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Biological degradation of MSW in a methanogenic reactor using treated leachate recirculation

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Abstract

With a methanogenic reactor using treated leachate recirculation, the effects of 12 effective microorganisms (EMs), isolated from Hangzhou Tianzhiling landfill, on the degradation of municipal solid waste (MSW) were investigated. The preliminary experiment indicated that the EMs increased the biodegradability of MSW, enhanced 24% of organic mass effluent from the landfill reactor, and shortened methane production period to about 91 days in the bioreactor landfill system. The total gas production volumes for the landfill only with leachate recirculation, the bioreactor landfill system with and without EMs inoculation were 65.7, 620.9 and 518.6 l, respectively, after 105 days operation. The average methane concentration of the gas formed in the bioreactor landfill system was above 70%. These showed that a combination of EMs and methanogenic reactors using treated leachate recirculation might be a good way to increase the degree of MSW stabilization, and enhance the rate and quality of gas production for energy recovery.

Keywords: MSW; Bioreactor landfill; Leachate recirculation; Methanogenic reactor; EMs

1. Introduction

Sanitary landfills represent a common, economical and environmentally acceptable method for the disposal of municipal solid waste (MSW). In 2000, approximately 118 million tones of MSW was generated in China and that only 41.5% of this material was disposed, mostly by burial in landfills [1]. During the stabilization of landfilled waste, most organic materials are broken down into simpler compounds by aerobic and anaerobic microorganisms, leading to the formation of landfill gas and leachate. The rate and characteristics of leachate and landfill gas generated from a landfill vary from one phase to another. If the methane fermentation phase does become established, biodegradable organic matter is consumed mainly by methane-forming consortia (methanogenic archaea) and converted into methane and carbon dioxide, while little released into leachate. Under a stabilized methanogenic condition, which is the stage of interest from a beneficial recovery perspective, landfill gas is composed of approximately 55–60% methane and 40–45% carbon dioxide with trace amounts of other gases [2]. Although quantities of methane high enough for commercial production are generally formed for only 5–20 years, residual production can last for more than 40 years [3]. And landfills are extremely heterogeneous microenvironments. Different areas of a landfill, even in close proximity, can be at different stages in the decomposition process concurrently [4]. Projects to recover landfill gas are frequently rejected due to the unpredictable process of gas production and low volume of landfill gas.

Some of the operational techniques identified as increasing the rate and quality of landfill gas production were elevated moisture content, leachate recirculation, shredded MSW, pH control/buffer addition, nutrient addition, anaerobic sewage sludge inoculation and elevation of the operating temperature [5,6]. Of these, leachate recirculation is a more efficient method of increased levels of biodegradation to

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obtain higher methane yields and lower treatment costs [2,4,7,8]. By keeping leachate in the landfill, this process would permanently sequester additional carbon, which results in higher levels of methane generation as well as leachate treatment in situ. However, if the MSW contains a high proportion of easily digestible materials, the increased level of biodegradation associated with leachate recirculation can result in the imbalance in the growth rates of fast-growing acidogenic bacteria and slow-growing methanogens in the first phase of MSW decomposition [9]. As a result, methanogenesis may be delayed, prevented, or inhibited [7].

Chynoweth et al. [10] and Yu et al. [11] reported that circulating leachate between a landfill and a methanogenic reactor (mature landfill) could take advantage of adapted microflora and high alkalinity of effluent in the methanogenic reactor to buffer pH and inoculate the landfill, which provided optimal environmental and nutrient conditions for acidogenic bacteria and methanogens, and improved the performance of landfill system. Many efforts have been focused on increasing the populations of microorganisms associated with refuse decomposition with a seed of well-decomposed refuse and digested sludge [12,13] that served to accelerate refuse decomposition and landfill gas production.

For most organic waste such as cellulose, hemicellulose and lignin, the hydrolysis is the rate-limiting step of the anaerobic digestion that prevails in landfills [14]. However, the hydrolytic bacterial and archaeal populations associated with refuse decomposition are lower [15,16]. The biochemical transformation or mineralization of landfilled refuse even readily degradable materials sometimes persists for surprisingly long time in landfills [17]. The addition of specialized microorganisms to degrade subsurface pollutants has proved to be successful [17,18]. But little attempt has ever been made to inoculate effective microorganisms (EMs) into landfills to increase biodegradability of landfilled waste.

The study was aimed at inoculating EMs into a simulated landfill reactor to increase the anaerobic biodegradability of MSW, and enhance the rate and quality of gas production for energy recovery. As compared with the landfill only with leachate recirculation, organic mass effluent from landfill reactor, the performance of methanogenic reactor, gas production and MSW stabilization were investigated to characterize the bioreactor landfill system with a methanogenic reactor using treated leachate recirculation.

2. Materials and methods

2.1. MSW Composition

MSW was mixed by 10 different components shredded into 2–4 cm pieces in the experiment. The physical composition of the synthetic MSW mixture, according to the investigation made in the city of Ningbo, was as follows (by weight): vegetables, 45.0%; fish, 2.5%; meat, 1.0%; fruit, 9.0%; paper, 7.4%; plastics and leather rubber, 11.9%; cellulose textile, 3.6%; brick sand and soil, 8.5%; metals and glasses, 7.6%; woods, 3.5%. The moisture content of the mixture was about 54% (w/w).

2.2. EMs inoculum

By means of the main component of MSW as sole source of carbon, the experimental microorganisms isolated from Hangzhou Tianzhiling landfill according to normal microbiological procedures [19]. Twelve culture media were used for microorganisms isolation. (a) Four media for bacteria isolation contained (g/l): the same inorganic nutrition: $(NH_4)_2SO_4$, 0.2; K_2HPO_4, 1; $MgSO_4{\cdot}7H_2O$, 0.2; and different carbon substrate: carboxymethyl cellulose (CMC), raw starch, olive oil or casein, 1, respectively. The medium pH was adjusted to 7.0–7.2. (b) Four media for actinomycetes isolation contained (g/l): the same inorganic nutrition: KNO_3, 1; K_2HPO_4, 0.5; NaCl, 0.5; $MgSO_4{\cdot}7H_2O$, 0.5; $FeSO_4$, 0.01; and different carbon substrate: CMC, raw starch, olive oil or casein, 1, respectively. The medium pH was adjusted to 7.0–7.2. (c) Four media for fungi isolation contained (g/l): the same inorganic nutrition: $NaNO_3$, 3; K_2HPO_4, 1; KCl, 0.5; $MgSO_4{\cdot}7H_2O$, 0.5; $FeSO_4$, 0.01; and different carbon substrate: CMC, raw starch, olive oil or citrus pectin, 1, respectively. The refuse samples were inoculated into the culture media at 30 °C for 5–7 days. Then 1.0 ml of this liquid culture was spread onto the different solid media using the dilution plate method, and incubated at 30 °C for 5–7days. The separated colonies were serially picked up and inoculated on the new plates repeatedly until pure isolates were obtained.

Twelve EMs with high degrading activities were selected from representative microorganisms derived from different media, and identified according to the descriptions in the *Bergey's Manual of Determinative Bacteriology*, eighth ed., 1974; *Microbial Taxonomy* [20] and *Manual of Determinative Common Bacteria* [21]. They were as follows: cellulose-degrading bacterium and olive oil-degrading bacterium as *Cellulomonas* sp., casein-degrading bacterium from casein-bacteria media as *Brevundimonas diminuta*, casein-degrading bacterium from casein-actinomycete media as *Bacillus anthracis,* starch-degrading bacterium as *Bacillus megaterium*, three actinomycetes (cellulose-degrading actinomycete, starch-degrading actinomycete and olive oil-degrading actinomycete) as *Streptomyces* sp. and four fungi (olive oil-degrading fungi, cellulose-degrading fungi, starch-degrading fungi and pectin-degrading fungi) as *Mucor* sp.

Sets of 250 ml of sterile flasks, containing the culture media used for isolate and the EMs, were incubated in an orbital incubator (120 rpm) at 30 °C for 7 days. The cultures were then centrifuged at 3000 rpm for 30 min, and the resulting pellet was resuspended to 50 ml with sterile distilled water in 100 ml of sterile flasks. The concentration of each suspension approximates 10^8 to 10^{10}cells/ml, determined by the most probable number (MPN) enumera-

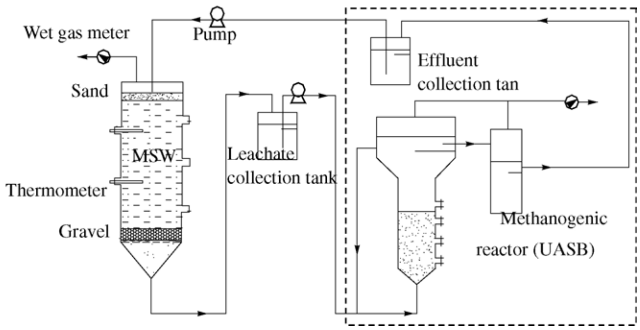

Fig. 1. Schematic diagram of the bioreactor landfill system in the experiment.

tion [19]. Then these suspensions were mixed and used as inoculum in the following experiment.

2.3. Experimental set-up

The experimental set-up used in this research is illustrated in Fig. 1. The simulated landfill reactor consisted of a 42-l cylinder made of PVC (28.7 cm i.d., 65 cm height). A polyethylene male adapter (about 0.8 cm) was installed at the bottom of each landfill reactor as a leachate drainage port. Two such adapters were installed in the lid of each landfill reactor for leachate recirculation and gas collection. Adapters were held in place with wax to provide a gas-tight system. The methanogenic reactor was operated in an upflow anaerobic sludge blanket (UASB) made of PVC (10 cm i.d., 80 cm height) with working volume of 5.50 l.

2.4. Experimental design and operation

Three simulated landfill reactors were used in the experiment. Landfill reactor R1 was constructed with leachate recirulation as used in bioreactor landfill practice. The schematic diagram of the bioreactor landfill is illustrated in Fig. 1 without the broken line part (the methanogenic reactor). Landfill reactor R2 was connected with methanogenic reactor R2 prior to leachate recirculation. The operational configuration of the laboratory-scale bioreactor landfill system with a methanogenic reactor is presented in Fig. 1. Landfill reactor R3 was operated with methanogenic reactor R3 using treated leachate recirculation, and initially inoculated with EMs.

The methanogenic reactors R2 and R3 were seeded with raw sludge procured from the Hangzhou citric acid factory and Hangzhou Shibao sewage treatment plant. 4.5 l of sludge was added to each of methanogenic reactors. The sludge was incubated with the synthetic water with a chemical oxygen demand (COD) of 30,000 mg/l for 10 days. Then it was acclimated by leachate with COD concentration of 1743–2436 mg/l and NH_4^+–N concentration of 242–359 mg/l from Hangzhou Tianzhiling landfill. The acclimated sludge in methanogenic reactors R2 and R3 contained total solids (TS) content of 87.7 and 95.2 g/l and volatile solids (VS) content of 27.0 and 29.0 g/l, respectively.

Prior to filling, a 5-cm thickness of gravel was placed at the bottom of each landfill reactor to retain refuse and stop small particles from leaching out. Then about 20 kg synthetic MSW mixture, which was added with deionized water to 75% moisture content, was filled into each landfill reactor and a specific height of 50 cm was attained. Finally, the waste mixture was covered with a 5 cm depth of sand. Leachate was recirculated through the top of the landfill reactor on a daily basis. The stimulated landfill reactors were operated at room temperature and the methanogenic reactors were carried out in a temperature-controlled room at 30 ± 1 °C.

2.5. Analytical methods

Leachate collected from each of landfill reactors was analyzed for COD, volatile fatty acids (VFA), NH_4^+–N and pH. COD, NH_4^+–N and pH in leachate were determined by conventional methods [22]. VFA was analyzed by acidified ethylene glycol colorimetric method [23]. Gas production rate was measured with a wet gas meter (model SQL, Shanghain, China). Dry weight of refuse was measured by drying the sample in an oven at 105 °C. VS was measured by ashing the dried waste in a furnace at 550 °C. Gas concentration (CO_2 and CH_4) was analyzed using a gas chromatograph (GC) (model 102G, Shanghai, China) equipped with a thermal conductivity detector (TCD) and a 2-m stainless column packed with GDX104 (60/80 mesh). The operational temperature of the column was at 40 °C, and carrier gas (N_2) at a flow rate of 30 ml/min.

3. Results and discussion

3.1. Organic mass (COD) effluent from landfill reactors

During the stabilization of landfilled waste, organic matter are released from landfills by the non-uniform and intermittent percolation of water through the refuse mass. Laboratory and field studies have shown that organic mass effluent from landfills varied with the quantity and quality of

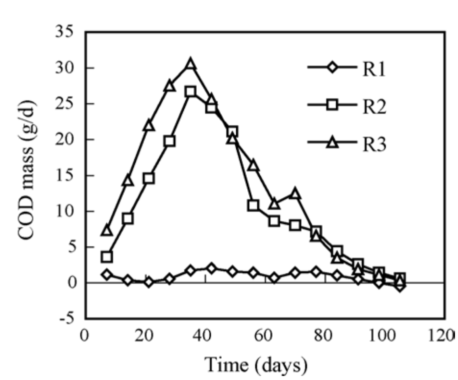

Fig. 2. Rates of COD mass effluent from landfill reactors.

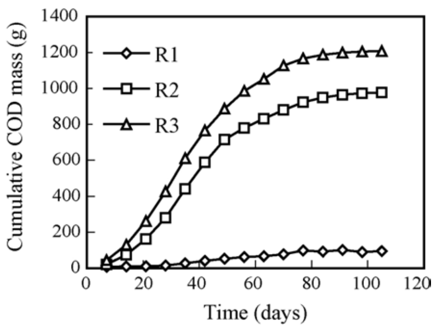

Fig. 3. Cumulative COD mass effluent from landfill reactors.

leachate [24]. Fig. 2 shows the rates of organic mass (COD) effluent from landfill reactors for 105 days. Due to the rapid release and hydrolysis of organics and an increase in leachate volume, the rates of COD mass leached from landfill reactors R2 and R3 with methanogenic reactors using treated leachate recirculation increased fast, and reached their peak values on day 35, then decreased and stabilized their lower level at the end of the experiment. This indicated that the stabilization of landfilled waste in landfill reactors R2 and R3 processed the mature phase on day 105. The rate of COD mass from landfill reactor R3 with EMs inoculation was the highest among the landfill reactors. This fact is also shown in Fig. 3 where cumulative COD mass coming from landfill reactor is plotted against time. The cumulative COD mass leached from landfill reactors R2 and R3 were 977 and 1208 g, which were 10.2 and 12.6 times higher than that from landfill reactor R1, respectively, after 105 days operation. EMs inoculation enhanced 24% of organic mass effluent from the landfill reactor. The results suggested that the EMs increased the biodegradability of landfilled waste and accelerated the hydrolysis of polymers, such as cellulose, hemicellulose, proteins, and lipids, into soluble sugars, amino acids, long-chain carboxylic acids, and glycerol.

3.2. *Peformance of methanogenic reactors*

Organic matter effluent from landfill reactors R2 and R3 was treated and converted into methane and carbon dioxide in methanogenic reactors. Fig. 4 illustrates the organic removal in methanogenic reactors R2 and R3. The COD removal efficiencies were maintained higher than 90% until day 49 and thereafter declined as the influent COD concentrations decreased, and reached very low levels at the final, due to the low biodegradability of organic in leachate from the old landfill [25]. The effluent COD concentrations of methanogenic reactors were maintained at 1–2 g/l throughout the study. The data was higher than reported by Kettunen et al. [26], a COD of less than 380 mg/l in leachate after anaerobic treatment. This might be attributed to the recirculation practice in the landfill and methanogenic reactors reintroduced non-biodegradable organic to the reactor keeping the high value.

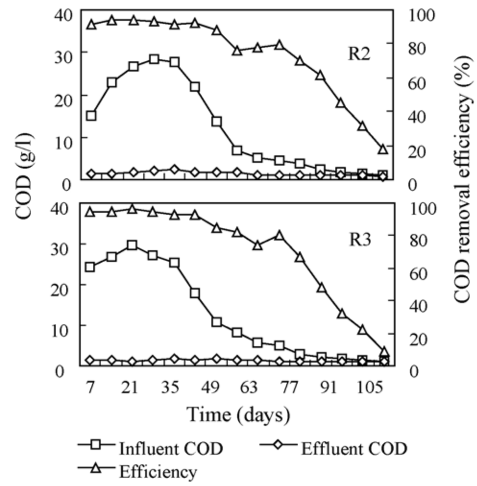

Fig. 4. Organic removal in methanogenic reactors.

The gas production rate as a performance of organic loading rate (OLR) is shown in Fig. 5. The gas production rates behaved similarly to the OLRs. Strong linear relationships between gas production rates and OLRs in methanogenic reactors R2 and R3 supported the affinity of gas production. The high gas production rate was observed in the first 70 days. After 91 days operation, no gas produced from methanogenic reactor R3 because readily available substrates began to be limited, while methanogenic reactor R2 ceased gas production on day 98. The gas production rate was much higher in methanogenic reactor R3 than in reactor R2 in the first phase. This meant EMs inoculation resulted in a higher peak of gas generation rate and a shorter gas generation period, which increased the potential for beneficial use of the gas.

Fig. 6 shows the relations between COD removal rates and gas productions in methanogenic reactors. The correlation factors in methanogenic reactors R2 and R3 were 0.4607 l gas/g COD ($R^2 = 0.9388$) and 0.4502 l gas/g COD ($R^2 = 0.952$), respectively. Similary, Keenan et al. [27] reported that daily gas production in anaerobic treatment of leachate was correlated with OLR. The components of gas generation from methanogenic reactors were principally methane and carbon dioxide (Fig. 7). The methane concentration was maintained at 73–78% until day 84 and declined slightly at the end of the study. Similar

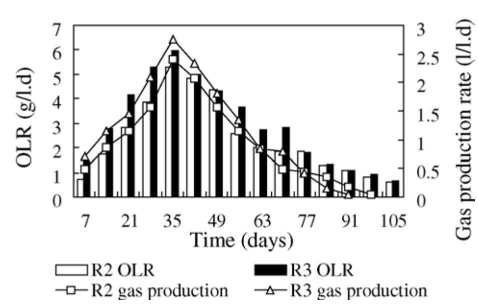

Fig. 5. OLRs and gas production rates in methanogenic reactors.

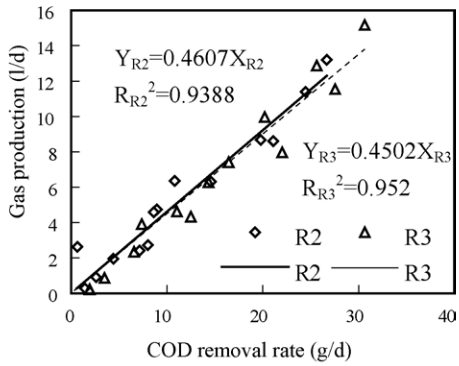

Fig. 6. Correlations of COD removal and gas production rate in methanogenic reactors.

finding was previously obtained during two-phase anaerobic digestion, where the methane concentration of the biogas generated in the methanogenic reactor was significantly higher [28].

3.3. Gas production

The cumulative gas productions for landfill reactors, methanogenic reactors and systems are shown in Fig. 8. The overall volume of gas production in the bioreactor landfill systems with methanogenic reactors using treated leachate recirculation was much larger than in landfill only with leachate recirculation. When the bioreactor landfill systems R2 and R3 produced 518.6 and 620.9 l of gas, respectively, the landfill reactor R1 produced only 65.7 l of gas. The gas production in the bioreactor landfill system occurred mainly in the methanogenic reactor, which produced over 88% of the overall gas volume. The average methane concentration of the gas formed in the bioreactor landfill system was above 70%, and thus energy yield from bioreactor landfill system gas (7 kW/m^3) would be higher than that from landfill gas (4–5 kW/m^3) [29].

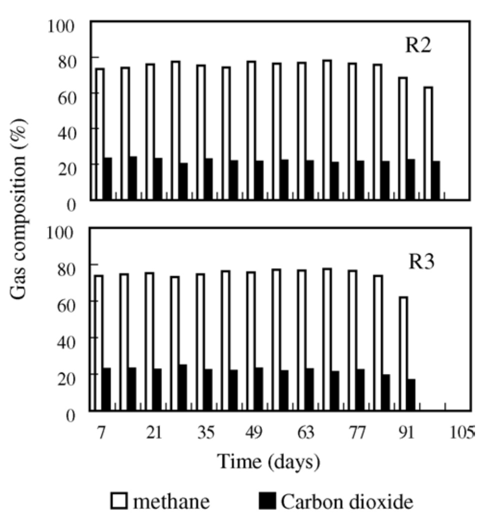

Fig. 7. Gas compositions for methanogenic reactors R2 and R3.

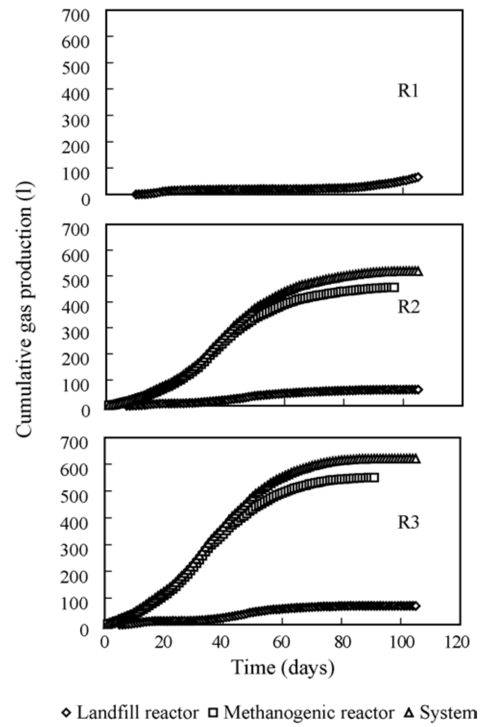

Fig. 8. Cumulative gas productions for different landfill systems.

EMs inoculation did not affect obviously the gas production from the landfill reactor, but it increased the amount of gas generated from the methanogenic reactor and shortened gas generation period to about 91 days. The increased gas production was directly related to the higher degree of waste stabilization in the bioreactor landfill system R3, which might be attributed to the combination of initial refuse inoculated with EMs and phase separation in the landfill. EMs inoculation increased the populations of microorganisms and hydrolytic enzyme activities associated with refuse decomposition [30,31], which accelerated waste degradation and increased gas production.

3.4. MSW stabilization

Table 1 shows the physical and chemical properties of different landfill reactors at the initial and final time. Among the landfill reactors, landfill reactor R3 with EMs inoculation had the highest degree of MSW stabilization according to the quantity of MSW degradation, leachate characteristics and settlement. The quantities of MSW degradation were 11, 27.5 and 30.5% in landfill reactors R1, R2 and R3, respectively, after 105 days operation. The rate of MSW degradation in landfill reactors R2 and R3 with methanogenic reactor using treated leachate recirculation was 2.5–2.7 times higher than in landfill reactor R1 only with leachate recirculation. Dry weight and VS of landfilled waste had a statistically significant decrease in landfill reactors R2 and R3 compared to landfill reactor R1 (Table 1; $P < 0.01$).

Table 1
Physical and chemical properties of different landfill reactors

Time (days)	Reactor	Landfilled waste (kg)			Leachate characteristics (mg/l)				Settlement (%)
		Wet weight	Dry weight	VS	COD	VFA	NH_4^+–N	pH	
Initial ($t = 0$)		20	8.47 ± 0.75	4.94 ± 0.52					
Final ($t = 105$)	R1	17.8	7.86 ± 0.74	4.45 ± 0.30	38734	19560	1457	5.23	12
	R2	14.5	$6.50 \pm 0.58^{**}$	$3.57 \pm 0.24^{**}$	1009	178	1287	7.40	24
	R3	13.9	$6.03 \pm 0.46^{**}$	$3.06 \pm 0.28^{**}$	1071	99	1212	7.39	27

$^{**}P < 0.01$.

The organic strength of leachate was much lower in landfill reactors R2 and R3 than in landfill reactor R1. The leachate COD and VFA concentrations in landfill reactor R1 were 38734 and 19560 mg/l, respectively, and the pH remained at a low value of about 5.23 on day 105. These indicated that the stabilization of landfilled waste processed the acid phase in the landfill reactor R1 only with leachate recirculation. The slow stabilization of refuse in landfill reactor R1 was attributed to low pH value and high VFA concentration in circulating leachate, which might cause inhibition in methanogenic archaea [7]. The NH_4^+–N concentration in leachate was high (above 1000 mg/l) in the three landfill reactors at the end of the study, but it was below 12–17% in landfill reactors with methanogenic reactors using treated leachate recirculation when compared to landfill reactor R1. This suggested that NH_4^+–N was produced during the decomposition of organic nitrogen and kept stable in anaerobic biological environment of landfills. NH_4^+–N typically accumulated in leachate with leachate recirculation [32]. Though a part of NH_4^+–N was consumed by the growth of microorganisms in methanogenic reactors, the NH_4^+–N removal was insignificant in the bioreactor landfill system. These results agree with the findings of Sponza and Ağdağ [33], the leachate recirculation management strategy recirculated within the reactors providing an increased opportunity for NH_4^+–N accumulation.

The corresponding cumulative settlements in landfill reactors R1, R2 and R3 were 12, 24 and 27% of the initial refuse height at the end of the study, respectively. Rapid settlement of waste in landfill reactor R3 was a function of refuse composition and density, refuse decomposition, compacting, height of cover soil, climate [34], and provided an opportunity to utilize valuable air space for refilling with new waste before closure of the compartment.

4. Conclusions

This work demonstrated that EMs inoculation resulted in an increase in the biodegradability of MSW and a high degree of waste stabilization. With a methanogenic reactor using treated leachate recirculation, the high organic mass effluent from landfill could be rapidly converted into methane and carbon dioxide, which alleviated inhibitory effects of hydrolytic products in landfills only with leachate recirculation, and provided optimally environmental and nutrient conditions for acidogenic and methanogenic organisms. The degradation of landfilled waste and organic pollutants in leachate was a two-phase degradation process within the bioreactor landfill system with a methanogenic reactor using treated leachate recirculation, where the hydrolysis-acidification of organic waste occurred mainly in the landfill reactor, and methanogenesis occurred chiefly in the methanogenic reactor. The gas production from the methanogenic reactor accounted for nearly 88% of the overall gas volume. And the methane concentration in the methanogenic reactor was maintained at 73–78%, although it declined slightly at the end of the study. A combination of EMs and methanogenic reactors using treated leachate recirculation could generate steadily a high volume of landfill gas and with a high methane content. Furthermore, it also produced the most stabilized and lowest strength leachate. The findings had important applications of EMs in landfills operated in ways conducive to methanogenesis and energy recovery from waste.

Acknowledgements

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