Journal of Environmental Sciences Vol.16, No.5, pp.802—807, 2004

ISSN 1001—0742
CN11—2629/X

Article ID: 1001-0742(2004)05-0802-06 **CLC number**: X703; X705 **Document code**: A

Removal of high concentrated ammonia nitrogen from landfill leachate by landfilled waste layer

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Abstract: The landfill of municipal solid waste (MSW) could be regarded as denitrification reactor and involved in ammonia nitrogen biological removal process. In this research, the process was applied to municipal solid waste collected in Shanghai, China, which was characterized by high food waste content. The NH_4^+ removal efficiency in the system of SBR nitrifying reactor followed by fresh and matured landfilled waste layer in series was studied. In the nitrifying reactor, above 90% of NH_4^+ in leachate was oxidized to NO_2^- and NO_3^-. Then high concentrated NO_2^- and NO_3^- were removed in the way of denitrification process in fresh landfilled waste layer. At the same time, degradation of fresh landfilled waste was accelerated. Up to the day 120, 136.5 gC/(kg dry waste) and 17.9 gN/(kg dry waste) were converted from waste layer. It accounted for 50.15% and 86.89% of the total carbon and nitrogen content of preliminary fresh waste, which was 4.42 times and 5.17 times higher than that of reference column respectively. After filtering through matured landfilled waste, BOD_5 concentration in leachate dropped to below 100 mg/L, which would not affect following nitrification adversely. Because the matured landfilled waste acted as a well methanogenic reactor, 23% of carbon produced accumulatively from fresh landfilled waste degradation was converted into CH_4.

Keywords: landfill leachate recirculation; ammonia biological removal; fresh landfilled waste; matured landfilled waste

Introduction

Biological removal of ammonia, i.e. nitrification-denitrification processes, is the most effective and economic method for treating wastewater with high ammonia nitrogen concentration, such as landfill leachate. But the leachate from matured landfill is characterized with low biodegradable carbon concentration. So the supplementary external carbon source has to be added to facilitate denitrification. Consequently, the operational cost increases (Carley, 1991).

There is not only abundant organic carbon, but also a great deal of anaerobic and facultative anaerobic bacterium in landfilled waste, especially in fresh waste layer. So landfill can be regarded as an anaerobic reactor to accomplish denitrification process. This process can reduce leachate treatment cost and accelerate waste degradation. Onay *et al*. conceived a three-component landfill including anoxic, anaerobic and aerobic zones for *in-situ* attenuation of ammonia, and conducted correlative lab-scale research (Onay, 1998). However, they used matured compost, not lanfilled waste, as the matrix of the simulated landfill reactor. So it did not faithfully reflect the process of high concentrated ammonia removal by landfilled waste. Additionally, Jokela and Burton studied the denitrificaton efficiency of the matured lanfilled waste and the mathanogenic landfilled waste respectively (Jokela, 2002; Burton, 1997). The studies on ammonia removal from leachate by landfilled waste are still at the initial stage, and most of the research objects are matured landfilled wastes.

In this study, the feasibility of ammonia nitrogen removal with the system of SBR nitrifying reactor followed by fresh landfilled waste layer and the matured landfilled waste layer was studied.

1 Materials and methods

The system used in this study consisted of a nitrifying reactor and two simulated-landfill columns. Schematic diagram of the system is shown in Fig.1.

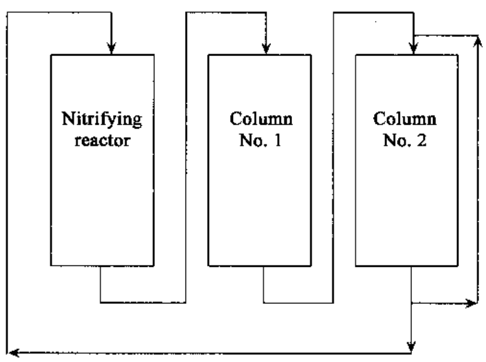

Fig.1 System schematic diagram of this study

The nitrifying reactor was operated in SBR mode, in which NH_4^+ was oxidized to NO_2^- and NO_3^-. No.1 simulated-landfill column was used to simulate the denitrification process of fresh waste layer. The synthetic waste loaded in No. 1 column was prepared according to a survey on municipal solid waste composition in Shanghai. Its composition and characteristics are shown in Table 1 and Table 2 respectively. No. 2 simulated-landfill column was used to simulate matured landfilled waste layer. No.2 column was loaded with the matured waste from Laogang Landfill in

Foundation item: The National Hi-Tech R&D Program(863) of China(No. 2001AA644010; 2003AA644020) and the Key Project of Shanghai Education Committee(No. 2001-44); * Corresponding author

Shanghai. The characteristics of waste are listed in Table 2.

Table 1 Composition of the synthetic waste

Composition	Mass content, %
Food waste	85.0
Plastic	5.0
Paper	7.0
Glass	1.6
Tissue	1.0
Metal	0.4
Total	100.0

The nitrifying reactor was fed once every day, aerated consecutively for 23.5 h and settled for 0.5 h, then discharged finally. The hydraulic load was kept at the level of 500 ml/d, 1/3 of the total volume of the reactor. The leachate was recycled into column No.1 and No.2 once every day. The hydraulic load of No.1 column was constant, about 27.8 ml/(kg wet waste·d). Due to the leachate produced by No.1 column was totally recycled to No.2 column, the hydraulic load of No.2 column changed with the variation of leachate production of No.1 column consequently.

Table 2 Physico-chemical characteristics of the experimental material

	Waste	Moisture content	Volatile solid	Element composition					
				C	H	N	S	O	Ash
Mass content *, %	Synthetic waste	70	88.00	27.27	3.59	2.06	0.27	34.59	32.24
	Matured waste	40	20.19	——					

Notes: * All the indices, except for moisture content, based on dry weight of waste

Additionally, a reference simulated-landfill column was used to simulate traditional sanitary landfill, without leachate recycle, but with simulated rainfall once a week, approximately 1—3 ml/(kg wet waste · week), which was decided based on the statistical data of monthly rainfall in Shanghai.

All simulated-landfill cylinder columns were made of plexiglass (D: 19 cm; H: 100 cm). The waste densities of No.1 simulated-landfill column and reference simulated-landfill column were both 0.85 t/m^3. The waste density of No.2 simulated-landfill column was 1.10 t/m^3.

The whole system was operated at 35℃. The analysis of the leachate was performed according to national standard methods(Xi, 1996).

2 Results and discussion

2.1 Nitrifying reactor

The operation of the nitrifying process could be separated into three successive stages, i.e. sludge cultivation stage, ammonia concentration increasing stage, and steady operation stage. The quality of the effluent during the whole experiment is shown in Table 3. Ammonia removal efficiency in the nitrifying reactor is shown in Fig. 2.

Table 3 Qualities of the SBR nitrifying reactor effluent

Indexes	pH	COD	BOD_5/COD	NH_3 - N	NO_3^- - N	NO_2^- - N
Concentration*	8.5—9.0	< 1000	< 0.1	< 20	560—1450	50—780

Notes: * Except for pH and BOD_5/COD, the units of the parameters were all in mg/L

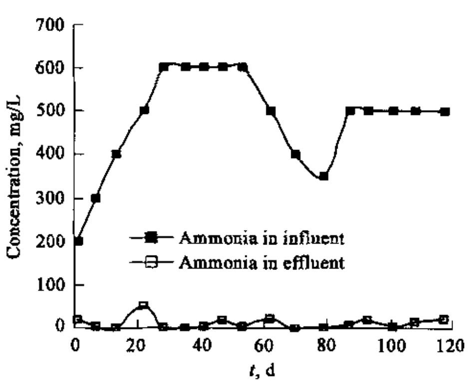

Fig.2 Ammonia removal in nitrifying reactor

2.1.1 Sludge cultivation stage

Inoculated sludge was taken from sewage treatment plant, and cultivated by adding NH_4Cl solution everyday, for nearly one month. The aerating rate was kept at 0.375 m^3/h. The pH value in the reactor was kept between 8.0—8.5 by adding Na_2CO_3 solution. After about two weeks, steady nitrification condition was established, for the ammonia concentration in effluent was close to zero. The proportion of NO_2^- concentration in NOx concentration(NO_2^- concentration plus NO_3^- concentration) was above 50%. But the ratio decreased along with the increase of cultivation time.

2.1.2 Ammonia concentration increasing stage

The variations of DO and ammonia concentrations are shown in Fig.3 and Fig.4 respectively.

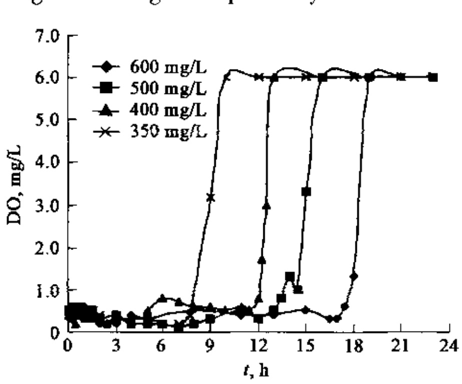

Fig.3 Variation of DO in nitrifying reactor

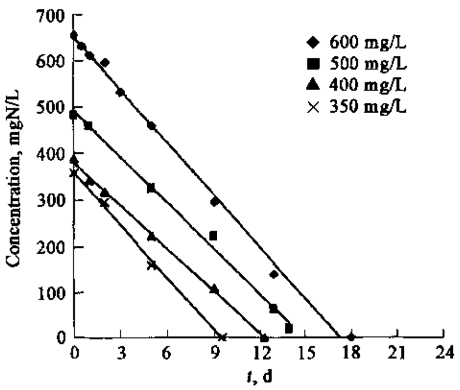

Fig.4 Ammonia concentrations variation vs. time

When initial ammonia concentration increased, time consumed to eliminate all the ammonia in reactor increased correspondingly. But the variations of ammonia concentration vs. time were similar, i.e. ammonia concentration descended almost linearly, and the slope changed little. It suggested that ammonia was removed in similar rate.

The variations of DO in different conditions were the same. Before ammonia was removed completely, DO was kept below 1.0 mg/L. As soon as ammonia concentration closed to zero, DO increased sharply to about 6 mg/L, and kept at such level until aeration was stopped.

The samples taken at certain interval in a cycle were analyzed. The results indicated that NO_2^- accumulation occurred in such low DO and high pH conditions. The nitrous transformation was defined as the ratio of the NO_2^- concentration to the initial ammonia concentration. As shown in Fig. 5, when ammonia was eliminated, all the nitrous transformation efficiency reached to 70%—80%. It indicated that NH_4^+ was transformed to NO_2^- in priority in such condition. So the nitrification in one whole cycle could be separated into two stages, most of NH_4^+ was transformed to NO_2^- firstly, and then NO_2^- was oxidized to NO_3^-. The proportion of NO_2^- in NOx in the final effluent was below 10%.

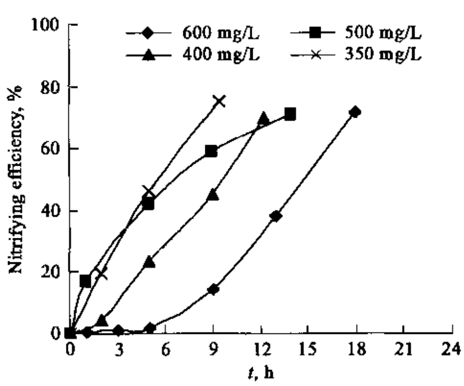

Fig.5 The nitrous transformation efficiency of NH_4^+

2.1.3 Steady operation stage

Initial ammonia concentration was kept at 500 mg/L, and the quality of effluent was steady.

2.2 No.1 simulated-landfill column

2.2.1 Variation of NO_3^- and NO_2^- concentrations

During the entire experimental process, NO_2^- concentrations of both No. 1 column and reference column were close to zero, and the timely evolution of NO_3^- concentrations are shown in Fig.6.

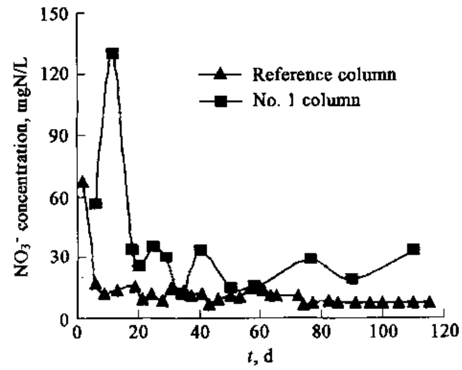

Fig.6 Evolution of NO_3^- concentration in leachate

In two weeks after waste was landfilled, there was still some oxygen remained in interspace of waste. So NO_3^- concentration in leachate from reference column rose up to 67.43 mg/L. After oxygen was used up, a reductive environment was established, and NO_3^- concentration decreased to near zero consequently.

Because of the effect of remained oxygen in waste and high NO_3^- concentration in recycled leachate, NO_3^- concentration in the leachate from No.1 column was higher than that of the reference column. However, NO_3^- concentration was kept below 30 mg/L, which indicated high denitrification efficiency.

2.2.2 Variation of pH

In the first 20 d, the aerobic condition in simulated column shifted to anaerobic or anoxic condition. In this stage, the dominant factor affecting the degradation of organic waste was oxygen in waste interspace. So the pH evolution of the leachate from No. 1 column and reference column was similar, as shown in Fig.7.

Hereafter, the pH value of reference column was about 5.70, close to that of volatile acid buffer system (Kim, 2001), which indicated the accumulation of volatile acid in the reference column. The pH value of No. 1 column was higher than that of the reference column. Because denitrification by facultative anaerobic bacteria in No. 1 column could produce alkalinity, which neutralized acids produced by hydrolysis and acidification of waste, pH elevated consecutively and was close to 7.0 in the end.

2.2.3 Net accumulated production of leachate

Net accumulated production of leachate was equal to accumulated leachate production minus accumulated quantity of recycled leachate. Most of net leachate production came

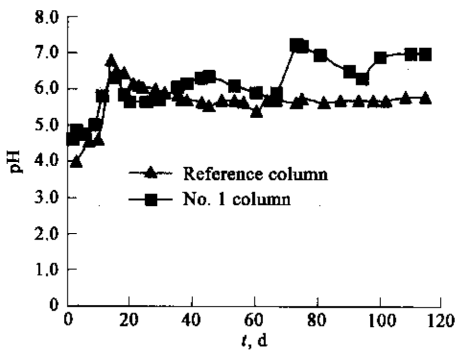

Fig.7 Evolution of pH vs. time

from waste biodegradation. So this index could reflect the biodegradation rate of waste in some extent. From Fig.8, it could be found that the net accumulated leachate production of No. 1 column was obviously higher than that of the reference column.

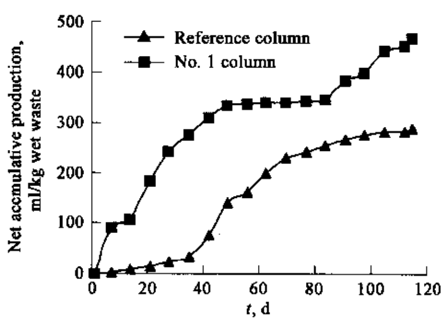

Fig.8 The net cumulative production of leachate

The net accumulated leachate production curve of No.1 column had a stagnant stage from the day 60 to day 80. During this period, waste density in the column became very high due to waste settlement. It hindered the infiltration of recycled nitrified leachate and the discharge of produced leachate, so there was some distortion in experimental results. On the day 80, the waste in column was reloaded, waste density was changed to 0.85 t/m^3, and the recycled manner changed from feeding intermittently and discharging continuously to both feeding and discharging performed intermittently. After the adjustment, the infiltration of recycled nitrified leachate and the discharge of produced leachate worked normally, so the net accumulated leachate production increased again.

2.2.4 COD concentrations and accumulated productions

The evolution of COD accumulated productions and concentrations are shown in Fig. 9. COD concentration of leachate from the reference column quickly increased to 70000 mg/L, which was far higher than the peak of COD concentration of No.1 column, because the simulated rainfall recycled into the reference column was much lower than the nitrified leachate quantity recycled into No.1 column. When contacted with the same quantity of waste, it was more easily for leachate from the reference column to reach to higher organic content. However, due to dilution effect, COD concentration of No.1 column was much lower.

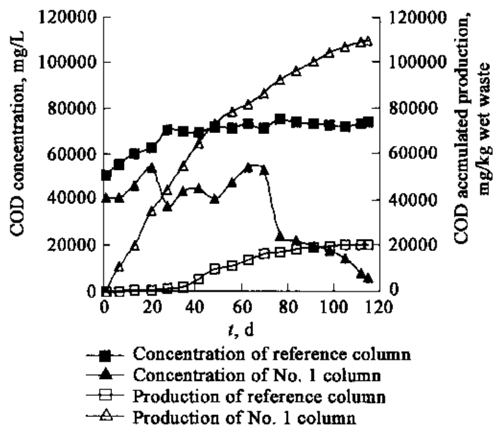

Fig.9 Evolution of COD concentration and accumulated production

For the reference column, because of low waste biodegradation rate and low COD washed-out rate, COD concentration kept constant in that level after it reached to peak value. For No. 1 column, due to large quantity of recycled leachate, both biodegradation rate and COD washed-out rate were accelerated. COD concentration began to decline from the day 70, and was close to 5800 mg/L on the day 120.

Though COD concentration of the reference column was lower than that of No.1 column, leachate production was in reverse. So COD accumulated production of No. 1 column was greatly higher than that of the reference column consequently. Up to the day 120, 20133 mgCOD/kg wet waste, i. e. 25.2 gC/kg dry waste, was produced from the reference column, which accounted for 9.26 percent of the total carbon content of preliminary waste. However, 109226 mgCOD/kg wet waste, i. e. 136.5 gC/kg dry waste, was produced from No. 1 column, which accounted for 50.15 percent of the total carbon content of preliminary waste. The latter was 4.42 times higher than the former.

2.2.5 KN concentrations and accumulated productions

Because the ratio of ammonia nitrogen to KN fluctuated very little (0.70—0.80) during the whole experiment, and the variation of KN could reflect the degradation of nitrogen-contained waste more directly, only the variation of KN concentrations and accumulated productions were discussed in this study.

The variations of KN concentration and accumulated production of No.1 column and reference column were similar to that of COD, as shown in Fig.10. It indicated that the biodegradation of different components of waste was all accelerated.

On the day 120, 880 mgKN/kg wet waste was produced accumulatively in the reference column, i. e. 2.9 gN/kg dry waste, which equaled to 14.08 percent of the total nitrogen

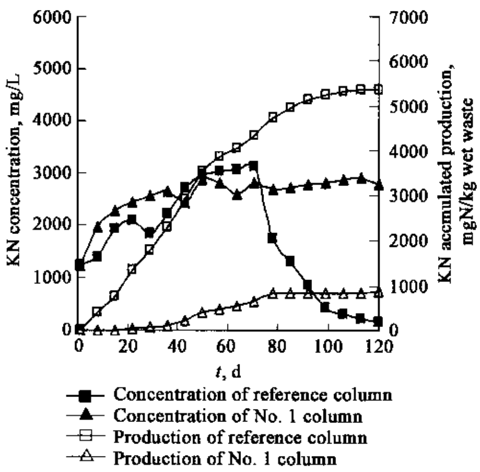

Fig. 10 Evolution of KN concentration and accumulated production

content of preliminary waste. However, the accumulated KN production of No.1 column was 5370 mgKN/kg wet waste, i.e. 17.9 gC/kg dry waste, which equaled to 86.89 percent of the total nitrogen content of preliminary waste. The latter was 5.17 times higher than the former.

Based on the above discussion, it was concluded that when nitrified leachate was recycled into the landfilled waste layer, abundant water and its movement helped to create favorable environment for waste biodegradation. Furthermore, due to high nitrite and nitrate concentrations in recycled leachate, waste could be decomposed by the facultative bacterium. The combination of the two mechanisms highly accelerated the waste degradation.

2.2.6 Accumulated production and composition of landfill gas

As discussed above, in the first 20 d, the environmental condition in simulated columns transited from aerobic condition to anoxic or anaerobic condition. So the evolution of accumulated gas production of both columns was similar, which could be observed in Fig. 11. After this period, the gas accumulated production of reference column increased very slowly, because waste degradation was still in acid formation stage and the relative bacterium had limited ability to transfer organic waste to gas phase (Bae, 1998). In this period, however, the accumulated gas production of No. 1 column increased almost linearly and the production was much higher than that of reference column, which indicated that recycled nitrified leachate could accelerate the transfer of organic carbon from solid waste to landfill gas.

During the entire experimental process, no methane was detected in the landfill gas of the reference column, which suggested that methane fermentation stage had not started yet. For No. 1 column, however, methane appeared in gas since the day 32. It indicated that the lag time of methanogenesis was shortened. But the methane volume content was only 17% in the end, which was far lower than normal methane volume content in steady methane

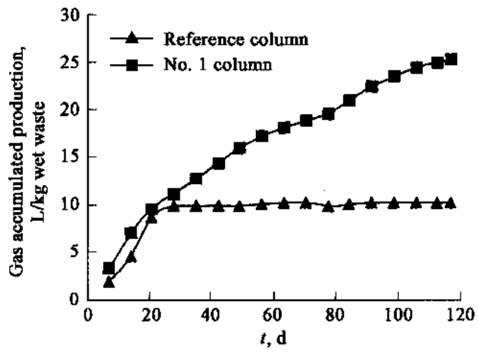

Fig. 11 Evolution of the gas cumulative production

fermentation stage, approximately 60% (Kim, 2001). It was reasonable because that the growth of methanogenic bacterium was inhibited by high-content nitrite and nitrate (Chen, 1993).

Due to denitrification in the column, nitrogen gas was detected in landfill gas of No.1 simulated landfill column. In one week after waste was landfilled, the N_2 volume content decreased from 75% to 10% sharply. It was the result of the landfill gas produced by waste degradation replacing the remained atmosphere in column. Hereafter, the N_2 volume content fluctuated between 10% and 20%. And the N_2 production approximately equaled to the removed nitrite nitrogen and nitrate nitrogen in denitrification process.

According to the above discussion, the total carbon accumulated production of No. 1 column and its distribution in leachate and gas phase could be calculated, shown in Fig. 12. Only 18% of total produced carbon was in gas phase, while the majority was transferred into leachate. The quantity of organic carbon transformed to CO_2 was 129.46 g C, which accounted for 17% of the total accumulated carbon production. And the quantity of organic carbon transformed to CH_4 was 6.85 g C, which accounted for 1% of the total accumulated carbon production.

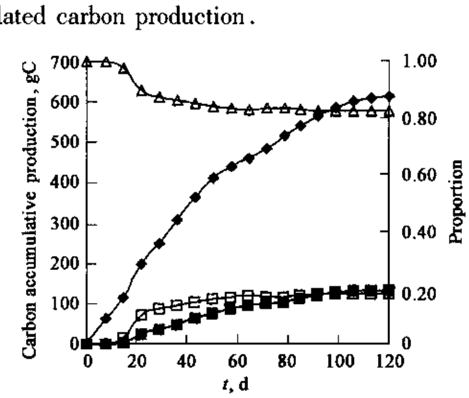

Fig. 12 Carbon cumulative production and distribution in No. 1 simulated-landfill column

Theoretically, reducing 1 g NO_3^--N needed 2.86 g BOD and reducing 1 g NO_2^--N needed 1.71 g BOD (Zhang, 1996). In this study, 57260 mg NO_3^--N and 19510 mg NO_2^--N were removed accumulatively. So the total quantity of carbon consumed by denitrification was 197.12 g BOD, i.e. 73.92 g C. It accounted for 10% of the total carbon accumulated production of No.1 column(744.95 g C), and accounted for 57% of the quality of organic carbon column transformed to CO_2 in No.1 column(129.46 g C).

2.3 No.2 simulated-landfill column

The waste loaded in No.2 simulated-landfill column was the matured landfilled waste, and the predominant bacterium in column was methanogenic bacterium. So after sufficiently contacted with the matured waste in No.2 column for long time, the organic pollutant in the leachate from No.1 column was almost removed entirely. Consequently, No.2 column could be treated as an anaerobic bacterium attached-growth reactor.

As shown in Table 3, there was almost no organic carbon in discharged leachate from No.2 column, and the ratio of ammonia to KN was closed to 1.0. Such leahcate was not able to cause the failure of nitrifying reactor.

Methane became the major content of the landfill gas, its volume content reaching to 70%.

Because of the existence of No.2 simulated-landfill column, the distribution of produced carbon in leachate and landfill gas changed vastly, shown in Fig.13. 53% of total carbon produced in whole system was transformed to landfill gas, which was 0.94 times higher than that of No.1 column. The quantity of organic carbon finally transformed to CH_4 was 140.74 g C. It accounted for 23% of the total accumulated carbon production, which was 19.55 times higher than that of No.1 column.

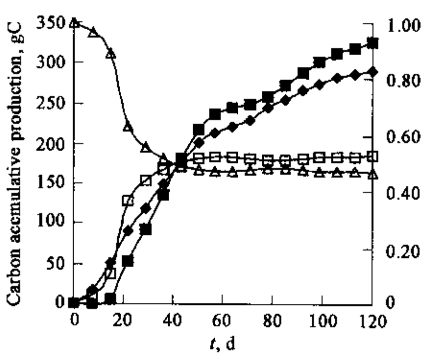

Fig.13 Carbon cumulative production and final distribution in whole system

As discussed above, it could be concluded that No.2 column had two functions. Firstly, leachate from No.1 simulated-landfill column was treated by it and met the requirements of nitrifying reactor for feed quality. Secondly, the organic pollutant in leachate was transformed into landfill gas by it as much as possible, so the resource-recovering ability of whole system increased dramatically.

3 Conclusions

Removal of high concentrated ammonia nitrogen in leachate by landfilled waste layer was feasible. The system of SBR nitrifying reactor followed by fresh and matured landfilled waste layer in series was capable of removing ammonia nitrogen effectively, and accelerating waste degradation at the same time.

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(Received for review October 31, 2003. Accepted March 17, 2004)