

Enhanced Landfill Biocell: Cost-effective Technology to Achieve Sustainability of Landfill Operations

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Abstract

Landfill biocell technology (LBT), a recent innovation, is a modification of the landfill bioreactor technology, where a waste cell is operated sequentially in anaerobic and aerobic bioreactor modes with subsequent mining to recover value-added products and space. Although, LBT technology is a desirable alternative waste management technique, technical issues prevent large-scale application in Canada and worldwide. Results from recent research on advancing the LBT field applications are presented. Laboratory-based recent research has shown that waste degradation rates in LBTs can be biologically enhanced through leachate augmentation, thereby reducing waste degradation cycle and make resource recovery more efficient. Adding lignin-degrading enzymes improve the performances of both aerobic and anaerobic stages of the LBT process. In laboratory batch-scale studies, orders of magnitude increase in biogas production was observed when experiments were conducted in anaerobic mode. The flow through-column experiments showed the feasibility of increasing biogas production in field-scale operations and thereby extending the biogas recovery phase. Research also showed that augmentation of leachate with lignin-degrading enzymes significantly increased the kinetic rates of aerobic waste degradation. In addition to resolving LBT design and operation issues, it is also important to accurately determine biogas production in LBTs; a critical performance indicator. This requires accurate determination of biomethane potential (BMP) of landfilled waste. Results from traditional BMP measuring techniques do not represent the field BMP values. Preliminary results from research being conducted to develop a site-specific BMP method provided promising results that could be used under any environmental condition.

Introduction

A novel concept of landfill waste disposal, the biocell (Hettiaratchi et al., 2007), is a variation of the landfill bioreactor approach and involves the operation of a waste cell under optimum conditions for gas recovery and resource/space recovery. The biocell operation consists of three stages. In the first stage, the biocell is operated as an anaerobic bioreactor with accelerated methane (CH₄) production. In the second stage, the biocell is converted to an aerobic bioreactor, and the operation is carried until most of the waste is decomposed. The third stage involves cell mining to recover degraded material/recyclables and space, thus making the waste cell operation sustainable (Hettiaratchi et al., 2007). Considering the difficulties in locating new landfills in or near urban centers, recovery of landfill space is one of the key benefits of the biocell approach (Hunte, 2010).

The success of using the biocell concept as a waste to energy recovery option and rapid waste stabilization could be hindered by the limitations related to waste properties and other issues. The presence of large amounts of lignin, a moderately degradable material, limits biogas production and waste stabilization. Lignin, the major component of the target waste in this research, is a three dimensional polymer connected by several acid resistant C-C linkages. Lignin could be only partly degraded to monomeric compounds by hydrolysis and is mostly degraded by oxidative attack on the C-C bonds (Higuchi, 2004; Martinez *et al.*, 2005). A special category of commercially available enzymes made from white rot fungi is peroxidases that potentially catalyze the lignin degradation process. (Jayasinghe et al., 2011; Higuchi, 2004).

Through enzymatic enhancement by leachate augmentation, lignin degradation could be accelerated, such that longer anaerobic energy recovery periods and shorter aerobic stabilization periods could be achieved. Leachate augmentation with enzymes before recirculation was first studied by Lagerkvist & Chen (1993). They conducted column experiments to investigate the effect of addition of cellulolytic enzymes to fresh MSW. The effect of enzymes on waste degradation was studied during acidogenic and methanogenic degradation stages separately by measuring changes in cellulose content and conversion of volatile solids (VS). The observed conversion of cellulose was 42-70% in cells with enzymes addition and 29% in cells without enzyme addition. The conversion of VS was approximately 40% to 50% in enzyme added cells (Lagerkvist & Chen, 1993).

However, past studies on leachate manipulation with enzymes only considered enhancing the degradation of fresh waste at early stages of landfill operation (Lagerkvist & Chen, 1993; Crine et al., 2008). Enhancing waste degradation at later stages of cell operation, when the waste consists primarily of lignin-rich materials has not been studied. The current study was designed to determine the viability of augmenting leachate with different peroxidase enzymes to increase the degradation rates of lignin-rich waste at later stages of anaerobic bioreactor

and early stages of aerobic bioreactor operation when cells are operated sequentially in anaerobic and aerobic modes with subsequent mining for resource/space recovery.

The benefits of methane recovery from a landfill has led to substantial research in determining ways to quantify and project methane recovery volumes. Investors in landfill methane recovery projects need to have foreknowledge of the amount of methane that would be generated from a municipal solid waste to be landfilled to make an informed decision of best ways of exploitation and profitability. A main analytical parameter in this decision-making process is the Biochemical Methane Potential (BMP). Benchmarking methane potential against theoretical calculations can be misleading. The experimental BMP results show variations in degradability of waste due to different experimental conditions and setups that can affect these results. The mathematical models for predicting methane yields are based on parameters with some uncertainties embedded in them. Studies have shown that using theoretical calculations to determine the ultimate methane potential of landfilled waste would most times overestimate actual values as landfilled waste consists of components with varying biodegradability. The experimental BMP tests is the most reliable estimate of ultimate methane potential (Karanjekar 2012). Nevertheless, conducting BMP assays for municipal solid waste (MSW) is challenging because of the heterogeneous nature of MSW. The conventional BMP assay has been originally developed for the anaerobic degradation of wastewater sludge; where the assay is boosted with the macro- and micro-nutrients. Several researchers have adopted this BMP method for MSW, providing ideal conditions for waste degradation (Owen 1979, Owens and Chynoweth 1993, Bilgili et al. 2009, De la Cruz and Barlaz 2010). Consequently, the quantity of methane generated from current BMP assay is not representative of gas production in actual landfills (Angelidaki et al. 2009, Raposo et al. 2011, Hidalgo and Martín-Marroquín 2015). Therefore, it is necessary to develop a landfill-specific BMP methodology. In such methodology, more details should be given on type of inoculum to be used in terms of form (granular, liquid or semi-solid), source (characteristics of treatment plant or initial substrate), ISR ratios (range to which researchers should work within) and pretreatment procedures for inoculum (Pre-incubated or not, filtered or not etc.). Also, there should be clarity as to whether nutrient media should be added to the BMP assay setup as practiced by some researchers.

This paper discusses our evaluation of the potential of enhancing landfill biocell technology with leachate augmentation. The paper also reports results from a preliminary study to establish the BMP assay using a model waste on methane yield in a different setup of inoculum to substrate (I/S) ratio, and moisture content, without the addition of nutrient supplements.

Materials and Methods

The experiments to study the effect of leachate augmentation were conducted in several scales: batch experiments, column experiments and lysimeter experiments.

Batch experiments – comparison of enzyme types

For the analysis of enzymatic enhancement of anaerobic and aerobic stages, the enzymes, lignin peroxidase (LiP) and manganese peroxidase (MnP from *Phanerochaete chrysosporium*), were used, as they are considered true lignin degraders due to their high potential redox value (Sanchez, 2009).

A two-factor, three-level factorial experimental design was used for both anaerobic and aerobic experiments. The experimental factors were enzyme dose and hydrogen peroxide (H₂O₂) dose. The two enzyme types were tested individually at three-levels of enzyme doses and three levels of H₂O₂ doses, resulting in 18 treatments. Each treatment had three replications. The three levels of enzyme doses were 0 (control), 0.1 and 0.15 mg/gDS and the H₂O₂ doses were 0, 0.01 and 0.02 mL/gDS. The different treatment combinations were labeled with levels of -1, 0, and 1. For example, experiments conducted with an enzyme dose of 0.1 mg/gDS and H₂O₂ dose of 0.02 mL/gDS was labeled as 0,1.

The anaerobic batch experiments were performed using 125 mL glass bottle reactors. The quantity of dry waste used in the experiments was 2 g. Pre-determined amounts of water, enzymes and H₂O₂ were added to the reactors. A sufficient amount of water was added to each sample to reach a moisture content (MC) of 160% of the field capacity (FC) of the waste. The amount of water added to the sample brought the final MC of the waste to about 60% (w/w) that is within the optimum moisture content for maximum waste degradation (Khanal, 2008).

One batch reactor was maintained as the control, with no enzyme or H₂O₂ addition. The reactors were sealed to prevent air entry and purged with pure nitrogen gas to create anaerobic conditions. The CH₄ concentration within the reactor was measured daily, over the monitoring period of 30 days, by collecting samples of headspace gas and analyzing using a VARIAN 4900 Micro gas chromatograph (Micro GC). The volume of CH₄ produced was calculated as described by Jayasinghe et al. (2011).

Flow-through column experiments – Anaerobic bioreactor simulation

The batch experimental study was designed to test various parameters in a simplified system, but the results are not directly applied in field-scale applications. The simplified batch system lacks both the necessary scale and processes such as leachate recirculation to reflect actual field conditions. Therefore, the process was scaled up to a set of flow-through column experiments to mirror landfill conditions for testing the enzyme augmentation concept in a cost-effective and controlled manner prior to field-scale application.

The height and the diameter of the columns were 100 cm and 14 cm, respectively. Each column consisted of leachate collection, sampling and recirculation, and gas collection and sampling. A layer of gravel and a screen were placed at the bottom of each column to prevent clogging of leachate outlet and pipes from fine particles. A gas measuring device was connected to each column, with control valves. A leachate collection tank was connected to each column to collect and store the leachate before recirculation.

Each column was filled with 9 kg of uniformly mixed representative MSW. The waste used was the same as in the batch experiments. Once the initial adjustment phase was completed, activated enzyme, MnP, identified as the most effective from batch experiments, was added to the leachate tank of the test column. The leachate was then recirculated. The quantity of produced gas was measured regularly. In addition, dissolved organic carbon (DOC) and other leachate parameters, biochemical oxygen demand (BOD), chemical oxygen demand (COD), total nitrogen and total phosphorous were measured using standard test methods.

Batch and flow-through column experiments - Aerobic experiments with MnP Enzyme

The objective of enzyme enhancement of aerobic landfill operation is to reach stable conditions rapidly. The ability of an MnP augmented system to reach stable conditions rapidly was measured using CO₂ production, nutrient content and cellulose and hemicellulose to lignin ratio (C+H/L). Two types of experiments were conducted; batch reactor experiments and laboratory-scale lysimeter experiments. Batch experiments were conducted in glass bottles and enzyme dose is used as the experimental factor. The response variables were CO₂ production and C+H/L ratio.

In the second series of experiments, a laboratory cylindrical plastic lysimeter bioreactor unit of wet waste was used. The waste was kept at field capacity. The lysimeter was designed such that leachate could be collected at the bottom and re-circulated to the top of the waste matrix using a leachate distribution system. The laboratory-scale lysimeter was operated without enzyme addition for a period of 28 days. At day 28, MnP was added to the leachate before recirculation in order to potentially enhance waste degradation.

BMP method development - Preliminary investigations

A sample of biosolids from anaerobic digesters was collected from Bonnybrook Wastewater Treatment Plant, Calgary, Alberta, Canada. Prior to use, the biosolid sample was centrifuged to concentrate the solids and remove excess water. Moisture content, water absorption capacity, total solids (TS), total volatile solids (TVS), and ash content of waste samples were determined. The characteristics of waste samples were determined according to the standard methods (APHA 2005). The volatile solids composition of waste and biosolids was used to determine how much waste should be added to each BMP assay. The volatile solids content of dry biosolids was 61% of total solids showing considerable methane potential. The percent C, H and N elemental composition analysis was carried out with Perkin Elmer Model 2400 C H N analyzer. The BMP assay was designed to understand the effect of inoculum to substrate ratio and moisture content.

The BMP assays were conducted in duplicate and one set of treatment was conducted in triplicate to estimate the experimental error. In addition, the background methane generation from biosolids, the inoculum, was determined separately by conducting a duplicate control treatment experiment without waste. Every three days, the bottles were removed from the temperature-controlled incubator and allowed to reach room temperature before removing headspace gas for analysis. The amount of biogas accumulation in the headspace was measured by water displacement method using an inverted burette. The volume of gas produced in the assay bottles was measured until the end of the experiment.

Results and discussion

Comparison of the performance of enzymes - from batch experiments

The results from batch experiments conducted to evaluate the enzyme type yielded similar results for both anaerobic and aerobic experiments. The experimental response, CH₄ yield, was used as the primary indicator of the effectiveness of enzyme addition on the lignin-rich waste degradation under anaerobic conditions. Significant increases in CH₄ yields were observed in all enzyme treated batch reactors compared to the control and the inactivated-enzyme (i.e., no addition of H₂O₂). Although H₂O₂ is a strong oxidant, our results show that H₂O₂ alone is not sufficient in degrading lignin-rich solid waste.

In the case of MnP treatment, the highest CH₄ yield was observed with an enzyme dose of 0.15 mg/g_{DS} and H₂O₂ dose of 0.02 mL/g_{DS}. The MnP treated reactors exhibited higher CH₄ yields than the LiP treated reactors on a consistent basis irrespective of the treatment. For example, after 30 days of operation, the MnP treated reactors

produced 1.5 times more cumulative CH₄ than the LiP treated reactors and 36 times more cumulative CH₄ than the control reactor. Furthermore, the statistical analysis showed that both the primary effects of enzyme and H₂O₂ doses on CH₄ yields are highly statistically significant (Hettiaratchi et al., 2014).

In aerobic experiments, the cumulative CO₂ yield was used as the indicator of the effectiveness of enzyme treatment. Both MnP and LiP treatments showed positive effects on the CO₂ yield, however, the MnP treated reactor showed the best performance in terms of the CO₂ yield at each enzyme dose. A maximum CO₂ yield of 660 mg/g_{DS} was observed in the reactor treated with a MnP dose of 0.1 mg/g_{DS}. This is more than a two-fold increase in CO₂ yield over that of the control reactor.

A primary reason for exhibiting higher CH₄ and CO₂ yields in the enzyme treated reactors compared to those of the control reactors was that peroxidase enzymes catalyze the depolymerization of lignin, thereby increasing the rate of hydrolysis of waste. Furthermore, LiP is known to oxidize only non-phenolic lignin units, whereas MnP is known to produce Mn⁺³ ions, which are capable of oxidizing both phenolic and non-phenolic lignin units (Gronqvist et al., 2003). The capability of MnP to oxidize both phenolic and non-phenolic lignin units may explain the observed higher gas yields in MnP treated reactors than the gas yields in LiP treated reactors.

Unlike in anaerobic reactors, the effect of H₂O₂ dose on the performance of the aerobic reactors was insignificant. Therefore, in aerobic bioreactor landfills, enzymes can be used directly without using an activator such as H₂O₂ (Hettiaratchi et al., 2014)

Biogas production in anaerobic flow-through column experiments

In anaerobic column experiments, the daily methane production and changes in DOC in leachate were used as response variables. Consistent with the expected effects of the enzyme treatment, the experimental columns demonstrated increased methanogenic activity, as indicated by daily gas production values. The primary reason is that the enzyme increased the hydrolysis rate of waste as shown with the increase in DOC in leachate. The microorganisms were able to degrade DOC rapidly, increasing the methane production rate.

The methane production rate and the DOC change over time were highly correlated (Figure 1). There was an initial lag phase (~ 10 days) of gas production. The DOC in leachate of enzyme modified lysimeter increased from 820 mg/L to 4215 mg/L during this lag period. In the meantime, the DOC in the control lysimeter increased from 820 mg/L to 1756 mg/L, possibly due to the moisture adjustments in the system through leachate recirculation. The DOC then decreased in both columns increasing gas production, indicating that the anaerobic microorganisms consumed the DOC while producing methane. The stabilization of methane production rates appears to be related to a decrease in DOC that began to show steady decrease after about day 100. These observations confirmed that the limiting factor in gas production, and thus methanogenic activity, was the availability of carbon in the system (Jayasinghe, 2013).

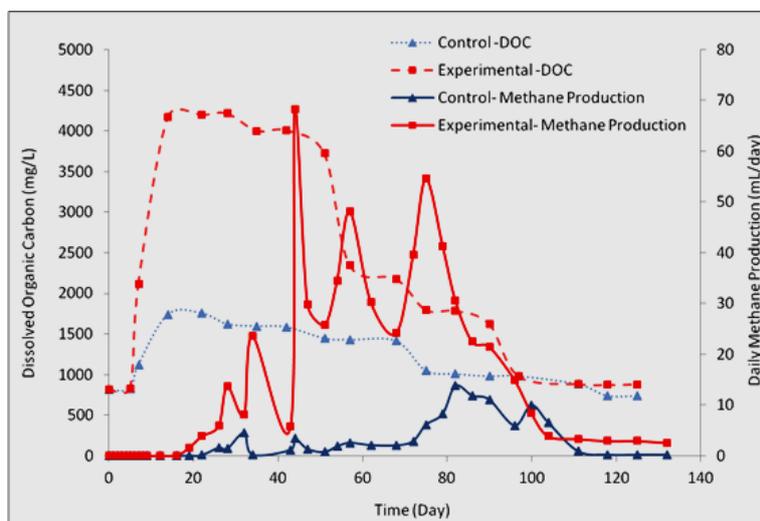


Figure 1: Daily CH₄ production rate and DOC variation in control and MnP augmented experimental column

Waste stabilization in aerobic flow-through column experiments

The batch experiments showed that enzyme added treatments have the capability to reach a stable C+H/L and lower CO₂ production rates, faster than the treatments without enzyme addition (Bartholameuz et al., 2016)

The enzyme enhancement increased biodegradability of waste; gas production increased more than two times (Figure 2) and there was clear evidence of increase in nutrient content (nitrogen, dissolved carbon) in leachate.

There was an immediate increase in nutrient content, but CO₂ production exhibited a delayed response. This could be due to the fact that although enzymes are capable of rapid hydrolysis of lignocellulose material, actual oxidation occurs at a lower rate. This slower overall degradation rate could be due to the presence of other limiting factors for microbial growth. Other nutrients, environmental conditions such as alkalinity or moisture content, could be factors that limit microbial growth. (Bartholameuz et al., 2016)

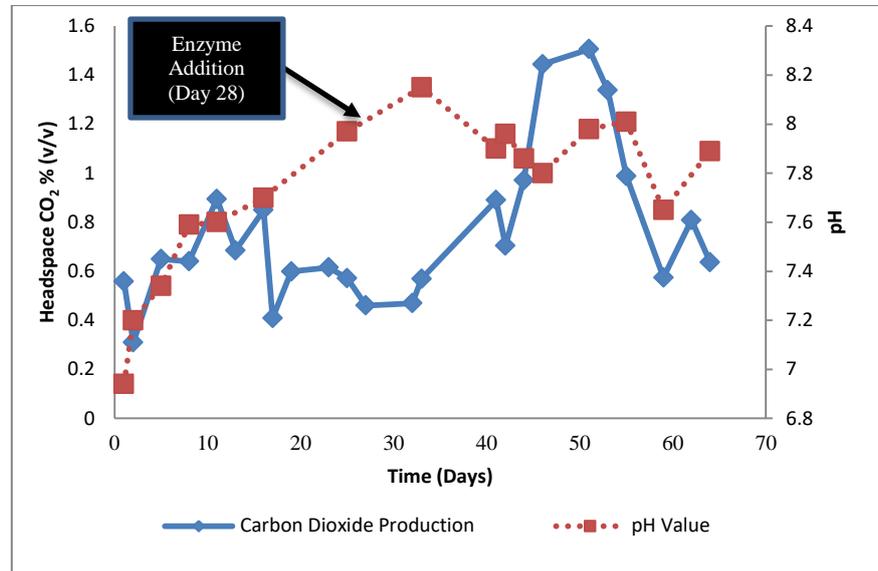


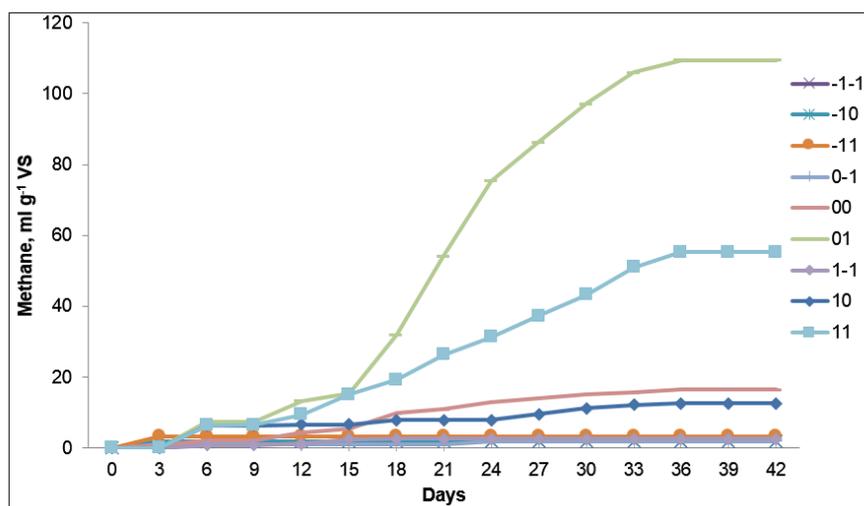
Figure 2: Aerobic lysimeter experiments with MnP enzyme – CO₂ production and pH changes

Parameters affecting results of BMP methodology

The cumulative methane generation was highest in the assay with 400 % moisture saturation and an inoculum to substrate ratio of two. Results revealed that increasing the inoculum to substrate ratios and moisture content significantly increases the methanogenesis of the substrate. I/S ratio less than two and low moisture content caused biochemical limitations, while high I/S ratio and moisture could promote methane generation. Since low I/S ratio treatments contained high substrate and less dilution, eventually the assay became too acidified and limited methanogenesis. Similar observations were reported by Raposo et al. (2006 and 2008).

At the beginning of the experiments, an initial lag phase was observed with negligible methane yield in all the assay bottles. The average lag phase was approximately 12 -18 days. The reason for reduced methane generation and long lag phase is not well understood because the anaerobic digestion process occurs in multiple stages: hydrolysis, acidogenesis, acetogenesis and methanogenesis. Furthermore, moisture, pH, temperature, carbon source, and nutrients are the major environmental factors affecting the anaerobic digestion process (Raposo et al. 2012). Since, the BMP assay was conducted without any nutrient supplements, assuming that the required microbial growth nutrients will be achieved from biosolids and waste materials. Therefore, the nutrient limitations and/or acidogenesis activity could be responsible for the long lag phase to establish the methane generation.

Based on the elemental analysis, an approximate molecular formula of the waste was calculated as C_{20.5}H_{40.6}O_{16.4}N; with a corresponding molecular weight of 563 g mol⁻¹. The theoretical ultimate methane potential (Lo_{th}) was estimated as 466 m³ per tonne of waste volatile solids (WVS), if all the WVS are biodegradable. However, the maximum observed ultimate methane potential (Lo_{exp}) from BMP assay was about 113 m³ per tonne of the WVS. Overall, the experimental methane yields were substantially lower than the theoretical values. Hidalgo and Martín-Marroquín (2015) also reported similar results with livestock and agri-food waste. This may be because incomplete waste degradation and/or accumulation of volatile fatty acids consequently inhibiting the methanogenesis reactions.



Variables	Levels		
	-1	0	1
A : Inoculum to substrate ratio (I/S)	1	2	4
B : Moisture content (%)	100	200	400

Figure 3: Effect of I/S ratio and MC on cumulative methane generation

Conclusion

The results show that the CH₄ yield and, thus the energy recovery, from partially degraded waste could be potentially increased by enzyme treatments in anaerobic landfill bioreactors. Furthermore, peroxidases have the ability to stabilize waste relatively quickly in aerobic landfill bioreactors. Among the tested enzymes, MnP showed the best performance. Unlike in anaerobic reactors, the effect of H₂O₂ dose on the performance of the aerobic reactors was insignificant. Therefore, in aerobic bioreactor landfills, enzymes can be used directly without using an activator such as H₂O₂. Preliminary BMP assay tests show that an inoculum to substrate ratio of two with high moisture content could be the most promising key parameter combination for conducting BMP assays.

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