Physical and geochemical transport modelling of pre-oxidised acid-generating tailings with a monolayer cover

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Abstract

This paper presents the results of an investigation on the efficiency of a monolayer cover placed on sulphidic tailings to control acid mine drainage. The study involved column tests and simulations conducted with a reactive transport model. A large column was filled with partially oxidised tailings covered with a single layer of relatively fine grained material. Monthly wetting and drying cycles were applied. The leachate was sampled for chemical analysis, including the pH, electrical conductivity, and the concentrations of sulphate and iron. Mineralogical analyses were also performed to obtain the parameters required for the simulations. The numerical model was used to simulate the flow of water and oxygen, as well as geochemical reactive transport in the column. The column model was calibrated using laboratory measurements. Additional simulations were conducted to extrapolate results over space and time. Alternative reclamation methods are also analysed.

1   Introduction

Sulphidic tailings stored in surface disposal sites can raise serious environmental issues when left exposed. The reactive minerals can interact with oxygen and water to generate acid mine drainage (AMD). The resulting leachate is typically characterised by high concentrations of sulphate and dissolved metals, a low pH and a high electrical conductivity (Jambor, 1994; Nordstrom, 2000). Most reclamation methods aim at decreasing availability of oxygen or water to the sulphide minerals to inhibit the oxidation reaction (MEND, 2001; Aubertin et al., 2002). For mine sites located in humid climatic regions, the preferred options are water covers (Yanful and Catalan, 2002; Romano et al., 2003), layered covers with capillary barrier effects (Nicholson et al., 1989; Aubertin et al., 1994, 2006; Bussière et al., 2003, 2006), and an elevated water table (Orava and Swinder, 1996; Ouangrawa et al., 2009). The latter is investigated here.

Reclamation work should be planned well ahead of the site closure and implemented near the end of operation or shortly thereafter to maximise efficiency. Old tailings sites that have been left exposed for some time typically include tailings that have been altered before the reclamation work starts. In such cases, the selected reclamation technique must take into account the contamination history of the site, especially when the pH of the leachate is low enough to maintain ferric ions in solution, which can then indirectly oxidise iron sulphide minerals (Nicholson, 1994; Nordstrom and Southam, 1997).

This study focuses on the reclamation of a partially oxidised tailings impoundment, where the pore water is already acidic. The reclamation method being considered consists of raising the water table and adding a 1 m (or more) layer of fine grained materials (i.e. non acid-generating tailings) over the surface of the reactive tailings. The specific goal of the experimental and numerical investigation is to assess the behaviour of the tailings-cover system, and determine the efficiency of the monolayer cover. The experimental data obtained from the laboratory tests are used to calibrate and validate the numerical approach. Key results are presented and discussed in this paper.

2   Materials and laboratory setup

The samples were collected in situ in 2007 from a heterogeneous disposal site, located in western Quebec (Canada) that had been abandoned for decades. The average pyrite content is about 12%, but values of up to 20% have been observed. The pH of the water within the tailings and around the site is very acidic (often < 2). The upper layer of the impoundment has been extensively oxidised. Cover material samples were also collected. Non acid-generating tailings, produced by a recently opened mine located in the vicinity, are used to cover the reactive tailings.

These materials have been extensively characterised in the laboratory to define their hydrogeological properties (Pabst et al., 2011). The water retention curves (WRCs) were determined using modified Tempe cells. The measured curves were combined with the Modified Kovács (MK) predictive model (Aubertin et al., 2003) to calibrate the WRC for varying grain size and porosity. Saturated conductivity k_{sat} was measured using flexible wall permeameters. These experimental results were compared with predictive estimates obtained from the Kozeny-Carman, KC (Chapuis and Aubertin, 2003) and Kozeny-Carman modified, KCM (Mbonimpa et al., 2002) models, which are also used to adjust the values of k_{sat} for varying material conditions. Oxygen diffusion and consumption parameters were also determined using tests in closed cells (Mbonimpa et al., 2003; Aachib et al., 2004). The detailed results are presented in Pabst (2011). Some of the main characteristics of the tailings and cover material are summarised in Table 1, together with WRC parameters based on the van Genuchten (1980) equation (Equation (2)).

Table 1   Material characteristics (D_r: specific gravity; $C_U = D_{60}/D_{10}$: coefficient of uniformity; k_{sat}: saturated hydraulic conductivity; θ_{sat} (satiated volumetric water content), θ_r (residual water content), α_v, n_v, and m_v: van Genuchten (1980) equation parameters)

	D_r	C_U	D_{10}	k_{sat}	α_v	n_v	m_v	θ_{sat}	θ_r
	(-)	(-)	(cm)	(m s^{-1})	(m^{-1})	(-)	(-)	(-)	(-)
Reactive tailings	3.136	12.7	0.056	$5.3.10^{-7}$	0.035	1.52	0.34	0.36	0.00
Cover material	2.769	10.9	0.038	$7.7.10^{-7}$	0.125	1.90	0.47	0.32	0.00

Mineralogical compositions at several locations along the column were determined using X-ray diffraction (XRD) analyses, before and after each of the columns tests. Total C and S contents were also measured (using a LECO furnace) to confirm those results. The main mineralogical composition of each material is summarised in Table 2.

Table 2   Mineralogy of the tailings and cover material, as used in the simulations (%weight)

	Reactive Tailings	Cover Material
Pyrite	12.0	n/d
Dolomite	n/d	5.5
Calcite	n/d	2.0
Quartz	57.0	21.5
Chlorite	5.0	6.6
Muscovite	22.0	7.4
Anorthite	3.0	10.0
Albite	1.0	47.0
Gypsum	n/d	n/d

As stated previously, the reactive tailings contain a fairly high proportion of pyrite (~12%), which is the source of AMD. Virtually no neutralising mineral has been detected in these tailings, so there is no buffer

capacity to slow down acid production. The cover material contains about 5.5% of dolomite and 2% of calcite.

Large instrumented columns (230 cm in height, 15 cm internal diameter) were used to study the hydrogeological and geochemical behaviour of the tailings–cover system. The physical model set-up follows a methodology developed over several years (Aachib et al., 1994, 1998; Bussière et al., 2004; Ouangrawa et al., 2010). The configuration was selected to reproduce some of the site conditions and provide representative results for long term analyses. Two large columns were mounted with tailings from this site (and two others for another site) (Pabst, 2011), but only one column test is presented here. The column was filled with 170 cm of reactive tailings, covered with 40 cm of cover material.

The tailings and cover material were put in place and consolidated under full saturation, layer by layer (15 cm thick). The top of the column was left open to the atmosphere during the wetting-drainage cycles, so evaporation could play a role during the tests. A ceramic porous plate was placed at the base of the column to control the water table position (using a flexible U-tube). The position of the water table was typically maintained 90 cm below the base of the column, i.e. about 300 cm below the surface of the monolayer cover. The U-tube at the base was also used to collect leachate for chemical analyses.

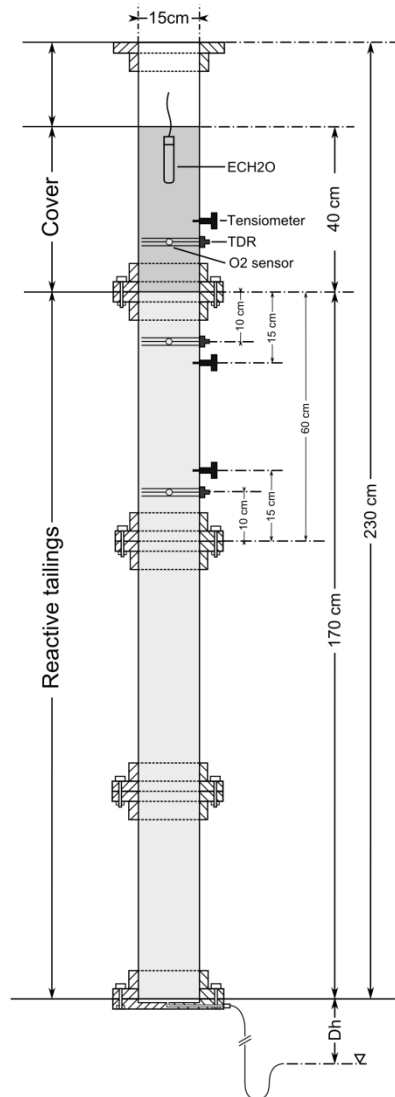A schematic view of the column set-up is presented in Figure 1.

Figure 1   Schematic view of the column-set up, showing the associated instrumentation

In addition, two small columns (50 cm in height, 10 cm in diameter) were also mounted with the reactive tailings to study their behaviour when left uncovered. These columns were also left open to evaporation. The suction applied at the base was about 2 m. The goal of the small column tests was to desaturate the tailings (between wetting cycles) to favour oxidation. These tests were specifically designed to help calibrate the geochemical models. Only one of these tests is considered here (with the small column filled with the same tailings as the large column).

Monthly wetting and drainage (drying) cycles were repeated to simulate, in a simplified (but controlled) manner, the site climatic conditions. Every 30 days or so (the cycles were not always of the same duration), 1,700 cm³ (about 10 cm) of deionised water was added at the top of the large column, and 500 cm³ (about 10 cm) into the small column. It usually took between 2 to 3 days before seepage was collected at the base of the large column (somewhat earlier for the small column). The infiltration of free (ponding) water into the surface of the cover lasted for about 5 to 6 days. A total of 19 cycles were applied to the large column (over a period of about 700 days), while 10 cycles were applied to the small column.

Two leachate samples of about 100 mL were collected during each wetting cycle: the first sample was taken at the beginning of drainage, and the second sample two (or three) days later. The values of the pH, electrical conductivity, and sulphate and Fe(II) concentrations were measured shortly after the samples were collected. Hydrochloric acid was added to the samples before the atomic absorption analyses to ensure all elements would remain in solution.

The experimental programme also included a complete hydrogeological analysis (Pabst et al., 2011; Pabst, 2011).

3   Numerical modelling

3.1   The Min3P code

The simulations were conducted using the Min3P code (Mayer et al., 1999, 2002). Min3P is a finite volume numerical model developed for simulating 3D reactive transport, including variably-saturated flow, diffusive gas transport, sulphide mineral oxidation, and kinetic or equilibrium controlled geochemical reactions. The model has been previously used in several studies of acid mine drainage (Jurjovec et al., 2004; Molson et al., 2005, 2008; Ouangrawa, 2007; Ouangrawa et al., 2009).

Min3P uses a control volume, global implicit and locally mass conservative solution method. Newton iteration is employed to linearise both the flow and transport equations. Richards' (1931) flow equation is solved by Min3P, assuming that the fluid is incompressible and the air phase is passive (no convection). In such conditions, the mass conservation can be written as:

$$S_r S_s \frac{\partial h}{\partial t} + \phi \frac{\partial S_w}{\partial t} - \nabla \cdot [k_{rw}\bar{k}\nabla h] - Q_w = 0 \qquad (1)$$

where S_r is the degree of saturation (-), S_s is the specific storage coefficient (m⁻¹), h is the hydraulic head (m), t is time (s), ϕ is the porosity (-), Q_w is a source–sink term (s⁻¹), k_{rw} is the relative permeability of the porous medium with respect to the water phase (-), and $\bar{k}$ is the saturated hydraulic conductivity tensor (m s⁻¹).

Equation (1) is nonlinear because S_r and k_{rw} both vary with head. A reduced (or effective) degree of saturation is used in the Min3P code, based on the van Genuchten (1980) equation:

$$S_{re} = \frac{S_r - S_{rw}}{1 - S_{rw}} = (1 + (\alpha_v \psi^{n_v})^{-m_v}) \qquad (2)$$

where ψ is the (negative) pressure head (m), α_v (m⁻¹), n_v (-) and m_v (-) are the van Genuchten (1980) equation parameters determined from the moisture-retention data (with $m_v = 1 - 1/n_v$), and S_{rw} is the degree of saturation at the residual water content (-).

The relative permeability is defined in Min3P by the Mualem, (1976) – van Genuchten (1980) equation:

$$k_{rw} = S_{re}^{\lambda} \left(1 - \left(1 - S_{re}^{1/m_v}\right)^{m_v}\right)^2 \qquad (3)$$

where λ is a parameter representing the degree of pore connectivity ($\lambda = 0.5$ is typically used).

In Min3P, the equation for advective-dispersive transport of the dissolved phase components is written as:

$$\frac{\partial}{\partial t}\left(S_r \phi T_j^w\right) + \frac{\partial}{\partial t}\left(S_g \phi T_j^g\right) + \nabla\left(q_w T_j^w\right) - \nabla\left(S_r \phi D_w \nabla T_j^w\right) - \nabla\left(S_g \phi D_g \nabla T_j^g\right) - \sum Q_j^{w,} = 0 \qquad (4)$$

where S_g is the degree of saturation of the gaseous phase (-), T_j^w and T_j^g are respectively the total water and gas phase concentrations of component j (kg m_{H2O}^{-3}), q_w is the Darcy fluid flux (m s^{-1}), D_w and D_g are respectively the dispersion tensors for the water and gas phase components (m^2 s^{-1}), and $Q_j^{w,}$ are the source-sink terms. The conservation law for the change in mineral mass is applied in the calculations.

Chemical reactions in the Min3P code can be equilibrium-controlled (law of mass action) or kinetically controlled. In the calculations presented in this paper, all minerals, except pyrite, use surface-controlled reactions.

Activity correction formulations and equilibrium constants for dissolved species originate from the thermodynamic equilibrium model MINTEQA2 (Allison et al., 1991) database. Additional rate data were obtained from various papers (Mayer et al., 1999, 2002). Mineral reactivity is assumed to be constant in the models presented in this paper.

The Min3P model uses the shrinking core model (Davies and Ritchie, 1986) to simulate direct oxidation using dissolved oxygen. However, when the pH is lower than about 3.5, Fe(III) may become the principal electron receptor in the reaction, and it can oxidise pyrite (even without oxygen) indirectly. Oxygen nonetheless remains a limiting reagent because it is needed to oxidise Fe(II) to Fe(III), so indirect oxidation can continue. This aspect is not included in Min3P. In the simulations conducted here, the pyrite reactivity was artificially increased to simulate this reaction (Pabst, 2011).

Oxygen transport through a partially saturated porous medium is described in Min3P as a Fickian diffusion process. For these calculations, the code was modified to take into account the bi-modal diffusion (in the water and gas phases) process described by Aachib et al. (2004). Oxygen advection in the aqueous phase is neglected.

For this study, the code was also modified to take into account hysteresis of the WRC, which may significantly affect the behaviour of the tailings and cover system. Monitoring of the column tests showed that after a few cycles the degree of saturation S_r of the tailings and cover material did not exceed 80% (which is much less than the 100% predicted from the drainage WRC) The consequences on oxygen flux can be drastic as the oxygen diffusion coefficient directly depends on S_r (Mbonimpa et al., 2003; Aachib et al., 2004). The maximum value at satiation $S_{w,max}$ ($\in$ [0 ; 1]) was thus defined in the code as an entry parameter. Min3P calculates the actual degree of saturation as $S_w.S_{w,max}$ and the air saturation as (1- $S_w.S_{w,max}$). More details on this aspect are presented in Pabst (2011).

Also, a few elements (Ni, Zn, Pb, Cu, Mn, Co, Cd) measured in the leachate were not linked directly to any minerals detected by XRD. These are deemed to originate from substitutions in the pyrite grains, as their concentrations tend to vary with sulphate concentrations. The pyrite oxidation model was thus modified to include these substitutions, by adding very small amounts (<0.05 mole/mole of pyrite) of selected metals as by-products of the oxidation reaction.

3.2   Modelling methodology

The first step of the modelling work was to compare and calibrate the chemical parameters using the small column test results. The presence of a single material (reactive tailings), their small size, and the more pronounced desaturation (inducing increased reactions) simplified the calibration. These results are then used directly to simulate the large column test (with the cover).

The numerical models were constructed as 1D columns having square cross-sectional areas of 1 m². The vertical size of each element is 1 cm for the small columns and 5 cm for the large one. The simulation time extends for just less than 700 days, using adaptive time stepping, with steps varying from a minimum of 10^{-20} day to a maximum of 1 day. Convergence tolerance is set at 10^{-4} for both hydrogeological and geochemical calculations (Newton iteration settings based on mass balance). Only the wetting WRCs are used in the calculations shown here. Min3P does not evaluate evaporation, but this factor cannot be neglected because it

affects water distribution and the concentration of the elements within the column. To approximate this phenomenon, a net influx was imposed at the top of the model, as calculated from the water added minus the evaporated amount (obtained from hydrogeological simulations conducted with Vadose/W (GeoSlope Inc.)). The pressure head imposed at the base of the column was obtained from calculations conducted with Vadose/W (Pabst et al., 2010, 2011; Pabst, 2011 for more details). The comparison between the hydrogeological results from Min3P and experimental measurements (of volumetric water content and suction) showed a very good agreement (not shown here).

For the geochemical boundary conditions, the demineralised water chemistry was used for the top influx, with concentrations slightly modified to ensure ionic neutrality. The chemical conditions remained opened at the outflow. The initial water quality within the columns was based on inductively coupled plasma – atomic emission spectrometry (ICP-AES) analyses on water samples taken during the set up of the columns.

The geochemical simulations of the tailings-cover response in the large columns were conducted in two steps. The water quality at the base of the cover was assessed first and then imposed as a boundary condition to the large column model containing the reactive tailings only. This partitioned method allows a better convergence of the models (when compared with simulations with both layers at the same time).

4   Results

4.1   Small column test

The results from the simulation have been compared with those obtained from the small column test. Some relatively minor adjustments were then implemented into the numerical model to better represent the observed data. The calibration essentially involved modifying the surface area of the minerals included in the model to fit the measurements. A few minerals were also added to the model, because the XRD analyses did not detect amorphous minerals (including some secondary precipitates). Min3P requires that minerals which may precipitate (like gypsum or ferrihydrite) be specified in the model.

It should be emphasised here that such a calibration is a delicate process, which involves maintaining an equilibrium between each mineral. A small modification can induce large variations on other components. Hence, some uncertainty exists as more than one combination of parameters may lead to the same response (for the specified conditions).

Figure 2 compares experimental data and numerical calculations for the small column with the reactive tailings, showing representative results (for more details see Pabst, 2011).
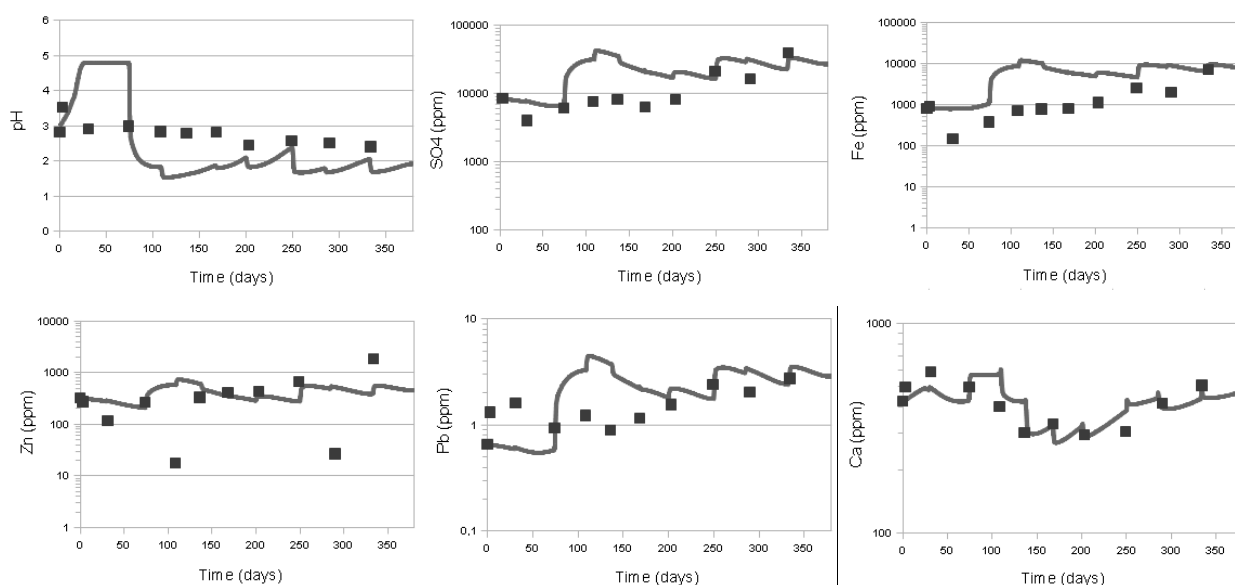

Figure 2   Geochemical measurements from the small column test and simulation results (points: experimental data; line: simulation)

The experimental data show that the tailings generate a very acidic leachate. After 10 cycles, the pH is below 2.5 and continues to decrease, while sulphates and iron concentrations are above 40,000 ppm and 8,000 ppm respectively, and still increasing. Metal concentrations also seem to increase regularly during the experiment. For instance, Zn and Pb follow the same trend as the sulphates, exceeding 1,800 ppm and 3 ppm respectively. The relatively high concentration of Ca (between 300 and 600 ppm) can be explained by anorthite dissolution at low pH.

As can be seen in Figure 1, the simulations reproduce fairly well the laboratory measurements. Some differences can be observed, especially during the first cycles, but in general, the trends are the same. The pH is underestimated, with values reaching 1.5 compared with measured values above 2.2. The calculations were extended a few more weeks after the end of the experiment, and the results confirm the general trend toward continued acidification and contamination.

Considering these encouraging results (and those obtained by others, as mentioned previously), it was inferred that Min3P could be used to simulate the behaviour of the tailings under more complex conditions. The next step thus consisted of implementing the calibrated parameters into the model of the large columns, and to compare the simulation results with the experimental measurements.

4.2   Large column test

The geochemical parameters, adapted from the small column modelling, are used here to simulate the response of the large column. Adjustments were made to take into account the addition of the cover material (not used in small column). The influx water geochemistry was set by modelling the flow of water within the cover alone, using basic parameters (with no possibility to precisely calibrate the model). It was observed that the influx of oxygen decreased due to the presence of the cover. However, as the cover was prone to desaturate, it could not effectively prevent oxygen migration (although oxygen partial pressure is lower than for exposed tailings). The early calculations with Vadose/W indicated that the concentration of oxygen at the top of the reactive tailings was about 15% (on average), which is close to actual measurements in the laboratory column (Pabst, 2011). The shrinking core model parameters (including the surface area and volumetric fractions of each mineral) remained unchanged from the small column model (Table 2).

The hydrogeological conditions were adapted for the different flow conditions (increased quantity of added water, different suction at the base), and the mesh (element) vertical size was increased from 1 to 5 cm. Calculations with the larger mesh size showed that this has no consequence on the results. Other numerical settings remained the same as those used for the small column simulation.

Figure 3 summarises the experimental measurements and simulation results for the large column test.
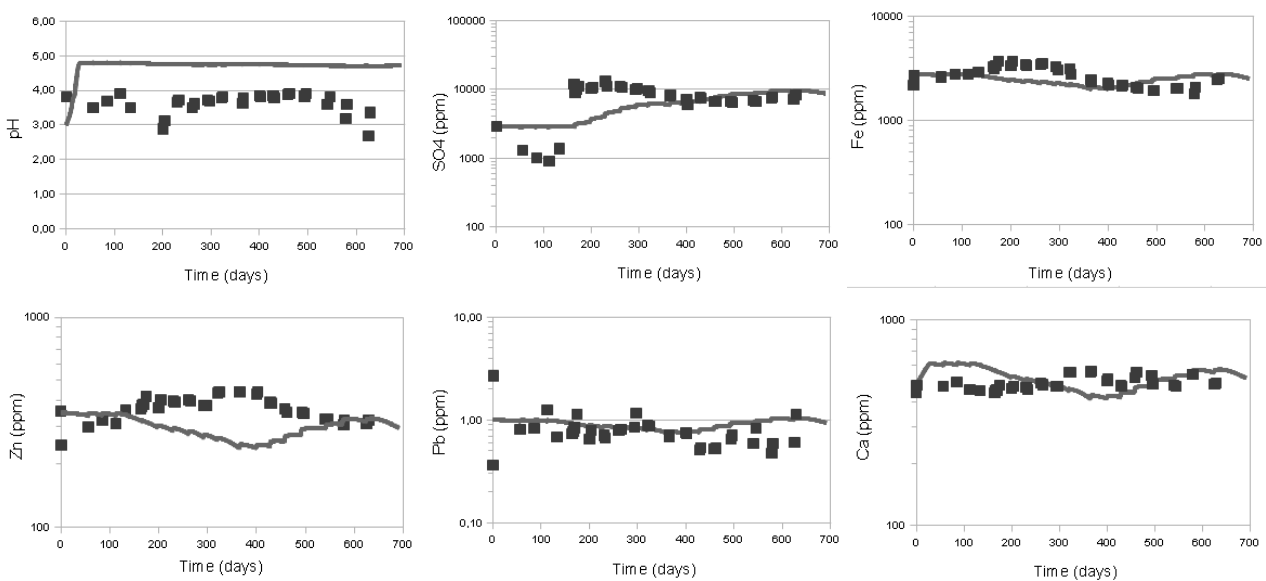

Figure 3   Experimental results for the large column test compared with numerical simulation results (tailings + cover; points: experimental data; line: simulation)

Experimental measurements show that the large column, filled with sulphidic tailings and covered with a non acid-generating tailing layer, still produces acid. The initial pH is around 4 and remains around 3.8 for some time. After 16 cycles, the pH decreases again to about 2.5, a value observed until the end of the test. Sulphate concentrations are initially below 3,000 ppm, but increase to 12,000 ppm at cycle 6, and then decrease smoothly until reaching a plateau around 7,000 ppm. Iron follows the same trend, except the increase at cycle 6 is not so abrupt. The concentrations of metals remained relatively constant during the test (including Zn at around 320 ppm). Calcium, produced by anorthite dissolution, remains around 500 ppm. The geochemical response appears to have reached a (pseudo) steady state, with tailings producing acidity primarily as a result of indirect oxidation (for pH < 3.5).

Despite showing the same trends, the numerical simulations show some discrepancies with the experimental results, especially at the beginning of the test. Nonetheless, fairly similar values are reached after the tenth cycle. The differences may be attributed to the complex reactions within the tailings, and also by the initial water chemistry and solid grain surface conditions (resulting from more than 20 years of exposure) which are difficult to capture in the simulation. It is seen that the simulated pH remains too high (at 4.8) compared with experimental results (values close to 3.8 were measured). This may be a consequence of not explicitly including the effect of indirect oxidation in the simulation. However, as sulphate and metal concentrations are fairly well reproduced, and the same trends are obtained, it can be assumed that the simulations reflect fairly well the general behaviour of the tailings, when the parameters calibrated from the small column tests are used for the larger column. These parameters are also being used to simulate field conditions and to asses alternate reclamation methods (such as layered cover systems).

5   Discussion and conclusion

A large column test was conducted to assess the geochemical behaviour of acid-generating tailings and to evaluate the efficiency of a monolayer cover made of non acid-generating tailings. A test on a small column, filled with the same reactive tailings, was used to define their geochemical parameters. Numerical simulations were conducted with Min3P to try to reproduce laboratory tests results.

Experimental results clearly indicate that the reactive tailings generate contaminated acid drainage for the imposed conditions, and that the cover configuration was not very efficient to prevent pyrite oxidation. As a consequence, the leachate sampled at the base of the large column contained high concentrations of sulphate and metals, showing a pH of 3.8.

The numerical simulations conducted using Min3P reproduced fairly well the experimental results obtained in this study, including sulphate and metal concentrations. Simulated long term trends were also similar to those in the laboratory experiment (despite some discrepancies). Field conditions differ from those imposed in the laboratory experiments, particularly in regards to the water table position (which may vary over time and location), material properties (not homogeneous) and cover thickness. The geochemical behaviour may consequently differ from these lab results. Further simulations have been conducted to take such variations into account, to help assess if the cover system is appropriate for this site. Alternate reclamation methods were also tested using the Min3P model; these results are presented in Pabst (2011).

Acknowledgements

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