Compressibility of a residual soil when percolated by acidic waters

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ABSTRACT
One dimensional loading tests were completed in samples of a residual soil (Botucatu Formation) after the percolation of distilled water and acidic waters. The objective of this study was to investigate potential changes in the compressibility of the soil due to the percolation of AMD, simulating in the laboratory the use of the proposed soil in the construction of liners or dykes in mining areas. The results show that percolation by acidic waters returned in higher settlements than in samples percolated only by distilled water for the same levels of loading. It was observed that the fine size particles of the samples migrated towards the flux direction during the tests with acidic waters which returned in higher porosity at the top of the samples.

RÉSUMÉ
Des essais sous sollicitation uniaxiale ont étés effectués sur des échantillons d´un sol résiduel (Formation de Botucatu) après percolation par eau distilée et/ou drainage minier acide – AMD. L´objective de cette étude était d´examiner des modifications éventuelles de la compressibilité du sol dues a la percolation de l´AMD, em simulant en laboratoire l´utilisation du sol proposé dans la constrution de soutènements ou digues dans lês zones minières. Les résultats montrent que la percolation par AMD s´est traduite par des tassements plus importants comparés aux échantillons percolés seulement par eau distillée soumis aux mêmes niveaux de sollicitation. Une émigration des particules fines des échantillons suivant la direction de l´écolement a été observée lors des tests avec AMD, ce qui sést traduit par une porosité plus élevée dans les couches supérieures des échantillons.

1 INTRODUCTION

Acidic drainage waters, containing high concentrations of dissolved metals, result from the oxidation of sulphide minerals within mines and mine wastes. The most abundant sulphide minerals in mine wastes are the iron sulphide minerals pyrite (FeS_2) and pyrrhotite ($Fe(1-x)S$). Because of their low commercial value, these minerals are excluded from concentrates in the ore beneficiation process and are deposited in waste rock piles and mine tailings impoundments (Blowes, 1997).

It is well known that mine deposits have high amounts of non-commercial mineral substances associated with the ore. During the process, elevated volumes of mining materials are rejected resulting in the production of large amounts of tailings with high mineral contents (including pyrite) with no commercial value (Knop, 2007). Due to the low technological process for extracting the minerals from the ore body, some of the abandoned mine tailings still have high concentration of copper, zinc, manganese, gold and other metals. The low investment in the mining industry, the low cost of some metal products due to the economic crisis, the abundance of mine bodies around the world and the high liability involved in the process are some of the reasons that reprocessing tailings bodies are not a viable business in most of the cases. Hopefully governmental incentives and the recovery of the economy would allow tailings areas to be reprocessed in the future which would associate environmental interests and profitability.

The generation of low-pH waters due to oxidation of the tailings and subsequent acidification of water bodies downstream of the tailings cause significant impacts in the environment around the world. Changes in pH, the equilibrium between soil-air-water-waste and the life in the surrounding areas also change. The most common environmental problem due to acidification of surface and groundwater is the consequent high solubilization rates of heavy metals and metalloids in water such as iron, manganese, arsenic, lead, zinc, copper and nickel, as well as high concentrations of sulphates.

The pH of the acid mine drainage from pyritic wastes ranges from 0.8 to 4.0 in most of the sites. During the summer, high evaporation rates and the exposure of the tailings to the atmosphere and moisture results in more concentrated AMD in terms of dissolved metals and low pH. Pyrite contains about 46% of iron and more than 50% of sulphur dioxide; in the 4% remaining, materials such as arsenic and nickel can be found (Daniels & Stewart, 1992). In tailings bodies, pyrite is associated with the metals that could not be extracted from the ore body due to low technology used in some extraction processes.

Pyritic wastes (tailings) have limited environmental impact if there is no water (e.g., humidity) and/or oxygen available for oxidation to occur (Nordstrom, 1979). Most of the mine closure activities in Canada and around the world involve soil covers to decrease the volume of water within the tailings and the oxygen and water covers, to reduce or even eliminate the presence of oxygen necessary to allow oxidation to occur.

The process of pyrite oxidation occurs according to the Equations 1 to 5:

$$4FeS_2(solid) + 14O_2(gas) + 4H_2O(liquid) \rightarrow 4Fe^{+2}(aqueous) + 8SO_4^{-2}(aqueous) + 8H^+ \quad [1]$$

$$4Fe^{+2}(aqueous) + O_2(gas) + 4H^+(aqueous) \rightarrow 4Fe^{+3}(aqueous) + H_2O(liquid) \quad [2]$$

$$4Fe^{+3}(aqueous) + 12H_2O(liquid) \rightarrow 4Fe(OH)_3(solid) + 12H^+(aqueous) \quad [3]$$

$$4FeS_2(solid) + 15O_2(gas) + 14H_2O(liquid) \rightarrow 4Fe(OH)_3(aqueous) + 8SO_4^{-2}(aqueous) + 16H^+ \quad [4]$$

Fe^{+3} hydrolysis reaction generally occurs in the initial stages of pyrite oxidation limiting the activity of free Fe^{+3} in the solution. However, as acidity accumulates in the environment and consequently pH decreases to values lower than 3.5, the hydrolysis reaction is limited, increasing the concentration of Fe^{+3}significantly. In such conditions, Fe^{+3} will act as an electron acceptor, becoming the main mechanism of sulphur oxidation, as shown in Equation 5.

$$4FeS_2 + 14Fe^{+3} + 8H_2O \rightarrow 15Fe^{+2} + 2SO_4^{-2} + 16H^+ \quad [5]$$

Once the pH of the environment is sufficiently acidic, Fe^{+3} becomes the main oxidizer of pyrite and reduces it to Fe^{+2}. In acid conditions corresponding to pH values less than 3, oxidation of pyrite due to Fe^{+3} occurs up to 100 times faster than due to O_2 (Jambor & Blowes, 1994, Knop, 2007). The process becomes capable of generating higher amounts of H^+. Under these conditions, other techniques should be considered in the remediation of tailings areas, such as pH improvements before or during the closure of the mine to reduce the oxidation by $Fe^{+3.}$

At several locations in Canada and around the world, tailings are/were disposed directly in the environment, and the acid mine drainage generated with time is treated with lime slurry or limestone channels, depending on the volume of water to be treated and concentration of metals and sulfates dissolved in the water. Typically the scenario involves the placement of tailings in depression areas over peat at locations near the mill site.

Due to the potential lack of borrow sources of clay at several locations around the world for constructing containment liners and the high cost of geosynthetics and budget limitations, the use of residual soils is considered one option (or the only option) for constructing bottom/cover liners. Thus, the objective of this research is to investigate some of the effects in a residual soil when percolated by AMD under different scenarios (hydraulic gradients, concentrations and soil amendments) through one-dimensional loading tests, as described in the experimental program – section 2 of this paper.

2 EXPERIMENTAL PROGRAM

2.1 Experimental Conditions

Well-controlled one-dimensional loading tests were completed on compacted soil samples permeated with neutral to acidic waters to analyze potential changes in the compressibility of the soil under different conditions. Each of the compacted soil types were permeated with distilled water that contained either 0, 2, 6 or 10% of sulphuric acid dissolved in distilled water under a hydraulic gradient of 10 and 20. Sulphuric acid was employed to decrease the percolate pH to represent the drainage waters emanating from oxidized pyrite waste rock. In total, six different loading tests were completed to understand the effect of the acidity in the compressibility of the samples. The experimental program is provided in Table 1. The pH of the solutions for 0, 2, 6 and 10% of sulphuric acid added to the solution was 5.7, 1.8, 1.1 and 0.7 respectively.

Table 1. Experimental Program.

Sulphuric acid added to the solution (%)	0; 2; 6; 10
Hydraulic gradient	10; 20
Bulk density of the sample (kN/m^3)	17.6
Initial porosity	0.34
Total of tests carried out	18

2.2 Soil Properties

The soil used for the experiments is a residual soil from Botucatu formation (BRS), sampled in a natural deposit in the metropolitan region of Porto Alegre, Southern Brazil. This soil is relatively well characterized as a result of recently completed studies (Heineck et al., 2005; Knop, 2003; Knop, 2007).

The soil has liquid limit of 23.8%, plasticity limit = 13% (ASTM D4318-05) and density of solids (G_s) = 26.5 kN/m^3 (ASTM D4892-89). Any changes in these soil index properties will be quantified by analyzing the Atterberg limits and grain size of the soil particles after the soil is permeated with acid solution.

Each soil sample was compacted to the maximum standard Proctor (ASTM D698-07), dry unit weight of soil mass (γ_d = 17.6 kN/m^3) and optimum water content ($w_{optimum}$ = 14.2%).

The material contains approximately 5.0% clay, 36.4% silt, and 58.6% sand (52.6% fine sand and 6.0% medium sand), as observed in Figure 1. Thus, the soil is classified as silty sand (SM) according to Unified Classification System (ASTM D2487-93).

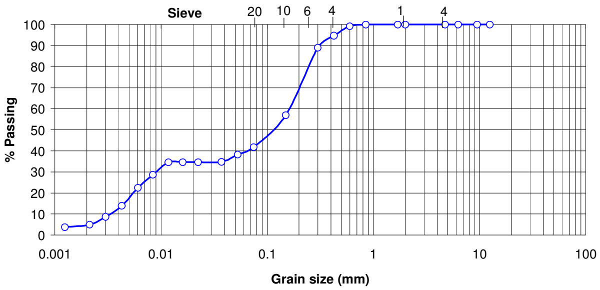

Figure 1. Grain Size Distribution – Botucatu Formation Residual Soil.

2.3 Equipment

A stainless steel column, which contains a 7 cm diameter and 10 cm long soil sample, was manufactured for this study (see Figure 2). The column is equipped with a top cap that allows liquid to enter the sample, as well as a piston entrance to transmit a load to a loading cap placed at the top of the sample. The column was installed in conventional consolidation equipment, as observed in Figure 3. An effluent exit was located at the base of the sample. A polypropylene filter was used at the bottom of the specimens to avoid the loss of soil out of the equipment.

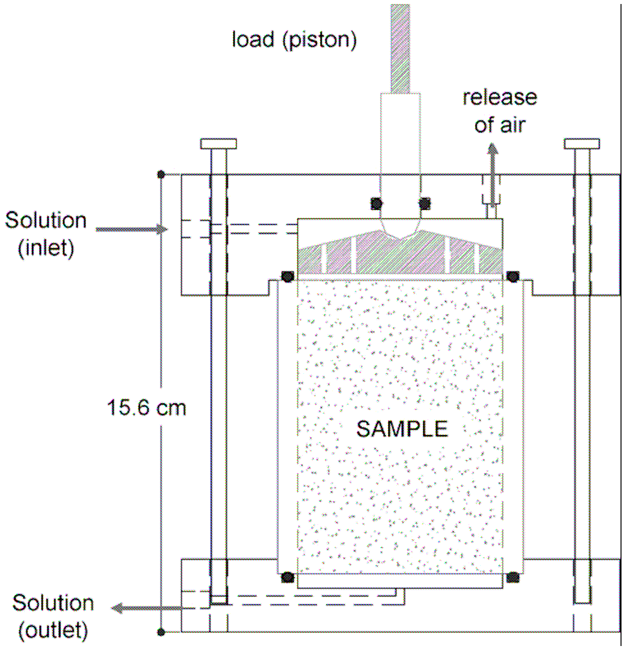

Figure 2. Modified odometer test project.

A digital displacement sensor was placed at the top of the loading piston to measure sample displacements during the percolation and loading phases (*Mitutoyo Corpotation, Digimatic Indicator*, Model ID-U1025, range 0.01-25.40mm).

2.4 Methods

The procedures for testing were divided into three phases: percolation (distilled water followed by acidic waters or not); loading and unloading.

After the soil was compacted into the column, the specimens were submitted to the percolation of distilled water until the electric conductivity and volume of water entering and exiting the column were similar. Electrical conductivity provides a measure of the ionic concentration and was used as a surrogate for a directly measured dissolved solute. After the specimen was percolated by distilled water until the electric conductivity was near zero, it was then submitted to the loading phase or to acid percolation, according to the experimental program.

The influent and effluent fluid samples were tested for electrical conductivity and pH with time using standard electrical probes (measuring conductivity in increments of 1.0µS/cm) connected to an analogical/digital converter. These electrical probes also allow the measurement of pH and temperature of the liquid. Measurements of the volume of solution entering and effluent exiting the specimens were also carried out. The loading phase on samples percolated by acidic waters was completed only after the pH and electric conductivity of the outlet solution was similar to the inlet solution. The percolation phase of the test was completed under a pre-loading of 4 kPa (due to the weight of the loading cap placed on top of the sample). The next loading was applied after the settlements had stabilized, respecting a minimum of 30 minutes per load.

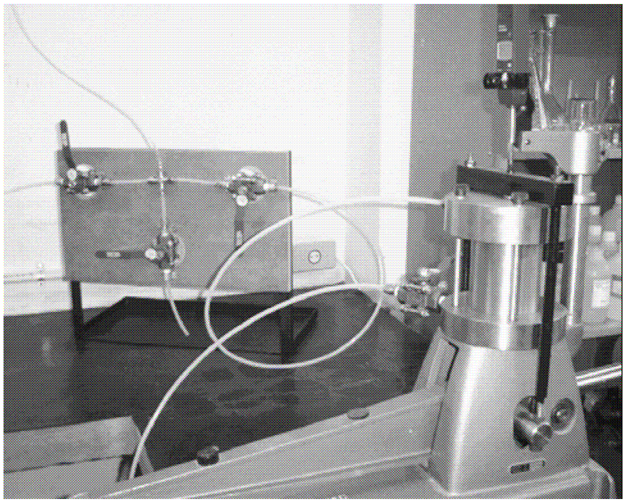

Figure 3. Modified odometer test.

The maximum pressure applied to the samples was approximately 4 MPa (maximum capacity of the odometer equipment). The unloading behaviour was also measured after the loading phase.

After each test, the samples were submitted to Atterberg Limit, grain size distribution and moisture content tests to investigate potential changes in the soil properties.

3 RESULTS AND DISCUSSIONS

Figures 4, 5, 6 and 7 present the results of one-dimensional loading tests completed in soil samples percolated by solutions with 0, 2, 6 and 10% of dissolved sulphuric acid respectively.

Figure 4 presents the results of one-dimensional loading tests carried out by percolation of only distilled water through a sample of soil (SRAB). The percolation phase of the test was considered completed after 48 hours, when the volume of water entering and exiting the sample was similar. The sample percolated under a hydraulic gradient 20 returned in slightly higher settlements for stresses of up to 1 MPa, if compared to samples under hydraulic gradient 10; similar stress-voids ratio curves were observed for both tests at higher stress levels. The final voids ratio (after the last loading was applied and stabilized at approximately 4 MPa) was 0.42 for both samples; after the unloading phase the average final voids ratio was approximately 0.44. Both samples were tested from an initial voids ratio of 0.485.

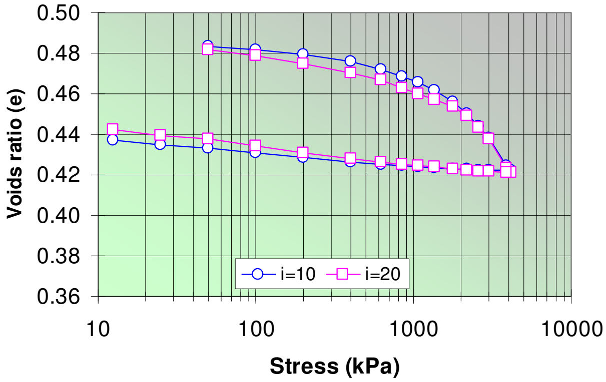

Figure 4. Voids ratio versus stress – samples percolated by a solution with 0% sulphuric acid.

In Figure 5 it is possible to observe higher compressibility, at the end of the loading phase for samples percolated by a solution with 2% of sulphuric acid, than observed in Figure 4 for samples percolated only by distilled water (no acid). As observed, the curves for the tests under different hydraulic gradients show different compressibility rates at medium stress levels (between 0.8 and 3 MPa) and similar voids ratio after the last load (approximately 4 MPa), where the voids ratio was approximately 0.415.

Figure 6 presents the results of one dimensional loading tests completed in samples after the percolation of acidic waters with 6% of sulphuric acid. It is possible to observe that the voids ratio at the end of the loading phase is smaller to what was observed in samples percolated by distilled water and with 2% of sulphuric acid. At this level, the hydraulic gradient during percolation became one of the variants of the test, once the samples percolated under hydraulic gradient 20 returned in higher compressibility of the samples if compared to samples percolated under a hydraulic gradient 10.

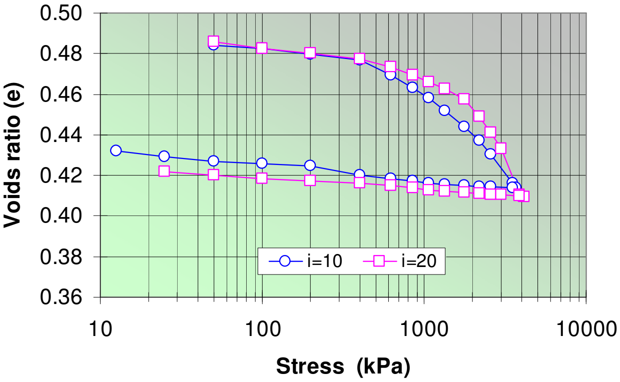

Figure 5. Voids ratio versus stress – samples percolated by a solution with 2% sulphuric acid.

In order to verify the repeatability of the results and confirm the influence of the hydraulic gradient during the phase of percolation, the tests were duplicated under the same conditions. The results presented a satisfactory repeatability of the results, with a small discrepancy in the results for samples submitted to percolation under hydraulic gradient 10.

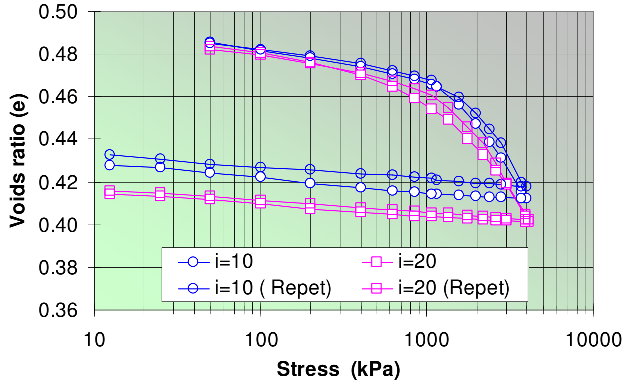

Figure 6. Voids ratio versus stress – samples percolated by a solution with 6% sulphuric acid.

As observed in Figure 7, the percolation of a solution with 10% of sulphuric acid resulted in soil samples with the higher compressibility than samples percolated with 0, 2 and 6% of sulphuric acid, for the same level of stresses. As observed in samples percolated by solutions with 6% of sulphuric acid dissolved in distilled water (Figure 6), loading tests completed after a percolation of a solution with 10% of sulphuric acid resulted in a higher compressibility for the sample percolated under hydraulic gradient 20 than 10. Under hydraulic gradient 20, the lowest voids ratio observed in the experimental program was achieved, approximately 0.395 after the application of the last load. The compressibility of the sample percolated under gradient 20 was higher than the sample percolated under gradient 10 at all stress levels.

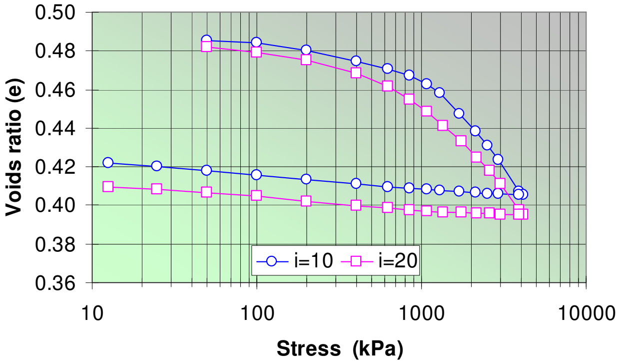

Figure 7. Voids ratio versus stress – samples percolated by a solution with 10% sulphuric acid.

Figures 8 and 9 present voids ratio versus stress curves for samples percolated by waters with 0, 2, 6 and 10% of sulphuric acid, under hydraulic gradients 10 and 20 respectively.

In Figure 8, it is possible to observe the increase in compressibility of the samples with the increase of the concentration of sulphuric acid dissolved in the water percolated through the samples under hydraulic gradient 10. In Figure 9 (hydraulic gradient 20) the discrepancy in the voids ratio at the end of the loading phase is more expressive than in samples percolated under hydraulic gradient 10. However, for both hydraulic gradients 10 and 20, higher concentrations of sulphuric acid dissolved in the solution returned in higher compressibility rates of the soil samples at the end of the loading phase.

Table 2 summarizes the results obtained in the one-dimensional loading tests. The coefficient of elastic rebound (C_r) ranged from 0.007 to 0.013 observed for three samples. The coefficient of compressibility (C_c) ranged from 0.099 to 0.179. The swelling coefficient (C_s) ranged from 0.004 to 0.009. The pre-consolidation pressure ranged from 1070 kPa for a sample percolated by a solution with 2% sulphuric acid and hydraulic gradient 10 to 2080 for a sample also percolated by a solution with 2% of acid, but percolated under a hydraulic gradient 20.

The soil used in this research is naturally acidic, with a pH of approximately 4.6. The percolation of acidic waters would potentially return in higher compressibility in alkaline soils other than in soils with a natural acidic pH, such as the soil adopted for this research. After the percolation of acidic waters through the samples, visual inspections completed of the samples confirmed that the fines of the soil migrated to the bottom of the sample, resulting in higher porosity at the top of the sample and consequent higher compressibility. In order to verify the magnitude of the variations in grain size distribution along the sample, grain size distribution tests were completed in a sample after the percolation of a solution with 6% of sulphuric acid under a hydraulic gradient of 10. The sample was divided in three sections, upper 1/3, medium 1/3 and bottom 1/3. The results are presented in Figure 10. The results showed that most of the fine particles of the sample migrated towards the bottom of the sample which resulted in higher porosity at the top of the sample.

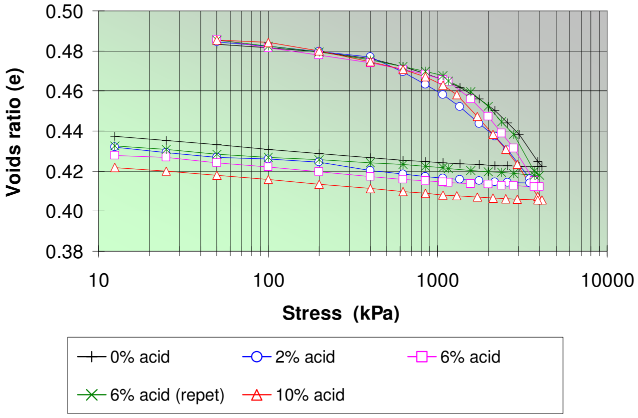

Figure 8. Voids ratio versus stress for samples percolated with acidic waters under hydraulic gradient 10.

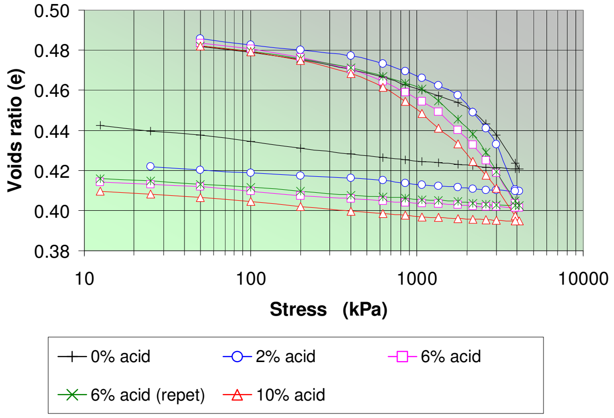

Figure 9. Voids ratio versus stress for samples percolated with acidic waters under hydraulic gradient 20.

Table 2. Summary of one-dimensional loading tests.

Hydraulic gradient	% sulphuric acid in the solution	C_r	C_c	C_s	Pre consolidation pressure - (kPa)
10	0	0.007	0.108	0.006	1710
	2	0.008	0.099	0.008	1070
	6	0.013	0.122	0.006	1410
	10	0.013	0.135	0.006	1505
20	0	0.013	0.112	0.009	1840
	2	0.010	0.179	0.005	2080
	6	0.012	0.116	0.004	1310
	10	0.012	0.120	0.006	1140

Potential changes in Atterberg limits of the soil were also investigated in all samples after the percolation by acidic waters. The results are summarized in Table 3. The liquid limit slightly decreased from 23.5% (sample percolated only by distilled water) to 17.8% after the percolation of a solution with 6% acid.

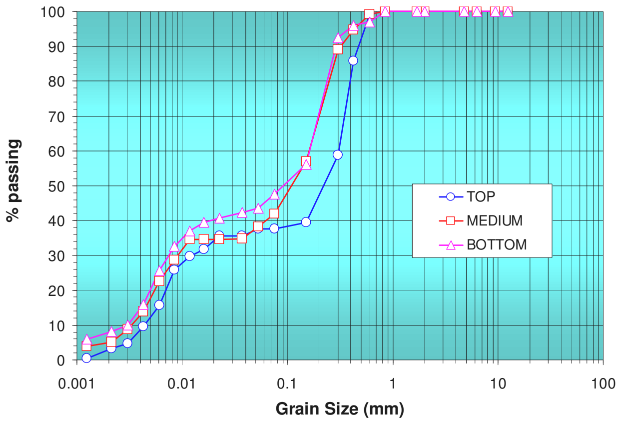

Figure 10. Grain size distribution along sample percolated by a solution with 6% of sulphuric acid under hydraulic gradient 10.

Table 3. Atterberg limits – test results

% sulphuric acid in the solution	Liquid Limit (%)	Plasticity Limit (%)	Plasticity Index (%)
0	23.5	13.0	10.5
2	19.8	10.6	9.2
6	17.8	9.9	7.9
10	19.0	11.2	7.8

The plasticity limit also decreased from 13.0% (only distilled water percolated through the sample) to as low as 9.9% for the sample percolated by a solution with 6% acid. It is assumed that only small changes were observed in Atterberg limits due to the small clay fraction presented in the soil.

4 CONCLUSIONS

The following comments summarize the tests completed in this study:

- Higher concentrations of sulphuric acid in the solution resulted in higher compressibility of the soil at same levels of stress.
- The influence of the hydraulic gradient in which the acidic waters percolated through the soil samples was evident in the compressibility of the soil for concentrations of sulphuric acid in the solution higher than 6%;
- The high acidity of the solution percolated through the soil samples affected the behaviour of the fines of the soil, which migrated to the bottom of the sample (towards the direction of the flow). This effect is recommended to be investigated in soils with higher clay content in order to determine the efficiency or not of the use of clayey soils in the containment of acidic waters/minerals, as observed in tailings areas.
- The Atterberg limits of the soil decreased after the contact with acidic waters. The slight changes in the Atterberg limits can be attributed to the small clay content of the soil.
- If a soil is intended to be used for containment of pyritic wastes or AMD, laboratory tests are suggested to be completed in order to determine the effects of the acidity and metal content of the AMD in the soil.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the financial and technical support of AECOM Canada Ltd., Brazilian Research Council (CNPq) and Federal University of Rio Grande do Sul, Brazil.

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