THE EFFECT OF WATER TABLE ELEVATION ON ACID MINE DRAINAGE FROM REACTIVE TAILINGS: A LABORATORY AND NUMERICAL MODELING STUDY¹

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Abstract. Laboratory column experiments and numerical simulations are used to evaluate the effectiveness of an elevated water table as a management and reclamation method for acid-generating mine tailings. The laboratory experiments involved monitoring oxygen and effluent chemistry from several columns of mine tailings with different water table elevations. Each column contained a layer of reactive tailings overlain by a sand cover and was subjected to transient recharge and flushing in fourteen 30-70 day cycles for 502 days. A free-draining control column with no sand cover was also used. The experimental results were compared with numerical simulations using the fully coupled reactive transport model MIN3P, which included infiltration, saturation-dependent oxygen diffusion, pyrite oxidation, multi-component geochemical reactions, and mineral buffering. The numerical results were consistent with the observed data which showed that an elevated water table can be very effective in reducing the production of acid mine drainage.

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Introduction

Over the past several years, the concept of an elevated water table has been considered as a possible option for management and closure of sulphidic tailings impoundments. The interest in this relatively new technology is related to its physical and economic advantages, including improved stability and lower costs (MEND, 1996). The concept is to maintain a high degree of water saturation within the tailings in order to reduce the rate of O_2 diffusion and sulfide oxidation. Oxygen diffusion rates through saturated or nearly saturated porous media, for example, can be several orders of magnitude less than under fully unsaturated (dry) conditions (Nicholson and Tibble 1995; Elberling and Nicholson 1996; MEND 1996; Mbonimpa et al. 2003; Dagenais, 2005). For optimal efficiency (i.e. to maintain saturation), the depth of the water table must be less than the air entry value (AEV) of the tailings (MEND 1996; Orava et al., 1997; Aubertin et al., 1999; Ouangrawa et al. 2005; see also Dagenais et al., this Conference).

Until recently, full water covers were often considered the most effective means of preventing acid mine drainage (AMD) from reactive tailings. However, the greatest difficulty related to their application is in maintaining functional infrastructure over the long term. They can be relatively expensive and susceptible to long term stability problems that can lead to failure. In the case of an elevated water table, the tailings are not flooded completely but saturation is maintained by the tailings' capillary properties.

The general objective of this study is to evaluate the effectiveness of an elevated water table for reducing sulfide oxidation. Using the highly acid generating tailings from the Louvicourt mine, Quebec, numerous laboratory column tests have been completed to establish the hydraulic and geochemical conditions which control system behavior. The observed behavior is compared with numerical reactive transport simulations. The simulated and measured data indicate that this approach can be very efficient in preventing the production of AMD.

Materials and Methods

A total of 15 cylindrical column experiments were undertaken in this study, each with different tailings material and/or negative (suction) pressures at the base. In this paper, the authors will consider only four of these test columns (Columns 1, 4, 6 $\&$ 12) which were prepared using the tailings sampled from the Louvicourt mine, located near Val-d'Or, Quebec, Canada. Figure 1 shows a schematic view of the columns. Columns 1, 4 and 6 contained 15 cm of reactive tailings overlain by 20 cm of non-reactive sand (acting as an evaporative barrier) and were equipped with a ceramic porous plate below the tailings to maintain the desired level of suction. Column 12, used for control, had no ceramic plate or sand cover and was underlain by a thin geotextile.

At the beginning of each cycle, seven centimetres of demineralised water ($pH \sim 6$) was added at the upper surface after which the columns were allowed to drain. Cycle 1 lasted 70 days, while cycles 2-14 lasted approximately 30 days each. The top of each column was open to the atmosphere and a port at the base of each column was used to adjust the suction (Fig. 1). Column 1 had the highest permanent base suction head $(\sim 209 \text{ cm of water})$ while column 6 had the lowest suction head (-61 cm) . The applied suction in column 12 was 0 cm from 0-256 days (cycle 1-6), then was increased to 75 cm from 256-322 days (cycle 7-8), then finally the column was subjected to free drainage over the remaining 6 cycles. The different suction values at the

base represented different water table depths below the tailings. The bottom port was also used to collect leachate for chemical analyses.

Figure 1. Typical experimental column and instrumentation

The column experiments used fresh (unoxidized) Louvicourt mine tailings which are sulfur rich (34-40 wt.% pyrite). They are composed primarily of silt and fine sand size particles with a percentage passing 80 µm of about 90 % (and 80 % passing 40 µm). The characteristic grain diameters, corresponding to 10 % (D_{10}), 50 % (D_{50}) and 60 % (D_{60}) passing on the cumulative grain size distribution curve are $2 \mu m$, 17 μm and $24 \mu m$, respectively. The tailings include various silicate minerals such as quartz (21 wt. %), chlorite (11%), muscovite (7%), and paragonite (6%). Feldspar minerals are also likely present but the fraction was unknown. In addition to pyrite, the sulphide minerals include trace amounts of chalcopyrite, sphalerite and galena. Carbonates are represented by dolomite $(4 \text{ wt. } %)$, magnesite (4%) and ankerite (1%) . The primary oxide mineral is magnetite (4 wt. %). The saturated hydraulic conductivity of the tailings is in the range 10^{-6} - 10^{-5} cm/s at a void ratio of 0.5 - 0.7 and the Air Entry Value (AEV) is approximately $300 \text{ cm H}_2\text{O}$.

Acid-Base Accounting tests (ABA test; see Lawrence, 1990) show that the tailings net neutralization potential (NNP) is approximately -558.3 kg $CaCO₃/tonne$ and the acid generation potential (AP) is 599.7 kg $CaCO₃/tonne$. These results show that the Louvicourt mine tailings can be interpreted as being highly acid generating with NNP values much lower than -20 kg of CaCO₃/tonne and a NP/AP ratio of 0.07 (e.g. SRK, 1989).

Additional details on material properties and testing procedures have been presented in Ouangrawa et al. (2005).

Experimental Results and Discussion

The observed effluent data are shown over the first 14 drainage cycles (about 500 days) in Fig. 2. In column 1 (with the highest continuous suction of 209 cm), the pH remained close to 7 for the first 250 days, then showed a slight decreasing trend (to a minimum of 6.6) over the last 250 days. In columns 4 and 6 (with lower suctions, therefore higher water saturation), the pH remained near-neutral (~ 7.5) for the entire 500 day period. In column 12 (control column), the pH remained between 7-8 for the first 322 days, then gradually decreased to 4.5 by 500 days. The pH decrease in column 12 coincided with the change to free drainage at 322 days.

Figure 2. Observed drainage water evolution during leaching of the 4 columns, showing (a) pH, (b) SO_4^2 and (c) total Fe. Water suctions refer to the suction head applied at the base of the column. Each point corresponds to the beginning of a drainage cycle (effluent concentrations were measured within 1-2hrs. after adding water).

Sulfate concentrations can be related to sulfide oxidation since these columns are relatively short and SO_4^{-2} precipitation is unlikely to occur. In column 1, sulphate concentrations remained between 2000-3000 mg/L throughout the study period, whereas in columns 4 and 6, the SO₄⁻²concentration gradually decreased from about 1500 mg/L to 400 mg/L after 500 days. The highest SO_4^2 concentrations were observed in control column 12 where the concentration increased to over 35,000 mg/L after free drainage conditions were applied.

Iron concentrations were somewhat more difficult to interpret because of geochemical reactions and Fe precipitation (for example, as ferri-oxyhydroxides). In columns 4 and 6 (lowest suctions), total Fe concentrations varied between 0.02 and 1 mg/L. In column 1 which had a higher suction (209 cm), total Fe concentrations were somewhat higher, between 1-10 mg/L. In column 12 from 0-322 days (suction $= 0-75$ cm), the total Fe increased to a maximum of about 2 mg/L. Following free-drainage conditions at 322 days, total Fe in column 12 increased to over 200 mg/L.

The observed data show that in general, higher suctions (e.g. column 1) lead to increased oxidation (lower pH, higher SO_4^2 and Fe) relative to the lower suction cases (columns 4 & 6). In all three of these columns, however, the degree of oxidation remained relatively low as shown by the fairly uniform concentrations of oxidation products in the effluent. The greatest change occurred in column 12 after cycle 8 (322 days) where the pH dropped from about 7.5 to 4.5 by 500 days and the SO_4^2 and total Fe concentrations increased by about 2 orders of magnitude. This change coincided with the application of free drainage, which likely caused significant desaturation at the top of the tailings. This suggests that these tailings would be acid-generating if they were exposed to natural conditions in the field (as predicted by static tests).

MIN3P Model Simulations

The experimental columns were simulated using the reactive transport model MIN3P (Mayer et al. 2002). MIN3P is a finite-volume based fully coupled reactive transport model capable of simulating saturated/unsaturated flow, oxygen diffusion, pyrite oxidation, and reactive transport with kinetic and/or equilibrium reactions. The model has been well proven for complex reactive systems including predictive modelling of mine closure (Bain et al., 2001) and for modelling the effects of capillary barrier covers on reactive tailings (Molson et al., 2004).

The conceptual model for the simulated column mineralogy was somewhat simplified from the observed system, however for the minerals included, the volume fractions were based on observed data. The upper layer of non-reactive sand was composed of quartz (60 vol. %), muscovite (5%) and k-feldspar (3%). The underlying reactive tailings included pyrite (1 3.7 vol. %, corresponding to 40 wt. %), dolomite (2.3 vol.%), magnesite (5%), k-feldspar (3%), muscovite (4%) and quartz (20%). Secondary minerals not initially present but which were allowed to precipitate included siderite, gibbsite, gypsum, ferrihydrite, and jarosite. Thirteen primary aqueous components $(Ca^{+2}$, K^{+1} , Cl^{-1} , H_4SiO_4 , Al^{+3} , CO_3^{-2} , H^{+1} , $O_2(aq)$, Fe^{+2} , SO_4^{-2} , Fe^{$+3$}, HS⁻¹, Mg⁺²) and 21 secondary aqueous species were included in each simulation. Pyrite oxidation was assumed governed by the shrinking core model assuming a uniform grain size of 17 μ m (= observed D₅₀). The van Genuchten (1980) model parameters controlling unsaturated flow behavior were fitted based on observed data (Table 1). A dispersivity of 0.5 mm was assumed for all simulations.

Material		θ_r	K_Z ⁽¹⁾	a_v , $n^{(2)}$	<i>AEP</i>
			$\lceil m/s \rceil$	$[m^{-1}]$, [-]	(cm H ₂ O)
Sand	0.44	0.05	1.02×10^{-3}	5.3, 10.7	14.
Tailings	0.43	0.10	3.13×10^{-8}	0.10, 2.6	318.

Table 1. Material characteristics used in the numerical model.

 $⁽¹⁾$ saturated hydraulic conductivity</sup>

 (2) van Genuchten (1980) parameters (l=0.5 for both materials)

The columns were simulated in 1D by resolving each column into 200 elements. The bottom flow boundary condition was a type-1 (Dirichlet) with a fixed (negative) water pressure to control the tailings saturation. The top boundary was a type-2 (Neumann) flux condition with cyclic monthly recharge events. At the beginning of each cycle, a recharge of 1.29×10^{-5} m/s was applied for approximately 1.5 hours (representing the lab-applied 7 cm recharge events), followed by a zero-flux condition for the remaining time of each cycle. The chemistry of the recharge water for each simulated cycle was based on measured concentrations and pH of the demineralised water used in the experiments.

The simulations shown here are conceptual; a more detailed calibration is currently in progress. For clarity, the modelling results are presented only for column 1 (high suction case) and column 6 (low suction case). Except for the applied suction at the base, each column is identical in geometry and mineralogy, and uses the same flow and transport parameters. A typical 13 component, 502-day simulation with adaptive time stepping took approximately 8 hours on a PIV 2GHz machine.

The simulation results for column 1, showing vertical profiles of water saturation, O_2 , pH, SO_4^{-2} , Fe, and pyrite volume fraction, are provided at selected times in Fig. 3. At the beginning of each cycle, water drains rapidly from the sand cover, while the tailings remain close to saturation (degree of saturation $S_w \sim 0.9$). Although the applied suction is higher in column 1 (209 cm) compared to column 6 (60 cm, see below), the tailings saturation remains high because of its fine grained structure. As the sand desaturates, oxygen diffuses rapidly into the column but diffusion into the underlying tailings is limited because of its higher saturation. Nevertheless, in this case there is sufficient diffusion to partially oxidize the tailings at the sand/tailings interface and the pH here drops locally to about 2-3 by the end of the sixth cycle (256 days). A limited oxygen supply (because of the higher water saturation) combined with dolomite and magnesite pH buffering limits the oxidation and prevents this low pH from advancing further down the column. The pH at the base therefore remains around 6-7, as observed during the column experiment. The sulphate and iron concentration profiles can be correlated to the pH drop. Since these components are more mobile however, the profile peaks tend to advance further down the column, and appear as pulses in the effluent (see Fig. 5 below). The pyrite volume fraction shows that the oxidation front has advanced about 1 cm after 500 days.

The simulation results for column 6 (Fig. 4) are similar: water drains rapidly from the sand while O_2 enters from above. However, the extent of oxidation is somewhat less in this case because the applied suction at the base of this column was less and the tailings therefore remained closer to full saturation ($S_w \sim 0.98$), which limits O_2 availability within the tailings. In

particular, the SO_4^2 and Fe concentrations are significantly lower and less pyrite has been oxidized.

Figure 3. Simulated vertical profiles for column 1 (suction = 209 cm) showing water saturation, O_2 , pH, SO_4^{-2} and Fe concentrations, and pyrite volume fraction at selected times (times in days unless otherwise noted).

The simulated transient behavior of the column effluent from columns 1 and 6 is provided in Fig. 5. The simulated effluent chemistry is characterised by monthly variations which correlate with the cyclic flushing, drainage and oxidation of the columns. At the beginning of each cycle, the addition of recharge water flushes the oxidation products which have accumulated from the previous cycle. This produces a brief drop in the pH at the base of the column (by at most 1 pH unit), and a corresponding increase in SO_4^{-2} and Fe concentrations. This "pulse" of oxidation products is immediately followed by a rapid pH increase (of up to 2 units) and a drop in SO_4^{-2} and Fe, all caused by a reduction in the rate of pyrite oxidation due to the higher moisture content remaining from the recharge water. As this water drains during the remainder of each cycle, and the saturation returns to its lower equilibrium level, the oxidation rate increases, and the pH gradually decreases from its cyclic peak. Likewise, SO_4^{-2} and Fe concentrations gradually increase over the drainage period, until the addition of the next pulse of recharge water.

The simulated effluent pH is similar in both columns, varying from 6-9, with the observed pH correlating best with the simulated pH minimum of each cycle (i.e. during the flushing events). The simulated effluent SO_4^2 varies between about 100 and 10,000 mg/L, with the observed data corresponding best to the simulated peaks, except for the period between 256 and 353 days where the observed data correspond better to the simulated minimums. Similar trends are observed for Fe.

Figure 4. Simulated vertical profiles for column 6 (suction head = 60 cm) showing water saturation, O_2 , pH, SO_4^{-2} and Fe concentrations, and pyrite volume fraction at selected times (times in days unless otherwise noted).

Figure 5. Simulated and observed evolution of column effluent quality (pH, SO_4^{-2} and Fe) for columns 1 and 6.

Although the simulated concentrations of the oxidation products are somewhat higher than those observed, the trends are similar and can be more closely matched with further refinement of the model components. Possible reasons for the differences include inherent assumptions of the shrinking core model (e.g. uniformly spherical grains) and an uncertain mineralogy (e.g. % kfeldspar was unknown). There was also some evidence of leakage or preferential flow at the tailings/column wall interface. The differences can also be attributed to the measurement time scales: while the simulated effluent chemistry is shown for every time step (generally on the order of $10^{-3} - 10^{-2}$ days, or ~1-15 minutes), the observed data represent an integrated response over several hours and may not capture the rapid response during drainage.

Conclusions

The main goal of this research project was to evaluate the effectiveness of the elevated water table technique in controlling the production of AMD from reactive tailings. The experimental and simulation results have shown that the most important factors which control the effectiveness of this technique are the hydraulic parameters and degree of saturation of the tailings. The observed data show that sulphide oxidation can indeed be controlled by maintaining a base suction which is less than the air entry value (AEV) of the tailings, i.e. when the tailings are kept saturated by capillary pressure. For example, pyrite oxidation was limited in columns 1 and 6 with suction heads of 209 and 60 cm (on tailings with an AEV of 300 cm H_2O), while oxidation increased significantly in column 12 when free drainage conditions were applied. The numerical model confirmed the conceptual interpretation and showed that the elevated water table concept can be effective in preventing acid mine drainage from reactive tailings.

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