Environmental, Geotechnical and Hydraulic Behaviour of a Cellulose-rich By-product Used as Alternative Cover Material

By

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Abstract: Deinking by-product (DBP), a cellulose-rich by-product produced in the early stages of the paper recycling process, has been used as alternative material for the construction of cover systems for municipal waste disposal facilities and acid-producing mine residues. Considering the high organic content of this material, covers constructed with it are susceptible to biodegradation, thus to changes in their properties with time. With the goal of identifying the biodegradation parameters that could influence the long term behaviour of DBP covers, an experimental laboratory program was developed and a series of 15 samples of DBP were monitored in biodegradation tests, for 400 days. Periods of intermittent water percolation allowed for collection of leachate. The evolution of gas and leachate production was monitored in terms of quality and quantity. According to the results obtained, the hydraulic and geomechanical properties of importance for a cover do not seem to be adversely affected by the level of biodegradation of the DBP or by mass loss.

Key words: Cover systems, deinking by-products, biodegradation, mass loss by gas production, mass loss by leaching
INTRODUCTION

Cover systems in municipal waste disposal facilities (MWDF) help control infiltration, whereas in the case of mining residues, the final cover must reduce the influx of oxygen that can lead to the generation of acid mine drainage (Cabral et al. 2000). In both cases, it is technically possible – and often an economic advantage – to use recycled materials or industrial by-products as alternative materials in the cover system, ultimately reducing their disposal in dedicated landfills (CQVB 1996; Wiegand and Unwin 1994). The beneficial use of deinking by-products (DBP), which are generated in large quantities by the pulp and paper industry and which used to be disposed of in dedicated landfills, is an example where an industrial by-product can replace fine-grained materials as infiltration control barrier, while helping to reduce landfilling costs.

DBP, also known as fiber-clay, are composed mainly of cellulose fibres, clay and calcite. The amount of fibres and the overall composition varies significantly with the type of paper recycled and the efficiency of the deinking process employed (Latva-Somppi et al. 1994). DBP have been characterized by many researchers, including Vlyssides and Economides (1997), Latva-Somppi et al. (1994) and Ettala (1993). This highly compressible by-product usually leaves the production plant with gravimetric water contents varying from 100 to 190%. The volatile matter content ranges from 48% to 60% and the relative density (Dr; this parameter is also know as specific gravity, whose symbol is Gs) is generally in the range of 1.8 to 2.0. These values are similar to data found in literature (Cabral et al. 1998; Kraus et al. 1997; Moo-Young and Zimmie 1996).

While composting remains an option in terms of beneficial reuse, the significant mineral and fibre content of this by-product allows other economically sound reuse alternatives. This is particularly the case when the material is used as replacement for fine-grained soils in the
construction of cover systems in MWDF and for mining tailings that produce acid mine drainage (AMD). Indeed, DBPs have been acknowledged to perform quite well as barriers to infiltration in sanitary landfills in the United States (Floess et al. 1995; Kraus et al. 1997; LaPlante 1993; Malmstead et al. 1999; Maltby and Eppstein 1994; Zimmie and Moo-Young 1995), Japan (Kamon et al. 2002), South Africa (Brown et al. 1998) and Canada (Audet et al. 2002). DBP have also performed quite effectively as an oxygen barrier layer to prevent AMD (Cabral et al. 2004; Cabral et al. 2000). The DBP barrier limits oxygen from reaching the tailings mass due to the high degree of saturation that can be maintained, but also due to aerobic degradation of its organic matter. Aerobic degradation implies oxygen consumption. If O$_2$ is consumed, less of it reaches the tailings under the DBP cover. Indeed, considering the high organic content of this material, such barriers are susceptible to biodegradation, thus to changes in their properties with time. There is, however, very little data concerning the evolution with time of biodegradation of DBP barriers. Several uncertainties also exist concerning prediction of its long term behaviour.

This paper presents and discusses the results of laboratory tests aimed at evaluating the long term biodegradation behaviour of DBP, as well as the implications of the evolution of DBP on key geotechnical parameters and properties.

MATERIALS AND METHODS

An experimental laboratory program was developed with the aim of identifying the biodegradation parameters that could influence the long term geotechnical and hydraulic behaviour of DBP. It included a series of 15 samples that were monitored during 400 days. Periods of intermittent water percolation (herein denominated leaching periods) allowed for lea-
chate collection. The evolution of gas and leachate production was monitored in terms of quality and quantity.

The initial degrees of saturation, density and water contents of the samples were similar to those found in actual DBP layers placed in the field (Table 1). However, 12 of the tests were performed under much higher temperatures (38°C) to accelerate the degradation process. This temperature is considered optimal for the activity of anaerobic (methanogenic) micro-organisms (Hartz et al. 1982; Zeikus and Winfrey 1976). Three control cells were prepared: two were tested at 21°C and one at 12°C. At 21°C, the prevailing temperature in the upper part of the DBP layer during the summer and fall was simulated, while at 12°C, the temperature in the middle and at the base of a DBP layer was simulated (Cabral et al. 1999).

After each leaching period, two of the cells placed at 38°C were disassembled. The direct mass loss of the disassembled cells was determined and several parameters obtained, such as water content, relative density and organic matter content (cellulose, hemi-cellulose and lignin). In addition, the following geotechnical parameters were obtained: void ratio, degree of saturation, relative density and density water content relationship (Proctor curve). The materials and methods employed are described as follows.

**Materials**

The compositions of the DBP used in the experiments described below are presented in Figure 1. It can be observed that a large portion of the material is composed of organic matter and that calcite dominates the inorganic fraction.
Experimental equipment

Fourteen 5 mm-thick PVC biodegradation cells were constructed, each with a volume of 9.5 litres. The size of the samples was determined based on the need to perform various geotechnical and physico-chemical characterization tests after the end of each accelerated biodegradation test. One of the control cells was constructed of Plexiglas with a total volume of 4.1 litres and placed at 12°C. A schematic drawing of the cell is presented in Figure 2. The dimensions in brackets correspond to those of the control cell at 12°C. The design was made bearing in mind that deaerated, deionized, distilled water was to be percolated through the samples at certain times and that leachate was to be collected. To do so, the cells were designed with two chambers, one on top and the other below the sample (Figure 2). Water intake took place at point A, leachate collection at point D and gas sampling at point B. At points A, C and D quick connect type valves were installed allowing for complete waterproofing of the cells. Biogas was collected from both chambers. Thermocouples were installed in five cells allowing for the temperature within the cells to be measured. The temperatures varied less than 0.5°C for the cells placed at 38 °C, whereas in the case of the control cells, variations of almost 3 °C were measured.

Sample preparation and testing

After mixing several samples delivered to the laboratory, random sub-samples were taken for physico-chemical characterization, which included: water content, relative density and organic matter content (cellulose, hemi-cellulose and lignin). Sample compaction was performed in a manner that aimed at reproducing field conditions as closely as possible in terms of density and water content (thus degree of saturation and void ratio). The sequence and energy prescribed in the ASTM D698 procedure (ASTM 2000) was employed. After compaction in 3 layers, the
cells were hermetically sealed, with a layer of silicon applied to the cover. The general conditions of the 15 samples are presented in Table 1. The averages and standard deviations of the different parameters are presented. The data referring to the control cell at 12°C (Plexiglas cell) are also presented in Table 1.

**Gas sampling and analysis**

As shown in Figure 2, bags were attached to the cells for storage and removal of the biogas generated. The bags, manufactured by Pollution Measurement Corporation, are made of laminated polyethylene with two outlets, one attached to the cell and the other permitting the removal of gas through a septum. The volume and biogas (actually O₂, CO₂ and CH₄) composition of the cells were determined every day or every three days, during the first months of the experiment. Afterwards, these analyses were done once a week. To collect gas samples, 60 ml syringes were used.

The concentrations of the three gases were measured with a LFG-20 apparatus (Triple Landfill Gas Analyser) from ADC-Analytical Company Ltd. The precision of the apparatus is ± 0.4% for oxygen. For CO₂ and CH₄, the precision varies depending on the magnitude of the readings: from 0 to 10%, the precision is ± 0.5%, whereas from 10 to 100% it is ± 3.0%. After calibration, the LFG-20 was purged with pure nitrogen and the gas in the syringe was injected in it. Gas measurements were done in triplicate. Once the composition of the gases had been analysed, the bags were emptied using a 500 ml syringe. The volumes were then corrected to the standard temperature and pressure.
Water percolation, sampling and characterization

The tests were divided into sequences of flow (water percolation) and no flow, as is the case in actual field conditions. During leaching periods, which occurred 20% of the time, approximately one equivalent volume of voids (EVV) was percolated through the sample under a hydraulic gradient close to unity. One EVV is equivalent to the porosity volume of the sample. In the present case, 1 EVV is equal to approximately 4 litres.

For each EVV collected, the following analyses were performed as described in APHA Standard Method (1995) (APHA or other methods are indicated in parenthesis): pH (4500-H+ B - electrometric method), total solids content (2540-B), total volatile solids (2540-E), dissolved solids (2540-C), dissolved volatile solids (2540-C), the chemical oxygen demand (5220-D), volatile organic acid contents (potentiometric titration – calculated based on alkalinity; DiLallo and Albertson 1961), and the calcium concentration (using a Varian AA1275 atomic absorption spectrometer).

Cell opening and sample exhumation

During the 400 days of the tests, four groups of two cells (all placed at 38°C) were opened after the leaching periods. The control cells, placed at 21°C and at 12°C, were also opened after 400 days. The remaining samples are still being monitored.

When the cells were opened, measurements were taken to calculate the final volume of the sample. Sub-samples were then taken from each of the three layers in which the samples had initially been compacted. The sub-samples were packaged in plastic bags and weighed. The water content and volatile solid content of each layer was determined. The water content was determined by drying at 110±5°C, in accordance with the CAN/BNQ 2501-070-M-86 (Bureau de Normalisation du Québec 1986b) procedure. The volatile matter content was determined.
according to the ASTM D2974-C (ASTM 1987) standard. Further details pertaining to the procedure of cell opening are described by Teixeira (2001).

The mineral composition of the material was performed using a Rigaku D max/B X-ray diffractometer and a 1410-X Ray Spectometer (fluorescence). A crosscheck of the mineral content was done by differential thermogravimetric analysis (TG/DTA). The organic composition (cellulose, hemi-cellulose and lignin) of the DBP used in the experiments was determined by acid hydrolysis, which solubilizes sugar monomers. The sugar monomers were analyzed by High Performance Liquid Chromatography (HPLC) using a Dionex DX-500 apparatus equipped with a Dionex PA-10 column, a PA-10 pre-column and a pulsating amperometric detector. The lignin content was determined from the same hydrolysed samples using a variation of the ASTM E1721-01 Klason lignin method (ASTM 2001).

**Evaluation of mass loss by leaching and gas production**

Mass loss by leaching was determined by multiplying the total solids concentration in the leachate by the volume collected. It is expressed in terms of absolute mass loss and as a percentage of the total mass of the sample. The mass loss by gas production was calculated for each gas sample collected, and from the volumes of dissolved CO₂ and CH₄ in the collected water. The volumes in the gas bags and of the gas dissolved in water were transformed into mass by using the perfect gas law and their molecular weight. The amounts of dissolved gases were calculated by applying Henry’s Law (Tchobanoglous et al. 1993). The mass losses due to gas production were also expressed in terms of absolute mass loss and percentage of the mass of the sample.
Basic geotechnical characterization

The geotechnical characterization includes the determination of water content, void ratio, degree of saturation, relative density and the density water content relationship (Proctor curve). This characterization was done for the fresh samples and for the samples submitted to controlled degradation, i.e. samples collected after cell opening. The Proctor Standard tests were conducted according to the ASTM D698 (ASTM 2000) standard. The specific weight of solids was obtained according to the procedure CAN/BNQ 2501-070-M-86 (Bureau de Normalisation du Québec 1986a).

RESULTS

Evolution of biogas production and composition

Figure 3 presents the total accumulated production of CO\textsubscript{2} and CH\textsubscript{4} as a function of time, for the tests at 38°C. The leaching periods and number of samples considered to calculate the averages are also indicated. In the case of the 1\textsuperscript{st} period, instead of 12 (n=12), only 10 cells (n=10) were considered due to a problem with the gas collection system for two cells.

A total average volume of 60.7 ± 5.4 litres of gas per kilogram of dry DBP (herein noted as l.kg\textsuperscript{-1}.j\textsuperscript{-1}-dry basis) was measured in 400 days. The total CO\textsubscript{2} production measured was 30.5 ± 2.4 l, whereas that of the CH\textsubscript{4} was 23.8 ± 2.7 l. On average, the production of CO\textsubscript{2} and CH\textsubscript{4} is responsible for 91% of the total volume of gas produced. The non characterized 9% very likely consists of other gases formed during anaerobic degradation, such as ammonia, hydrogen, sulphites, mercaptans, and others (Tchobanoglous et al. 1993).

Observation of Figure 3 shows that gas production begins following a rate of 0.5 l.kg\textsuperscript{-1}.j\textsuperscript{-1}-dry basis. CO\textsubscript{2} is by far the most important component. The period between the 15th and 17\textsuperscript{th}
days, that precedes the start of \( \text{CH}_4 \) production, is characterized by a significant slowdown in gas production. Starting on the 27\textsuperscript{th} day, gas production resumed with the appearance of \( \text{CH}_4 \). From the 27\textsuperscript{th} day, the rate of production lowered (0.1 l kg\textsuperscript{-1} j\textsuperscript{-1}-dry basis), as is shown in Figure 3. The rate of \( \text{CH}_4 \) production remained constant until the 400\textsuperscript{th} day.

Figure 4 presents the percentages of \( \text{O}_2 \), \( \text{CO}_2 \) and \( \text{CH}_4 \) in the gas produced over time. It can be observed that the oxygen was rapidly consumed at the beginning of the test, while \( \text{CO}_2 \) production was intense. Around the 20\textsuperscript{th} day, \( \text{CO}_2 \) represented approximately 90 \% of the total gases produced. Methane production started around the 27\textsuperscript{th} day, following a short transition period during which gas production stalled. Its concentration rapidly increased and around the 60\textsuperscript{th} day \( \text{CH}_4 \) represented 40\% of the biogas produced. Around the 90\textsuperscript{th} day, the concentration of \( \text{CH}_4 \) surpassed that of \( \text{CO}_2 \). It is important to note that the leaching periods are responsible for the oscillations of \( \text{CO}_2 \) concentration, due to the dissolution in water.

Figure 5 presents total gas production measured over time, for different temperatures. As in other biological processes, anaerobic degradation is affected by temperature (Christensen and Kjeldsen 1989). As expected, gas production was the highest at 38 \(^\circ\)C. At 21\(^\circ\)C, around 40 l of gas per kg of dry DBP were produced. This production represents two thirds of the production at 38\(^\circ\)C. At 12\(^\circ\)C, the production was clearly lower.

Figure 6 presents the variation over time of the volumetric concentration of \( \text{CH}_4 \) for each testing temperature. Methane appearance occurred on the 27\textsuperscript{th} day for tests at 38\(^\circ\)C, on the 63\textsuperscript{rd} day for tests at 21\(^\circ\)C and on the 158\textsuperscript{th} day for the tests at 12\(^\circ\)C. Once the \( \text{CH}_4 \) production had begun, the concentrations for the tests at 21\(^\circ\)C remained below those observed for the tests at 38\(^\circ\)C. In the case of the tests at 12\(^\circ\)C, the concentration increased with time and rose to around 18\% after 400 days.
Comparison between potential and measured biogas production

Potential biogas production was calculated to establish a production threshold and to be able to compare it to the production measured during the 400 days of the study. This potential was estimated according to the reaction R1, while putting forward the hypothesis that all the cellulose and hemicellulose convert to gas (Barlaz 1997; Barlaz and Ham 1989; Christensen and Kjeldsen 1989; McBean et al. 1995; Tchobanoglous et al. 1993).

\[ C_aH_bO_cN_d + \left( \frac{4a-b-2c+3d}{4} \right)H_2O \rightarrow \left( \frac{4a+b-2c-3d}{8} \right)CH_4 + \left( \frac{4a-b+2c+3d}{8} \right)CO_2 + dNH_3 \]

where the parameters a, b, c and d vary depending on the molecule considered.

Cellulose represents the greatest source for biogas production from DBP, followed by hemicellulose (Figure 1). Considering that lignin is recalcitrant in anaerobic conditions (Young and Frazer 1987), the production of CO\(_2\) and CH\(_4\) resulting from lignin was considered negligible. As a consequence, its potential of biogas production, which represents 6.6 \% of the dry mass of DBP, was not taken into account. As shown in Table 2 the cellulose and hemicellulose masses present in each sample, in the beginning of the tests, can supply 387.5 l of CO\(_2\) and 299.4 l of CH\(_4\). In other words, 1.0 kg (80 \% cellulose, 20 \% hemicellulose) can supply 687 l of gas (CO\(_2\) and CH\(_4\)). A comparison between the total calculated potential and the actual production obtained during the tests is presented in Table 3. The results presented show the accumulated average volumes of CO\(_2\) and CH\(_4\) produced and the respective rates of production for each testing temperature. It is noted that the recovery rates (percentage of total potential) are low and decrease as temperature decreases. The rightmost column in Table 3 shows the total accumulated quantities expressed as a percentage of the estimated potential of gas production.
after 400 days (18.3%, 12.0% and 0.3% for the tests, at 38, 21 and 12°C, respectively). Although temperature influences gas production rates, it is not the only parameter affecting the degradation of DBP; otherwise the gas production rate at 38°C would have been higher. Other limiting conditions affecting gas production rates are discussed in the following.

The tests were designed to reproduce compaction conditions in the field. The implications of compaction on gas production are two fold. First, it leads to a low hydraulic conductivity of the DBP samples (Cabral et al. 2002), which means that the pore water is purged out slowly during the leaching periods. Second, it creates a less effective contact between the solid and liquid phases, as compared to suspensions. Boman and Bergstrom (1985) conducted anaerobic degradation studies of DBP in reactors, where 0.4 g of solids were placed in one liter of water. During the 15 days of the study, Boman and Bergstrom (1985) obtained a biogas production rate of 480 l for each kg of volatile solids, which is nearly five times higher than the 100 l/kg produced in 400 days. However, the solids to liquid ratio in the present study was 1:1.7, i.e. three orders of magnitude lower. This creates quite different degradation dynamics resulting in slower gas production rates.

Other factors can be mentioned to explain the relatively slow gas production rates. One is the high C:N ratio, typical of DBP (129:1, in the case of this study). However, Jackson and Line (1997) did not experience any problems composting DBP with C:N ratios as high as 218:1. Another possible inhibitor would be the presence of toxic elements. Several characterization studies performed with DBP (e.g. Ettala 1993; Teixeira 2001; Trépanier et al. 1996; Vlyssides and Economides 1997) have, however, concluded that DBP cannot be considered toxic.
Evolution of leachate production

Figure 7 shows the variation over a period of 400 days of pH, COD, volatile organic acid (VOA) content in the leachate, total solids (TS) and total volatile solids (TVS). The leaching periods and the number of samples (n) considered in the calculation of the averages are also indicated. The pH increased from 5.5 in the beginning of the 1st leaching period (Figure 7a) to approximately 6.5 early during the percolation of the 2nd EVV. COD and VOC (Figure 7a), as well as TS and TVS (Figure 7b), show the same evolution, i.e. a significant decrease after the leaching of the 1st EVV. After the leaching of the 2nd EVV, the concentrations remained the same until the end of the tests.

During leaching of the 1st EVV, the total solids concentration in the leachate quickly decreased from its initial value of approximately 25 000 mg/l to 11 000 mg/l. At the beginning of the second leaching period, undertaken after an interval of 40 days without water percolation, the total solids concentration was as low as 4 000 mg/l and decreased even further to around 1 400 mg/l at the end of the 2nd EVV. According to Teixeira (2001), approximately 93% of the total solids are found in the dissolved form.

COD and VOC also decreased between the 1st and 2nd leaching periods. This decrease is explained by the use of dissolved carbon sources in the leachate by micro-organisms, resulting in gas production. Once CH₄ started to be produced, the total gas production rate remained constant (Figure 3), although the proportion of CH₄ in the gas quickly increased (Figure 4).

As can be observed in Figure 7, the ratio of volatile solids to total solids was 0.53 at the beginning of the 1st leaching period. In other words, a little over half of the solids consisted of volatile solids. By the beginning of the 2nd leaching period, the concentration of TS and TVS had decreased to approximately a third of the value obtained at the end of the 1st period and the
TVS/TS ratio slightly decreased to 0.5. The loss of volatile solids can be explained by gas production (Figures 4 to 6), whereas the decrease in non-volatile solids can be partly associated with carbonate precipitating out from the leachate due to an increase in pH and decrease in VOC (Figure 7b). In addition, carbonates in the leachate are still transformed into CO₂ due to the slightly acidic pH. The TVS/TS ratio reached a value of 0.25 at the end of the leaching of the 4th EVV.

Figure 8 shows the evolution of the pH, TS, COD and VOC as a function of time testing temperature. At the beginning of the tests at 38°C, the pH was close to 5.5 (the lowest value) then increased to reach approximately 6.5. For the tests at 21°C and 12°C, the starting pH was higher than at the beginning of the tests at 38°C. However, near the end it remained a little bit lower (around 6.0). A decrease in TS and TVS after leaching of the 1st EVV was observed for all temperatures. The greatest drop was observed for the cells at 38°C, where, at the end of the 4th leaching period, the TS concentration was 1400 mg/l, whereas for the tests at 21°C and 12°C, the TS concentrations remained respectively at 8800 mg/l and 11000 mg/l. COD and VOC followed the same change pattern observed for the TS with respect to the influence of temperature.

**Mass loss by leaching and gas production**

Figure 9 presents the evolution of the total mass loss. At the end of the 1st leaching period, the mass loss due to leaching (4 %) represents 67 % of the total loss (6 %). After the 1st leaching period, mass loss by leaching becomes negligible (Figure 9). This implies that non-volatile solid loss also becomes negligible. At the end of the 400 days, the total mass loss was approximately 12%. The loss from gas production increased from 2%, during leaching of the 1st
EVV to 8% after 400 days. Therefore, it represents 66% of the total loss. The loss from gas production occurred even during periods without leaching.

Figure 10 presents the total mass loss as a function of the testing temperature. The results show that there is no marked difference between the total mass loss at 38°C and 21°C. The loss at 12°C represents approximately half of those observed for the cells at 38°C and 21°C. Considering the low gas production (Figure 5), the mass loss in the cell at 12°C can be considered principally caused by leaching.

The mass loss obtained by leaching and gas production was compared to the mass loss obtained directly, i.e. by subtracting the total dry weight of the sample at the opening of the cells from its weight in the beginning. Figure 11 presents the mass losses obtained from tests performed at 38°C, a much higher temperature than what is experienced within DBP covers, in temperate climates. As a consequence, the tests at 38°C represent a “worst case scenario” in terms of mass loss that might affect the performance of the DBP layer. As shown in Figure 11, the losses are expressed in terms of variation of dry mass and of percentage of dry mass lost from DBP. The indirect mass loss accounts for an average of 78% of the mass loss calculated directly. The latter represents the maximum possible (or 100%) loss at any stage. Several potential sources of error may explain the origin of these differences, including leakages from the gas bags. According to the mass report following a laboratory study on the degradation of domestic wastes, Barlaz and Ham (1989) identified errors associated with various measurements (volume and gas composition), precision of physico-chemical analyses and unaccounted carbon converted to cell mass.
Impact of biodegradation on geotechnical parameters and properties

For mineral soils, volume changes are related to changes in void ratio by the following equation:

\[
\frac{\Delta V}{V} = \frac{\Delta e}{1 + e_0}
\]

However, the results presented in Table 4 show that the equality in Equation 2 does not hold for DBP. Given that the samples were not subjected to any confining load, small increases in volume were observed (less than 2% in all but one case). These volume changes were accompanied by a 10 to 25% decrease in density and 17 to 30% increase in void ratio. The values obtained for the right-hand side of Equation 1 are many times greater than those found for \(\Delta V/V\). Increases in void ratio were greater for samples that were subjected to biodegradation for a longer period of time. The increase in void ratio without a corresponding change in volume can only be explained by the loss of solid matter due to leaching and gas production associated with biodegradation (Figure 10).

From these results, it can be concluded that without confinement of the deinking by-product layer, an increase in its hydraulic conductivity with time can be expected. In several sites in Quebec where DBP have been used as part of the cover system, a 30-cm thick soil layer (equivalent to a 10 kPa overburden) was placed over the DBP layer (Audet et al. 2002; Cabral et al. 2002). Field results (unpublished data) and laboratory experiments with large (0.6 x 0.6 x 0.5 m) samples (Gharbi 2002) showed that such level of confinement is sufficient to induce a continuous compression of the DBP layer with time, thus resulting in a reduction in hydraulic conductivity.
Figure 12 shows that the decrease in volatile matter content resulting from the degradation of DBP causes an increase in the water content of the samples, therefore a decrease in the dry mass of the DBP. Each point represents the average of 18 points (9 points per cell, two cells open at a time). The values for t = 0 represent the averages of the eleven cells (Table 4). Considering that the percentage of variation of the water content is always greater than the percentage of the void ratio increment, the degree of saturation must increase accordingly. In all cases, the relationship $e_{Sr} = Dr.w$ is respected. Furthermore, the relative density increases very little, with the exception of cells 9 and 10, for which the greatest variations in mass, volume, density and void ratios were observed. In this case, the loss in organic matter (less dense than the loss of mineral matter) could be at the origin of this increase in relative density.

The density-water content relationship (Standard Proctor curve) for sample KF 08/06/99 is presented in Figure 13, where the data obtained for the different samples tested after their exhumation can also be found. It can be observed that the biodegradation of the samples did not significantly influence the density-water content relationship, since the values obtained remain similar to that determined for fresh DBP.

GENERAL DISCUSSION ABOUT THE EVOLUTION OF BIODEGRADATION AND ITS IMPLICATIONS FOR A DBP COVER

The evolution of biogas production and of the quality of the leachate collected during the tests was qualitatively compared to the behaviour dictated by the models routinely used to establish the evolution of anaerobic degradation in sanitary landfill sites. According to these models, the stabilization process is divided into four or five phases described below (Barlaz and Ham 1989; Farquhar and Rovers 1973; Fuller et al. 1979; Tchobanoglous et al. 1993). It must be
noted, however, that the conditions under which degradation occurs in cover systems and in MWDF are rather different. Indeed, DBP are compacted to minimize the hydraulic conductivity and are placed with a much higher degree of saturation than solid wastes. Under more saturated conditions, gas exhaustion is slowed, which in turn slows the degradation process itself, thus the amount of biogases generated.

Figure 14 presents the evolution of the gas and leachate composition during the first 150 days of the series of tests at 38°C. The same degradation behaviour of solid wastes buried in MWDFs (phases I to IV) is observed. During Phase I (aerobic), which lasted 5 days, organic substances, such as sugars and volatile fatty acids, are easily metabolized by fungi and aerobic bacteria resulting in quick oxygen consumption. During Phase II (acidic anaerobic; hydrolysis and fermentation), which took place between the 6th and 37th day, there was an intense production of CO\textsubscript{2} and the production of a leachate with a high VOC content. Phase III (unstable methanogenic), which began around the 37th day and ended on the 113th day, represents the transition between the acid and stabilization phases of CH\textsubscript{4} production. During this phase, the volumic concentration of CH\textsubscript{4} increased rapidly and around the 75th day, represented 40 % of the gas formed. From the 113th day, it can be observed that the process had reached Phase IV (stable methanogenic), during which the CH\textsubscript{4} concentration is greater than that of CO\textsubscript{2} and the biogas production rate due to degradation of the DBP remains constant and the concentration of organic matter in the leachate is low. This low concentration of dissolved organic matter seems to indicate that the consumption of organic substances occurs as it is solubilized. Thus, it appears that there would be no volatilization from DBP. However, the methanogenic phase does not represent the end of hydrolysis and fermentation. Hydrolysis and fermentation continue, but the methanogenic population increases until the point where the consumption rate of the final products
approaches the hydrolysis rate. In other words, “methanogenic syntrophy” is reached, i.e. the point in time from which the degradation process steps are in equilibrium (McBean et al. 1995).

Considering the steady gas production after 400 days of the study, it is clear that Phase V, which is characterized by the end of the degradation process, i.e. the end of CH₄ and CO₂ production, has not yet begun. Regarding the tests at 21°C and 12°C, it is possible to state that the controls at 21°C have reached Phase IV (see Figures 6 and 14a), but that they are slightly delayed compared to the tests at 38°C. There still remains a certain “accumulation” of dissolved substances (Figure 8) and the gas production is lower than that obtained for the cells at 38°C (Figure 5). The cell at 12°C produced a negligible amount of gas (Figure 5). Since there is no transformation of the dissolved substances to gas, the leachate remains loaded even after the leaching periods. It can be suggested that hydrolytic and fermentative bacteria are less sensitive to temperature and continue to hydrolyze as long as the DBP are leached out of the system.

The degradation of organic materials, such as DBP, can be explained, in a simplified way, as a transfer of mass between three phases or physical states: solid, liquid and gas. This transfer also summarizes in a certain way, the methanogenic syntrophy phases (Augenstein and Pacey 1991; Barlaz and Ham 1989; Farquhar and Rovers 1973; Fuller et al. 1979; Rees 1980). The solid phase is the source of carbon and nutrients for the development of micro-organisms, while the liquid phase (water) occupies the voids. As the degradation advances, solubilization of the solid components of the DBP occurs by degradation and/or chemical solubilization. The solubilization changes the characteristics of the liquid phase, which becomes a leachate loaded with organic and dissolved inorganic substances. The third phase, known as the gaseous phase, is highly dependent on the liquid phase, since CH₄ production needs simple carbon sources, which become available through hydrolysis and fermentation.
As shown in Figure 15, the micro-organism-mediated interactions between these three phases (gaseous, liquid and solid) result in mass loss over the long term, both through leaching and gas production. In the beginning of the degradation process, the mass loss is mostly caused by the solubilization of the substances in the solid phase. This loss causes an accumulation of dissolved substances in the leachate, which is characteristic of the acidic phase of anaerobic digestion. In the case of the laboratory tests, a part of the accumulated substances was leached out. In the case of the 1st leaching period, the mass loss was significant (Figure 9). In the absence of percolation, mass loss occurs by gas production (Figure 9).

**Biodegradation in the field**

Analysis of unpublished field results by Teixeira (2001) show that the pore water is not purged as fast as in the laboratory. According to calculations made by Teixeira (2001) using lysimeter data (Table 5), if only vertical percolation by advection is considered, it can be estimated that the pore water would have advanced around 0.8 m in 4 years, i.e. 40 % of the total thickness of a 2-m thick DBP layer above the lysimeter. The mass loss (dry basis) would be approximately 0.6 % per year, clearly indicating that mass loss by leaching is a slow process. It can, therefore, be evaluated that at least 10 years would be necessary for one EVV to be completely leached out of the system in the field. This would correspond to an accumulated mass loss of 6 %.

In Table 5, the flow rate considered (35 l/year) was taken from calculations made by Cabral et al. (2002). The percentage of mass loss equals the ratio between the mass lost and the total solid mass in the DBP column from the top of the lysimeter until the surface. The
concentration of total solids, 30 000 mg/l, corresponds to the concentration found at a depth of 1.0 m in observation wells, in the beginning of the monitoring period (Teixeira 2001).

Gas production rates in DBP covers constructed in Quebec have not been determined and the study of dynamics of mass loss under field conditions requires further investigation. But, given the slow pace of leaching, the high CH$_4$ concentrations obtained below actual DBP layers (from 40 to 60% according to Audet et al. 2002, i.e. within the range that is characteristic of phases III and IV of anaerobic digestion), and the decrease in concentration of several parameters mentioned above, it is suggested that mass loss in compacted DBP layers would be mainly associated with gas production.

Gas production in the field can be roughly estimated using the rate of production determined in the laboratory for the test at 12°C, which is the one that best represents actual conditions within a DBP cover layer. According to Teixeira (2001), the rate of gas production at 12°C is in the vicinity of 0.8%/yr (CH$_4$ and CO$_2$ combined), whereas for tests performed at higher temperatures, a rate of up to 4.2%/yr was obtained. Given the fact that temperatures can reach 25°C at a depth of one meter inside a DBP layer, it would be reasonable to consider a rate of production between 0.8%/yr and 4.2%/yr.

CONCLUSIONS

This paper presented a laboratory testing program whose main goal was to evaluate the accelerated biodegradation behaviour of DBP, a material rich in organic matter that is increasingly used in cover systems at mining tailings and landfill sites. The experimental program included a series of tests performed under three different temperatures: 12, 21 and 38°C. The first simulated field conditions, the second simulated the temperature that prevails in the
upper part of the layer of DBP during summer and fall, whereas the last (38°C) is the ideal
temperature to accelerate the anaerobic degradation process. The evolution of gas and leachate
production (quality and quantity) was monitored for 400 days. During four periods of
approximately one month, deaerated, deionized, distilled water was leached through the samples.

Overall, the results obtained showed that the biodegradation evolution of the DBP placed
as part of a cover system may result in: oxygen consumption, production of a leachate loaded
with organic and dissolved inorganic substances, gas production, mass loss (from the DBP
material itself), changes in void ratios and changes in water content.

The production of an organic rich leachate is an important issue, as far as the use of DBP
in mine tailing reclamation projects is concerned, both in terms of the longevity of the barrier
and its effectiveness as O₂ barrier. According to the results obtained, the level of biodegradation
reached in the laboratory after 400 days of testing (expressed, among others, in the form of mass
loss) does not seem to adversely affect the behaviour of a DBP cover.

The evolution of the production of biogas and leachate during the DBP degradation tests
followed the behaviour of anaerobic degradation in sanitary landfill sites. In the beginning
(Phase I), O₂ was consumed quite rapidly. During Phase II, there was a significant accumulation
of dissolved substances in the leachate. This concentration tended to decrease with time (Phases
III and IV), as a result of gas production and leaching. Once the process reached equilibrium, a
greater accumulation of dissolved substances in the leachate, due to the transformation to gas,
(cell at 38°C and 21 °C) was observed. In fact, the laboratory tests at 38 °C and at 21 °C
demonstrated that, even during the periods without leaching, there was a decrease in the
concentration of dissolved materials, which was caused by gas production at rates that quickly
became constant. The final stabilization phase (Phase V), where degradation stops, was not
reached during the 400 days of monitoring. Consequently, it wasn’t possible to evaluate the extent of the ultimate degradation.

At the end of the tests, the mass losses by leaching (between 5% and 6%) were similar for all the tests, without respect to temperature. In the long term, at 38°C, the mass loss due to gas production was greater than the loss from leaching (8% for the former, 5% for the latter). The mass loss from leaching and gas production corresponds to approximately 78% of the loss calculated by the difference of the solid mass before and after each test. This recovery rate can be considered satisfactory given the numerous potential sources of error associated with this type of study. Among the identified sources of error are those associated with measurements (volume and gas composition), uncertainties associated with the physico-chemical analysis methods and limitations of the methods for estimating dissolved gas concentration, potential gas leaks, etc.

Monitoring of the evolution of selected geomechanical and hydraulic parameters showed that the degradation caused an increase in water content, which was expected. There was a slight swelling of the DBP (due to the absence of a minimum overburden) and a substantial increase in void ratio at the end of the tests. The increase in void ratio was greater for the tests that lasted longer. The increase in void ratio without a corresponding change in volume can only be explained by loss of solid matter due to leaching and gas production. Normally, an overburden of around 10 kPa would cause a decrease in the void ratios, thus of hydraulic conductivity.

ACKNOWLEDGEMENTS

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Table 1. Average conditions of the DBP placement in the cells

<table>
<thead>
<tr>
<th>Parameter</th>
<th>PVC cells (14 cells)</th>
<th>Plexiglas cell (12°C) (1 cell)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total height (cm)</td>
<td>17.9 ± 0.1</td>
<td>15</td>
</tr>
<tr>
<td>Sample volume (cm³)</td>
<td>5 628.4 ± 32.2</td>
<td>2717.6</td>
</tr>
<tr>
<td>Dry mass (g)</td>
<td>2 317.7 ± 24.9</td>
<td>1091</td>
</tr>
<tr>
<td>Dry density (kg/m³)</td>
<td>411.8 ± 5.8</td>
<td>401.5</td>
</tr>
<tr>
<td>Water content (%)</td>
<td>172.3 ± 3.1</td>
<td>180.6</td>
</tr>
<tr>
<td>Degree of saturation (%)</td>
<td>89.9 ± 0.6</td>
<td>91.3</td>
</tr>
<tr>
<td>Void ratio</td>
<td>3.7 ± 0.1</td>
<td>3.9</td>
</tr>
</tbody>
</table>
Table 2. Stoichiometric potential of CO$_2$ and CH$_4$ production per kg of cellulose and hemicellulose in the DBP placed in the cells

<table>
<thead>
<tr>
<th></th>
<th>Cellulose</th>
<th>Hemicellulose</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass (kg)</td>
<td>0.8</td>
<td>0.2</td>
<td>1.0</td>
</tr>
<tr>
<td>CO$_2$ (l)</td>
<td>304.5</td>
<td>83.0</td>
<td>387.5</td>
</tr>
<tr>
<td>CH$_4$ (l)</td>
<td>237.8</td>
<td>61.6</td>
<td>299.4</td>
</tr>
<tr>
<td>Total CO$_2$ + CH$_4$ (l)</td>
<td>-</td>
<td>-</td>
<td>686.9</td>
</tr>
<tr>
<td>Sample</td>
<td>Volume ( \text{CO}_2 ) (l)</td>
<td>( \text{CO}_2 ) (l/kg dry material)</td>
<td>Volume CH(_4) (l)</td>
</tr>
<tr>
<td>------------------------</td>
<td>-------------------------------</td>
<td>--------------------------------------</td>
<td>---------------------</td>
</tr>
<tr>
<td>Total potential for sample at start of tests</td>
<td>387.5</td>
<td>-</td>
<td>299.4</td>
</tr>
<tr>
<td>(Equation 3-1)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Samples at 38 °C</td>
<td>70.7</td>
<td>30.5</td>
<td>55.2</td>
</tr>
<tr>
<td>Samples at 21 °C</td>
<td>51.9</td>
<td>22.4</td>
<td>30.5</td>
</tr>
<tr>
<td>Sample at 12 °C (^1)</td>
<td>2.0</td>
<td>0.9</td>
<td>0.2</td>
</tr>
</tbody>
</table>

\(^1\) Values already adjusted considering that the mass of the sample tested at 12°C is lower than the mass of the other samples.
Table 4. Variation of geotechnical parameters in function of cell opening

<table>
<thead>
<tr>
<th>Cell opening (elapsed time in days)</th>
<th>Cell numbers</th>
<th>V initial (V final) cm$^3$</th>
<th>ΔV/V initial (%)</th>
<th>Cell opening numbers</th>
<th>Initial density (final) kg/m$^3$</th>
<th>$e_0$ (e final)</th>
<th>$\Delta e/(1+e_0)$ (%)</th>
<th>w initial (w final) (%)$^1$</th>
<th>Sr initial (Sr final) %</th>
<th>Dr initial (Dr final) $^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1$^{st}$ (54)</td>
<td>2</td>
<td>5.600 (NM)</td>
<td>416 (NM)</td>
<td>3.691 (NM)</td>
<td>200.1 (NM)</td>
<td>170.1 (NM)</td>
<td>89.8 (1.96)</td>
<td>1.95</td>
<td>1.95</td>
<td></td>
</tr>
<tr>
<td></td>
<td>11</td>
<td>5.663 (NM)</td>
<td>408 (NM)</td>
<td>3.777 (NM)</td>
<td>172.6 (NM)</td>
<td>196.8 (NM)</td>
<td>89.1 (1.95)</td>
<td>1.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2$^{nd}$ (146)</td>
<td>5</td>
<td>5.632 (5.734)</td>
<td>412 (361)</td>
<td>3.736 (4.408)</td>
<td>172.3 (205.0)</td>
<td>89.9 (1.95)</td>
<td>(1.95)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>5.663 (5.742)</td>
<td>409 (359)</td>
<td>3.763 (4.462)</td>
<td>172.1 (207.1)</td>
<td>89.2 (1.95)</td>
<td>90.6 (1.95)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3$^{rd}$ (272)</td>
<td>3</td>
<td>5.585 (5.679)</td>
<td>416 (350)</td>
<td>3.687 (4.589)</td>
<td>171.8 (203.2)</td>
<td>90.9 (1.95)</td>
<td>(1.96)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>7</td>
<td>5.648 (5.719)</td>
<td>415 (349)</td>
<td>3.694 (4.678)</td>
<td>169.7 (212.1)</td>
<td>89.6 (1.95)</td>
<td>(1.96)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4$^{th}$ (400)</td>
<td>6</td>
<td>5.663 (5.692)</td>
<td>402 (346)</td>
<td>3.851 (4.694)</td>
<td>178.6 (211.8)</td>
<td>90.4 (1.95)</td>
<td>(1.96)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Control at 21°C (400)</td>
<td>9</td>
<td>5.600 (5.610)</td>
<td>418 (351)</td>
<td>3.669 (4.694)</td>
<td>169.4 (224.5)</td>
<td>89.9 (1.95)</td>
<td>(2.0)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>5.632 (5.837)</td>
<td>418 (341)</td>
<td>3.670 (4.954)</td>
<td>168.0 (225.6)</td>
<td>89.3 (1.95)</td>
<td>(2.03)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Control at 12°C (400)</td>
<td>15</td>
<td>2.718 (2.765)</td>
<td>401 (358)</td>
<td>3.858 (4.502)</td>
<td>179.0 (211.0)</td>
<td>91.28 (1.95)</td>
<td>(2.03)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^1$ Mean of 9 test results per cell

$^2$ Mean of 1 test result per cell

NM: not measured
Table 5. Lysimeter data (Eustis site; lysimeter L1) used to make preliminary estimations of mass loss in the field.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Data</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry density in place (kg/m$^3$)</td>
<td>400</td>
</tr>
<tr>
<td>Height of the DBP column above the lysimeter (m)</td>
<td>2.0</td>
</tr>
<tr>
<td>Lysimeter area (m$^2$)</td>
<td>0.23</td>
</tr>
<tr>
<td>Total volume of DBP above the lysimeter (m$^3$)</td>
<td>0.46</td>
</tr>
<tr>
<td>Solid mass (kg)</td>
<td>184</td>
</tr>
<tr>
<td>Total solid concentration in the water (mg/l)</td>
<td>30 000</td>
</tr>
<tr>
<td>Accumulated volume of water in one year (l)</td>
<td>35</td>
</tr>
<tr>
<td>Operational hydraulic conductivity ($k_{op}$) (cm/s)</td>
<td>$5 \times 10^{-7}$ cm/s</td>
</tr>
</tbody>
</table>

Note: Hypothesis of 1-D flow through a totally operational lysimeter.
List of Figures

Figure 1. Average composition of the DBP used in the experimental program (% by weight)

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Figure 6. Evolution of the volumetric concentration of CH4 in the biogas for different testing temperatures

Figure 7. Evolution of: (a) COD, VOA, and pH; (b) TS, TVS and the ratio TVS/TS

Figure 8. Evolution of the: (a) pH; (b) TS; (c) COD; and (d) VOA in the leachate as a function of the testing temperature

Figure 9. Evolution of the total mass loss (gas + leaching)

Figure 10. Evolution of the total mass loss (gas + leaching) as a function of the testing temperature

Figure 11. Comparison between measured total mass loss (gas + leaching) and mass loss obtained by weighing the samples before and after the tests at 38 ºC

Figure 12. Evolution of the water content and of the volatile matter

Figure 13. Density-water content relationship (Proctor curve) for the DBP tested

Figure 14. Evolution of (a) gas composition; and (b) pH, COD, VOC and Ca++ in the leachate

Figure 15. Schematic representation of the evolution of mass loss
39.7% NON VOLATILE

- Calcite
- Talc 1.4%
- Metakaolin 3.9%
- Rutile 1.9%
- Residual alumine 2.6%
- Other 0.7%

60.3% VOLATILE

- Cellulose 34.0%
- Hemicellulose 9.2%
- Lignin 10.5%
- Other 6.6%
- Other 0.7%
- Residual alumine 2.6%
The graph shows the total accumulated CO$_2$ and CH$_4$ (l/Kg dry material) over time (days). The data is represented with error bars, and the graph includes the following information:

- Total gas
- CO$_2$
- CH$_4$

The leaching period is indicated by a horizontal line on the graph.

The trend lines are given by the equations:

- y = 0.52x + 2.18 with $R^2 = 0.972$
- Average: y = 0.14x + 7.70 with $R^2 = 0.991$

 (*) Two cells not considered due to problems with the gas collection system.
(*) Two cells not considered due to problems with the gas collection system
(*) Two cells not considered due to problems with the gas collection system
The graph illustrates the volumetric composition of CH₄ (% over time) at different temperatures (38 °C, 21 °C, and 12 °C) during the leaching period. The data points are denoted as follows:

- **38 °C**: Squares (■)
- **21 °C**: Crosses (×)
- **12 °C**: Triangles (△)

The graph shows the time in days along the x-axis and the volumetric composition of CH₄ (%) along the y-axis. The leaching period is indicated by a horizontal line on the graph.

(*) Two cells not considered due to problems with the gas collection system.
(a) pH vs. time for leaching periods at 38 °C, 21 °C, and 12 °C.

(b) Total solids (TS) vs. time for leaching periods at 38 °C, 21 °C, and 12 °C.

(c) Chemical oxygen demand (COD) vs. time for leaching periods at 38 °C, 21 °C, and 12 °C.

(d) Volatile solids (VOA) vs. time for leaching periods at 38 °C, 21 °C, and 12 °C.
Mass loss (%) by gas production and leaching

- 38 °C
- 21 °C
- 12 °C

leaching period

Time (days)

n = 12  n = 10  n = 8  n = 6

1st EVV  2nd EVV  3rd EVV  4th EVV

Gas only
(a) Water content, $w$ (%)

(b) Dry density, $p_d$ (kg/m$^3$)

- fresh
- exhumed from cell 2
- exhumed from cell 11
- exhumed from cell 12
- exhumed from cell 5
- exhumed from cell 3
- exhumed from cell 7
- exhumed from cell 6
- exhumed from cell 9
- exhumed from cell 10
- exhumed from cell 4

$Sr = 80\%$

$Sr = 100\%$
Graph (a) shows the gas volumetric composition (%) with different phases labeled as follows:

I. Aerobic
II. Acidic
III. Unstable methanogenic
IV. Stable methanogenic

Graph (b) presents the concentration in the leachate (mg/l) over time (days), with key markers labeled as follows:

- $\text{CO}_2$
- $\text{CH}_4$
- $\text{O}_2$

The leaching period is divided into two EVV stages.

The pH values are indicated with a range from 4.0 to 7.0.
Deinking by-product

Mass loss

Leaching

Gas

Time