Anaerobic digestion of landfill leachate: A modified approach

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A partially bi-phasic, laboratory-scale, anaerobic digester was designed and utilized for optimum generation of methane, using a high strength landfill leachate, with chemical oxygen demand (COD) of approximately 15,500 milligrams per liter (mg/L) at a loading rate of 200 mL/day. The test apparatus comprised of a down flow semi-continuous digester, operating at mesophilic temperature (37 ± 0.5 degree Celsius [°C]) where the hydraulic retention time was maintained at 20 days. Modification to the conventional anaerobic digestion was accomplished by creating an aerobic condition two days prior to the effluent’s entry into the anaerobic system. This modification gave the digester adequate buffering capacity to withstand limited environmental stress and prevented generation of excess acid that could lead to development of a sour condition, resulting in the failure of the system. Experimental results showed that the bi-phasic digester was capable of withstanding high loading rate and minor environmental shocks and could be used successfully to treat landfill leachate with high COD. The experimental data showed a total COD removal of approximately 79% and a methane yield of approximately 0.032 cubic meter per kilogram (m³/kg) of COD removal. Methane content of the biogas varied between 35 and 39%.

Key words: Anaerobic digestion, landfill leachate, leachate digestion, anaerobic digester, modified anaerobic digestion, bi-phasic anaerobic digester, methane recovery

INTRODUCTION

Disposal of municipal solid waste (MSW) by landfilling is one of the oldest and widely used methods. According to the most recent MSW disposal data available from the United States Environmental Protection Agency (U.S. EPA), in 2010 approximately 54.2% of the total MSW generated in the United States was disposed in the landfills, which amounts to approximately 135.5 millions of tons of MSW (U.S. EPA, 2011a). Although over the years, shortage of open lands and stricter regulations governing proper construction and/or management of landfills (Code of Federal Regulations 40 CFR, subtitle D, 1993) has led to a steady decrease in the number of operating landfills in the United States, however, elsewhere around the world, especially in the developing countries, landfilling still remains the most preferred among the MSW disposal practices. In 1988 there were 7,924 operating landfills in the United States, whereas in 2010 there are only 1,908 operating landfills in the United States (U.S. EPA, 2011b).

However, the present landfills are more sophisticated in terms of design and management and are of much larger in capacity compared with the earlier landfills. MSW landfills can be considered as large bioreactors that generate mainly two types of products from aerobic and anaerobic catabolism of MSW: (a) leachate and (b) biogas. Leachate is a liquid that is produced due to percolation of water through emplaced refuse (or waste), carrying with it soluble compounds and transportable organic and inorganic materials as well as micro-organisms. Leachate from landfills is typically high in biochemical oxygen demand (BOD₅), chemical
oxygen demand (COD), and inorganic constituents, all of which pollute soil and groundwater as they come in contact. Various field observations indicate that even small municipal landfills generate sufficient quantity of leachate that may impact groundwater (Bagchi, 1987). Since many of the newly constructed landfills are sealed at the sides and bottom, especially in the United States, Germany, Italy and the United Kingdom, leachate management has become an even greater problem and must be removed for treatment by physical-chemical and/or biological methods (Stegmann, 1983).

The collection and treatment of leachate are two of the most important problems associated with operation of landfills. Chemical processes were reported to be inefficient as they produce large amount of chemical sludge (Lin, 1991). The aerobic biological process is considered impractical because of high dilution requirements. The anaerobic process is considered to be suitable for treating high strength organic wastewater and has been popular in the wastewater treatment field because of its advantages, such as high treatment efficiency and methane producing ability, i.e., energy recovery potential (Lin, 1991).

During anaerobic decomposition process organic matter is degraded in the absence of oxygen (O$_2$) yielding primarily cell biomass, methane (CH$_4$) and carbon dioxide (CO$_2$) gases.

The main advantages of anaerobic digesters over the aerobic ones are: (a) anaerobic digesters can handle higher organic loading rates; (b) generate less biomass leading to less generation of digester sludge; (c) can degrade some difficult to degrade organic chemicals; and (d) produce energy in the form of CH$_4$ gas as an useful by-product. Methane-forming bacteria utilize very little energy obtained from the degradation of volatile acids for biomass production, while most of the energy released from the volatile acids is transferred to methane (Gerardi, 2003). During anaerobic digestion process approximately 80 to 90% of the organic matter is converted to gases (Eckenfelder, 1989). Therefore, bacterial growth or sludge production is low and optimum operational conditions must be maintained for satisfactory rates of solids destruction and methane production. However, due to slow growth rate of anaerobic microorganisms, it requires longer time to produce enough biomass leading to longer digester start up time. Another critical drawback of the process is, anaerobic digesters exhibit low kinetic rate of removal of organic matters at low temperatures. In order to promote faster kinetic rate, external source of energy (in the form of heat) is generally required, which could be quite expensive.

Owing to successful development of high treatment rate reactors, most of the industries prefer to set anaerobic digesters in their plant to prevent soil and water pollution. But most of them have problems of restart and interruption that result in reduced efficiency of the system. The causes of these problems are non-availability of waste throughout the year, shock loading, abnormal concentrations of chemicals etc. Anaerobic microorganisms are very sensitive to the pH of the media. A pH in the range of 6.6 to 7.6 is generally suitable for anaerobic microbial growth (Eckenfelder, 1989). Any drastic change of pH outside the favorable condition may cause low microbial activity to complete inhibition of microbial processes leading to digester failure. During anaerobic decomposition sometimes toxic hydrogen sulfide (H$_2$S) gas is produced from sulfate reduction and/or protein decomposition. Excess generation of H$_2$S may produce toxic effects to anaerobic microorganisms and also may lower the pH of the media, leading to digester failure. The problems are indicated either by delayed restart of the anaerobic reactor or failure of biomethanation (Kanavade and Kapadnis, 2000).

Despite some of the disadvantages and drawbacks of the anaerobic digestion process, as earlier discussed, anaerobic digestion is a promising alternative for the treatment of landfill leachate (Henry et al., 1987).

However, concentration of many of the organic materials, especially xenobiotic organic compounds, are reported be much higher in certain waste streams, including those in landfill leachate (Kanavade and Kapadnis, 2000). Patel et al (1991) found that methanogenesis may not be the problem in the anaerobic digestion of dilute wastes, however, it becomes so when treating more concentrated wastes, such as landfill leachate.

The main objective of this laboratory study was to evaluate if a modified digestion approach could resolve the digester souring issue, which was noted in many previous studies (Glassman, 1948; Kodicek, 1949; Neiman, 1954; and Kabara et al., 1977) to be a common problem during anaerobic digestion of waste due to excess acid generation. Also, this study evaluated the methane generation potential of the modified digestion approach using landfill leachate as the source waste material. Digester performance was further evaluated under environmental stress conditions.

**MATERIALS AND METHODS**

In order to conduct the laboratory scale anaerobic digestion of landfill leachate a modified design of the conventional anaerobic digester was developed, a schematic presentation of which is shown on Figure 1. In the conventional digester, there is only one digestion vessel, called the anaerobic digester, within which anaerobic condition is maintained. In our modified approach, an aerobic phase (also referred to as hydrolyzing phase/digester) was introduced at the start-up for the first 35 days of digester operation, developing a bi-phasic digestion process.

The leachate was collected from an operating MSW landfill located near Kansas City metropolitan area.
The landfill is operating since 1987 and has nearly 7 million tons of emplaced waste. This landfill generates on an average 400,000 gallon per month (high: 780,000; low: 120,000) of leachate, which is collected and stored in a centralized collection tank from where it is discharged under-ground into deep injection wells.

In this experimental bi-phasic digestion process, for the first 35-days of operation, everyday 200 milliliters (mL) of fresh leachate was introduced into the hydrolyzing digester, which had a retention time of two days. The hydrolyzing digester comprised a one-liter open mouth glass bottle where the aeration took place under atmospheric diffusion process. On third day onwards, everyday 200 mL of the leachate was drained from the hydrolyzing digester into the anaerobic digester under gravity flow. At the beginning, every day 1% weight-by-volume of cow manure was added with the fresh leachate to supply the required anaerobic (specially methanogenic) microbial population to the hydrolyzing digester. When the digester started generating gas, addition of cow manure was discontinued.

The anaerobic digester was a five-liter airtight conical flask, which was connected at the top with the hydrolyzing digester and at the bottom a valve was provided to drain out the digested liquid (Figure 1). Another inlet was provided at the top, which was used for direct sample introduction (200 mL of fresh leachate every day) after the initial 35-days, when the hydrolyzing/ aerobic phase was discontinued permanently. After the first 23 days when the anaerobic digester was filled to its capacity (4.6 liters), everyday 200 mL digested wastewater was allowed to drain through the bottom outlet of the anaerobic digester.

At the top of the anaerobic digester there was a gas collection outlet, which was connected with an airtight one-liter glass bottle, completely filled with acidic water...
RESULTS AND DISCUSSION

In the conventional anaerobic reactor, the digestion process is carried out by obligate anaerobes. So, presence of oxygen at the initial stage could be detrimental to the performance of the anaerobic process. However, the first phase of anaerobic digestion is an aerobic one (or at least facultative) where the aerobic or facultative aerobic microorganisms degrade the higher-molecular-mass organic constituents of the waste to generate the simple or lower-molecular-mass intermediate compounds (Tchobanoglous and Burton, 1996) from which the ultimate product of methane and carbon dioxide are generated.

The first groups of aerobic organisms are responsible for hydrolyzing organic polymers and lipids to basic structural building blocks such as monosaccharides, amino acids, and related compounds. After the hydrolyzing phase the facultative and obligate anaerobic bacteria come into the scene that degrades the compounds to produce organic acids (commonly acetic acid) under anaerobic condition. The third group of microorganisms, commonly called methanogens, converts the hydrogen and acetic acid formed by the acid formers to methane gas and carbon dioxide. The microaerophilic condition that exists within a conventional anaerobic digester ultimately slows down the initial hydrolyzing step.

In order to overcome this problem, in this study a biphasic, partly aerobic and anaerobic, digester setup was designed to study the bio-methanation potential of the landfill leachate. The hydrolyzing phase was separated from the anaerobic phase until the stabilization of the digester system. It is also known fact that the aerobic processes are very fast and once the lower molecular-mass compounds are formed, it is not necessary to maintain aerobic condition. So after the hydrolyzing phase, the partially digested leachate was subjected to anaerobic condition, where the acidogenic microorganisms started working. As the retention time required for the anaerobic digestion to complete is longer than the aerobic one, so to provide a longer retention time, the digester volume was increased. This also gave the added advantage of larger volume of gas generation, which was easy to detect. Detectable changes in the water level in the gas collection bottle was noticed on and after 28th day from the start of the digestion process, which indicated collection of considerable amount of gases from the anaerobic process.

Periodic analysis of digester gas was conducted using infrared spectroscopy, which showed that the gas was mainly composed of methane, carbon dioxide, and water vapor. The first two gas samples were also found to contain some hydrogen sulfide, which was not found in later samples. This confirms the fact that during initial stages of anaerobic digestion, sulfate reduction takes place which leads to hydrogen sulfide generation.
Quantitative analysis of the gas samples using gas chromatography found the average methane content to be approximately 37±2%. Methane yield was calculated to be approximately 0.032 cubic meter per kilogram (m³/kg) of COD removal. These results showed that the digester was working satisfactorily.

According to the U.S. EPA's guidelines, if the methane content exceeds 25% in a gas, it can be used as a fuel. Therefore the digester gas with about 37% methane can easily sustain the flame. In order to establish the digester performance various physiochemical parameters were monitored during the course of the treatment process which are summarized in Table 1. Graphical representations of reduction in the key parameters during the treatment process are presented on Figure 2 and Figure 3 for data from Set I and Set II, respectively.

BOD₅ test is one of the most commonly used quantitative parameter to evaluate the wastewater characteristic. BOD₅ measures the usage of dissolved oxygen by microorganisms in the biochemical oxidation of organic matter over a 5-day period (Tchobanoglous and Burton, 1996). In the first treatment set (referred as Set I) pre-treatment average BOD₅ value was noted to be 7,642 mg/L, which was reduced to 965.5 mg/L after the completion of the anaerobic digestion process (Table 1 and Figure 2). Similarly, in the second treatment set (referred as Set II) pre-treatment average BOD₅ value was noted to be 7,304 mg/L, which was reduced to 1,092 mg/L after the completion of the anaerobic digestion process (Table 1 and Figure 3). In the first treatment set COD showed an average 87.4% reduction in values, while in the second set BOD₅ was reduced 77.4% (average). The BOD₅/COD ratio also is a good indicative parameter for evaluation of the wastewater treatment efficiency. The BOD₅/COD ratio in the untreated leachate for both treatment sets I and II were determined 0.49, which reduced to 0.30 and 0.32 respectively in the treated leachate. As the average BOD₅/COD ratio has decreased considerably from the pre-treatment to the post-treatment landfill leachate, it indicates higher removal of the biodegradable organic matter from the wastewater, which directly supports the efficiency of this modified treatment approach.

The final BOD₅, COD test and BOD₅/COD ratio indicate conclusively that the modified anaerobic digestion approach was effective in treating the high strength landfill leachate, and improved the wastewater characteristic considerably. The TS and TSS values of the post-treatment landfill leachate showed decrease from the pre-treatment leachate (Table 1, and Figures 2 and 3), which was most likely due to precipitation of the suspended solids as digester bottom sludge and consumption of part of the suspended and dissolved solids by the microorganisms for their metabolic activities. Another critical parameters monitored during this study was pH of the leachate, which showed an increasing

### Table 1. Physico-chemical analysis data of the untreated and treated sample.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Set I Untreated leachate</th>
<th>% Reduction</th>
<th>Set I Treated leachate</th>
<th>% Reduction</th>
<th>Set II Untreated leachate</th>
<th>% Reduction</th>
<th>Set II Treated leachate</th>
<th>% Reduction</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>6.71 ± 0.10</td>
<td>-</td>
<td>7.48 ± 0.13</td>
<td>7.67 ± 0.16</td>
<td>7.45 ± 0.08</td>
<td>-</td>
<td>7.19 ± 0.08</td>
<td>-</td>
</tr>
<tr>
<td>TS*</td>
<td>2939 (73.2)</td>
<td>61.3</td>
<td>1138 (70.0)</td>
<td>3015 (13.1)</td>
<td>1228 (57.1)</td>
<td>60.5</td>
<td>1105 (38.1)</td>
<td>51.0</td>
</tr>
<tr>
<td>TSS*</td>
<td>1992 (66.0)</td>
<td>46.8</td>
<td>1060 (96.6)</td>
<td>2163 (19.2)</td>
<td>1061 (38.1)</td>
<td>51.0</td>
<td>1054 (38.3)</td>
<td>50.0</td>
</tr>
<tr>
<td>COD*</td>
<td>15575 (1067.5)</td>
<td>79.3</td>
<td>3225 (178.3)</td>
<td>7304 (82.9)</td>
<td>3380 (161.9)</td>
<td>77.4</td>
<td>3379 (162.1)</td>
<td>77.4</td>
</tr>
<tr>
<td>BOD₅*</td>
<td>7642 (88.8)</td>
<td>87.4</td>
<td>965.5 (79.0)</td>
<td>7304 (82.9)</td>
<td>1092 (52.7)</td>
<td>85.9</td>
<td>1109 (38.1)</td>
<td>51.0</td>
</tr>
<tr>
<td>BOD₅/COD</td>
<td>0.49</td>
<td>-</td>
<td>0.30</td>
<td>0.49</td>
<td>0.32</td>
<td>-</td>
<td>0.32</td>
<td>-</td>
</tr>
<tr>
<td>ECª</td>
<td>5.14 ± 0.26</td>
<td>6.6</td>
<td>4.80 ± 0.20</td>
<td>4.97 ± 0.17</td>
<td>4.75 ± 0.23</td>
<td>4.4</td>
<td>4.75 ± 0.23</td>
<td>4.4</td>
</tr>
<tr>
<td>Alkalinity*</td>
<td>227 (2.1)</td>
<td>6.6</td>
<td>212 (7.42)</td>
<td>226.9 (3.1)</td>
<td>210.9 (4.1)</td>
<td>7.1</td>
<td>210.9 (4.1)</td>
<td>7.1</td>
</tr>
<tr>
<td>Hardness*</td>
<td>253 (4.2)</td>
<td>22.5</td>
<td>196 (3.5)</td>
<td>254.8 (5.1)</td>
<td>195 (2.9)</td>
<td>23.5</td>
<td>195 (2.9)</td>
<td>23.5</td>
</tr>
</tbody>
</table>

*a Values noted in milligrams per liter (mg/L)
ª Values noted in milli-Siemens per centimeter (mS/cm)
- Values in parenthesis indicates the standard deviation (n = 10)
trend from acidic to the neutral or slightly alkaline range in pre-treatment to post-treatment phase (Table 1).

The increase of pH was most likely due to the uptake of anions from the leachate and accumulation of microbial metabolic products into the solution. Most anaerobic bacteria, including methanogens, show optimal microbial activity within a pH range of 6.8 to 7.2 (Gerardi, 2003). The increase in the pH of the post-treatment leachate proved to be in agreement with the optimal pH range of operation and proved to be helpful in maintaining the digester stability against development of the acidic condition and resulting digester failure during the treatment process. Elaborate statistical analysis of the data was performed including Student’s t-test, which showed strong statistical significance (p < 0.05) among observed data.

Hydraulic overload, organic overload, pH changes, temperature fluctuations, toxicity, large withdrawal of sludge, and sudden changes may cause upsets or unstable anaerobic digester and resulting digester failure (Gerardi, 2003). The decrease in biogas and methane production, decreases in alkalinity, pH, and volatile solids destruction, and increases in volatile acid concentration and percent carbon dioxide in the biogas are some of the indicators of unstable anaerobic digesters. One of the main objectives for this modified digester design was to evaluate the digester response under environmental stress. When the digester was subjected to the environmental shocks, i.e., when the digester was subjected to laboratory conditions without temperature control and when no sample was added or removed for 10-day period, the digester stopped generating gas and pH went up to a slightly higher values, 7.75 ± 0.24; COD of the drained liquid from the digester was found to be 4,690 mg/L (standard deviation 157.2, n = 10).

On the basis of the observations, it was concluded that the optimum pH for methane generation was 7.45 ±0.15; a pH higher than 7.6 was detrimental to the methanogenic consortia that resulted in decreased methane production. When the fresh sample addition and digested sample removal were not performed on a regular basis, the pH of the digester was found to go up above 7.6 and digester ultimately stopped generating gas. When organic and hydraulic overloading were attempted the methane percentage in the generated gas dropped to around 10% and pH decreased to a value of 7.1 ± 0.2. One of the important successes of this digester was that it never turned sour and the digestion process never failed. Fongastitkul and Lo (1994) reported a combination of hydraulic and organic overloading of M-UASB (methane phase-upflow anaerobic sludge blanket) reactor as a major cause of process failure which led to MLVSS (mixed liquor volatile suspended solids) washout,
Figure 3. Set 2 representation of some parameters

Conclusions

The modified design of the bi-phasic digestion process not only worked well but was also found to be quite economical from the energy recovery point of view. Addition of the aerobic phase at the initial stage proved useful in stabilizing the digester in the long run and provided the digester enough buffering capacity to withstand minor environmental and overloading shocks. The improvement in wastewater quality indicates that the digested water after further treatment and dilution could be discharged into the common sewage. As the modified digester never failed when using landfill leachate, the same approach should be tried with other wastewaters before making it a common step in all digesters. Due to the time and funding constraints no analysis of anaerobic and especially methanogenic microbial population was carried out. However, understanding the digester microbial population profile may provide helpful insight into the digester microbial population dynamics and various inter- and/or intra-community interactions that are observed at different stages of wastewater digestion. Overall, this project bears the possibility of further modification of the age-old conventional digestion system.

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