

Treatment of landfill leachate using modified anaerobic baffled reactor

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Received 13 February 2019; Accepted 23 November 2019

ABSTRACT

Landfill leachate is a highly concentrated organic wastewater with complex compositions. It is a major source of pollution potentially threatening the quality of groundwater, surface water, and life forms. The treatment of landfill leachate consists of various combination processes such as biological, chemical and physical methods. In this study, raw leachate was subjected to an anaerobic treatment using a modified anaerobic baffled reactor (MABR). Initially, the start-up of the MABR system was accomplished using meat extract as synthetic feed. The start-up of the reactor was carried out by maintaining a low chemical oxygen demand (COD) of 350 mg/L at an organic loading rate (OLR) of 0.0875 kg COD/m³/d. Once the reactor attained 99% COD removal, real wastewater (landfill leachate) was gradually fed into the MABR. The OLR was increased to 0.175, 0.375, 0.75 and 1.40 kg COD/ m³/d, respectively. The process performance of the reactor was evaluated in terms of pH, COD, color, volatile acid (VA), biogas production and heavy metal removal. Results showed that an average COD removal efficiency of 79.3% was observed when the OLR was 1.4 kg COD/m³/d. The VA concentration showed a stable profile with a very little value (38.9 mg/L of HOAc) in the effluent of the reactor for all the OLR studied. The color removal was 32%, 46%, 45.1% and 78.2% when the OLR was increased to 0.175, 0.375, 0.75 and 1.40 kg COD/m³/d, respectively. As, Cr and Fe removal was 87.5%, 88.8%, and 87.8%, respectively, when the reactor was operated at an OLR of 1.4 kg COD/m³/d. The heavy metals removal efficiency provides further evidence that heavy metals can be degraded in anaerobic environments.

Keywords: Landfill leachate; Modified anaerobic baffled reactor (MABR); Heavy metals; Anaerobic granular sludge

1. Introduction

Waste reduction, reutilization, and recycling are widely practiced to achieve sustainable waste management [1]. Sustainable landfilling is a necessary part of an efficient integrated waste management system. Sanitary landfilling is one of the most common methods for dispersion of municipal solid waste (MSW) [2]. Modern sanitary landfills are designed, constructed, and maintained to minimize the adverse environmental impacts from waste disposal over both the short and long terms. After landfilling, solid waste undergoes physicochemical and biological changes. Consequently, the degradation of the organic fraction of the wastes in combination with percolating rainwater leads to the

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production of "leachate". Leachates may contain enormous quantities of organic contaminants, ammonia, suspended solids, heavy metals, and inorganic salts, phenols, and phosphorus [3]. The complexity of these characteristics makes the leachate more difficult to manage [4,5]. If not treated and carefully disposed of, leachate may enter the surrounding soil, groundwater, or surface water [6,7]. Regulations concerning leachate discharge into receiving waters are becoming more and more stringent [8]; therefore, it requires a more proper and efficient treatment system.

To reduce the negative impact of discharged leachate on the environment, several techniques of treatment have been used. The technologies that were developed for the treatment of landfill leachate could be classified as physical, chemical and biological [9,10]. The characteristics of the leachate directly govern the implementation of the most suitable technique for the treatment of leachate. Leachates from different landfills vary considerably in their chemical compositions [11]. The current primary leachate treatment method is by aerobic treatment combined with chemical and physical treatment. The aerobic treatment mechanism mainly consists of microorganisms degrading the pollutants in the leachate using aeration. However, this method is still insufficient to remove some toxic pollutants in the leachate. Accordingly, a combination of treatment methods such as biological, coagulation, activated carbon, sand filter, and membrane filters are used for the complete removal of pollutants [12,13].

The anaerobic treatment is a biological process of decomposition of organic matter which does not only remove most pollutants but also generates valuable by-products, namely biogas, in the form of methane [14]. The significant advantages of anaerobic treatment are no aeration required, deficient excess sludge production, biogas production with high energy content, low nutrients requirement, and application of high organic loading [15]. Anaerobic baffled reactor (ABR) is described as a series of up-flow anaerobic sludge blanket reactors in which the wastewater is forced to flow under and over of a series of vertical baffles as it passes from the inlet to the outlet [15]. The compartmentalization of the reactor prevents horizontal movement of the biomass, and thus a high amount of active biomass is retained in each compartment. This feature provides an excellent contact between the contaminants and the microorganisms, longer biomass retention times and better resilience to organic and hydraulic shock loadings [16]. Some of the bacteria also move horizontally down the reactor at a relatively slow rate, giving rise to the cell retention time of 100 d at hydraulic retention time (HRT) of 20 h [17]. Therefore, the wastewater can come into intimate contact with a large amount of active biomass as it passes through the ABR with short HRTs (6-20 h), while the effluent remains relatively free of biological solids.

This study aimed to investigate the start-up of a modified anaerobic baffled reactor (MABR) system using meat extract and to investigate the treatment of landfill leachate containing heavy metals (As, Cr, and Fe). The MABR is an enhancement of the existing ABR where each compartment was further divided by slanted baffles to encourage mixing within the compartment and for better pollutant removal efficiency. The conventional ABR has some limitations such as the accumulation of volatile fatty acid and low pH, exposing sensitive bacteria in the front compartments, nutrient limits in the final compartment and elimination of phase separation [18]. Furthermore, it has also had some drawbacks such as a high quantity of solids washout, inactive and stagnant sludge at the bottom and poor performance for some recalcitrant wastewater. In our previous study [18], the fundamental of treatment performance of MABR using synthetic wastewater (glucose) was described thoroughly. In this study, the performance of MABR was evaluated using landfill leachate containing heavy metals. Most of the previous studies on the treatment of landfill leachate using anaerobic reactor concentrated on the removal of chemical oxygen demand (COD), ammoniacal nitrogen and color, but neglected the heavy metal degradation in the process. Moreover, to date, there is no reported study on the MABR system treating landfill leachate. The treatment of landfill leachate using MABR may lead to the effective treatment solution of landfill leachate as compared to available aerobic treatment. It will be of immense practical significance in terms of effluent characteristics of landfill leachate.

2. Materials and methods

2.1. Modified anaerobic baffled reactor (MABR)

The MABR is a laboratory-scale plexiglass reactor having 28 L capacity containing four uniform compartments (7 L capacities) and each having a slanted baffle (45°), heater and sludge and gas sampling ports (Fig. 1). The length, width, and height of the reactor are 82, 17 and 32 cm, respectively. Each compartment was installed with a heater to maintain the temperature at 37°C. A digital temperature probe located in each compartment provided the constant operating temperature. Other detail information's about the MABR was extensively described in our previous study [18].



Fig. 1. Design of MABR system.

2.2. Landfill leachate

The landfill leachate was taken from Jeram Sanitary Landfill, Selangor (Worldwide Landfills Sdn Bhd). The characteristics of landfill leachate were analyzed and tabulated in Table 1. The composition of landfill leachate varies from region to region; however, it depends mainly on the nature of deposited wastes, soil characteristics, rainfall patterns and age of landfill. The characteristics of the landfill leachate show that it is highly contaminated with regard to COD (5,678 mg/L) and various heavy metals. The biochemical oxygen demand (BOD)/COD ratio of the leachate is 0.2, which shows that the landfill leachate is intermediate. According to the leachate classification in Malaysia, a BOD/COD ratio between 0.1 to 0.5 represents an intermediate category of leachate, which shows the moderate biodegradability of the substance. The characteristics of landfill leachate indicate that the concentrations of COD, color and heavy metals that are considered in this study are quite high (according to Environmental Quality Regulations, 2009, Department of Environment, Malaysia) and may be harmful to the surface or groundwater.

2.3. MABR operations

The reactor was seeded with anaerobic granular sludge. The sludge is a naturally occurring biocatalyst having wide application as inoculation material for the start-up of bioreactors for the anaerobic treatment of wastewater. Granular sludge-based bioreactor technologies are recognized worldwide as cost-effective and efficient for the anaerobic treatment of industrial and municipal wastewater [19]. Granular sludge having high density is advantageous

| Table 1 | |
|--------------------------------------|--|
| Characteristics of landfill leachate | |

| Parameter | Results | Units |
|--------------------------|---------|--------------------|
| рН | 8.3 | _ |
| Temperature | 26.5 | °C |
| COD | 5,678 | mg L ⁻¹ |
| BOD ₅ @ 20°C | 1,135 | mg L ⁻¹ |
| Total suspended solids | 220 | mg L ⁻¹ |
| Oil & grease | 0.6 | $mg L^{-1}$ |
| Ammoniacal nitrogen (AN) | 90.5 | mg L ⁻¹ |
| Arsenic (As) | 0.28 | mg L ⁻¹ |
| Aluminum (Al) | 0.47 | mg L ⁻¹ |
| Boron (B) | 1.83 | mg L ⁻¹ |
| Cadmium (Cd) | 0.43 | mg L-1 |
| Chromium (Cr) | 0.26 | mg L ⁻¹ |
| Fluoride (F) | 0.36 | mg L-1 |
| Formaldehyde (FA) | 8.60 | mg L ⁻¹ |
| Iron (Fe) | 25.2 | mg L ⁻¹ |
| Manganese (Mn) | 0.86 | mg L-1 |
| Nickel (Ni) | 0.19 | mg L ⁻¹ |
| Zinc | 0.46 | mg L-1 |
| Color | >500 | ADMI |

in controlling sludge washout, which was a drawback in the conventional ABR. Table 2 shows the properties of the granular sludge. The granular sludge was taken from an anaerobic digestion plant that processes food waste. Three liters of granular sludge was fed into each compartment of MABR, and the remaining volume was filled with tap water. The density of the granular sludge and its aggregate nature are the properties, which are helpful in the retainment of the sludge in the respective compartments of MABR. The pH of the sludge is also favorable for the anaerobic digestion of landfill leachate.

This amount of sludge substantially contributed to the solid requirement in the reactor system after settling. After seeding, the head plate was attached, and the headspace above the sludge in each compartment was flushed with nitrogen gas to displace residual air in the system before introducing the feed. The reactor was left to stabilize at 37°C for 7 d without further modification. The start-up of the reactor was carried out using meat extract as synthetic feed by maintaining a low COD of 350 mg/L at an organic loading rate (OLR) of 0.0875 kg COD/m³/d. Once the reactor attained a steady-state condition (99% COD removal), real wastewater (landfill leachate) was gradually fed into MABR. The COD was increased (700-5,600 mg/L) by reducing the synthetic feed until 100% leachate was fed with the OLR values increased from 0.175-1.40 kg COD/ m³/d (Table 3). The concentration of influent COD was increased gradually to avoid shock loading, and the HRT is maintained as 4 d throughout the treatment so that the wastewater and microbes get ample time to acclimatize in the MABR system, which will subsequently lead to the efficient treatment of landfill leachate.

| Table | 2 | | |
|-------|---|---|--|
| D | | ~ | |

| Properties | s of granu | lar sludge |
|------------|------------|------------|
|------------|------------|------------|

| Aggregation | Thick fluid (sediment solids) |
|-------------------|---|
| Odour | Soil-like or light smell of rotten eggs |
| Color | Grey-black |
| Solubility | Pellet-like particles |
| Boiling point | Like water |
| Coagulation point | -2°C |
| Density (kg/L) | 1.0–1.1 |
| Acidity (pH) | 7–9 |
| Toxicity | |

Table 3

Operating conditions during the treatment of landfill leachate

| Influent COD (mg/L) | HRT (d) | OLR (kg COD/m³/d) | Duration (d) |
|------------------------|------------|----------------------|-----------------|
| 350 (Start-up) | 4 | 0.0875 | 0–28 |
| 700 | 4 | 0.175 | 28-60 |
| 1,500 | 4 | 0.375 | 60-80 |
| 3,000 | 4 | 0.75 | 80–96 |
| 5,600 | 4 | 1.40 | 96–112 |

2.4. Sampling and analysis

Sample analysis included pH (Fisher Scientific Accumet AP61 Model, USA), COD and volatile acid (VA) all were conducted according to Standard Methods [20] by using DR 6000 Spectrophotometer. The total biogas volume was determined using an optical gas bubble counter [16,18]. The gas produced from the MABR was captured and measured in liters per day. The gas was captured and passed through a unique device (regulator) [16,18] which creates a uniform bubble with a specific volume. The detected number of bubbles is digitally displayed on the electronic circuit, and the amount of gas can be calculated. The biogas composition was determined using a portable gas analyzer (GA2000, Geotechnical Instruments, USA). Heavy metals are analyzed in an inductively coupled plasma-optical emission spectrometry, an analytical technique used for the detection of chemical elements (Perkin Elmer/Optima 8300, USA). Color is analyzed using a UV-VIS (Merck/Spectroquant Pharo 300, USA) on the ADMI scale.

3. Results and discussion

3.1. MABR performance during start-up

Microbial groups involved in anaerobic degradation have a specific pH region for optimal growth. Values outside this range can be quite detrimental to the process, particularly to methanogens. Therefore, maintaining a suitable and stable pH within the digester should be one of the priorities for ensuring efficient methanogenic digestion. The optimum pH range in an anaerobic digester is 6.7–7.2; however, the process can tolerate a range of 6.5–8.0 [21]. For anaerobic digestion, the alkaline phase is preferred to maintain stable microbial populations in the sludge. It is vital to maintain suitable alkalinity in the reactor. The alkalinity was kept in the reactor at 1,000–2,000 mg/L as CaCO₃ using sodium hydroxide (NaOH).

The start-up of MABR lasted 28 d. During the start-up, a fixed concentration of feed COD (350 mg/L) was maintained until a steady-state COD removal and pH were achieved. The OLR and HRT were also maintained at 0.0875 kg COD/ m³/d and 4 d, respectively. By maintaining a steady feed concentration with long detention time, the reactor would achieve enhancement of solids build-up, encouragement of methanogenic populations and rapid recuperation to hydraulic shock [22]. Fig. 2a illustrates the pH profile during the start-up period. The pH levels during reactor start-up using the synthetic wastewater were generally stable in all compartments of the MABR until day 12 (pH 7.0-8.1), where a slight reduction was observed from days 16-20 (pH 6.3-6.7). However, this was temporary, as the pH levels in all compartments recovered to stable conditions on day 28 (pH 6.9-7.2). Fig. 2b shows a steady increase in the COD removal efficiency during start-up from day one up to day 28. The COD removal efficiency increased at a steady rate from 58% to 99%, confirming that the reactor was working



Fig. 2. MABR performance during start-up with synthetic feed; (a) pH, (b) COD, (c) VA, and (d) biogas production.

efficiently. Even though low pH levels were noted during the initial operational period (e.g. day 16–20), the high COD removal efficiencies confirm the ability of the MABR system configuration to overcome the adverse effect of pH.

VA is a useful indicator to evaluate the anaerobic reactor performance. In the current study, acetic acid (HOAc) was assessed for the concentration of the acid in the reactor due to its main desire for methanogenic digestion. A VA value of less than 150 mg/L in an anaerobic reactor indicates that the reactor is operating under stable conditions [23]. In general, if the pH of the reactor system is high (e.g. pH 7-7.5), the VA should be lower [24], and in the current study, this trend was observed (Fig. 2c). The VA concentration in all compartments during reactor start-up using the synthetic wastewater was stable throughout the study (40-95 mg/L of HOAc), confirming a stable operation of the reactor. Fig. 2d shows the total biogas production during reactor start-up. In can be seen that the gas production increased steadily from 0.013 to 0.021 L/d during the operational period, confirming that the biodegradation of the substrate is taking place.

3.2. MABR performance during the treatment of landfill leachate

3.2.1. pH

pH plays a significant role in the process of AD because the acid-forming bacteria are more tolerant of low pH concentrations than methanogens [25]. Acidogens responsible for VA production cannot survive either in extreme acidic (pH 3) or alkaline (pH 12) environments and maintenance of appropriate acidogenic pH is essential to maximize VA production [26]. After the start-up of MABR (until day 28), the pH dropped subsequently due to the rapid production of VA's resulting from increased acidogenic activity (day 32) (Fig. 3a). When the OLR was raised to 0.175 kg COD/m³/d, the pH recovered (day 36 onwards) in all the compartments of the reactor (pH 7.3-8.3). Further increase of the OLR to 0.375, 0.75 and 1.40 kg COD/m3/d, resulted in variation of the pH profile (7.9-8.4) in all the compartments, signifying the stable condition of the reactor. It is known that in the ABR system, the first compartments were populated mostly with the fast-growing acidogens [27]. In the later compartments, the slow-growing methanogens were predominated, and this causes the difference in the pH profile across the reactor system. The methanogens are very sensitive to low pH in anaerobic digestion and required neutral pH (e.g. pH 7) for its growth. The ability to grow faster was limited in the first two compartments due to low pH, which is referred to as the slow-growing methanogens. The ability of the methanogens to grow faster was accomplished in the later compartments as the pH increases.

3.2.2. Chemical oxygen demand

Fig. 3b shows the COD removal profile of the reactor system at various OLR. When the OLR was 0.175 kg COD/m³/d, the average COD removal efficiency was 87%. Further increase of the OLR to 0.375 kg COD/m³/d reduces the COD removal efficiency to an average value of 78%. However, this is not permanent as the removal increased slightly to an

average value of 81% when the OLR was 0.75 kg COD/m³/d. An average COD removal efficiency of 79.3% was observed when the OLR was 1.40 kg COD/m³/d. The results indicated that not much changes in the COD removal efficiency when the OLR was increased from 0.375 to 1.40 kg COD/m³/d (removal efficiency was between 78% to 81%). This also shows that the reactor can withstand the increase in the OLR and have the ability to recover and adapt to the new conditions [28].

3.2.3. Volatile acid

Higher OLRs results in the accumulation of organic acids and favors the acidogenic process of AD [29]. Besides OLR, substrate complexity in terms of higher COD is also a significant factor that could disturb the process performance. The profile of VA concentration in Fig. 3c shows that the reactor was stable with very little VA (average 38.9 mg/L of HOAc) in the effluent of the reactor (C4) for all the OLR studied. Moreover, even in compartment 1 (C1) of the reactor system, the average VA concentration is below 84 mg/L HOAc, suggesting the utilization of these acids by the microorganisms. The trend in VA variation in the reactor system was in the order of C1 > C2 > C3 > C4. As a process performance indicator, VA concentration is probably the most sensitive parameter to monitor. They can be inhibitory of the digestion process which, can lead to system failure. In a correctly designed and well-operated digester, the concentration of total VA is typically below 500 mg/L HOAc [30]. The VA that is produced by acidogenic and acetogenic bacteria reflect a kinetic uncoupling between the acid producers and consumers.

3.2.4. Biogas

Biogas production was monitored in all compartments of the MABR throughout the operation. Fig. 3d shows the profile of biogas production in the MABR. The biogas composition profile follows the COD removal efficiency, whereby it is increased when COD removal increased, soluble COD in digestion converted to sludge and biogas. In the beginning, the produced biogas volume is low, and after a short period, due to the enhancement of microorganisms and reactor circumstances compatibility, the gas volume increases. During the start-up period (0.0875 kg COD/ m³/d), the biogas production rate was 0.02 L/d and increased gradually to 0.036, 0.053, 0.065, and 0.1 L/d when the OLR was increased to 0.175, 0.375, 0.75, and 1.40 kg COD/m³/d, respectively. The results indicated that when the OLR increased, the amount of biogas produced in the MABR was also increased.

3.2.5. Color

Color is one of the parameters in determining water quality, which was due to the low biodegradability of dissolved organic constituents in the leachate. The high concentration of color in landfill leachate is due to the presence of high organic substances. In general, leachate produced by an old landfill with low biodegradability is classified as stabilized leachate. Stabilized leachate contains prominent levels of



Fig. 3. Treatment performance of landfill leachate by MABR; (a) pH, (b) COD, (c) VA, (d) biogas production, and (e) influent and effluent color concentration.

organic substances such as humic and fluvic compounds, which can be indicated by leachate color [31,32]. It can be observed that the color removal increases (except for OLR 0.75 kg COD/m³/d) when the OLR was increased. The color removal was 32%, 46%, 45.1%, and 78.2% when the OLR was increased to 0.175, 0.375, 0.75 and 1.4 kg COD/m³/d, respectively. Fig. 3e displays the concentration of the color in the influent and effluent of the reactor at different values of OLR.

3.2.6. Heavy metal removal

The removal of heavy metals was investigated during the treatment of landfill leachate in the MABR system. It is well known that heavy metal toxicity during anaerobic digestion is a major concern [5]. The stability of MSW correlates to leachate quality in terms of heavy metals, ammonia, and COD. Physical, chemical and microbial processes all affect the heavy metal concentrations in landfill leachate. Table 4 shows the removal of heavy metals during the treatment of landfill leachate. It can be seen clearly that the heavy metals, As, Cr and Fe were removed to a certain extent in the treatment system at various levels of the OLR. When the OLR was 0.175 kg COD/m3/d, the As removal was 44%, after that it remains constant at OLR of 0.375 kg COD/ m³/d. However, when the OLR was further increased to 0.75 and 1.40 kg COD/m³/d, respectively, the As removal increased to 81.8 and 87.5%. The degradation of heavy metals in the MABR indicates that the anaerobic microorganisms could have utilized the pollutants or that it was accumulated in the sludge [33]. Dong et al. [34] reported during anaerobic digestion, the concentration of heavy metals such as Zn, Pb, Cu, Ni, and Cr, increased during high-solid anaerobic digestion of sewage sludge. However, Thanh et al. [35] found that methanogenic bacteria require trace elements for methanogenic activity and indicate that without the presence of heavy metal, the methanogenic activity decreased. Moreover, they also claim that heavy metal was utilized in the transport system of the methanogenic bacteria for the conversion of CO₂ to CH₄. For example, Fe is utilized in the transport system of the methanogenic bacteria for the conversion of CO₂ to CH₄ and functions both as an electron acceptor and as a donor. Fe also acts as a binding component in sulfide precipitation as it is often supplemented into anaerobic reactors, not only to precipitate the formed sulfide but also to control the level of hydrogen sulfide in the biogas [35]. Similar results were also obtained for Cr removal in the MABR system. When the OLR was 0.175 kg COD/ m³/d, the Cr removal was 37.5% and increased 61%, 70%, and 88.8% when the OLR was gradually increased to 0.375,

Table 4 Removal of heavy metals during the treatment of landfill leachate

| OLR (kg COD/m ³ /d) | As removal (%) | Cr removal (%) | Fe removal (%) |
|-----------------------------------|-------------------|-------------------|-------------------|
| 0.175 | 44 | 37.5 | 90.6 |
| 0.375 | 44 | 61 | 82.5 |
| 0.75 | 81.8 | 70 | 98.6 |
| 1.40 | 87.5 | 88.8 | 87.8 |

0.75, and 1.40 kg COD/m³/d, respectively (Table 4). Fe degradation in the MABR system shows the removal rate at increasing OLR and influent and effluent concentrations, respectively. The removal profile fluctuated when the OLR was increased, a trend that was not seen in the As and Cr removal. When the OLR was 0.175 kg COD/m³/d, the Fe removal was 90.6%, and then decreased to 82.5% when the OLR was 0.75 kg COD/m³/d. The removals achieved its highest at 98.6% when the OLR was 1.40 kg COD/m³/d, before reducing slightly to 87.8% (Table 4).

4. Conclusions

This research was initiated to evaluate MABR performance during reactor start-up (meat extract) and treatment of real wastewater (landfill leachate containing heavy metals). From the experimental results, there appears to be considerable potential for the MABR system to be implemented on-site for the treatment of landfill leachate. The performance of the MABR system showed the stable operation of the reactor leading to efficient treatment of landfill leachate. However, a techno-economic feasibility study of the treatment system should be conducted in the future to evaluate the actual performance of the process at the landfill site. Moreover, the microbiological aspect of the granular sludge should be conducted in the future to analyze the bacterial composition profile in the reactor during the operational period.

Acknowledgments

The authors thank Worldwide Landfills Sdn Bhd, Jeram, Selangor, Malaysia for giving access to the landfill site for leachate collection. The authors also thank Universiti Teknologi Malaysia and the Ministry of Education Malaysia for funding this research under the Research University Grant (Matching Grant) Vote number: Q.K130000.3040.01M17 and Fundamental Research Grant Scheme Vote number: R.K130000.7856.5F049.

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