Degradation of Refuse in Hybrid Bioreactor Landfill[1]

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Objective To explore the process of refuse decomposition in hybrid bioreactor landfill. **Methods** The bioreactor landfill was operated in sequencing of facultative-anaerobic and aerobic conditions with leachate recirculation. pH, COD, and ammonia in the leachate and pH, biodegradable organic matter (BDM), and cation exchange capacity (CEC) in refuse were detected. **Results** CEC increased gradually with the degradation of refuse, which was negatively correlated with BDM. COD and ammonia in the leachate was declined to 399.2 mg L^{-1} and 20.6 mg N L^{-1}, respectively, during the 357-day operation. The respective concentrations of ammonia and COD were below the second and the third levels of current discharge standards in China. **Conclusion** The refuse is relatively stable at the end of hybrid bioreactor landfill operation. Most of the readily biodegradable organic matter is mineralized in the initial phase of refuse degradation, whereas the hard-biodegradable organic matter is mainly humidified in the maturity phase of refuse degradation.

Key words: Refuse; Degradation; Hybrid bioreactor landfill

INTRODUCTION

The landfill is widely used in solid waste management over the world, especially in developing countries, due to its simple and relatively cheap operation. Bioreactor landfill, in particular, has a promising future in solid waste management because undergoes a faster degradation process and leads to a greater extent of degradation in comparison with the conventional one by optimizing *in situ* conditions. So far, four types of bioreactor landfill (anaerobic, aerobic, facultative, and hybrid systems) have been developed with different operating schemes to obtain optimal results. Furthermore, each type of bioreactor is a patented process[1]. The biggest advantage of bioreactor landfill is that it reduces the time for biological stabilization. Processes such as adsorption, ion change and mechanical filtration with leachate recirculation, greatly decrease the organic strength, which has been proven by reducing the COD half-life in landfills. The anaerobic landfill based on leachate recirculation has been extensively investigated. Aerobic landfill has also been used in Japan and Korea[2]. However, hybrid bioreactor landfill has been less studied and is still in its early stage. Hybrid bioreactor landfill involves both aerobic and anaerobic conditions. Two types of hybrid system have been explored, one of which short-term cycling of air injection into the landfill, the other is sequencing of aerobic and anaerobic conditions. A few studies about the effect of cyclic air injection on the performance of hybrid bioreactor landfill are available, showing that the cyclic air injection system stabilizes leachate in a shorter time than the purely aerobic system[3-5]. In addition, a recent study demonstrated that the refuse can be stabilized more quickly in the hybrid bioreactor landfill with cyclic air injection than that without air injection[6]. Dong *et al.*[7] reported that cyclic air injection system can reduce a mass of contaminants in a shorter time. However, few studies are available on the sequencing air-injection system. *In situ* aeration can improve the stabilization of refuse in old and anaerobic landfill, reduce the organic matter, especially which cannot be

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degraded in anaerobic conditions, increase *in situ* temperature, accelerate settlement and reduce gas emissions such as methane. *In situ* aeration has been successfully used to reduce odors and methane concentration in landfill mining projects[8-9]. However, leachate recirculation has not been utilized in these studies.

The cation exchange capacity (CEC) is a physico-chemical property to reflect the fertilization and buffer capacity of media. CEC of composts has been used to evaluate the degree of humidification and/or maturity[10]. Additionally, several studies have found the mobility of heavy metals closely related with CEC in soil and sediments. However, no report is available on use of CEC in landfill.

This study was to explore the process of refuse degradation in hybrid landfill as well as the sequencing of facultative-anaerobic and aerobic conditions and leachate recirculation.

MATERIALS AND METHODS

Experimental Design

In order to create sequencing of anaerobic and aerobic conditions, bioreactor landfill was operated for two stages. The first stage aimed at creating anaerobic condition for bioreactor landfill. In this stage, the two-phase bioreactor landfill consisting of methanogenic reactor (MR) and the fresh-refuse filled reactor (LR) was used. Schematic configurations are shown in Fig. 1 with a real line, in which LR is not aerated. The second stage aimed at creating aerobic condition for bioreactor landfill. In this stage, only LR was operated, air was aerated into the top layer of refuse in LR. Schematic configurations are shown in Fig. 1 with a broken line. All reactors were constructed with PVC. LR has a diameter of 28.7 cm and a length of 100 cm, while MR has a diameter of 28.7 cm and a length of 65 cm.

Simulated Bioreactor Landfill Loading and Characteristics of Refuse

The refuse was cut into 3-cm thick pieces and mixed before loading. MR was loaded with 26.5 kg of aged refuse at a wet density of 940 kg m^{-3}. The aged refuse was excavated from Hangzhou Landfill in China with a placement time of 6-7 years, and its characteristics were as follows: TN=4191 mg N kg^{-1} dry refuse, VS (w/w)=17.3%, BDM (w/w)=8.2%, pH=7.59, moisture content=38.2%.

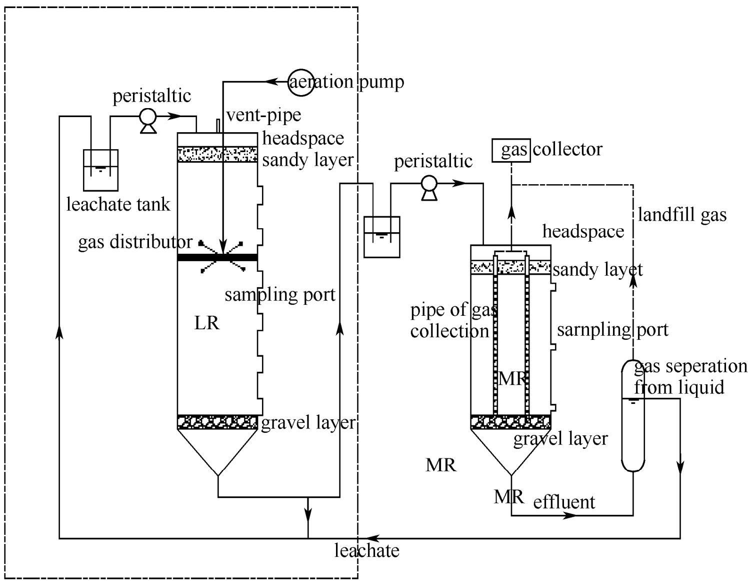

FIG. 1. Schematic configurations of hybrid bioreactor landfill.

LR was packed with 27.4 kg of fresh refuse at a wet density of 680 kg m^{-3}. The fresh refuse was collected from Caihe Transfer Station in Hangzhou, where the physical composition of fresh refuse is (by wet weight, w/w) residues of kitchen (57.9%), various kinds of paper (7.0%), plastics (5.0%), cellulose textile (3.0%), glass (2.6%), woods (7.7%), brick and soil (16.8%). The initial characteristics of the fresh refuse were: TN= 12330 mg N kg^{-1} dry refuse, VS (w/w)= 60.8 %, BDM (w/w)= 41.9%.

Both MR and LR were sealed with a gasket and silicone sealant after refuse loading. The moisture content of fresh refuse in LR was adjusted to 70% at beginning of operation. In addition, in order to ensure the initiation and development of microbial activity, MR was started up with synthesizing sewage before operation.

Operation of Bioreactor Landfills

LR and MR were placed at room temperature and semi-continuously operated during the experiment.

In the first stage, the leachate from LR and the effluent from MR were respectively recirculated into MR and LR with a peristaltic pump within 8 h daily. The surface loadings ranged 1.3 mm d^{-1} −10.1 mm d^{-1}. Landfill gas was collected from MR with a vacuum bag connected to two pieces of perforated pipes placed in MR. The first stage lasted for 200 days.

When MR stopped producing methane and the pH value of leachate from LR was above 7.0 in the first stage, cross recirculation of the leachate and effluent was stopped. From days 201-205, only the effluent from MR was collected daily to feed into LR.

In the second stage, MR was discarded from the bioreactor landfill. LR was daily aerated for 1 h at a flow rate of 30 L h^{-1}. Meanwhile, the leachate from LR was directly recirculated into itself within 8 h daily. As all the leachated effluents from MR were transferred into LR, its surface loading increased sharply at a range of 60.1 mm d^{-1} – 38.4 mm d^{-1}.

Analytical Methods

The pH values of leachate and effluent were analyzed daily, while COD, VFA, and ammonia were determined weekly. The pH values were measured with a PHS-digital pH meter (DELTA 320). COD and NH_4^+-N analyses were conducted following standard methods. Inorganic COD was also determined with the same method as COD without 2-h heating.

The BDM of refuse was analyzed according to the analysis manual of municipal solid waste. The pH of refuse samples was measured in 1:5 (v/v) suspensions using a pH-meter (DELTA 320). CEC was determined with 1 mol/L ammonium acetate at pH=7[11]. Gas production was collected into a vacuum bag and measured with water-draining method. Gas (CH_4) concentrations were analyzed using a gas chromatograph (GC) equipped with a thermal conductivity detector (TCD) and a 2-m stainless column packed with GDX104 (60/80 meshes). The operational temperature of column was 40 ℃, and carrier gas (N_2) at a rate of 30 mL min^{-1}.

Correlations between parameters were statistically analyzed using the software SPSS 13.0.

RESULTS

Characteristics of Leachacte and Effluent in the First Stage

The characteristics of leachate and effluent in the first stage are illustrated in Figs. 2a and 2c. With the rapid hydrolysis of readily biodegradable organic matter in refuse, the pH decreased while the COD and ammonia accumulated in leachate during the first 36 days. The pH of leachate reached its lowest level of 5.49 on day 29, and the COD and ammonia level increased to 55 614 mg $L^{-1}$and 929.8 mg L^{-1} respectively on day 22. From day 36, anwards the ammonia of leachate reduced steadily until day 155, and then increased slightly. The pH was gradually increased to 7.0 on day 118. The COD decreased consistently until the end of the first stage. However, the decrease of COD was apparently stagnated from day 57 to day 85. At the end of the first stage, the pH of leachate reached 7.42 and the level of COD and ammonia was 1255.4 mg L^{-1} and 359.1 mg N L^{-1}, respectively.

MR was used to treat leachate from LR. As shown in Figs. 2a and 2c, the pH of effluent from MR was above 8.0 throughout the experiment, even higher than 9.0 in the first 29 days. The COD of leachate was dramatically reduced by MR. The COD concentration in effluent was even below 200 mg L^{-1} during the first 43 days. However, it gradually increased over time against the decrease of COD in leachate. The ammonia in effluent gradually increased from 2.11 mg N L^{-1} to 491.1 mg N L^{-1} during the first 162 days, then decreased slightly. At the end of the first stage, the level of COD and ammonia was 477.9 mg L^{-1} and 411.1 mg L^{-1}, respectively.

Production of CH_4 from MR in the First Stage

The landfill gas produced in MR was collected into a vacuum bag to determine CH_4 concentration. The initial CH_4 concentration was 62%, and increased to 80% on day 24, then decreased slowly. The variations in CH_4 were correlated with COD in the leachate recycled into MR. MR stopped producing CH_4 from day 151 anwards, when the loading rate of COD in MR was less than 0.054 g d^{-1} L^{-1}.

Characteristics of Leachate in the Second Stage

The characteristics of leachate in the second stage are shown in Figs. 2b and 2d. Compared with the situation at the end of the first stage, the COD and ammonia in leachate increased slightly at beginning of the second stage, whereas the pH decreased slightly. As the second stage operation went on, the

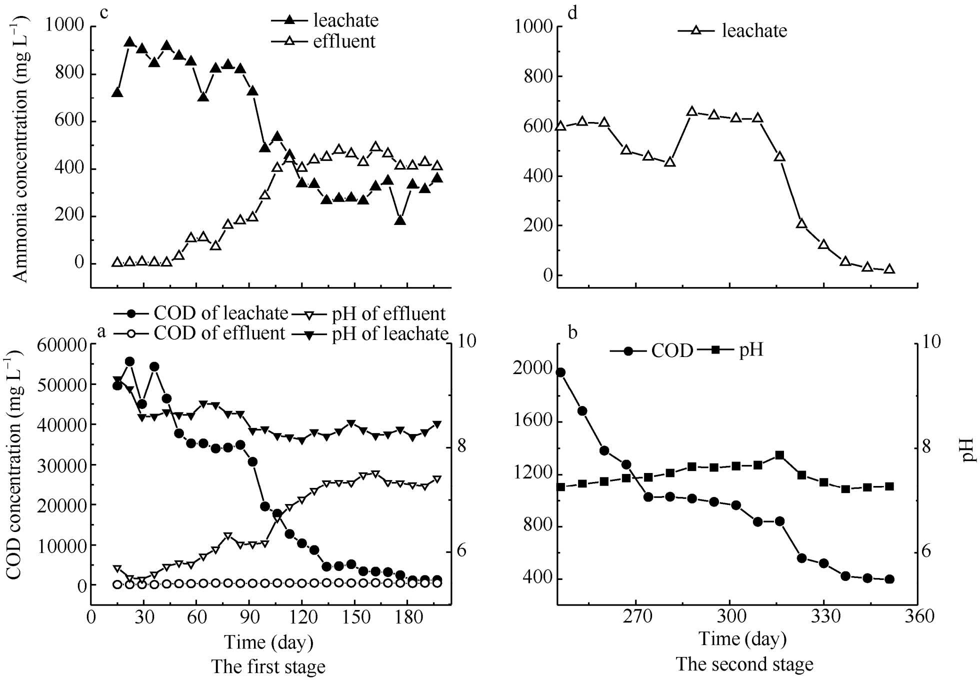

FIG. 2. Changes of COD, Ammonia and pH in effluent and leachate. A and c represent the first stage, b and d represent the second stage.

COD in leachate reduced quickly, whereas the ammonia increased slightly and then decreased rapidly. Additionally, the pH value also followed an increasing-decrease curve. At the end of experiment, the level of ammonia, COD, and inorganic COD was 20.6 mg N l^{-1}, 399.2mg L^{-1}, and 284.6 mg L^{-1}, respectively.

Attenuation of COD in the Hybrid Landfill

The COD attenuation rate was defined as follows:

$$\frac{dC}{dt} = -kt$$

where *C* is the COD concentration in leachate at different time points, *t* is the time, *k* is the attenuation rate. Consequently, the attenuation of COD could be formulated as:

$$C_t = C_0 e^{-kt}$$

where C_t represents the COD concentration at a certain time point, C_0 represents the COD concentration at beginning, *t* represents the time, *k* represents the constant attenuation rate.

The attenuation of COD in the first 85 days displayed a trend which was different from that from day 85 on wards in the first stage. The data were fitted into the formulation and three equations were obtained. The results are listed in Table 1 (R^2 means goodness of fit, *P* denotes probability). It was apparent that the COD decreased at a slowest speed during the first 85 days. Although air was introduced into LR in the second stage, the attenuation of COD was still slower than that from day 85 to day 200 in the first stage.

TABLE 1

Attenuation Model of COD over Time[a]

Item	Equation	R^2	*P*
Before Day 85 in the First Stage	COD (mg L^{-1}) = $59511e^{-0.007t}$	0.814	0.000
After Day 85 in the First Stage	COD (mg L^{-1}) = $22372e^{-0.028(t-85)}$	0.953	0.000
The Second Stage	COD (mg L^{-1}) = $1855.0e^{-0.014(t-245)}$	0.946	0.000

Note. [a] Unit: d as time.

Characteristics of the Refuse from LR and MR in the First Stage

To further investigate the performance of refuse degradation in the hybrid bioreactor landfill, pH, BDM, and CEC in the refuse were monitored. The results are illustrated in Figs. 3a and 3c. The time evolution of BDM in the refuse generally followed a decreasing curve except for a spine on day 100. The pH of refuse varied with BDM. During the first 30 days, the pH declined with the decrease of BDM, then increased in general but decreased slightly on day 100 when BDM increased at a certain degree.

Noticeably, the pH of refuse was always above 7.0. Generally, the CEC in refuse increased gradually in the first stage. However, the increasing rate before day 100 was obviously slower than after day 100. Furthermore, the CEC changed little from day 60 to day 100 when BDM increased at a certain extent.

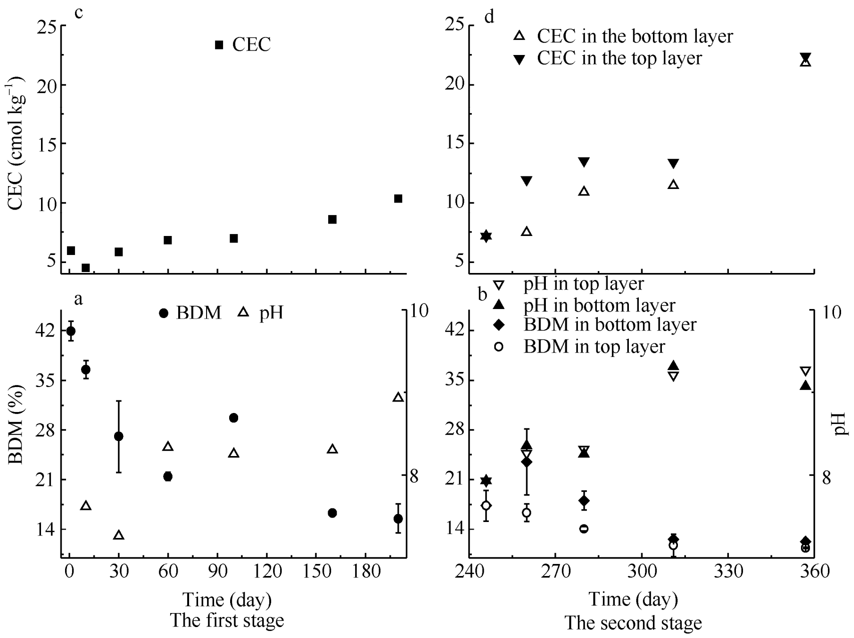

FIG. 3. Profiles of pH, BDM, and CEC in refuse. A and c represent the first stage, b and d represent the second stage.

The BDM of aged refuse changed little in MR, fluctuating between 10.8% and 14.2%. The CEC increased slightly from 9.62 cmol kg^{-1} to 15.65 cmol kg^{-1}. The pH value decreased slightly from the initial 9.47 to the terminal 8.98.

Characteristics of the Refuse from LR in the Second Stage

The refuse at the top and bottom layers was analyzed in the second stage. The results are demonstrated in Figs. 3b and 3d. Similar to the COD of leachate, the BDM of refuse increased, whereas the pH and CEC decreased at beginning of the second stage. The BDM reduced significantly with the operation of LR, and the decomposition rate of refuse was faster at the top layer than at the bottom layer. The BDM of refuse at the top layer quickly decreased from 17.4% to 11.4%, whereas that at the bottom layer reduced gradually from 17.4% to 12.3%. The pH value at both top and bottom layers increased slightly. The CEC enhanced significantly in the second stage. The BDM at the top and bottom layers increased from 7.18 cmol kg^{-1} to 22.42 cmol kg^{-1} and 21.81 cmol kg^{-1}, respectively.

Degradation of the Refuse and Relation between BDM and CEC in the Hybrid Landfill

Although the BDM content fluctuated over the time, it declined in general with the operation of landfill. According to previous works, the variation of BDM with placement time could be expressed with the following equation:

$$\text{BDM (\%)} = -k\text{Ln}(t) + C_0$$

where C_0 represents initial biodegradable organic matter, k is the product of degradation rate and time. The data were fitted into the formulation and the results are listed in Table 2 (R^2 means goodness of fit, *P* denotes probability). The decrease of BDM in the first stage and at the top layer in the second stage fits well the equation. Furthermore, the degradation rate of refuse was faster in the first stage than in the second stage. Although the decrease of BDM at the bottom layer of refuse did not fit the equation well, it was evident that the refuse was stabilized slower than at the top layer. The CEC increased with the decrease of BDM. The correlation between CEC and BDM was significant (Pearson correlation coefficient = -0.686, *P* = 0.003, 2-tailed test).

TABLE 2

Regression Equation of BDM over Time[a]

Item	Equation	R^2	*P*
The First Stage	BDM (%) = -4.29Ln(t) +44.04	0.819	0.005
Top Layer in the Second Stage	BDM (%) = -1.31Ln(t-245) +18.15	0.827	0.032
Bottom Layer in the Second Stage	BDM (%) = -1.17Ln(t-245) + 20.30	0.221	0.424

Note: [a]Unit: d as time.

DISCUSSION

Degradation of Refuse and COD Attenuation of Leachate in the Hybrid Bioreactor Landfill

Commonly, the bio-degradable process of refuse undergoes three basic phases[7]. At the first phase, the organic matter in solid phase (COD_S) decays fast, namely the large molecular organic matter degrades into small molecular organic matter. At the second phase, the small molecular organic matter in solid phase dissolves into the liquid phase (COD_L) and subsequently hydrolyzes into the smaller molecular organic matter in the liquid. At the third phase, the smaller molecular organic matter is transformed into gas (CO_2/CH_4) and biomass. At the first and second phases, the biodegradation cannot be observed in the leachate, but the BDM of refuse reduces quickly. Furthermore, the COD of leachate would accumulate due to the imbalance between the fast-dissolving refuse into leachate and the slow-degradating COD in leachate (or slow washout from the refuse). In the experiment, the COD of leachate was mainly degradated in MR. Thus, the fact that the BDM of refuse decreased sharply, whereas the COD of leachate increased slightly during the first 30 days is mainly attributed to the slow washout of COD_L from the refuse.

Over a refuse column, balance of leachate COD could be stated as follows[12]:

$$\text{COD out} = \text{COD}_{\text{solid dissolved into liquid}} + \text{COD in} - \text{COD consumed}$$

where the consumed COD includes the organic carbon assimilated by microbes, transformed into gas phase and lost in sampling. The COD attenuations of leachate underwent two processes, and the degradation rate changed significantly in the first stage, which might be explained by the equation. The composition of refuse is complicated, some parts of them are readily biodegradable, moderately biodegradable, and hard biodegradable while the other part cannot be degraded by microorganisms. During the degradation of refuse, the readily degradable organic matter is first consumed, then the moderately and hard degradable organic ones are sequentially hydrolyzed. The BDM of refuse decreased rapidly in the first 60 days, the COD of leachate reduced dramatically from day 36 to day 57 when the readily degradable organic matter was quickly degraded. In this period, the COD consumed by microbial assimilation and transformed into CO_2 was little, and could be negligible according to the well established principle which stated pH<6.0 in leachate, was an effective inhibitor of methanogenesis[13]. Meanwhile, the input of COD was also negligible because leachate was treated by MR and the recirculation effluent contained little organic matter, suggesting that the attenuation of COD in leachate depends on the interactions of refuse dissolution and recirculation dilution. However, the biodegradation process is complicated, with the large molecular BDM degraded first into small molecular BDM, thus possibly increasing the total BDM quantity until the point where both larger and small molecular BDM are degraded[14]. The BDM of refuse rebounded from day 60 to day 100, whereas the COD of leachate decreased at a relatively slower speed from day 60 to day 85, indicating that the moderately biodegradable organic matter began to hydrolyze. Compared to the readily biodegradable organic matter, the moderately biodegradable organic matter is decomposed slowly. In addition, leachate recirculation from MR into LR may provide inoculums and is accelerated to establish a microbial population with the proper balance between acidogenic and methanogenic organisms[15]. The COD consumption and dilution by recirculation play a primary role in the COD attenuation of leachate. That is why the COD attenuation rate was faster after day 85 than before day 85 in our study.

Compared with the situation at end of the first stage, the COD and ammonia of leachate and the BDM of refuse increased slightly at beginning of the second stage, which was probably due to the pause of leachate recirculation, leading to the decreased removal from day 201 to day 245.

In this study, the readily and moderately biodegradable organic matters were consumed in the first stage, the hard degradable organic matter was sequentially degraded in the second stage. Thus, although the air was introduced into LR in the second stage, the BDM decreased slower than in the first stage. The air addition can improve the stabilization of old refuse[9], which may explain why the BDM decreased faster at the top layer than at the bottom layer in the second stage in our study. Introduction of air can produce a significant effect on the quality of leachate[16]. The COD and ammonia of leachate decreased quickly in the second stage. However, the inorganic and refractory organic matters were gradually accumulated in leachate with operation of the landfill. Hence, the COD attenuation slowed down in the second stage. At the end of experiment, the ammonia concentration in leachate declined to 20.6 mg N L^{-1}, which is below the second level of the current discharge standards in China (NH_4^+-N< 25 mg N L^{-1}, GB16889-1997, State Environmental Protection Agency of PRC, 1997). The total COD concentration in leachate was 399.2 mg L^{-1}, which is below the third level of the current discharge standards (COD < 500 mg L^{-1}). Furthermore, about 284.6 mg L^{-1} of COD was contributed by the inorganic COD.

Effect of MR

The effect of leachate recirculation is maximized when a landfill reaches its stable phase[16]. The aged refuse contains a wide spectrum and a large number of microorganisms, and hence have a strong ability to decompose the refractory organic matter in some wastewater[13]. When the leachate is treated within a bioreactor landfill, not only biodegradation but also such processes as adsorption, ion change, and mechanical filtration proceed to significantly decrease the organic strength[1]. MR is highly-effective in reducing the organic matter in leachate. Some parts of the organic matter can be used to maintain the growth of microorganisms and the others are mostly transformed into CH_4. In this study, MR stopped producing CH_4 from day 151, implying that the COD of leachate is insufficient to meet the requirments of microbial growth and CH_4 production. The ammonia was also reduced effectively in the initial period of MR. However, the removal efficiency decreased gradually over time and the ammonia level was even higher in the effluent than in the leachate from day 120, which did not occur in the investigation[17] that utilized UASB as MR to reduce COD, but occurred in the study used well-decomposed waste as a bulking agent to remove COD[12], indicating that delayed ammonia in effluent mainly accounts for “trapped” (e.g. adsorption) by and “release” (e.g. desorption, washing out) from the aged refuse[18].

Mineralization and Humidification of the Refuse in the Hybrid Bioreactor Landfill

Changes in C/N ratio and CEC over time have been proved to be reliable indicators of the progress of composting process for establishing biological stability and compost maturity[19]. Increased CEC during composting can be attributed to the accumulation of materials bearing the negative charge, such as lignin-derived products[20], and increased carboxyl and/or phenolic hydroxyl groups might have contributed to the higher values of CEC in the composts. CEC also increased gradually with refuse degradation in our experiment, which is in accord with previous findings[21]. Furthermore, CEC was negatively correlated with BDM. CEC was gradually increased as BDM was sharply decreased in the first 60 days. In addition, CEC increased faster from day 100 to day 200 than in the first 60 days, in the second stage than in the first stage, at the top layer than at the bottom layer, suggesting that mineralization of labile organic compounds mainly occurs during the first 60 days when the readily biodegradable organic matter is reduced, whereas humidification prevailes over mineralization during the degradation of hard biodegradable organic matter. Introduction of air can improve the stabilization of refuse in old landfills[9], which can explain why CEC increased faster at the top layer than at the bottom layer in our study. The aged refuse of MR increased gradually while BDM only fluctuated in a narrow range, which is another evidence of humidification prevailing over mineralization during the maturity phase.

In conclusion, refuse is relatively stable through the hybrid bioreactor landfill operation. Introduction of air can improve the stabilization of refuse. Most of readily biodegradable organic matters are mineralized during the initial phase of refuse degradation, whereas the hard biodegradable organic matters are humidified mainly during the maturity phase.

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