOC in Q1, H1 and Y1

	Q1	H1	Y1
VFAs ($mg\,Cl^{-1}$)	1170	1980	2740
TOC ($mg\,Cl^{-1}$)	1820	2550	3070
VFAs/TOC	0.64	0.78	0.89

altered the hydrolyzed products in the leachate (Fig. 7). The VFAs concentration for Q1 peaked on day 40, and acetic acid reached $800\,mg\,Cl^{-1}$, and the propionic acid reached less than $240\,mg\,Cl^{-1}$. The acetic acid in H1 peaked at $1200\,mg\,Cl^{-1}$ on day 24, and both propionic and butyric acids declined from $800\,mg\,Cl^{-1}$. The acetic acid production for UFB effluent could reach up to $1600\,mg\,Cl^{-1}$ on day 10, and butyric acid reached $1530\,mg\,Cl^{-1}$. Acetic acid had a higher concentration than propionic acid and butyric acid over the first 70 days in Q1, H1, and Y1, with the butyric acid having a slightly higher concentration than propionic acid. The acetic acid concentration dropped to zero faster that of butyric acid, and the concentration of butyric acid dropped faster than that of propionic acid. Hence, the feed solutions impacted the hydrolysis rate (Fig. 2) as well as the composition of the fermentative products. The high acetic acid concentration and a high pH buffer capacity produced a favorable environment for methanogenesis.

Carbon transfer ratio to gas phase was defined as follows:

gas transfer ratio

$$= \frac{\sum \text{carbon in gas}}{\sum \text{carbon in leachate} + \sum \text{carbon in gas}} \times 100$$

(2)

Up to day 116, gas transfer ratios for columns fed with SBR effluent and UFB effluent were 23% and 38%, respectively. While gas transfer ratio for the columns fed with tap water was only 4%. The strong leachate noted in typical landfill sites was produced by an intense hydrolysis reaction without sufficient methanogenesis in the refuse. The ratio of carbon transfer to gas phase was used as a preliminary quantitative index to assess the effects of feed solutions. The capability to neutralize pH with high alkalinity and a reduced oxygen level increased the conversion ratio by 19% (4–23% for SBR). The presence of methanogenic bacteria and the further depletion of oxygen in the feed solution increased the conversion ratio to 23–38% with UFB solution.

These results clearly revealed that the leachate characteristics in a bioreactor landfill should be modified to improve degradation of organic matter. The hydrolysis and methanogenesis in the top layer of refuse that receives the recycled or fed leachate would be directly influenced, as would the hydrolysis rates and the hydrolyzed products, with the environments of the lower layers subsequently affected. When the refuse contains a great proportion of easily hydrolyzed components, such as in the case of Shanghai's refuse, the pre-treatment of leachate becomes an indispensable step for successful
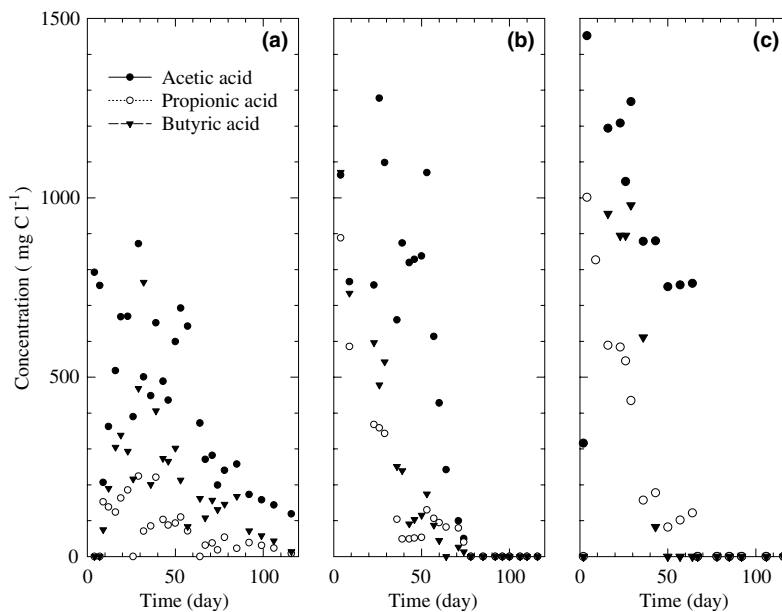

Fig. 7. Time evolutions of acetic acid, propionic acid, and butyric acid in (a) Q1, (b) H1, (c) Y1.

operation of a bioreactor landfill. In this study, anaerobic pre-treatment was the best option among those investigated since it produced high alkalinity for neutralizing the pH value of the leachate, and provided methanogenic bacteria for early methanogenesis in the top refuse layer.

5. Conclusions

This study examined the effects of feed solutions on the hydrolysis and methanogenesis in a refuse landfill. The alkalinity, dissolved oxygen level, and presence of methanogenic bacteria in a feed solution considerably affected the hydrolysis rate and establishment of methanogenesis in the top refuse layer. The leachate that flowed out of the first layer influenced the layers below. For the columns fed with dechlorinated tap water, the hydrolysis rate of refuse followed a bell curve and peaked following the sequence of Q3 (top column), Q2 and Q1 (bottom column); the peak TOC values for Q3, Q2 and Q1 were $6800 \, \text{mg} \, \text{C} \, \text{l}^{-1}$, $4600 \, \text{mg} \, \text{C} \, \text{l}^{-1}$ and $2800 \, \text{mg} \, \text{C} \, \text{l}^{-1}$, respectively. The pH value for Q1 ranged from 5.0 to 5.8, while the pH values for Q2 and Q3 fluctuated between 4.7 and 5.0. Hence, methanogenesis was inhibited and a strong leachate was discharge from the bottom layer. Direct adding tap water to the refuse layer did not assist in the refuse stabilization because of low buffer capability of leachate. The columns fed with aerobically pre-treated leachate (H1–H3) yielded a higher hydrolysis rate. After day 40 the pH value over the three columns started to increase, reaching 8 for H1, and 7 for H2 and H3 on day 65 and afterward. The methane content in biogas increased to 40–60%. The corresponding hydrolysis rates for anaerobically pre-treated leachate (Y1–Y3) were the highest among the three feed solutions. The strong buffer capacity of the UFB solution yielded a pH value of 8 for Y1, Y2, and Y3 on day 28, 32, and 57, respectively. The corresponding methane content increased to about 70% after day 80.

The chemical environment in the top refuse layer was dramatically affected by the characteristics of the feed solutions. The hydrolysis rates for the top layer were much faster than those for the lower layers. Without an appropriate buffer capacity, the methanogenic reaction in the lower columns was inhibited. Once the leachate's pH was neutralized with a high-alkalinity feed solution, early methanogenesis occurred and stimulated methanogenesis in the lower refuse layers. The VFAs accounted for 40–60% of the TOC in Q1–Q3; while those for H1–H3 for 60–80%; and Y1–Y3, up to 70–90% of TOC, respectively. The VFA production rate followed acetic acid > butyric acid > propionic acid, and Y1 > H1 > Q1. Therefore, different feed solutions also changed the hydrolyzed products in the leachate. After 120 days of testing with these three feed solutions, the carbon in refuse was converted to gas phase by 4%, 23%, and 38%, respectively. The success of bioreactor landfill depends on whether the recycled leachate could produce a favorable methanogenic environment at the top refuse layer or whether an appropriate pre-treatment has been adopted to modify the leachate's characteristics.

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