



Comparison of leachate treatments in the simulated landfill bioreactors with different operation modes

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ABSTRACT

In this study, the leachate treatments were compared in two simulated landfill bioreactors with different operation modes. In one reactor, the leachate was circulated between a landfill and a methanogenic reactor, while the other reactor was operated using direct recirculation of the leachate. The pH, Chemical Oxygen Demand (COD), volatile fatty acids (VFA), NH_4^+ -N of leachate, and the biologically degradable material (BDM), the enzymatic activities of municipal solid waste (MSW) were analyzed to achieve the essential characterization of the landfill. The results revealed that the loss of organic materials from the landfill occurred in an active methanogenic environment in the later period, while the environment was acidic due to a high concentration of VFA and contained a large volume of BDM during the early stage. In addition, the dehydrogenase and polyphenol oxidase activities of refuse were majority higher in the bioreactor landfill that was connected to a methanogenic reactor. Furthermore, the efficiency of leachate treatment was enhance at least 20%, and the stabilization process was accelerated over 50% in the landfill that was operated in conjunction with the methanogenic reactor when compared to the landfill in which there was direct leachate recirculation.

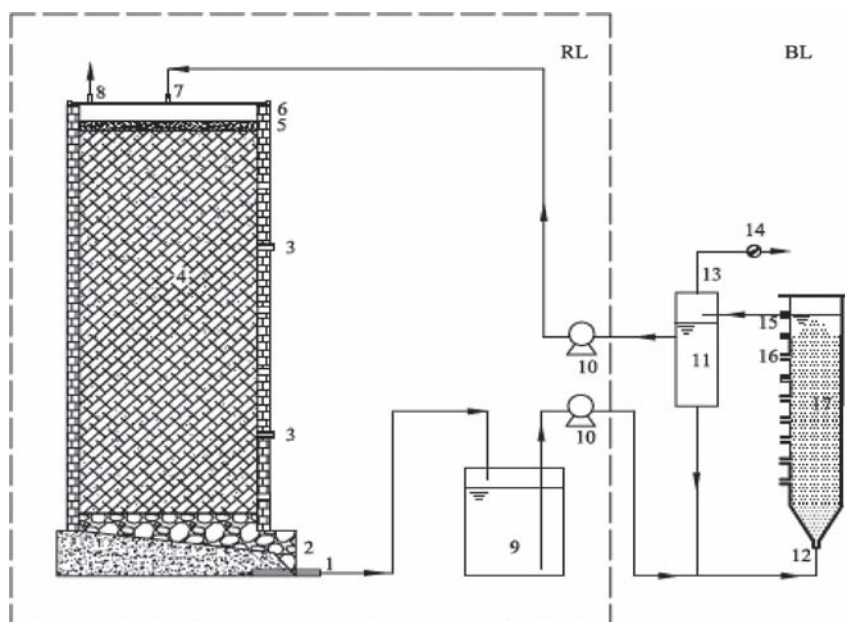
Keywords: Simulated landfill bioreactor; Leachate treatment; Leachate recirculation; Municipal solid waste; Methanogenic reactor

1. Introduction

Sanitary landfill is the primary method for the disposal of municipal solid waste (MSW). In 2006, approximately 148.4 million tons of MSW was generated in China and that 70% of this material was disposed by burial in landfills [1]. Landfills pass through typical phases soon after waste is deposited [2]. First, the landfill becomes anaerobic due to the depletion of oxygen, during which organic

compounds are hydrolysed and fermented to primarily volatile fatty acids (VFA). In the second phase, methanogens begin to proliferate, and acetogenic bacteria convert the VFA to acetic acid, hydrogen and carbon dioxide. In the final and longest phase, these biodegradation intermediates serve as substrates for the production of methane via methanogens. Throughout this process, organic materials are released by degradation of the waste, which gives rise to a high level of organic matter in the leachate. Liu and Li [3] reported that there were 77 kinds of organic matters in the leachate.

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(1) Leachate outlet (2) Gravel layer (3) MSW sampling port (4) Landfill site (5) Sandy layer (6) Headspace (7) Vent-port (8) Gas outlet (9) Leachate collection tank (10) Peristaltic pump (11) Separation gas from liquid (12) Leachate inlet (13) Gas outlet (14) Wet gas meter (15) Outlet (16) Sludge sampling port (17) Methanogenic reactor Recirculation Landfill (RL), the small frame part; Bioreactor Landfill (BL), the whole system

Fig. 1. Schematic diagram of the landfill bioreactor system.

Sanitary landfills that employ operational techniques such as leachate recirculation represent an economical and environmentally acceptable method for the disposal of MSW [4]. Leachate recirculation is capable of permanently reserving additional carbon in the landfill, which results in higher levels of methane generation as well as leachate treatment in situ. Liu *et al.* [5] reported that the leachate circulation regulated the water content in refuse, improved the microorganism circumstance and strengthened the biodegradation of organic materials. However, if the MSW contains a high proportion of easily digestible materials, the increased level of biodegradation associated with leachate recirculation can result in an imbalance in the growth rates of rapidly-growing acidogenic bacteria and slow-growing methanogens during the first phase of MSW decomposition. Such an imbalance can result in methanogenesis being delayed or inhibited [6].

Many studies have been conducted to evaluate the use of treated leachate recirculation to accelerate refuse decomposition [7–9]. The results have indicated that bioreactor landfills that circulate the leachate between the landfill and a methanogenic reactor take advantage of adapted microflora and the highly alkaline effluent of the methanogenic reactor to buffer the pH and inoculate the landfill. The addition of this effluent to the landfill can create optimal environmental and nutrient conditions for acidogenic bacteria and methanogens, thereby improv-

ing the overall performance of the system. He *et al.* [4] found that a combination of effective microorganisms and methanogenic reactors using treated leachate recirculation might be a good way to increase the degree of MSW stabilization. In addition, it was found that nitrate in recycled leachate had a negative effect on refuse decomposition [10, 11]. However, these studies mostly focused on the leachate characteristics to achieve the landfill stabilization process. Rare studies were conducted to evaluate the leachate treatments by combined analysis of leachate and refuse.

This study was conducted from the viewpoint that bioreactor landfills provide an advantage for the transformation of organic materials. Accordingly, the pH, Chemical Oxygen Demand (COD), VFA and $\text{NH}_4^+\text{-N}$ of leachate from simulated landfill bioreactors were analyzed. In addition, the biologically degradable material (BDM) and the enzymatic activities of MSW were analyzed to achieve an essential characterization of the landfill. Finally, the efficiencies of leachate treatments in landfill bioreactors operated with different modes were compared.

2. Materials and methods

2.1. Experimental set-up

Diagrams of the simulated landfill bioreactors used in this study are shown in Fig. 1. The bioreactor landfill (BL)

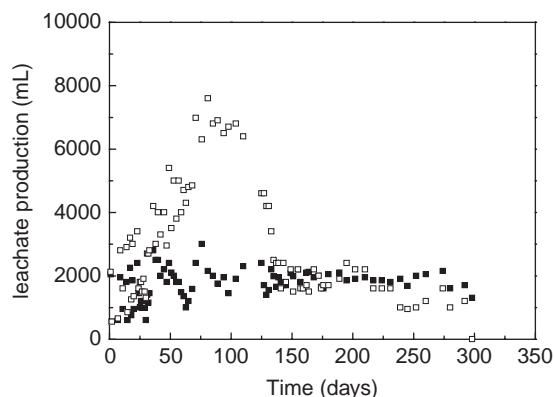


Fig. 2. Variation of leachate volume in RL & BL. RL (■); BL (□).

was comprised of a methanogenic reactor that received leachate from the landfill. In this system, the leachate was subjected to methanogenesis in the methanogenic reactor, after which it was recycled into the landfill. A leachate direct recirculation landfill (RL) was used as a control. All leachate was pumped by peristaltic pump within 8 h daily in sequence.

Both landfill bioreactors were constructed of brick-concrete and had the same effective size of 0.55 m × 0.55 m × 2.0 m (L × W × H). A plexiglass male adapter was installed at the bottom of each landfill bioreactor as a leachate drainage port. In addition, two such adapters were installed in the lid of each landfill bioreactor to enable leachate recirculation and gas collection. Furthermore, two MSW sampling ports were installed on one side of the landfill. The methanogenic reactor, which was constructed of plexiglass and had a working volume of 15 L, was seeded with 12 L of raw anaerobic sludge obtained from the Hangzhou Sibao sewage treatment plant. The sludge was incubated with synthetic water that had a COD of 3000–5000 mg/L for 10 days to activate it, after which it was acclimated to the leachate from the Hangzhou Tianziling landfill. The acclimated sludge in the methanogenic reactor had a total solids (TS) content of 90.5 g/L and a volatile solids (VS) content of 27.0 g/L.

Fresh refuse was collected from the Kaixuan transport station in Hangzhou. The physical composition of the refuse (by weight) was as follows: kitchen waste, 61.5%; plastics, 11.6%; paper, 10.3%; sand and soil, 7.1%; cellulose textile, 1.3%; glasses, 6.4%; metals, 0.6%; rubber, 0.6%; wood, 0.6%. Prior to adding the refuse to the landfill bioreactor, a 5 cm thick layer of gravel was placed at the bottom of the reactor to retain refuse and prevent small particles from leaching out. In addition, larger particles of the collected refuse were shredded

into 2 cm approximately. The refuse was then thoroughly mixed and loaded into the landfill bioreactors. The average wet density of the refuse compacted in the landfill bioreactors was 600 kg/m³. The moisture content of the refuse was adjusted to 75% by adding tap water to the MSW after loading the reactor. After the water was added, the MSW was covered with a 5 cm layer of sand so the leachate would be well-distributed when it was recycled. Finally, the bioreactors were sealed air-tight.

2.2. Sampling procedure

Leachate samples were collected from the landfill leachate drainage port and methanogenic reactor outlet (Fig. 1) daily to determine the pH. In addition, the leachate volume and the COD, VFA, NH₄⁺-N of the leachate samples were determined weekly. Furthermore, BDM and enzymatic activities of refuse samples collected periodically from the landfill were also determined. Both simulated landfill bioreactors were operated at room temperature for 300 days.

Leachate samples were collected from the sampling point using a glass tank. The leachate samples were then immediately transferred to brown glass bottles and analyzed. Refuse samples were further cut and ground in several steps, after which they were freeze dried and stored at –20° until analysis. All analyses of parameters of the leachate and refuse were conducted in triplicate to ensure the validity of the results.

2.3. Analytical methods

The pH, COD and NH₄⁺-N in the leachate were determined using the standard methods [12]. VFA was analysed using the acidified ethylene glycol colorimetric method [13]. The BDM of the MSW was analysed using the potassium dichromate method [14]. The MSW enzymatic activities of catalase, dehydrogenase and polyphenol oxidase were analysed following the method of Guan [15].

3. Results and discussion

3.1. Characteristics of leachate during waste decomposition

Leachate volume kept about 2000 mL/d in the RL during the experiment. In contrast, leachate volume increased sharply to 7600 mL/d with waste decomposition in the first 80 days, then decreased to 2000 mL/d on day 135 and kept this low level till the end of the experiment in the BL (Fig. 2). This showed that a rapid biodegradation of organic materials occurred in the BL with the

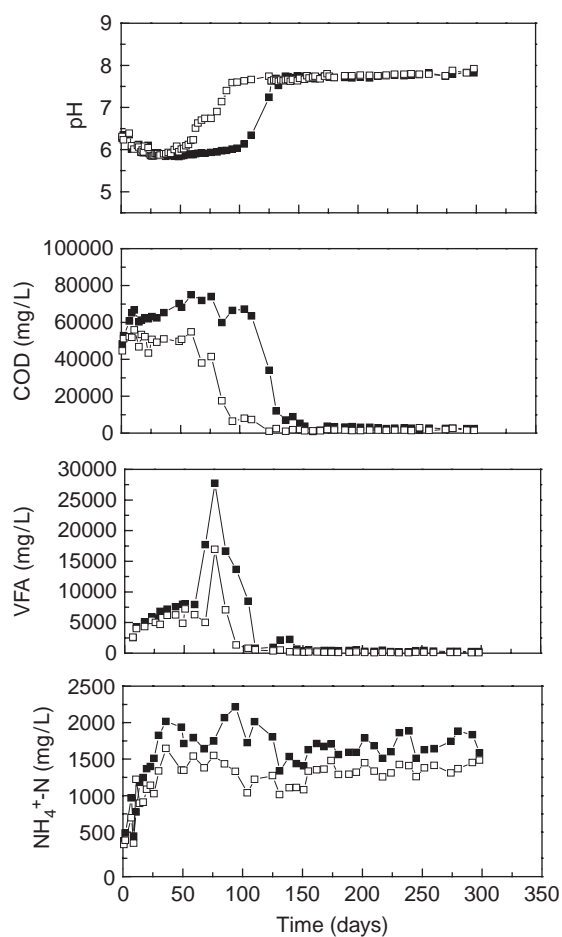


Fig. 3. Variation of COD, VFA, $\text{NH}_4^+\text{-N}$ concentrations and pH values of leachate from RL & BL, RL (■); BL (□).

combined effects of the methanogenic reactor and the landfill reactor.

The leachate characteristics are known to mirror the biodegradation of the organic refuse and the process of landfill stabilization [16]. Changes in the pH, COD, VFA and $\text{NH}_4^+\text{-N}$ of the leachate from the two bioreactors over time are shown in Fig. 3. The pH of the leachate from the RL and BL increased from acidic to approximately neutral after 120 and 85 days, respectively. In addition, the COD concentrations of the leachate from the RL were higher than those from the BL during the early experimental period, and they stabilized at approximately 2500 mg/L after 150 and 120 days respectively. These findings suggested that circulating the leachate between a landfill and a methanogenic reactor may accelerate waste stabilization. A similar phenomenon was observed when the VFA was evaluated. Specifically, the VFA concentrations of the leachate from the RL and BL increased to 27720 mg/L and 16940 mg/L after 75 days, respectively. These levels resulted in the

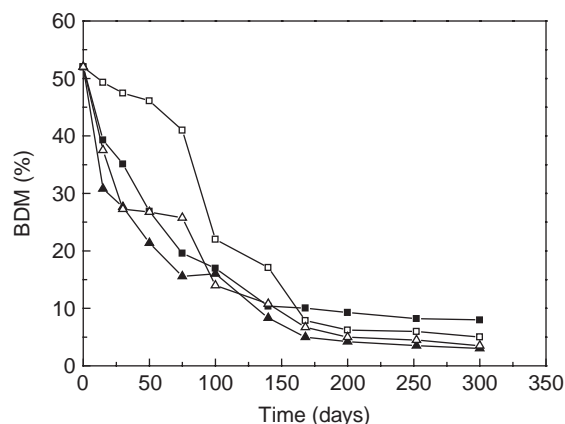


Fig. 4. Variation of BDM of refuse from the upper and lower layers of RL & BL. RL upper (■); RL lower (□); BL upper (▲); BL lower (△).

pH of the leachate from the RL and BL being 5.9 and 6.8, respectively. After 140 day, the VFA concentration had decreased to less than 200 mg/L in the leachate from the BL, indicating that the BL was completely methanogenic at that time [17]. However, the concentration of VFA was still fluctuating between 200 and 450 mg/L in the RL at 140 days. The $\text{NH}_4^+\text{-N}$ concentration of leachate from the RL increased to above 2000 mg/L on day 36 and maintained a high level till the end of the experiment, while the $\text{NH}_4^+\text{-N}$ concentration of leachate from the BL were lower during the whole experiment. Taken together, these results indicate that the pH values of the leachate from both bioreactors approached neutral and the COD concentrations were maintained at a low level for a long time during the latter period. These findings suggest that the two bioreactors had stabilized, but the environment in the BL was more stable than the environment in the RL.

3.2. BDM and enzymatic activities of MSW in the bioreactors

Of the parameters evaluated in this study, the BDM best reflects the degree of biodegradation of the MSW [18] Fig. 4 shows the variation in the BDM of the MSW from the upper and lower layers of both bioreactors. The degradation rate of MSW from the upper layer of the BL was higher than that of MSW from the upper layer of the RL. Indeed, by the time the COD concentrations of the leachate had risen to their highest value, the BDM value of the upper layer refuse had decreased from 52.0% to 19.6% and 15.6% in the RL and BL, respectively. This finding may indicate that the environment of the upper layer refuse was more suitable for degradation by the predominant microbes in the BL than by those in the RL. The BDM

in the lower layer of refuse in the RL was much higher than that of the lower layer of refuse in the BL during the early period. This finding may be ascribed to re-adsorption of the organic substances by the lower layer refuse when the raw leachate was recycled in the RL. After 175 days, the BDM was maintained at a low level, which suggests that both of these bioreactors had entered into the stabilized phase. However, the BDM of refuse from the BL was lower than that of the RL during the later period, which indicates that the BL had a better degradation environment and stabilized earlier than the RL.

In the biodegradation system, organic materials have an influence on the microbiota, and the enzyme activity is one way of describing the general condition of the environment [19, 20]. The MSW enzymatic activities of catalase, dehydrogenase and polyphenol oxidase are shown in Fig. 5. Catalase is an oxidoreductase associated with microbial activity [21]. Dehydrogenase activity is a useful indicator of anaerobic microbial activity [22]. Polyphenol oxidase is responsible for the transformation of aromatic compounds [23]. Dehydrogenase activity increased sharply (500%) when the landfills were acidic

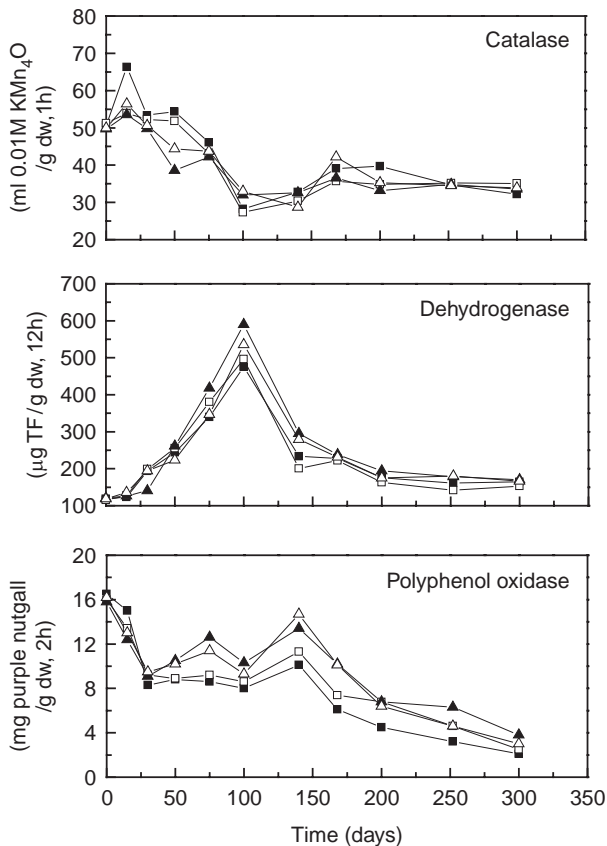


Fig. 5. Variation of enzyme activities of refuse from the upper and lower layers of RL & BL. RL upper (■); RL lower (□); BL upper (▲); BL lower (△)

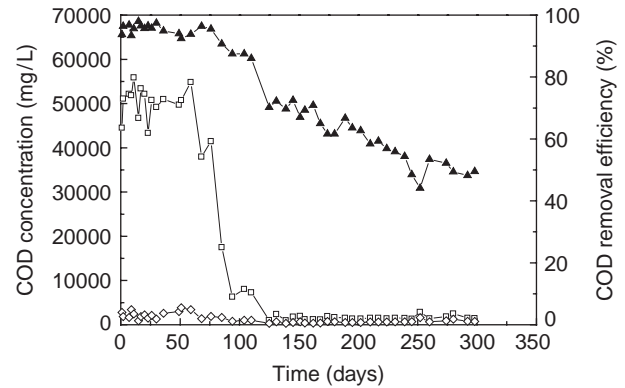


Fig. 6. COD concentrations and removal efficiencies in the methanogenic reactor. Influent (□); Effluent (○); Removal efficiency (▲).

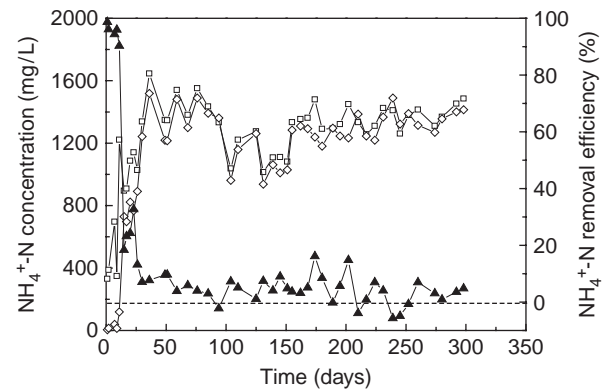


Fig. 7. $\text{NH}_4^+\text{-N}$ concentrations and removal efficiencies in the methanogenic reactor. Influent (□); Effluent (○); Removal efficiency (▲).

on day 100. This may indicate that the degradation of organic materials was enhanced by anaerobic microbe when the landfill turned to acidic. Conversely, catalase activity decreased during the experiment. This may indicate that some aerobic microbe died when the landfill became anaerobic gradually. Polyphenol oxidase activity was fluctuated in the early days, and then decreased till the end of the experiment. This likely occurred due to the depletion of oxygen and continuous degradation of the aromatic compounds. In addition, dehydrogenase and polyphenol oxidase activities of refuse from BL were higher than those from RL, which indicates that the BL had a higher biodegradation activity and was more suitable for the organic refuse decomposition than the RL.

3.3. Comparison of leachate treatments in the bioreactors

As mentioned above, the waste was degraded more rapidly in the BL than in the RL. This may have occurred

Table 1
Comparison of leachate treatments in the bioreactors.

Bioreactor	Time of the parameters of leachate becoming stable (day)			Settlement of the MSW		
	pH	COD	VFA	Initial height (cm)	Final height (cm)	Settlement (%)
RL	120	150	200	185	163	11.9
BL	85	120	140	185	140	24.3

due to the combined effects of the methanogenic reactor and the landfill reactor. Throughout the entire experiment, there were no detectable VFA in the effluent of the methanogenic reactor, and the pH values were all approximately neutral.

Fig. 6 shows the COD concentrations and COD removal efficiencies in the methanogenic reactor. The COD removal efficiencies were maintained at great than 90% until day 95. However, the efficiency declined as the influent COD concentrations decreased, with the efficiency being approximately 50% at the end of the experiment. This likely occurred due to the low biodegradability of organic materials in the leachate from the old landfill [24].

As showed in Fig. 7, the influent $\text{NH}_4^+\text{-N}$ concentration rose from 329 to 1645 mg/L in the methanogenic reactor in the first 36 days, due to the degradation of organic nitrogenous compounds in the landfill reactor. The $\text{NH}_4^+\text{-N}$ removal efficiency dropped from 98% to less than 10% as the $\text{NH}_4^+\text{-N}$ loading rate increased greatly in the methanogenic reactor. Only small amounts of $\text{NH}_4^+\text{-N}$ were removed under the anaerobic condition owing to the utilization of $\text{NH}_4^+\text{-N}$ through assimilation of anaerobic bacteria for cellular growth [25]. In the later period, no significant $\text{NH}_4^+\text{-N}$ removal was observed in the methanogenic reactor. On the contrary, the effluent $\text{NH}_4^+\text{-N}$ concentrations sometimes exceeded the corresponding influent $\text{NH}_4^+\text{-N}$ concentrations, due to the ammonification of organic nitrogenous compounds under anaerobic condition.

The results presented above demonstrate that the organic matter had already degraded to some extent when the leachate was fed into the methanogenic reactor. Once in the reactor, the methanogens produced enough alkalinity to buffer the acidic conditions caused by VFA in the landfill. Conversely, the refuse in the RL stabilized slowly due to the low pH value and high VFA concentration in the circulating leachate, which may have inhibited methanogenesis. Taken together, these findings indicate that the landfill surrounding was more suitable for the degradation of organic substances in the BL, which resulted in an increase in the biodegradability of MSW and a high degree of waste stabilization.

As showed in Table 1, the time of the pH values COD and VFA concentrations of the leachate from the BL becoming stable were all earlier than those of the RL, which indicates that the efficiency of leachate treatment was enhanced by at least 20% in the BL when compared to the RL. In addition, the corresponding cumulative settlements in the RL and BL were 11.9 and 24.3% of the initial refuse height at the end of the study (Table 1). This indicates that the stabilization process of the landfill was accelerated over 50% by circulating the leachate between a landfill and a methanogenic reactor.

4. Conclusions

The results of the analysis of the characteristics of leachate and MSW from two simulated landfill bioreactors indicate that the combined effects of the methanogenic reactor and the landfill reactor led to an increase in the biodegradability of MSW and a high degree of waste stabilization. The loss of organic materials from the landfill was much higher in an active methanogenic environment than in an acidic environment with a high VFA concentration, BDM and low enzymatic activities. In addition, leachate treatment efficiency was enhanced by at least 20% in the bioreactor landfill that was connected to a methanogenic reactor. Finally, the stabilization process was accelerated over 50% in the landfill with the methanogenic reactor than in the landfill with direct leachate recirculation. MSW is disposed of to landfills every day. Thus, there may be a constant discharge of leachate from MSW landfills to the surrounding environment. Additional methods on enhancing organic materials degradation in anaerobic environments and improving efficiency of leachate treatment should be further investigated.

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