Evaluation of the Use of Sulphide Paste Rock as Cover Material in Mine Reclamation

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Abstract
Today’s mining industry faces challenges related to mine site reclamation with increasing impacted footprints and the quantity of materials required to put covers in place. This research is complementary to other technical pre-reclamation studies to evaluate the application of paste rock, a mixture of waste rock and mill tailings, available at the LaRonde mine (Agnico Eagle Mines Ltd), with or without a limestone amendment and/or compaction. This study on the use of paste rock presents the results of various material characterizations, as well as hydrogeological and geochemical data from laboratory-based column tests.

Keywords: Mine site reclamation, reclamation covers, paste rock, cover with capillary barrier effects

Introduction
Tailings deposited in surface impoundments are prone to oxidation when they contain sulphide minerals and are exposed to water and atmospheric oxygen. In many cases, this can lead to the formation of acid mine drainage (AMD), and thus, appropriate measures must be taken to mitigate the associated environmental impacts. Covers with capillary barrier effects (CCBEs) can be used to control oxygen migration and, as a result, AMD generation (Aubertin et al. 1995; Bussière et al. 2003). This type of cover relies on a phenomenon called the capillary barrier effect, which occurs when a fine-grained material overlies a coarse-grained material due to the contrast between the materials’ unsaturated hydrogeological properties. In a CCBE, capillary barrier effects allow for one of the layers, i.e., the moisture-retaining layer (MRL), to be maintained at a high degree of saturation, thereby limiting the diffusion of oxygen from the atmosphere into the reactive mine wastes.

Cover systems used to limit acid generation in tailings storage facilities (TSF) are often comprised of natural materials. However, when the required volume of such materials is significant, concerns can arise related to the availability of suitable materials, as well as land preparation and transportation costs. As a result, the use of mined materials in covers has become an interesting, more sustainable alternative to natural soils. One such alternative is paste rock, which is a homogeneous mixture of waste rocks and mill tailings that are generated by routine mine operations. This mixture is offers significant environmental advantages because it retains both the geotechnical characteristics of waste rock and the hydrogeological characteristics of mill tailings (Wickland 2006; Wilson 2008).

LaRonde mine (Agnico Eagle Mines Ltd) is a polymetallic mine located in the Abitibi region of Québec (Canada). The mine has more than 180 hectares of TSF that contain sulphide-rich, acid-generating tailings. Because of the TSF area and the low availability of suitable natural soils, paste rock is currently being evaluated as a potential cover material for the reclamation the TSF, with or without a limestone amendment and/or compaction.

This study presents an assessment of paste rock (made with mined materials available at LaRonde mine) as a cover system for controlling the generation of AMD. More specifically, the performances of limestone-amended
and unamended paste rocks were assessed using laboratory-based column tests. Four columns were mounted and monitored for their hydrogeological and geochemical behavior over a period of eighteen months. Permeability tests, hydrogeological behavior (suction, ψ; volumetric water content, VWC, profiles), and effluent water quality were used to assess the paste rocks’ performances.

**Materials and Methods**

Both the waste rocks and tailings used in this study were sampled at LaRonde mine. The grain-size of the waste rock samples was truncated to 50 mm before being transported to the laboratory. Based on studies by Wickland (2006) and Wilson (2008), the ideal ratio in dry weight to make an efficient paste rock mixture should be between approximately 4:1 and 5:1 (waste rock: tailings). Several tests with ratios ranging from 3.6:1 to 5.1:1 were produced in the laboratory using a small cement mixer. Permeability tests, as described below, were used to select 4.3:1 as the optimal ratio for the LaRonde materials.

**Material Characterization**

The grain-size distribution (GSD) of the tailings was obtained using a Malvern Mastersizer laser particle size analyzer. Grain-size distributions for the waste rock and paste rock were determined by sieving for the coarse fraction (according to ASTM standard D422; ASTM, 2007), and by the laser particle size analyzer for the fine fraction (< 0.425 mm). The specific gravity (Gₛ) of each material was determined using an immersion basin for particles over 5 mm and with a helium pycnometer (according to ASTM standard D854-10; ASTM, 2012) for smaller particles.

The saturated hydraulic conductivity (kₛ) was determined for the tailings, the paste rock mixture, and waste rocks. For the tailings, the kₛ was evaluated using a rigid-wall permeameter (according to ASTM standard D5868-95; ASTM, 2007), then compared to values predicted by a model described by Mbonimpa et al. (2002a). Because of their GSDs, kₛ values for the waste rock and paste rock were evaluated through variable-head permeability tests in large high-density polyethylene (HDPE) columns (80 cm in height and 30 cm in diameter; see Peregoedova et al. 2013 for more details) as per standard ASTM D2434-68 (ASTM, 2006). The values were then compared to values from the predictive models proposed by Shepherd (1989) and Chapuis et al. (2004). The tailings’ water retention curve (WRC) was determined using a pressure cell (Tempe Cell) following ASTM standard D6836-02 (ASTM, ). A larger pressure cell test, which uses the same mechanism as a Tempe Cell and follows a similar protocol to ASTM standard D6836-02 (ASTM, ), was developed to determine WRCs for coarse-grained materials that contain fine, silt-like particles, as is the case for paste rock mixtures. The materials used in this study were also tested for their geochemical and mineralogical characteristics. Total sulfur (S) and carbon (C) contents were measured using an induction furnace; these values were used to estimate the acid generation and neutralization potential of the tailings and waste rocks. Concentrations of metal(loid)s (As, Be, Bi, Sb, Se, and Te) were measured using ICP-AES following complete digestion. Semi-quantitative estimates of the tailings’ and waste rocks’ mineralogical compositions were obtained using X-ray diffraction (XRD). For further details on material characterization methods, see Kalonji et al. (2017).

**Column Construction and Instrumentation**

Column tests were used to assess the performance of the paste rock cover in controlling AMD generation in the LaRonde tailings. Each column was constructed using black HDPE with an internal diameter of 0.30 m and a height between 0.9 and 1.2 m, depending on the thickness of the cover system. A 0.1 m headspace was left at the top of each column to allow for oxygen consumption (OC) tests and monthly wetting-drying cycles. These OC tests allowed for the in-situ estimation of diffusive oxygen fluxes migrating through the cover to the LaRonde tailings; results related to these tests can be found in Pouliot (2019). At the base of the columns, a ceramic plate was installed that connected to a flexible tube and a bottle; this produced suction by simulating the water table level below the base of the column.

The parametric settings of the four laboratory columns (C1, C2, C3, and C4) are...
summarized in Table 1. The moisture retaining layer of each cover was comprised of paste rock, with a ratio of 4.3:1 waste rock to tailings used in all columns. Two of the columns (C1 and C2) assessed the paste rock cover in a CCBE configuration with a waste rock capillary break, while the other two columns (C3 and C4) tested a bilayer cover configuration. Because of the high sulfur content of the LaRonde materials, the paste rock used as an MRL in columns C2 and C3 was amended with a fine limestone gravel as a neutralizing agent. The waste rocks used for the top draining layer were identical in all four columns (0 - 50 mm) and were all compacted.

Various instruments were installed throughout the experimental columns, including: GS-3 probes to determine the VWC in each layer, and Watermark probes and tensiometers to measure ψ values. Further details on the use of these instruments in covers are available in Kalonji et al. (2016).

### Results

#### Material Characteristics

Table 2 shows the important characteristics of the LaRonde tailings, paste rock mixture, and waste rocks that relate to their hydrogeological and geochemical behaviors. The materials’ Gs values are influenced by their sulphide contents, with the tailings (3.29) showing higher values as compared to the waste rocks (2.73). Based on the grain-size distribution, the tailings are classified, according to the USCS classification system, as a plastic silt (ML) and the waste rocks as a well-graded sand (SW) with variable proportions of fine particles. The tailings’ ksat varied between $3.9 \times 10^{-8}$ and $5.7 \times 10^{-8}$ m.s$^{-1}$, while ksat values for the waste rock varied between $1.6 \times 10^{-5}$ and $7.1 \times 10^{-4}$ m.s$^{-1}$, and the ksat of the paste rock was $2.8 \times 10^{-8}$ m.s$^{-1}$. The air entry value (AEV) of the two tested tailings samples were approximately between 20 to 30 kPa (Tempe Cell measurements). The AEV

### Table 2 - Chemical, physical, and hydrogeological properties of materials used in experimental columns.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Tailings</th>
<th>Paste rock</th>
<th>Waste rock</th>
</tr>
</thead>
<tbody>
<tr>
<td>%S</td>
<td>19.08</td>
<td></td>
<td>1.065</td>
</tr>
<tr>
<td>%C</td>
<td>&lt; 0.05</td>
<td></td>
<td>0.44</td>
</tr>
<tr>
<td>D$_{10}$ (mm)</td>
<td>0.00437</td>
<td>0.180</td>
<td>0.11</td>
</tr>
<tr>
<td>D$_{50}$ (mm)</td>
<td>0.02526</td>
<td>20</td>
<td>9.11</td>
</tr>
<tr>
<td>D$_{60}$ (mm)</td>
<td>0.03475</td>
<td>24</td>
<td>13.50</td>
</tr>
<tr>
<td>Cu$<em>{15}$ (D$</em>{60}$/D$_{10}$)</td>
<td>0.00795</td>
<td>0.133</td>
<td>122.73</td>
</tr>
<tr>
<td>k$_{s,i}$ (m.s$^{-1}$)</td>
<td>$3.9 \times 10^{-8}$ to $5.7 \times 10^{-8}$</td>
<td>$2.8 \times 10^{-4}$</td>
<td>$1.6 \times 10^{-5}$ to $7.1 \times 10^{-4}$</td>
</tr>
<tr>
<td>Porosity</td>
<td>0.42 - 0.46</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gs</td>
<td>3.29</td>
<td></td>
<td>2.73</td>
</tr>
<tr>
<td>AEV, ψ (kPa)</td>
<td>20-30</td>
<td>n.d.</td>
<td>0.4*</td>
</tr>
</tbody>
</table>

* Kalonji et al. (2017)
for the waste rock was measured at 0.4 kPa in a prior study (Kalonji et al., 2017). Using the measured total S and C values, the net neutralization ratio (NP/AP) was calculated at 0.007 for the tailings and 1.1 for the waste rock, demonstrating that both materials are potentially acid-generating (PAG; NP/ AP < 2). For this reason, a 6 mm limestone gravel was added to the paste rock mixture in columns C2 and C3 to increase the neutralization potential and make the material non-acid-generating. Semi-quantitative XRD analyses showed that the tailings are mainly composed of silicates such as quartz (42%), albite, and muscovite, but also included 30% pyrite. Analyses of the waste rocks demonstrated that it is rich in quartz (50%), carbonates (4.6%), and sulfides (1.35%).

Hydrogeological Results

Figures 1 and 2 show the evolution of the VWC (θ) and suction for two amended columns, C2 and C3, respectively. Data for the unamended columns, C1 and C4, are not shown because the hydrogeological results were similar to the amended columns. For the CCBE columns (C1 and C2), suction and VWC sensors were placed at 100 cm, 80 cm, 50 cm, and 20 cm from the base of the column. For bilayer columns (C3 and C4), suction and VWC sensors were placed at 70 cm, 50 cm, and 20 cm from the base of the column.

In columns C1 and C2, which had CCBE configurations, higher VWCs were observed in the moisture-retaining layer made of paste rock. Suction values in the moisture retaining layer (MRL) were below the paste rock’s AEV; this allowed for the material to maintain saturation (Figure 1). For the bilayer columns without a capillary break, C3 and C4, the VWCs measured near the top of the MRL were lower than those measured near the bottom. Additionally, suction values at the top of the MRL in the bilayer columns were slightly higher than in the CCBE columns, rising close to 40 kPa (Figure 2).

When the results for the amended columns are compared to the unamended columns (not shown here), lower VWC values and higher suction values were observed in the unamended scenario. This can likely be explained by the fact that the addition of the limestone amendment increases the density and decreases the porosity of the paste rock mixture. This improves water retention characteristics and allows for higher VWCs for a given suction.

Figure 1 - Volumetric water content and suction for column 2 (amended CCBE).

Figure 2 - Volumetric water content and suction for column 3 (amended bilayer).
**Geochemical Results**

The leachate samples from each column following each monthly rinse cycle were analyzed for pH, Eh, electrical conductivity, alkalinity, acidity, total sulfur, and several metals. The objective was to assess the capacity of each cover configuration to limit AMD generation and metal leaching. Some of the results, which were representative of the overall trends observed, are shown in Figure 3.

The unamended columns, C1 and C3, drop in pH and alkalinity over time. Column C1 performed slightly better than C3; this was likely due to its CCBE configuration more efficiently reducing oxygen migration. The amended columns, C2 and C4, maintained pH values above the limits set by the Canadian Mining Metal Effluent Regulation (MMER), however, a downward trend developed at around 400 days. Leachates from all columns showed negligible alkalinity by 500 days. This suggests that, at that point, the limestone amendment in columns C2 and C4 was not able to counterbalance the acidity generated by the tailings materials.

Concentrations of nickel (Ni) and zinc (Zn) were lower in the amended columns up to 400 days. Because of the limestone amendment, there was a downward trend in Ni and Zn for both columns C2 and C4. Concentrations of Ni were below the regulatory limit following the first flush, while concentrations of Zn decreased until 325 days, but did not fall below the regulatory limit. After alkalinity was depleted and the pH began to drop, the concentrations of both Ni and Zn started to increase.

**Conclusions**

An experiment using laboratory-based columns was performed to evaluate the use of paste rock as a cover material for the prevention of acid mine drainage. Hydrogeological monitoring of the columns demonstrated that the paste rock cover in a CCBE configuration that was amended with limestone was most successful in maintaining adequate volumetric water content and suction levels to limit the migration of oxygen through the cover. However, geochemical monitoring of the leachates showed that the limestone amendment was able to maintain a circumneutral pH for a period of approximately 325 days in the tested column.

To further interpret the results obtained in this study, data obtained from the columns will be compared to a full season of results from a field-scale experimental cell (CR4) with a paste rock cover that was constructed at LaRonde mine; this cover was designed
based on previous work by Bussière et al. (2007). The field cell is instrumented with volumetric water content sensors and suction sensors positioned in each of the different layers of material. A pore gas sampler was also installed and connected to oxygen sensors in each of the cell’s layers; this will allow for the measurement of vertical oxygen concentration profiles. A comparison between the covered cell and a control cell will be performed to assess the efficiency of the system. Results will be presented in Pouliot (2019).

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