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Conversion of organic carbon in the decomposable organic wastes in anaerobic lysimeters under different temperatures

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Abstract: The quantitative fractions of conversion of organic carbon in the decomposable organic wastes with initial moisture of 70% sorted from municipal solid wastes (MSW) in lysimeters into biogas, leachate and solid residue were characterized, under temperatures of 25, 30 and 41℃, respectively, and circulation of leachate generated within the lysimeters. It is found that 27% of organic carbon in the wastes are conversed into gases, 0.8% into leachate, and the other 72% remained in the decomposable solid residues, after 180 days' degradation at 41℃. Higher temperature will lead to more rapid degradation and result to higher conversion of the organic carbon to biogas and lower to both solid residues and leachate, while the pollutant concentrations in leachate will be lower at a higher temperature and the values of COD are quite consistent with TOC.

Keywords: refuse; organic wastes; lysimeters; organic carbon; conversion

Introduction

The proportion of decomposable organic wastes such as food origin wastes and backyard wastes, in municipal solid wastes in Shanghai, China, has been increasing rapidly, as the living standard of inhabitants in the city are remarkably improving in recent years. For example, around 1300 tons of food origin organic wastes with moistures of 70%—90% are being generated daily in Shanghai alone from over 200 large scale hotels and restaurants and other relatively smallerbars, etc., which are currently mixed with the municipal solid wastes (MSW) and placed in landfills. Traditionally, Chinese food is always over-served at hotels and restaurants, i.e., food will be ordered and bought more than needed by the guests. As a result, at least 10%—20% of food will become waste after the guests leave. Before 1999, these organic wastes were collected and transported to farms to use as feed for pigs and other livestock. However, it seems that such wastes are no longer healthy for livestock breeding as more and more infected viruses are found in the wastes. Hence, many cities in China prohibited the use of organic wastes as animal feed since 2000. The wastes generated should be dewatered as thoroughly as possible and then sent to landfills with charge. Nevertheless, it is soon found that such wastes do not meet the operational requirements in landfill as the moistures are still too high to compress by compressor (Zhao, 1999; 2000). Moreover, the pollutant concentrations in leachate seem to increase remarkably since the landfill receives the organic wastes.

Another issue facing landfill operators is the rapid increase of backyard wastes generated from the increasing quantity of private houses with a garden and several newly constructed large scale parks located around the city. It is evaluated that a net increase of around 300—400 t/d of backyard organic wastes consisting of tree leaves, shrubs, bamboo, wood, etc., may be presented in the placed MSW (Zhao, 2001). The introduction of these newly appeared organic wastes has apparently changed the stabilization process of the landfills (Zhao, 2000b; 2001). Consequently, the quantities of landfill gas, pollutants in and quantities of leachate, etc., all increase. Meanwhile, the chemical compositions in the remaining residues also change, which results to different settlement mode in the surface of landfill cover (soil). Hence, the increase of decomposable organic wastes in the MSW placed in landfill has unexpectedly changed the original physical, chemical and biological processes of landfill stabilization.

One of solutions to the increase of these decomposable organic wastes is to treat the wastes separately. However, it seems that landfilling is still the most practical treatment measure to the wastes, as the other measures such as composting and incineration are just at the stage of planning or under construction in most China cities. In order to understand the biologically

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decomposition mechanism so that the landfill can be better managed, the biological degradation of the organic wastes in landfill should be studied so that the fundamental knowledge for the improvement of landfill management can be provided (Zhao, 2000; 2001b).

Many researches have been done on the biochemical processes of refuse in landfill (Balarz, 1989; Belevi, 1989; El-Fadel, 1996). However, what they used were the mixed wastes, i.e., the mixtures of decomposable and non-decomposable organic wastes. The data thus obtained represent the concomitant effects of the mixed wastes, though such conduction may be more similar to the real landfill. It is reasonable to propose that the biodegradation of the decomposable organic wastes in a landfill should receive more attention, from which both biogas and leachate are largely generated (Leckie, 1979; Miller, 1994; Nancy, 1995; Zhao, 2000; 2001b).

Refuse degradation is a slow and long-term process (Belevi, 1989; Dean 1995; Pohland, 1980; Wang, 1999; Zhao, 2000; 2001b). It is quite difficult to study the real landfill for a long period (Ham, 1993). In our previous work, the similarity of biological degradation processes in different scale laboratory lysimters and real landfill was compared (Zhao, 2001c). It was found that the absolute values for all parameters such as pollutant concentrations in leachate, chemical compositions of solid residues, surface settlement, etc., were varied with the scale of lysimeters used, but the change patterns of relationships among these parameters and time (age of wastes in the lysimeters) were quite similar. Hence, laboratory lysimeters are useful and powerful tool to simulate the real landfill (Attal, 1995; Pohland, 1980; Somjai, 1995). In order to accelerate degradation, the recycling of leachate within the landfill simulators is often practiced (Pohland, 1980; Townsend, 1996).

Production of methane, a main component in the biogas, from refuse landfill, has drawn more and more attention worldwide (Balaz, 1989; Young, 1989; 1984). The yield and generation rate from a landfill may be theoretically evaluated, when the basic composition of the refuse is available. Many empirical formulae for methane production rate and quantity are also developed (Balaz, 1989; Wang, 1999). However, a big gap may be presented when compared the evaluated value with the real generation yield available *in situ*. Though the biological degradation of organic matters and dissolution of inorganic substance in refuse has been studied intensively (Young, 1989), the accurate conversion of carbon in refuse into biogas is unknown. Moreover, kinetics of refuse degradation process and substance partition in the solid residue, gases and leachate would be helpful for the prediction of these parameters, especially in the first phase of biodegradation (Zhao, 2000; 2001b; 2001c). In this work, the conversion of organic carbon in the decomposable organic wastes to and generation of leachate, landfill gas and solid residue were studied in the laboratory scale lysimeters in a period of 6 months.

1 Experimental

1.1 Composition of refuse used

The refuse used in this work was directly taken from Shanghai Refuse Landfill, 65 km away from city downtown in Shanghai. Refuse generated in households, commercial centers, business centers, etc., is sent to collection points located in every corner in Shanghai, and then transported to transition stations by trucks. The refuse may stay in these stations for one to three days, depending on the quantity stored. There are over 1000 stations in the downtown. The refuse was then transferred to boats and ships from the stations and transported to the landfill, which may take two to three days. The refuse in the boats and ships will be unloaded and placed in the landfill immediately as soon as they arrive. Hence, around a week may take for the refuse to reach the landfill since it is generated.

Table 1 Refuse composition used in this work in dry base

Composition	Weight percentages, %
Cooking wastes	60.7
Paper	3.3
Woods	2.23
Cloth	0.64
Fruits and backyard wastes	31.2
Metals	0.23
Class	0.9
Other	0.8
Total	100
Water (moisture)	70

The aim of this work is to explore the conversion of organic carbon into biogas, leachate and solid residues. Hence, all the large articles and non-degradable matters such as large cloths, shoes, glass bottles, stone, rubber, sand, building wastes, etc., were sorted out from the refuse by hand. What put in the lysimeter were the decomposable organic wastes such as fruit, cooking wastes, vegetables, tree leaves, schrubs, and small pieces of woods. As a result, the composition of refuse used in the lysimeters was shown in Table 1, which was different to the original. All the experiments were carried out at the same time starting from 19 April 1998, and terminated after 180 days. All the refuse weight concerned in this paper was calculated in dry basis unless indicated.

1.2 Lysimeter

It is essential that the quantity and composition of gas, solid residue and leachate generated in the lysimeters must be determined accurately. Hence, totally, 18 lysimetrs were used, with a decomposable refuse weight (in dry basis) of 14 kg in each lysimeters. The moisture of decomposable refuse was around 70%, as the cooking wastes and vegetable and fruits occupied the main components. Hence, the weight of wastes in every lysimeter was actually 46.0 kg. They were organized into three parallel groups (named as Groups Ⅰ, Ⅱ and Ⅲ), 6 lysimeters a group, as shown in Table 2, and put in the thermostatic room with 25, 30 and 41℃, respectively for a duration from 1 to 6 months. The height and diameter of lysimeter were 70 cm and 25 cm. N_2 gas was passed through all the lysimeters to escape the entrapped air as soon as the wastes were installed in the lysimeters and sealed. The biogas were collected from the upper and their volume was monitored by flowmeters and analysed by gas chromatograph. Leachate was circulated within each lysimeters once a day by peristaltic pump until it was stopped as scheduled (Table 2). The first lysimeters in every group were terminated for analysis of leachate, biogas and solid residues in the 30th day; the second lysimeters were terminated in the 60th days and so on. Around 25 L leachate, 55% of the initial weight of wastes, was generated in the last lysimeters terminated on the 180th day.

1.3 Analysis of samples

COD, BOD and NH_3-N in leachate were determined by conventional chemical methods. Gas chromatograph, TOC analyzer and elemental analyzer were respectively used for determinations of composition of biogas, TOC in leachate, and organic carbon in the initial refuse and solid residues. The data were all calculated on the dry basis.

2 Results and discussions

2.1 Temperature effect on the quantitative transformation of refuse

Table 3 gives the quantity of leachate, biogas, and solid residues, which vary with temperature and time. Quantity of leachate increases gradually at room temperature, while increases at the first 3 months and then decreases at 41℃, but always decreases at 30℃. In contrast, the accumulated volume of biogas always increase, and the weight of solid residues decrease, over the three temperatures.

Table 2 Layout of lysimeters (weight of organic wastes 46 kg, with 70% moisture)

			Group Ⅰ, 25 ℃			
No. of lysimeters	1	2	3	4	5	6
Time for stop of the lysimeter, d	30th	60th	90th	120th	150th	180th
			Group Ⅱ, 30 ℃			
No. of lysimeters	7	8	9	10	11	12
Time for stop of the lysimeter, d	30th	60th	90th	120th	150th	180th
			Group Ⅲ, 41℃			
No. of lysimeters	13	14	15	16	17	18
Time for stop of the lysimeter, d	30th	60th	90th	120th	150th	180th

The quantities of leachate and solid residues were weighed and those of gases theoretically evaluated for each lysimeter in the period of tests. The sums of these forms of matters are divided by the original weight of wastes, and thus the recovered rates are obtained and given in Table 4. It varies from 96% to 105%. Actually, the weights of both leachate and solid residues could be determined exactly, nevertheless, those of gases could only be evaluated using the standard state of gases and the methane contents. It was found that the water content in the biogas was nearly saturated. From Table 3 it can be seen that around 20% of the organic carbon in decomposable organic wastes may be conversed into gases or vapor over a period of 180 days' degradation in the lysimeters.

The composition of biogas was nearly identical with those reported in the other literature(Balaz, 1989). Methane content in the gases increased over time and then reached the maximum value of around 60% after 2—3 months.

Table 3 Refuse degradation of decomposable organic wastes

No. of lysimeters	1—18						Fresh wastes
Wet wastes, kg	46	46	46	46	46	46	
Moisture, %	70	70	70	70	70	70	70
Dry wastes, kg	13.8	13.8	13.8	13.8	13.8	13.8	
Content of organic carbon, dry basis, wt.%	20.21	20.21	20.21	20.21	20.21	20.21	20.21
Monitoring time, d	30	60	90	120	150	180	0
	25℃ (Group Ⅰ)						
No. of lysimeters	1	2	3	4	5	6	
Volume of leachate, L	21.3	23.2	24.1	24.8	25.3	25.4	
Weight of leachate, kg	22.3	24.4	25.2	25.8	26.0	25.9	
Volume of gases, L	6.4	63.6	143.7	197.9	466.7	624.7	
Solid residue, dry basis, kg	13.4	13.3	13.2	13.0	12.8	12.5	
Organic carbon, dry basis, wt.%	18.0	18.4	19.8	21.2	21.9	22.4	
	30℃ (Group Ⅱ)						
No. of lysimeters	7	8	9	10	11	12	
Volume of leachate, L	28.2	26.4	22.1	21.8	21.6	21.1	
Weight of leachate, kg	29.3	27.4	22.9	22.2	22.0	21.9	
Volume of gases, L	41.1	77.6	202.1	346.3	682.9	1051.4	
Solid residue, dry basis, kg	13.5	13.5	13.6	13.5	13.2	12.8	
Organic carbon, dry basis, wt.%	19.06	19.10	20.61	17.93	17.55	19.04	
	41℃ (Group Ⅲ)						
No. of lysimeters	13	14	15	16	17	18	
Volume of leachate, L	27.2	30.0	34.7	30.3	29.9	28.4	
Weight of leachate, kg	28.9	31.9	35.8	32.0	31.0	29.8	
Volume of gases, L	122.4	187.4	351.9	493.1	1144.1	1149.9	
Solid residue, dry basis, kg	13.2	13.0	13.1	13.0	13.2	12.9	
Organic carbon, dry basis, wt.%	18.1	19.25	18.05	17.58	15.67	14.48	

2.2 Leachate quality

TOC in leachate dependence on temperature is given in Fig. 1. Remarkable differences can be seen at the initial 4 months of degradation. However, such a difference disappears at 180th days, while higher TOC in leachate appears at 41℃, indicating the rapider degradation of wastes at higher temperature. Similar phenomena can be found for the cases of COD and BOD concentrations in leachate as shown in Figs. 2 and 3. Compared Fig. 2 with Fig. 1, it can be found that TOC and COD nearly overlaps, especially after 150th days. Hence, TOC can be used as a good indication of COD in leachate. Both COD (Fig. 2) and TOC in leachate decreases to around 600 mg/L after 150 days, with slightly lower for BOD (Fig. 3).

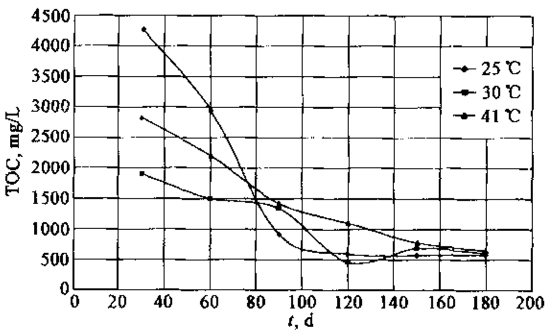

Fig. 1 Variation of TOC concentrations in leachate with time

2.3 Conversion of organic carbon

It is known that organic matters in refuse will be conversed into biogas, leachate and solid residues. The contents of TOC in biogas, leachate and residues were determined, and their percent fractions can be thus calculated respectively. Results are shown in Figs. 4—6. It can be seen that the percentages of organic carbon in biogas (Fig. 4) increase, while those in leachate(Fig. 5) and solid residues(Fig. 6), decrease with time. Moreover, the fractions of organic carbon in leachate only occupy a relatively low ratios, with lower than 4% at the starting stage, and 1% after 180 days, while those in gases may be as high as 27% at 41℃ after 180 days. That is to say, after 180 days' degradation at 41℃, around 27% of organic carbon in the wastes are conversed into gases, 0.8% into leachate and the other 72% remain in the residues.

Table 4 Mass balances of decomposable organic wastes

Time, d	Total refuse recovered at 25℃, %	Total refuse recovered at 30℃, %	Total refuse recovered at 41℃, %
30th	104.41	103.47	101.38
60th	103.64	101.63	96.75
90th	104.45	105.23	96.27
120th	99.86	103.84	99.08
150th	104.65	101.27	100.14
180th	96.74	105.61	106.28

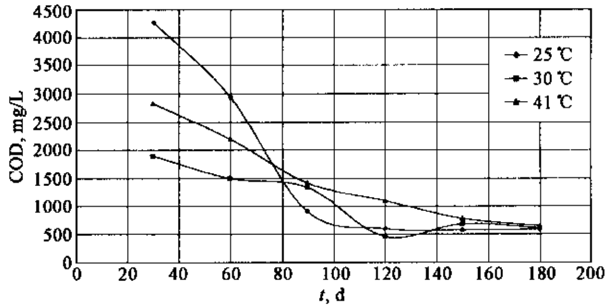

Fig.2 Variation of COD in leachate with time

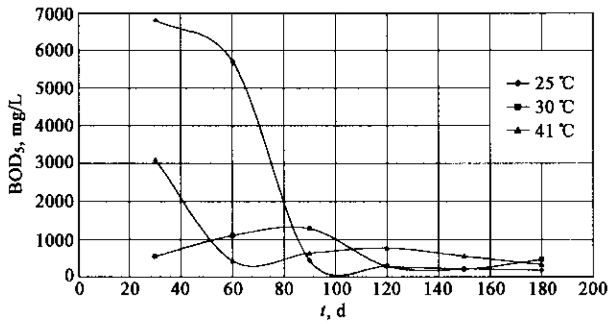

Fig.3 Variation of COD in leachate wih time

The recovered rates for organic carbon in the three temperatures are given in Table 5. It varies remarkably from 92% to 107%, which may be mainly contributed to the errors caused from the estimation of carbon contents in gases.

2.4 Kinetics of refuse degradation

It is generally considered that refuse degradation in landfill follows the first rate reaction, i.e., $\ln(C_0/C_t) = kt$, where C_0 is the initial organic carbon content in refuse (20.1%), and C_t is the organic carbon of solid residues at placement time of t(d). Using the data given in Table 2, it is easy to calculate the values of $\ln(C_0/C_t)$ and their linear relationship with time (t). The results are shown in Table 6. Values of k are calculated to be 0.0006, 0.0009 and 0.0016 at 25, 30 and 41℃ respectively.

According to Arrhenius equation: $k = A\exp(-E_0/RT)$, or $\ln k = -E_0/RT + \ln A$, using the values of k and T in Table 5, it is easy to obtain the following equation: $\ln k = -4677.9/T + 8.4337$, $r = 0.9998$ and values of E_0: $E_0 = 38.89$ (kJ/mol ℃).

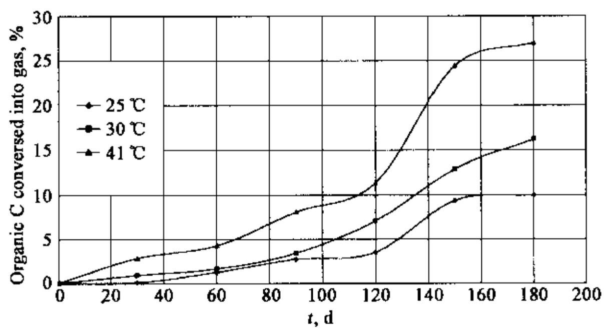

Fig.4 Fraction of organic carbon conversed into landfill gas

It can be seen that higher temperature will lead to larger k values, where the refuse degrades faster. In addition, the activation energy obtained in this work (38.89 kJ/(mol·℃)) seems quite high, compared with the value of 12.98 kJ/(mol·℃) reported in literature (El-Fadel, 1996). This difference may be contributed to the composition and water content of refuse used.

Table 5 Mass balances of organic carbon in the decomposable organic wastes

No.	Time, d	Total organic C recovered at 25 ℃, %	Total organic C recovered at 30℃, %	Total organic C recovered at 41℃, %
1	30	92.53	96.92	90.28
2	60	96.75	94.17	93.06
3	90	102.57	107.91	89.07
4	120	106.11	95.38	91.08
5	150	103.18	103.85	100.47
6	180	101.64	107.40	91.99

3 Discussions

It is surprised to observe that 10% (25℃) to 27% (41℃) of organic carbon in the fresh refuse are conversed into landfill gas, while less than 5% into leachate, regardless of temperature, over the monitoring

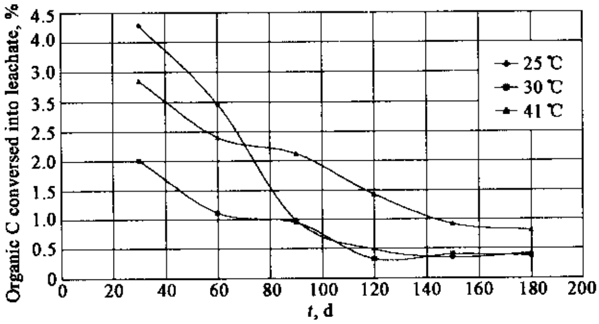

Fig.5 Fractions of organic carbon conversed into leachate

period of 180 days (the first phase of degradation). As refuse degradation proceeds, conversion ratios of organic carbon for gas increase and for leachate decrease.

Quantity of landfill gas generated in a landfill may be estimated by its contents of organic carbon in fresh refuse. Nevertheless, organic carbon conversion is dependent on temperature, re-circulation of leachate and humidity in refuse, according to the results obtained in this work. Perhaps the maximum possible accumulated quantity of biogas may be predicted by supposing that 40% of organic carbon in fresh refuse may be conversed into gases when the refuse stabilized after decades of placement. In this work, there is 200 g of organic carbon in 1 kg of refuse (in dry base), then ultimately, 80 g of organic carbon (ca. 7 mol C) would be changed into biogas. In general, around half of decomposable organic carbon would be conversed into methane, and another half into carbon dioxide. Hence, 1 kg of dry refuse used in this work would generate 3.5 mol methane, i.e., 78 L methane in standard conditions. In areas with high humidity and heavy rain, biogas are actually generated in 7—10 years. Hence, in average, 7—8 L of methane would be generated per year per kg of dry refuse. Adding to carbon dioxide, the total generation rate of gases may be 14—16 L per year per kg of dry refuse. But the biogas generation rate is heavily dependent on refuse composition, moisture, temperature, cover, and height of refuse in landfill. The values reported in literature ranged from 1—14 L per year per kg of dry refuse in actual landfill and 13—37.5 L per year per kg of dry refuse in the most lysimeters (Zhao, 1999). Suppose that refuse contains 50% moisture, then for a landfill with a disposal capacity of 1000 t/d (like in middle scale city in China), 15 × 500 × 1000 = 7500000 L (7500 m^3) landfill gas would be generated daily.

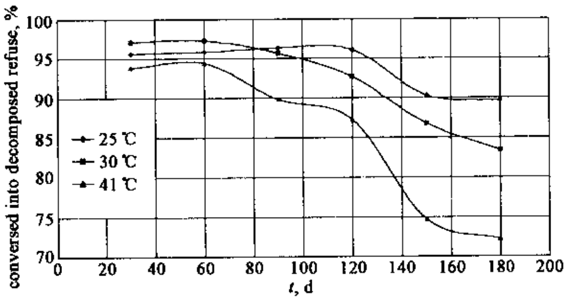

Fig.6 Fractions of organic carbon remained in the solid residues

Table 6 Kinetic parameters for refuse degradation in lysimeters

25℃	30℃	41℃
$\ln(C_S^0/C_S^t) = 0.0006t$, $r = 0.8745$	$\ln(C_S^0/C_S^t) = 0.0009t$, $r = 0.9374$	$\ln(C_S^0/C_S^t) = 0.0016t$, $r = 0.9430$

In practical situation, leachate quantity in a landfill located in drought area is much lower than that in a humid and rainy area. In reverse, concentrations in leachate in a landfill with poor cover may be usually higher than those with excellent cover, as in situ justified by many researchers (Balaz, 1989; Ham 1989; Young, 1989; Zhao, 1999) and the observations in Shanghai Refuse Landfill and Shenzhen Refuse Landfill. The former landfill is not covered daily and leachate is not drained well, and the latter is much better. Consequently, the average COD and BOD values in leachate generated in the operating landfill units (under the first phase degradation) in Shanghai Refuse Landfill are around 10000 and 2000 mg/L respectively, while those in Shenzhen Refuse Landfill are around 30000 and 5000 mg/L respectively. Hence, one of the most important measures to reduce leachate quantity is to cover the refuse surface well when closed, even the daily cover may not be practiced in landfill with limited soil sources. The price for good cover is an increase of concentrations in leachate. Obviously, leachate treatment technology selection, leachate quantity treated, concentrations in leachate, are closely related to the percolation capability of soil cover.

In most cases, the leachate quantity is estimated according to the maximum precipitation over a period of 20 years. From this work it can be seen that around 60% of moisture in refuse would be conversed into leachate over 6 months. However, around 90% leachate would be released within the first month. For a landfill with a capacity of 1000 t/d, 30000 t/m, with moisture of 50%, some 13500 tons (450 t/d in average) of leachate would be generated due to refuse degradation. Though this amount may be much lower in a real landfill, both due to the evaporation and slower release of water from the refuse, such proportion of leachate originated from the refuse could not be neglected when estimates the leachate quantity to be treated in design. Hence, the traditional estimation based on the maximum precipitation is not very reliable, which has been confirmed from several landfills investigated in China.

In our previous work, an experimental sanitary landfill was constructed in situ in Shanghai Refuse Landfill, and found that COD, BOD, NH_3-N, SS, Cl^- concentrations in leachate fluctuated and kept at high values before 4—6 months and then decreased fast to a much low values. Finally, concentrations in leachate decrease gradually and slowly in the long term process of stabilization (Wang, 1999; Zhao, 2000; 2001b; 2001c). These results are comparable with those obtained in this work. The absolute value of concentrations in leachate may vary with refuse composition, percolation and climate in a landfill, and scale of lysimeter if studied in laboratory, but the trend of concentrations in leachate against placement time seems to be similar as reported in several publications (Zhao, 1999; 2000; 2001b). Hence, much attention must pay to leachate generated in intensive reaction phase when a landfill is designed and managed. Holding tanks for leachate are always needed to adjust the leachate quantity and quality. Meanwhile, it is also found that parameters such as concentrations in leachate and volatile solid content in the solid residues are closely related to scales of lysimeters in the initial 0—4 months and seem to be independent on scales after 4—6 months of placement (Zhao, 2001c).

4 Conclusions

The fractions of organic carbon in the fresh refuse conversed into gas, leachate and decomposed refuse were monitored. It was found that about two thirds of organic carbon was remained in the solid residues and one third into landfill gas, while the fraction of organic carbon conversed into leachate was quite low. Higher temperature is favorable to the conversion of the organic carbon into biogas and negative to leachate. The kinetics of biodegradation is studied and the activation energy is calculated. It is considered that the estimation of leachate based on the maximum precipitation is not very reliable and should be modified according to the moisture in refuse treated.

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