Available online at www.sciencedirect.com
ScienceDirect
Journal of Environmental Sciences 20(2008) 499–504
JOURNAL OF ENVIRONMENTAL SCIENCES
ISSN 1001-0742
CN 11-2629/X
www.jesc.ac.cn

Evolution on qualities of leachate and landfill gas in the semi-aerobic landfill

HUANG Qifei[1,*], YANG Yufei[1,2], PANG Xiangrui[1,3], WANG Qi[1]

1. Research Institute of Solid Waste Management, Chinese Research Academy of Environmental Sciences, Beijing 100012, China. E-mail: huangqf@vip.sina.com
2. Civil and Environment Engineering School, University of Science and Technology Beijing, Beijing 100083, China
3. School of Municipal and Environmental Engineering, Harbin Institute of Technology, Harbin 150001, China

Received 26 June 2007; revised 30 July 2007; accepted 23 August 2007

Abstract

To study the characteristics of stabilization in semi-aerobic landfill, large-scale simulated landfill was constructed based on the semi-aerobic landfill theory. Consequently, the concentrations of chemical oxygen demand (COD), ammonia nitrogen, and nitrite nitrogen, and the pH value in leachate, as well as the component contents of landfill gas composition (methane, carbon dioxide, and oxygen) in landfill were regularly monitored for 52 weeks. The results showed that COD and ammonia concentrations declined rapidly and did not show the accumulating rule like anaerobic landfill, and remained at about 300 and 100 mg/L, respectively, after 48 weeks. Meanwhile, the descending rate reached 98.9% and 96.9%, respectively. Nitrate concentration increased rapidly after 24 weeks and fluctuated between 220–280 mg/L after 43 weeks. The pH values were below 7 during the first 8 weeks and after that leachates appeared to be alkaline. Carbon dioxide was the main composition in landfill gas and its concentration remained at a high level through the whole stabilization process. The average contents of carbon dioxide, oxygen, and methane varied between 19 vol.%–28 vol.%, 2 vol.%–8 vol.%, and 5 vol.%–13 vol.%, respectively. A relative equilibrium was reached after 48 weeks. The highest temperature in the landfill chamber could amount to 75.8 degrees centigrade.

Key words: semi-aerobic landfill; stabilization; leachate; landfill gas

Introduction

Sanitary landfilling is still one of the principal methods of municipal solid waste disposal at present. However, current landfill disposal techniques consider landfill site as a passive waste storage system, and waste in landfill is isolated from water and gas. The biodegradation of organic compounds is restricted for low moisture in landfill, which cannot provide microorganism with feasible conditions (Bookter and Ham, 1982). The dumped refuse will biodegrade slowly, which lengthens the process of landfill stabilization. Accompanied by different settlement, landfill gas and leachate were discharged, which can last for decades or even centuries and result in a significant negative impact on the environment. Therefore, several scholars started to seek new landfill techniques to accelerate the landfill stabilization process and to weaken leachate pollution.

In the 1970s, Pohland (1980) demonstrated that with leachate recirculation, landfill can be used as a relatively controlled anaerobic filter to treat leachate, provide accelerated waste stabilization, and reduce the volume of leachate by maximizing evaporative losses during recirculation. After that, the bioreactor landfill technique was studied and then applied by several countries in North America and Europe. Carson (1995) pointed out that the bioreactor landfilling technique has been widely used in the United States. Robison *et al*. (1983) reported that leachate recirculation can not only accelerate biodegradation of organic compounds, but also shorten the time required for stabilization from several decades to 2–3 years. In a leachate recirculation test conducted in America Sonoma County landfill site, the general range of decrease in the area recirculated by leachate can reach 20% of the landfill site depth, while in contrast just 8% (Leckie *et al*.,1979). The study of Yazdani *et al*. (1977) showed that the capability of accommodation and lifespan of bioreactor landfill were 12.6% higher than the conventional landfills. Raynal *et al*. (1998) reported that in two-phase bio-reactor, as the activity of producing methane was weakened, more fermented products that were deoxidized more easily were produced. Thereby, advantageous conditions to accelerate landfill stabilization were created. Chugh *et al*. (1999) reported that in the leach-bed process bioreactor landfill, the time taken for the leachate's pH value to reach 7 was shortened, and landfill stabilization was accelerated.

Bioreactor landfill technology has several advantages, such as: increased moisture content; enhanced activities of

* Corresponding author. E-mail: huangqf@vip.sina.com.

microorganism; accelerated stabilization process; reduced content of organic pollutants in landfill considerately. However, leachate recirculation can lead to the inhibition of methanogenesis as it may cause high concentrations of organic acids (low pH), which are toxic for the methanogens (Liliana *et al*., 1997). Ensiling problem may occur in a landfill cell, particularly at a high level of recirculation volumes resulting in an imbalance between acidogenesis and methanogenesis (Lay *et al*., 1997). The study performed by Pohland (1980) showed that recirculation at landfills is practiced to accelerate the decomposition of readily degradable organic compounds. However, Leachate ammonia nitrogen concentration may accumulate to higher level than during conventional single pass leaching.

In the late 1960s, Japanese scholar presented the concept of semi-aerobic landfilling (Masataka *et al*., 1986). In semi-aerobic landfill site, air entered the leachate collecting pipe, which was not full of leachate and air flow was strengthened for the temperature difference between the inner and outside of the landfilling pile; therefore, some aerobic zones in landfilling pile existed. After that, a series of researched results have shown that semi-aerobic landfilling can not only accelerate the landfill stabilization process and reduce organic substance concentration in leachate and the production of methane in semi-aerobic landfill, but also reduce ammonia concentration in leachate. Masataka reported that about 90% organic substance in waste were converted into gas phase (mainly as CO_2 and N_2) in semi-aerobic landfilling treatment, while 90% organic substance were converted into leachate phase in anaerobic landfill treatment after three years (Yasushi and Masataka, 1997). Compared with an anaerobic landfilling treatment, the concentrations of BOD and COD in leachate were slightly lower in the semi-aerobic landfilling treatment (Basri *et al*., 2000). Onay and Pohland (1998) simulated a series of landfill cells operated under methane producing, nitrifying, and denitrifying environments. Leachate concentrations of 920–1400 mg/L nitrite were generated in a refuse reactor that was supplied with air and the leachate was cycled to a denitrification reactor where ammonia removal ranged from 91% to 93%. They concluded that the use of leachate recirculation in simulated landfill bioreactors was feasible for the *in situ* removal of ammonia nitrogen. Wang Qi's lab research showed that adoption of import air into the landfill layers by engineering measures during the engineering design for the Sanitary landfill site can greatly shorten the time for stabilization of the landfill site, favorable for quick utilization of the landfill site (Dong *et al*., 2000).

At present, several researches have focused on the variations of leachate quality in semi-aerobic landfilling technology; however, the results on the characteristics of stabilization in semi-aerobic landfill have not been reported. Therefore, the purpose of this research was to study the characteristics of stabilization in semi-aerobic landfill systematically by means of constructing a large-scale simulated semi-aerobic landfill, and to study the characteristics of leachate quality, and variation of landfill gas component contents and temperature in landfill.

1 Materials and methods

1.1 *In situ* scale simulated semi-aerobic landfill construction

To study the characteristics of the semi-aerobic landfill stabilization process, *in situ* simulated semi-aerobic landfill was constructed based on the theory of semi-aerobic landfilling. The schematic configuration of simulated semi-aerobic landfill is presented in Fig.1. The dimensions of simulated landfill were 21 m × 3.8 m × 6 m. High density polyethylene (HDPE) membrane was laid on the landfill floor, and the leachate collecting pipes of 20 cm in diameter were installed on the HDPE membrane. Then pipes were covered with gravels. Three erect windpipes of 20 cm in diameter were fixed on the part of the leachate collecting pipe. The erect windpipes were perforated and protected by gravel cages (40 cm in diameter) to vail air flowing and penetrating into land layer. After that, municipal solid waste was filled in, and landfill gas collecting sets and temperature sensor were installed in landfill layer at different altitude. The simulated landfill was sealed with plastic membrane finally.

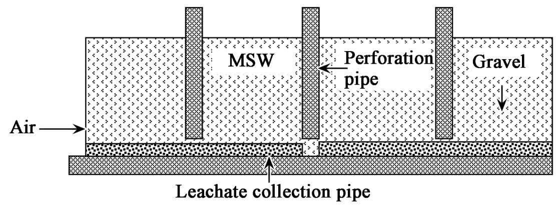

Fig. 1 Schematic of semi-aerobic landfill. MSW: municipal solid waste. MSW: municipal solid waste.

1.2 Municipal solid waste used in experiments

The simulated semi-aerobic landfill was filled with 250 t of shredded (diameter in the range of 5–20 cm) and compacted municipal solid waste. The composition of waste used in the experiments is given in Table 1.

Table 1 Components of experimental municipal solid waste

Component	Proportion (%)
Food	50.81
Yard waste	2.30
Paper	4.56
Textiles	1.21
Plastics	8.74
Metal	0.19
Putty	5.38
Dust	26.81

1.3 Analytical methods

Simulated rainfall was based on local annual rainfall records, about 1 m^3 per week. Leachate collected from landfill was analyzed weekly for pH, chemical oxygen demand (COD), ammonia-nitrogen and nitrate-nitrogen concentration, which is in accordance with the national standard of water quality. Landfill gas collected from landfill was analyzed for methane, oxygen, and carbon dioxide concentration once per week. The temperature of landfill layer was monitored on line and data were collected once per hour.

2 Results and discussion

2.1 Variations in leachate quantity

2.1.1 Dynamic variations of COD

A change from anaerobic to partial aerobic environment took place in semi-aerobic landfill, resulting in an increased carbon conversion during *in situ* aeration, occuring with the aerobic degradation of organic compounds and their release into gas phase (mainly as carbon dioxide) (Pohland and Yousfi, 1994). As a result, the highest concentration of COD was 26,486 mg/L in the test, which was considerably lower than that in anaerobic landfill (Kjedsen *et al*., 2002). It may also be because the components of waste in the test were different from others and leachate quality analysis was delayed by one month.

Since the bioavailable waste components content was high early, and organic substances were in the aerobic biodegradation, the COD concentration descended quickly in the former 8 weeks from 26,486 to 8,550 mg/L, reducing 67.7% (Fig.2). In the aerobic biodegradation process, large molecule organic compounds were decomposed into small molecule organic compounds by microorganism in the use of enzyme, and then, these small molecule organic compounds continued to be decomposed by aerobic microorganism and were finally converted into carbon dioxide, water, nitrate, and sulfate (Robert and Morton, 1996).

The variation of COD concentration in semi-aerobic landfill leachate has presented different characteristics from anaerobic landfill. A stable descending trend was observed as time went by, while in anaerobic landfill, it was accumulated and ascended in prophase (Elevi and Baccini, 1989). The cause of these phenomena may be that oxygen was available in anaerobic landfill in prophase, and organic compounds biodegraded under aerobic conditions. As time passed, oxygen in anaerobic landfill was exhausted and then anaerobic microorganism began to adapt to anaerobic condition and be in domination; most complex organic compounds were degraded into small molecule organic compounds, which could be dissolved in leachate, thus resulting in the COD concentration increase (Morris *et al*., 2003). As large-scale aerobic area existed, the circumstance had small change in semi-aerobic landfill, and therefore, had little effect on the activity of microorganism, and at the same time, the degree of aerobic degradation was high, not so much little molecule organic compounds were dissolved in leachate as in anaerobic landfill, and the COD concentration in semi-aerobic landfill presented a stable descending trend. The COD concentration descending trend can be divided into three stages, the first stage lasted from the 1st to the 8th week; in this stage, the COD concentration reduced rapidly because of the high content organic compounds in waste and the high ratio of organic compounds to microbe; microorganisms were in logarithm mount stage and propagated quickly, which led to organic compounds biodegrading bloom. The second stage lasted from the 9th week to the 24th week; in this stage, the COD concentration descended slightly as large quantity of microorganism propagated, the content of organic compounds in waste decreased, microorganism propagation came to an attenuation stage, and the organic compounds biodegradation weakened (SEPA, 2002). The stable phase started after the 24th week, and the COD concentration ranged around 300 mg/L. Since the organic compounds content reduced more, microorganism propagation came to an endognesis respiration stage and microorganism began to metabolize cell substances deposited by itself, the gross of microorganism reduced swiftly, and the degradation almost stopped. About 30%–60% of the dissolved organic carbon in leachate was hardly or non-degradable matter (Debra, 1996).

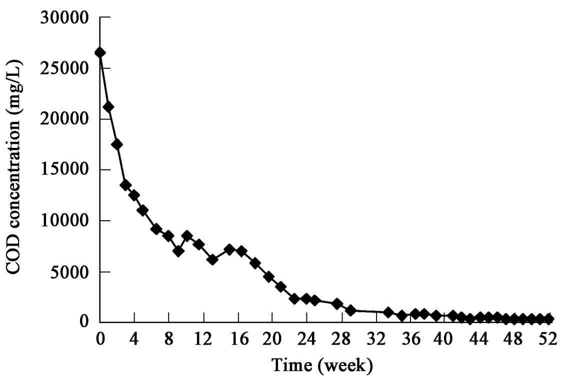

Fig. 2 Variation of COD concentration in leachate.

2.1.2 Variations of ammonia nitrogen, nitrate nitrogen, and pH in leachate

Ammonia nitrogen in leachate was mostly from the biodegradation of organic compounds containing nitrogen in landfill waste. One remarkable character of leachate is high ammonia concentration, which will restrain microorganism activity and slow waste biodegradation (Robinson, 1992).

Ammonia concentration in leachate fluctuated before the first four weeks and presented a stable descending trend after four weeks, which was different from that in anaerobic landfill in which the ammonia concentration in leachate will accumulate for a long time (Fig.3a). As a result of unavailable oxygen in anaerobic landfill, ammonia was very difficult to be transformed to nitrate in the function of nitrobacteria. However, semi-aerobic landfilling structure could make the landfill maintain better aerobic condition and these were convenient for nitrification, and at the same time, partial landfill were in anaerobic condition, therefore, denitrification also existed. Thus, ammonia in landfill can be transformed to gaseity nitrogen because of nitrification and denitrification, and then, ammonia concentration in leachate was reduced.

The maximum of ammonia concentration in test was 3,200 mg/L and descended quickly from the 4th week to

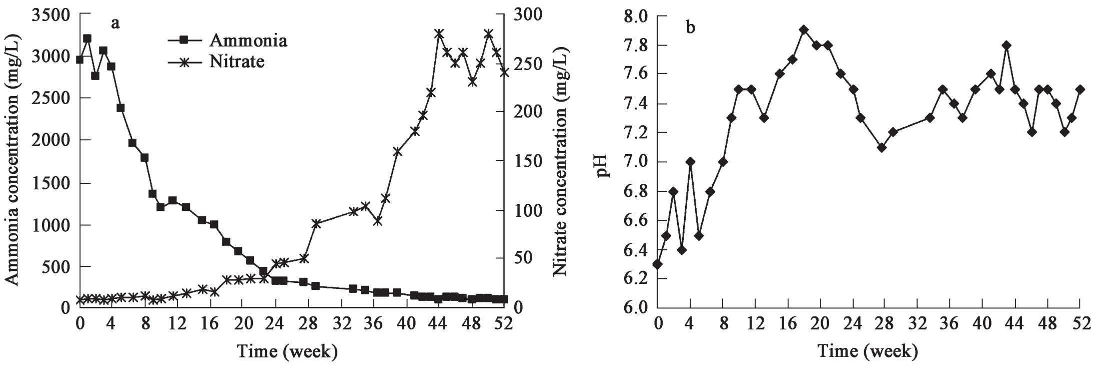

Fig. 3 Variation of ammonia, nitrate concentration (a) and pH (b) in leachate.

the 10th week; the reducing rate began to decrease after 10 weeks and the concentration ranged approximately 100 mg/L after 48 weeks, reducing 96.6%. Ammonia had a high concentration and a swiftly varied character, which was because a large quantity of ammonia was discharged from nitride as waste biodegraded and nitrobacteria was just in the propagation stage; also, nitrobacteria was self-nutrition bacteria, and its propagation was restrained by high content of organic compounds, therefore, the nitrification was very weak in prophase. On the other hand, very high ammonia concentration restrained the activity of microorganism and it made biodegradation unstable (Robinson, 1992). With nitrobacteria adapting to condition and the organic compounds concentration descending, nitrification began to strengthen and more ammonia was transformed to nitrate, and the ammonia concentration began to decrease gradually. The nitrite concentration variation curve also showed that the nitrite concentration began to ascend after the 4th week, and it proved that nitrification had strengthened. Nitrite maintained low concentration in prophase, because nitrification was restrained by high organic compounds concentration; on the other hand, it was the result of denitrification. Nitrite concentration fluctuated from the 10th week to the 29th week, but remained at a high level, because nitrobacteria's adapting to condition gradually and organic compounds concentration reducing led to strengthening nitrification; however, nitrite concentration reached maximum (270 mg/L) in the 44th week. Since organic compounds content in landfill descended greatly and carbon resource required in denitrification was little, denitirification weakened, which led to nitrite accumulation (Robinson, 1992).

Variations of pH values of the leachate in semi-aerobic landfill are given in Fig.3b. The pH values were all higher than 7 except for the initial 8 weeks. These may be because high degree of aerobic degradation resulted in less acidification dissolved in leachate. The pH value increased swiftly from 6.3 at the beginning of the test to 7.9 in the 19th week and after that, a slightly decrease was observed and remained constant at 7.4 after 33 weeks.

2.2 Variations of gas composition average content

The average contents of gas composition are given in Fig.4. It shows that methane's average content ranged between 7 vol.%–13 vol.% in semi-aerobic landfill, considerably lower than the typical values (50%–70%) (Bagchi, 1994). As oxygen was available in most parts of semi-aerobic landfill and methanogenic bacteria only existed in strict anaerobic condition, methane could not come into aerobic sphere. The results showed that methane cannot be detected in several sampling positions because of aerobic environment, and methane can be detected in some sampling positions where the oxygen cannot be always detected. Carbon dioxide average content varied between 19 vol.% and 28 vol.%, and it was the main gas composition in the whole stabilization process, which was different from that in anaerobic landfill, in which methane was the main landfill gas composition in methanogentic stage. The reason may be that organic compounds degraded in aerobic condition, and most organic substance in waste were converted into carbon dioxide and nitrogen gas, while, organic acid and hydrogen were transformed into methane by methanogentic bacteria in methanogentic stage in anaerobic landfill (Robert and Morton, 1996). The oxygen average content varied between 3 vol.%–8 vol.%. As can be seen, most parts of landfill were full of oxygen, which ensures that most wastes were under aerobic degradation in semi-aerobic landfill.

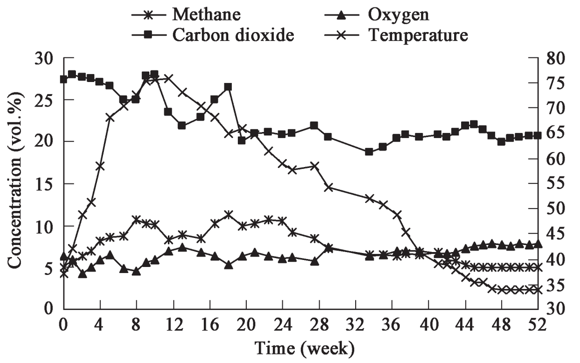

Fig. 4 Variation of landfill gas composition and temperature.

The variations of gas composition average content are shown in Fig.4. Carbon dioxide concentration had slightly fluctuated and presented a decrease trend before the initial 20 weeks, then remained at a constant level, which was different from the carbon dioxide variation in anaerobic landfill. The variations of methane and oxygen presented a contrary trend. Before the 28th week, methane concentration was higher than oxygen; after that, it was just on the contrary. At the initial stage, organic compound content was comparatively high and biodegradation was more intensive, which would demand large quantity of oxygen; thus, there was more anaerobic sphere in landfill, which resulted in methane concentration increase. Along with the stabilization process, organic compounds content decreased and biodegradation also weakened; then, the speed of oxygen entering into landfill was faster than the speed at which biodegradation consumed oxygen and reached balance at the 28th week. After 28 weeks, aerobic sphere extended and methane concentration decreased and oxygen concentration increased. Oxygen remained at a relatively constant concentration as the regulating function of oxygen concentration difference and temperature difference between landfill inner and outside.

2.3 Relation of leachate quality, landfill gas, and temperature

Landfill stabilization was a complex process accompanied with physical, chemical, and biological reactions. Organic compounds in wastes were degraded by microorganism, some were transformed into little molecule compounds, which were dissolved in leachate, others into sphere matter, and a great deal of heat was discharged. Thus, leachate quality, landfill gas, and temperature were not isolated but related mutually.

The intensity of biodegradation and degree of landfill stabilization can be reflected by landfill inner temperature. Landfill body temperature variation is shown in Fig.4. The temperature in semi-aerobic landfill remained at a high value; even in the late stage, it was higher than 33°C and the highest reached up to 75.8°C. Temperature fluctuated swiftly in the former 42 weeks and increased quickly in the initial 8 weeks and reached the highest value in the 12th week. There was an obvious decreasing process after the 12th week, which reached a constant stage after 44 weeks.

There existed partial aerobic areas in semi-aerobic landfill and high oxygen concentration in landfill gas (Fig.4); organic compounds were degraded under aerobic condition, therefore, the COD concentration descended quickly (Fig.2). Since partial aerobic areas existed in landfill, ammonia in landfill will be transformed into nitrite and nitrogen gas by nitrification and denitrification. Thus, ammonia concentration not only did not accumulate but also declined rapidly and nitrate concentration increased rapidly in the late stage (Fig.3a). The temperature in landfill increased most quickly in the initial 8 weeks and the reducing rate of organic compounds pollutants in leachate was maximal corresponding. The oxygen concentration was lower initially and maintained a constant level later, which maybe correlated to microorganism's violent activity initially and the temperature difference between landfill inner and outside. Owing to microorganism's violent activity and low temperature in landfill body, the velocity of consuming oxygen was higher than that of transferring oxygen from outside to landfill body. With the degradation weakening, the velocity of consuming oxygen decreased, which resulted in oxygen concentration's increase in landfill. As the velocity of consuming oxygen and the velocity of transferring oxygen reached balance, the oxygen concentration in landfill remained at a constant level, and as a result, the oxygen content in landfill gas remained constant.

3 Conclusions

On the basis of the experimental results obtained during the investigation, the following conclusions are drawn:

The concentrations of COD and ammonia not only did not cause accumulation as in anaerobic landfill but also declined rapidly, and remained 300 and 100 mg/L respectively after 48 weeks. Simultaneously, the reducing rate reached 98.9% and 96.9%, respectively. Nitrate concentration increased rapidly after 24 weeks and remained 250 mg/L after 41 weeks.

Carbon dioxide was the main composition in landfill gas, and its concentration remained at a high level through the whole stabilization process. The contents of carbon dioxide, oxygen, and methane varied between 19 vol.%–28 vol.%, 1 vol.%–8 vol.%, 5 vol.%–13 vol.%, respectively. All these remained at a stable state after 48 weeks. Carbon dioxide concentration had slightly fluctuated and presented a decrease trend before 20 weeks and then remained at a constant level. Before the 28th week, Methane concentration was higher than oxygen, and after that, it was just on the contrary.

The temperature in semi-aerobic landfill chamber remained relatively high; even in the later stage, it was above 33°C and the highest could go up to 75.8°C. Relations in variation of leachate quality, landfill gas, and temperature showed that the semi-aerobic landfilling structure could speed up the landfill stabilization process. Pollutants concentration in leachate decreased rapidly in the process, which was beneficial to the treatment of leachate. Low methane concentration in landfill gas also added a significant meaning to the problem of global warming.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (No. 50508042).

References

Bagchi A, 1994. Design, Construction and Monitoring of Landfill. John Wiley and Sons Inc, New York: 135–137.

Basri D R H, Mohamed A A K, Aziz H A, 2000. The effectiveness of cockle shells in treating leachate organic pollutants. In: ISWA International Symposium and Exhibition On Waste Management in Asian Cities. 23–25.

Bookter T, Ham R, 1982. Stabilization of solid waste in landfills. *Journal of Environmental Engineering*, 26(4): 1089–1100.

Carson D A, 1995. The municipal solid waste landfill operation as a bioreactor. Seminar Publication: Landfill bioreactor design and operation. US Environment Protection Agency, Washington. 124–135.

Chugh S, Chynow D P, Clarke W, Pullammanappallil P, Rudolph V, 1999. Degradation of unsorted municipal solid waste by a leach-bed process. *Bioresource Technology*, 61: 103–115.

Debra R R, 1996. Full-scale experiences with leachate reciculation landfillS: case studies. *Waste Management and Research*, 14(14): 347–365.

Dong L, Wang Q, Li Y, Huang S H, 2000. The research of acceleration for stabilization in the landfill site. *China Environmental Science*, 20(5): 461–464.

Elevi H, Baccini P, 1989. Long term behavior of municipal solid waste landfills. Waste Management and Research, 7: 43–56.

Kjedsen P K, Barlaze M A, Rooker A P, 2002. Present and long-term composition of MSW landfill leachate: A review. *Critical Reviews in Environmental Science and Technology*, 32(4): 297–336.

Lay J J, Li Y Y, Noike T, Endo J, Ishimoto S, 1997. Analysis of environmental factors affecting methane production from high-solids organic waste. *Water Science and Technology*, 36(6-7): 493–500.

Leckie J O, Pacey J G, Halvadakis, 1979. Landfill management with moisture control. *Journal of Environmental Engineering Division*, 105(2): 337–355.

Liliana B, Lqpz I, Anido C, 1997. Hydrolysis constant and VFA inhibition in acidogenic phase of MSW anaerobic degradation. *Water Science and Technology*, 36(6-7): 479–484.

Hanashima M, Matsufuji Y, Higuchi S, 1986. A method of designing leachate treatment systems for landfill sites. *Cities and Waste*, 6(12): 26–35.

Morris J W F, Vasuki N C, Baker J A, 2003. Findings from long-term monitoring studies at MSW landfill facilities with leachate reciculation. *Waste Management*, 23(6): 653–666.

Onay T T, Pohland F G, 1998. *In situ* nitrogen management in controlled bioreactor landfills. *Water Research*, 32(5): 1383–1392.

Pohland F G, 1980. Leachate recycle as landfill management option. *Journal of Environmental Engineering*, 106(6): 1057–1069.

Pohland F G, Yousfi B A, 1994. Design and operation of landfills for optimum stabilization and biogas production. *Water Science and Technology,* 30(12): 117–124.

Raynal J, Ddlgen J P, Moletta R, 1998. Two-phase anaerobic digestion of solid wastes by a municipal liquefaction reactors process. *Bioresource Technology,* 65: 97–103.

Robert J F, Morton A B, 1996. Hudrogen sulfide production during decomposition of landfill inpuls. *Journal of Environmental Engineering*, 124(4): 353–361.

Robinson H D, 1992. Leachate collection, treatment and disposal. *Water and Environmental Management*, 6(3): 321–332.

Robioson H D, Peter J M, 1983. The treatment of leachates from domestic wastes in landfills-I: Aerobic biological treatment of a medium-strength leachate. *Water Research*, 17(11): 1537–1548.

State Environment Protection Administration of China. Technical Specifications Requirements for Monitoring of Surface Water and Waste Water (HJ/T91-2002).

Yasushi M, Masataka H, 1997. Characteristic and mechanism of semi-aerobic landfill on stabilization of solid waste. Proceedings of the first Korea-Japan Society of Solid Waste Management. 87–94.

Yazdani R, Augensein D, Pacey J, 1977. EPA project XT: Yolo county's accelerated anaerobic and aerobic composting project. Management of gas and leachate in landfills proceedings of the third annual municipal solid waste research symposium, EPA-600/9-77-026. 73–86.