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Effectiveness of various cover scenarios on the rate of sulfide oxidation of mine tailings

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Abstract

Long term environmentally sound disposal of the millions of tons of mining residue is a serious challenge to the international mining industry. This paper evaluates, through a numerical investigation, the potential performance of desulfurized tailings as a cover material for the reduction of acidic drainage from sulfidic tailings. This evaluation is facilitated through a comparison of various cover types as decommissioning options. The cover types considered consist of a desulfurized tailings material cover exposed to ambient climate conditions, a water cover (flooded tailings), and a combination cover type. As part of the evaluation of cover performances, the effect of climatic variability on the potential rate of sulfide oxidation in tailings with an open ground surface, was also assessed. The numerical analysis was conducted using the model PYROX, which was modified to allow for variably-saturated conditions, time varying moisture contents, and to account for the temperature dependence of Henry's law and gas diffusion. In the case study presented here, the benign cover material consists of a low sulfide waste stream (cassiterite float tails, CFT), a by-product of the production of tin concentrate (cassiterite, SnO_2). Modelling results after a simulation period of 100 years indicate that a water cover alone or an exposed CFT cover alone are both less effective options than the combined cover type. A water cover alone leads to a reduction of approximately 99.1%, in the oxidation rate relative to uncovered tailings while the combined cover type results in the lowest potential extent of sulfide oxidation after mine closure-an approximately 99.8% reduction. Importantly, a CFT cover exposed to ambient environmental conditions can still substantially reduce the sulfide oxidation rate, by approximately 75–82% over a 100-year time period, relative to uncovered tailings. Variability in precipitation (and hence percent saturation of the surface layer) had less of an effect on the potential sulfide oxidation rate than did the cover type. The performance of the exposed CFT cover varied by less than 10%, within the range of climatic conditions expected at the Renison Bell mine site in southwest Tasmania, Australia. Although the modelling results indicate that the combined water and CFT cover is the best option, this approach achieves only a minor improvement over the water cover alone.

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1. Introduction

Acid mine drainage (AMD) is of worldwide environmental concern for the mining industry. It is produced when sulfide minerals are exposed to oxygen and water to release protons, sulfate and dissolved metals (Lowson, 1982; Skousen and Ziemkiewicz, 1995). Formation and uncontrolled release of AMD can cause considerable damage to the environment, and prevention and remediation of this problem is of concern to mining companies as well as governments. It is by far the most technically challenging issue facing the international mining industry today (Feasby et al., 1997).

Sulfide mineral oxidation is due to the exposure of sulfide minerals to atmospheric oxygen. As the sulfide minerals in shallow portions of the tailings become depleted due to their oxidation, oxygen migrates further down into the tailings profile, leading to the propagation of the oxidation reaction front. The advancement of this front is primarily limited by the rate of oxygen diffusion through pore spaces and within tailings particles (Davis and Ritchie, 1986). Oxygen ingress due to gas advection can also be significant in mine waste (Ritchie, 1994). Both diffusion and advection transport processes are significantly affected by the moisture content of the waste material and oxygen ingress under saturated conditions is strongly inhibited (Ritchie, 1994). If oxidation takes place under saturated conditions, the flow velocity of the groundwater and its oxygen content determines how quickly a reaction front can advance (e.g. Lichtner, 1992; Engesgaard and Kipp, 1992). In addition, the propagation and sharpness of the reaction front will be affected by the rate of oxygen consumption (i.e. the rate of sulfide oxidation), and the availability of sulfide minerals (Ritchie, 1994).

Various disposal/remediation options for mine tailings have been proposed by researchers, such as blending and layering of mine waste (Mehling et al., 1997; Delaney et al., 1997), flooded open-pit disposal (Orava et al., 1997), use of water treatment sludge to neutralize AMD (Aubé and Payant, 1997; Zinck, 1997), and covering of tailings with low permeability benign material (Woyshner and Yanful, 1995). In some cases bulk tailings are being desulfurized (sulfide component removed) to provide sufficient material with appropriate geotechnical and geochemical properties for a surface cover layer (Aubertin et al., 1997; Hanton-Fong et al., 1997). The effectiveness of desulfurised tailings covers as close-out remediation options for the reduction of AMD is the topic of this paper.

A primary objective of many tailings cover designs is to inhibit the influx of oxygen into the tailings, thus minimizing the sulfide oxidation rate. A second objective may be to limit the infiltration of rainwater or surface water, which results in the migration of existing oxidation products already in the tailings mass.

‘Wet’ covers designed to minimize rainwater infiltration while maintaining a high saturation of water in the cover layer to minimize oxygen diffusion into the underlying tailings have been proposed as remedial options for sulfidic tailings (Swanson et al., 1997; Wilson et al., 1997; Woyshner and Swarbrick, 1997). Alternatively, water covers which maintain full saturation in the underlying tailings have been proposed (Li et al., 1997; Amyot and Vézina, 1997). These covers have the potential to significantly inhibit the ingress of oxygen into the tailings, primarily because of the low solubility (8.6 mg l^{-1} at 25 °C) and low diffusion coefficient (2×10^{-9} m^2 s^{-1}) of oxygen in water, and also because of the possible generation of an organic-rich anaerobic layer on the surface of the flooded tailings (FT) following establishment of aquatic vegetation. A water cover alone, however, may increase infiltration into the tailings. In addition, the construction and maintenance of an engineered water cover over tailings is a complex task, suggesting this alternative may be more costly. A third alternative consists of a cover layer of suitable benign material of low permeability overlain by a permanent water cover. This cover scenario addresses both the problem of surface water infiltration and oxygen ingress. However, this option may be the most expensive given that it involves the placement of a layer of cover material in addition to maintaining within an appropriately engineered structure a depth of water above the tailings.

The technical feasibility of proposed close-out options for three tailings dams at the Renison Bell tin mine in southwest Tasmania, Australia is currently being assessed (Jones et al., 1997). Renison Bell is the world's second largest underground tin mine.The mine site is at 42° south latitude and the climate is sub-temperate, with rainfall greatly exceeding evaporation throughout most of the year (mean annual precipitation 2.21 m, mean annual evaporation 0.70 m). The tailings at Renison Bell contain between 10 and 30% sulfur as sulfide, which is largely in the form of the reactive mineral pyrrhotite. The sulfides contain only low concentrations of other metals, which means that acidity and concentration of iron and sulfate are the main solute species in runoff and seepage. Cassiterite flotation tailings (CFT), a fine-grained relatively non-reactive material produced as a byproduct of the production of tin concentrate, has been proposed as a cover material. The CFT contains low concentrations of sulfides (less than 1.5%), is aluminosilicate rich, contains up to 5% siderite, and has a net negative acid generating potential (Jones et al., 1997). Greenhouse and small-scale field trials have indicated that this material is also a good substrate for plant growth.

Details of the hydrological, hydrogeochemical and water balance assessments of the tailings dams have been published elsewhere (Ellerbroek and Jones, 1997; Jones et al., 1997). Unsaturated flow modelling of the surface layer of the proposed CFT cover has shown that the water content of the CFT cover would be highly variable through time and in the vertical profile due to changing climatic conditions at the site. Thus steady-state approximations for predicting oxygen diffusion and sulfide oxidation would be of limited utility for predictive modelling of cover efficacy, since oxygen diffusion rates would vary substantially through time as a result of changes in cover saturation (Reardon and Moddel, 1985).

The propogation of oxidation reaction fronts in unsaturated mine tailings has previously been investigated by a number of researchers (e.g. Davis and Ritchie, 1986; Blowes and Jambor, 1990; Wunderly et al., 1996). In addition, numerical studies of advancing redox fronts in aquifers have been performed (e.g. Lichtner, 1992; Engesgaard and Kipp, 1992; Bain et al., 2001). These studies were helpful in estimating both the rate of propagation of the fronts and changes in water composition along the groundwater flowpath. However, the effect of transient moisture conditions in unsaturated tailings on sulfide mineral oxidation rates have not been investigated previously.

This paper evaluates, through a numerical investigation, the potential performance of desulfurized tailings as cover material under variable recharge conditions. This is accomplished through a comparison of the potential effectiveness of various types of cover configurations in reducing the rate of sulfide oxidation in the underlying tailings. The cover scenarios considered here utilize either a cover of CFT material, a water cover, or a combination of these cover types. In addition, the effect of climatic variability on the rate of sulfide mineral oxidation is evaluated in tailings exposed to ambient environmental conditions at ground surface through the inclusion of transient moisture contents in the unsaturated zone.

PYROX, a transient oxygen diffusion-sulfide oxidation model developed by Wunderly et al. (1996), is used to investigate the cover performance and compare the scenarios. The model also computes the reaction front position over time and has been applied previously to assess sulfide oxidation at various field sites. (e.g. Wunderly et al., 1996; Al., 1996). In the framework of this study, modifications were made to PYROX which allow analysis of sulfide mineral oxidation under both variably and fully saturated conditions, with temporally-spatially changing moisture content conditions to mimic the effect of natural variability in precipitation, and to account for the temperature dependence of oxygen diffusion and solubility. These modifications increase the versatility of PYROX and enhance its applicability to mine tailings problems.

2. Model development

PYROX (Wunderly et al., 1996) is a numerical model that couples oxygen diffusion and sulfide-mineral oxidation for the simulation of acid generation in unsaturated tailings. It allows for the spatial variability of various input parameters, including moisture content. This feature is especially beneficial for the simulation of sulfide oxidation in the unsaturated zone. PYROX uses a one-dimensional finite element scheme to simulate the transport of oxygen by diffusion, and a shrinking-core model (Levenspiel, 1972) to quantify the oxidation of sulfide-mineral grains. The flow of groundwater, advective oxygen transport, and the transport of reaction products from sulfide oxidation is not considered. This formulation implies that the rate of sulfide-mineral oxidation is limited by diffusive transport of oxygen to the reaction site.

The direction of oxygen diffusion in the model domain is vertically downwards from the ground surface toward the watertable. At ground surface, the concentration of oxygen is assumed to be constant and equal to the atmospheric concentration. Therefore, the model incorporates a constant oxygen concentration (Type 1 boundary condition) at the top boundary and a zero concentration gradient (Type 2 boundary condition) at the bottom boundary, representing conditions where oxygen has been depleted.

For this study, PYROX was modified to allow for fully saturated conditions, time-varying moisture contents, as well as other provisions required to better characterize conditions in the tailings. The modifications made to PYROX are described below.

The original version of PYROX computes a bulk diffusion coefficient, D_b ($m^2\ s^{-1}$), that is based on an empirical expression developed by Reardon and Moddel (1985):

$$D_b(x,t) = 3.98 \times 10^{-9} \times \left[\frac{\theta_a(x,t) - 0.05}{0.95} \right]^{1.7} \times (T(x) + 273.15)^{1.5} \tag{1}$$

where x is the spatial coordinate, t is time, θ_a is the air-filled porosity ($m^3\ m^{-3}$), and T is the temperature (°C). However, this expression is limited to the unsaturated zone and oxygen diffusion into the pore water is neglected. Consequently, two alternative expressions for bulk diffusion coefficient that account for oxygen diffusion in both the air-filled and the water-filled pore space are incorporated into PYROX. The first is a theoretical expression taken from Šimunek and Suarez (1993) with the tortuosity factors in both phases defined in a manner similar to Millington and Quick (1961) m:

$$D_b(x,t) = D_a^o \frac{\theta_a(x,t)^{10/3}}{\phi(x)^2} + HD_w^o \frac{\theta_w(x,t)^{10/3}}{\phi(x)^2} \tag{2}$$

where ϕ is the porosity ($m^3\ m^{-3}$), θ_w is the moisture content ($m^3\ m^{-3}$), H is Henry's law constant (dimensionless) defined as the concentration of oxygen in the water phase divided by the concentration of oxygen in the gas phase at equilibrium, and D_a^o and D_w^o are the diffusion coefficients of oxygen ($m^2\ s^{-1}$) in pure air and pure water, respectively. The second is a semi-empirical expression for sandy tailings from Elberling et al. (1993):

$$D_b(x,t) = (0.273 \pm 0.08) D_a^o \left(\frac{\theta_a(x,t)}{\phi(x)} \right)^{3.28 \pm 0.4} + HD_w^o \left(\frac{\theta_w(x,t)}{\phi(x)} \right)^{3.28 \pm 0.4} \tag{3}$$

All three expressions (Eqs. (1)–(3)) are interchangeable and any one expression can be selected in the input file for PYROX. Following a preliminary analysis, Eq. (2) was selected for this study.

The original version of PYROX is based on a mass balance equation governing bulk oxygen diffusion into only the air-filled pore-space:

$$\underbrace{\theta_a(x,t) \frac{\partial C_a(x,t)}{\partial t}}_{\text{STORAGE TERM}} = \underbrace{D_b(x,t) \frac{\partial C_a(x,t)}{\partial x^2}}_{\text{DIFFUSION TERM}} - \underbrace{q(x,t)}_{\text{SINK TERM}} \tag{4}$$

where C_a is the concentration of oxygen in the pore space ($kg\ m^{-3}$) and q is the sink term due to oxygen consumption by the particles in the bulk tailings ($kg\ m^{-3}\ s^{-1}$) (Wunderly et al., 1996). Since the bulk diffusion coefficient expression of Eqs. (2) and (3) account for oxygen diffusion in both the air-filled and water-filled pore space, the storage term of Eq. (4) requires an adjustment to account for the storage of oxygen in the water-filled pore space, thereby expanding the applicability of the mass balance equation to the saturated zone (Šimunek and Suarez, 1993):

$$(\theta_a(x,t) + H\theta_w(x,t)) \frac{\partial C_a(x,t)}{\partial t} = D_b(x,t) \frac{\partial C_a(x,t)}{\partial x^2} - q(x,t) \tag{5}$$

Other modifications to PYROX include the incorporation of an expression to determine the temperature-dependent Henry's law constant, H, based on the Van't Hoff equation (e.g. Appelo and Postma, 1994):

$$\log K_H = \log K_{H(25C)} - \frac{\Delta H_r^o}{2.303R} \times \left(\frac{1}{(T(x) + 273.15)} - \frac{1}{298.15} \right) \tag{6}$$

$$H = K_H R (T(x) + 273.15)$$

where K_H is Henry's law constant ($K_{H(25C)} = 10^{-2.8996}$ M atm^{-1} at 25 °C), R is the universal gas constant (8.314 J K^{-1} mol^{-1}), and ΔH_r^o is the reaction enthalpy for the dissolution of oxygen in water (− 10 kJ mol^{-1}).

In addition, expressions to determine temperature-dependent oxygen diffusion coefficients in the pure phases are included as:

$$\begin{aligned} D_w^o &= (0.0559 \times T(x) + 0.9864) \times 10^{-9} \\ D_a^o &= (0.0119 \times T(x) + 1.7764) \times 10^{-5} \end{aligned} \tag{7}$$

These expressions are based on regression analysis of diffusion coefficients at various temperatures taken from Glinski and Stepniewski (1985) and are valid over a temperature range from 0 to 30 °C.

Moisture contents in the vadose zone respond to changes in precipitation and evaporation. PYROX was modified such that moisture contents along a vertical profile can be updated at regular time intervals to simulate infiltration events in the tailings material, provided a moisture content database (i.e. set of moisture content profiles at different times) is available.

The bulk oxygen concentration in the subsurface, C_T (kg m^{-3}), is defined as the sum of the oxygen concentration in the air phase and water phase of the pore space:

$$C_T(x,t) = \theta_w(x,t) C_w(x,t) + \theta_a(x,t) C_a(x,t) \tag{8}$$

where C_w and C_a are the concentrations of oxygen in water (kg m^{-3}) and air (kg m^{-3}), respectively, and are related through Henry's law constant:

$$C_w(x,t) = H C_a(x,t) \tag{9}$$

which assumes equilibrium between the air and water phase. This assumption is adequate considering that the time scale of oxygen consumption by sulfide oxidation is slow in comparison to the time scale of mass transfer processes between the gas phase and the aqueous phase (Davis and Ritchie, 1986).

Updating the moisture contents in the vertical profile can lead to mass balance errors because PYROX neglects the advective displacement of soil gas due to changes in moisture content. Mass conservation of oxygen can be enforced by adjusting the dissolved and gaseous oxygen concentrations to satisfy:

$$\begin{aligned} &\theta_w(x,t) C_w(x,t+\Delta t) + \theta_a(x,t) C_a(x,t+\Delta t) \\ &\quad = \theta_w(x,t+\Delta t) C_w^{\text{adjusted}}(x,t+\Delta t) \\ &\qquad + \theta_a(x,t+\Delta t) C_a^{\text{adjusted}}(x,t+\Delta t) \end{aligned} \tag{10}$$

where $t + \Delta t$ denotes the new time at which the moisture contents are updated. Theoretically, after completion of a time step, the oxygen concentration in air can be adjusted ($C_a^{\text{adjusted}}(x, t+\Delta t)$) to preserve the bulk oxygen mass by combining Eqs. (9) and (10):

$$\begin{aligned} &C_a^{\text{adjusted}}(x,t+\Delta t) \\ &\quad = C_a(x,t+\Delta t) \frac{(\theta_a(x,t) + H\theta_w(x,t))}{(\theta_a(x,t+\Delta t) + H\theta_w(x,t+\Delta t))} \end{aligned} \tag{11}$$

However, mass conservation becomes problematic during infiltration. Moisture content increases may cause $C_a^{\text{adjusted}}(x,t+\Delta t)$ to exceed the atmospheric concentration. Oxygen concentrations greater than atmospheric cannot occur under normal circumstances. Because the direction of air flow and also the occurrence of air entrapment during infiltration events cannot be determined explicitly, mass conservation of oxygen is not accounted for by PYROX.

The percent relative error in bulk oxygen mass due to the neglect of mass conservation when moisture contents are updated can be expressed as the oxygen mass balance error divided by the mass of oxygen consumed due to sulfide mineral oxidation:

$$E_{\text{rel}}(\%) = \frac{E_{\text{abs}}}{M_{O_2\ \text{cons}}} \times 100 \tag{12}$$

The oxygen mass balance error, E_{abs} (kg), is calculated as the change in oxygen mass caused by a change in moisture content, integrated over the vertical profile:

$$E_{\mathrm{abs}} = \sum_{x=0}^{x_{\max}} C_{\mathrm{a}}(x,t+\Delta t)V(x)[(\theta_{\mathrm{a}}(x,t)+H\theta_{\mathrm{w}}(x,t)) - (\theta_{\mathrm{a}}(x,t+\Delta t)+H\theta_{\mathrm{w}}(x,t+\Delta t))] \qquad (13)$$

where $V(x)$ is the element volume (m^3) of bulk porous medium, determined from the grid spacing. The mass of oxygen consumed, $M_{\mathrm{O_2\ cons}}$(kg), can be determined from the total quantity of sulfate produced over the profile as a result of pyrite or pyrrhotite oxidation:

$$M_{\mathrm{O_2\ cons}} = \sum_{x=0}^{x_{\max}} C_{\mathrm{SO_4}}(x) \times \varepsilon \times \mathrm{gfw}_{\mathrm{O_2}} \qquad (14)$$

where $C_{\mathrm{SO_4}}$ (x) is the number of moles of sulfate produced at every node, ε is the ratio of oxygen to sulfur consumed based on stoichiometry and $\mathrm{gfw}_{\mathrm{O_2}}$ is the gram formula weight of oxygen (kg mole^{-1}). Eq. (12) is incorporated into PYROX so that the relative error in bulk oxygen mass (summed yearly and cumulative from start) is computed in relation to the yearly mass of oxygen consumed and the cumulative mass of oxygen consumed from the start of a simulation.

3. Description of numerical simulations

The four scenarios modelled by PYROX in increasing order of technical/engineering complexity were: 1) exposed tailings (ET), 2) tailings covered by 1 m of exposed CFT (EC), 3) a permanent water cover (FT), and 4) tailings covered by 1 m of CFT overlain by a permanent water cover (FC).

All scenarios are simulated to an end-time of 100 years to determine the long-term effectiveness of the cover. All input parameters, excluding moisture content, used to describe the CFT cover and the tailings mass are presented in Fig. 1. The diffusion coefficient for the oxidized rim of all particles (D2) was assigned a value of $1 \times 10^{-14}\ \mathrm{m}^2\ \mathrm{s}^{-1}$, based on the results of several previous studies (Johnson, 1993; Al, 1996). The remaining parameters in Fig. 1 were obtained from previous investigations (Wright and Nefiodovas, 1997; Ellerbroek, 1998) of the aforementioned mine tailings dams.

Unsaturated flow modelling by Ellerbroek (1998) using the WAVES model (Dawes and Short, 1993) was used to calculate the relative saturation through 1 m of exposed CFT material (unvegetated surface) overlying 5 m of tailings in response to low, medium and high annual rainfall which spans the range of climatic conditions at the mine site. WAVES is a one-dimensional soil-water model that uses Richard's Equation to redistribute water in the unsaturated zone. It simulates the movement of water, energy and solute in a vertical profile. The energy available for evaporation is determined using a radiation balance based on Beer's Law, and evaporation at the soil surface is estimated using the Penman-Monteith combination equation (Monteith, 1965; Penman, 1948). The Broadbridge

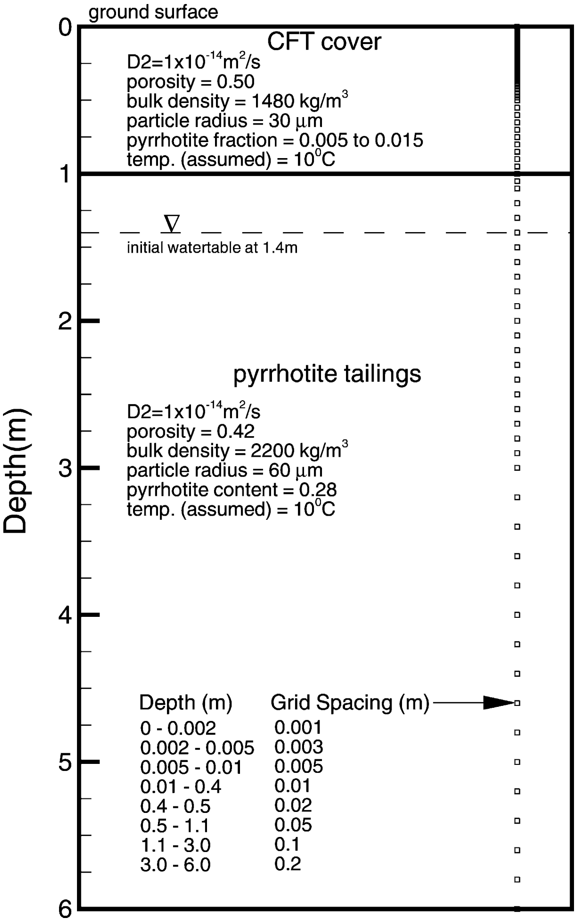

Fig. 1. Model domain, grid spacing, and input parameters for Scenario EC and FC. These specifications also apply to Scenario ET and FT excluding the 1 m CFT cover which is replaced by tailings to ground surface.

and White (1988) function is used to describe the relationship between soil-water pressure (Ψ), volumetric water content (θ_w) and hydraulic conductivity (K):

For $K_n/K_s < 4C(C-1)$,

$$\Psi(S_e) = -C^{-1}\lambda_s\left\{\ln\left(\frac{C-S_e}{C-1}\right)+\frac{1}{2}\ln\left(\frac{K_s(C-1)}{K(S_e)(C-S_e)}\right)+\frac{K_n-2C(C-1)\Delta K}{\kappa}\times\left[\arctan\left(\frac{K_s-(2C-1)\Delta K}{\kappa}\right)-\arctan\left(\frac{K_s-[2S_e(C-1)+1]\Delta K}{\kappa}\right)\right]\right\} \quad (15)$$

For $K_n/K_s \geq 4C(C\text{-}1)$,

$$\Psi(S_e) = -\lambda_s\left(\frac{1}{C}\ln\frac{C-S_e}{C-1}+\frac{C(C-1)}{r_1-C}\frac{\Delta K}{K_n}\ln\left|\frac{r_1-S_e}{r_1-1}\right.\times\left|-\frac{C(C-1)}{r_2-C}\frac{\Delta K}{K_n}\ln\left|\frac{r_2-S_e}{r_2-1}\right|\right)\right. \quad (16)$$

where

$$S_e = \frac{\theta_w-\theta_n}{\theta_s-\theta_n}$$

$$(K-K_n)/\Delta K = S_e^2(C-1)(C-S_e)$$

$$\Delta K = K_s - K_n$$

and

$$\kappa = \{[(2C-1)^2\Delta K - K_s]K_n\}^{1/2}$$

$$r_1 = \{K_n/[2\Delta K(C-1)]\}[1+(1-4C(C-1)\Delta K/K_n]^{1/2}$$

$$r_2 = \{K_n/[2\Delta K(C-1)]\}[1-(1-4C(C-1)\Delta K/K_n]^{1/2}$$

where θ_n and θ_s are the initial and saturated water contents, respectively, K_n and K_s are the hydraulic conductivities at θ equal to θ_n and θ_s, respectively, and C and λ_s are curve fitting parameters that can be adjusted to fit the water retention model (i.e. Eqs. (15) and (16)) to laboratory data. The hydraulic parameters used by Ellerbroek (1998) for the CFT cover and the tailings are presented in Table 1.

Table 1
Hydraulic parameters for the CFT cover and tailing, from Ellerbroek (1998)

Hydraulic parameter	CFT cover	Tailings
Saturated hydraulic conductivity (K_s)	0.144 m day^{-1}	0.029 m day^{-1}
Porosity (max. water content, θ_s)	0.5	0.42
Residual water content (θ_r)	0.02	0.09
λ_s	0.25	0.50
C	1.01	1.03

Three sets of time-series moisture content data were generated using WAVES, each consisting of daily moisture content measurements along a vertical profile over a 365-day period in response to low (1.87 m), medium (2.28 m), or high (2.48 m) annual rainfall conditions. The grid spacing applied to the PYROX simulations, shown on Fig. 1, is based on the spatial distribution of the moisture content measurements in these data sets. The grid spacing is smallest near ground surface to account for rapid changes in moisture content due to infiltration events and steep oxygen concentration gradients, and it generally increases with depth.

In the case of Scenario ET and Scenario EC (CFT cover), the effect of low, medium, and high annual rainfall conditions on the rate of sulfide oxidation were assessed by specifying moisture contents from one of the time-series moisture content data sets described above. Therefore, three simulations were conducted for Scenario ET, each based on one of the three rainfall conditions. However, six simulations were conducted for Scenario EC to account for rainfall variation and to assess the sensitivity to pyrrhotite content in the CFT cover. A 0.5% pyrrhotite content was assigned to the CFT for the first three simulations, each using one of the three rainfall conditions. A 1.5% pyrrhotite content was used for the CFT for the last three simulations. The two values used for sulfide content span the range of concentrations measured in samples of CFT.

As indicated above, the time-series data sets consist of moisture contents for a one-year period only. PYROX was set up to re-use the specified data set for every simulated year until the end time is reached. The data sets generated by Ellerbroek (1998) for the exposed CFT cover in Scenario EC are also used for the ET in Scenario ET because equivalent data sets for ET were not available. This approach requires the top 1 m of ET for Scenario ET to have a porosity equivalent to the CFT cover (0.50). The resulting saturations for ET are thus representative of a layered tailings mass with fine-to-medium grained tailings overtop of relatively finer tailings. This approach is representative for the covered tailings scenario (EC), and provides a worst case for Scenario ET. The generation of AMD for Scenario ET may therefore be slightly overestimated.

In the case of Scenario FT and FC, the moisture contents throughout the vertical profile were set to equal the porosity to simulate fully saturated conditions that prevail under a water cover. To assess the effect of variation in pyrrhotite content of the CFT material two simulations were conducted for Scenario FC. The first simulation used 0.5% pyrrhotite in the CFT, while the second used 1.5% pyrrhotite in the CFT. Domain dimensions, grid spacing, and input parameters, excluding moisture content, for Scenario EC and FC are shown in Fig. 1. These same specifications apply to Scenario ET and FT with the exception of the 1 m CFT cover which is replaced by tailings material to ground surface.

Fluctuations in barometric pressure can cause the advective transport of gases in systems with an open ground surface (Massmann and Farrier, 1992). Relatively small gradients in total pressure changes can result in advective gas fluxes, which are much larger than diffusive fluxes (Alzaydi and Moore, 1978; Thorstenson and Pollock, 1989a,b). The effect of barometric pumping of air into the exposed CFT and ET (Scenarios EC and ET, respectively) is not considered in this paper. It is unlikely that the long-term rate of sulfide oxidation is affected significantly by this process because atmospheric pressure changes are short-lived and may lead to the infiltration of oxygen into deeper areas when atmospheric pressure increases, but also to the removal of oxygen from the subsurface, when atmospheric pressure decreases. The extent of barometric pumping is also dependent on the permeability of the material, and the permeability of the CFT and tailings is moderately low (1.7×10^{-6} and 3.3×10^{-7} m s^{-1}, respectively). In addition, the depth of gas migration is very limited if the unsaturated zone is shallow (Massmann and Farrier, 1992), as is the case with tailings considered here.

The simulation results for the fully saturated cases (Scenarios FT and FC) are valid only when water loss through the tailings material is not excessive. If the flow through the tailings is substantial, sulfide mineral oxidation rates may be significantly higher than predicted in the simulations because dissolved oxygen is transported into the tailings material by advection at a faster rate than delivered by diffusion.

4. Results

The oxygen flux entering the tailings is the principal limitation on the rate of oxidation of sulfide minerals. The amount of oxygen entering is almost equivalent to the amount of oxygen consumed due to the development of a quasi-steady state condition (i.e. the change in oxygen stored in the system will be negligible in comparison to the flux). The mass of the reaction products sulfate and iron is proportional to the amount of oxygen consumed (Eq. (14) and either product is directly indicative of the effectiveness of the cover scenarios in minimizing sulfide oxidation and of the AMD potential after mine closure. Cumulative and yearly plots of sulfate production are used to compare cover performance for all simulations (Fig. 2).

The plots in Fig. 2 indicate that the effectiveness of the cover scenarios vary significantly. Scenario ET results in the greatest quantity of sulfate production (ranging from 831 to 970 kg m^{-2} after 100 years for the three rainfall conditions) while Scenario FC (combined cover), results in the lowest quantity of sulfate produced (0.9 to 1.6 kg m^{-2} after 100 years for 0.5 and 1.5% pyrrhotite content in the CFT). In comparison to Scenario ET, the mass of sulfate generated after 100 years is on average a factor of 4 to 5 (75 to 82%) less for Scenario EC (CFT cover), a factor of 114 to 133 (99.1%) less for Scenario FT

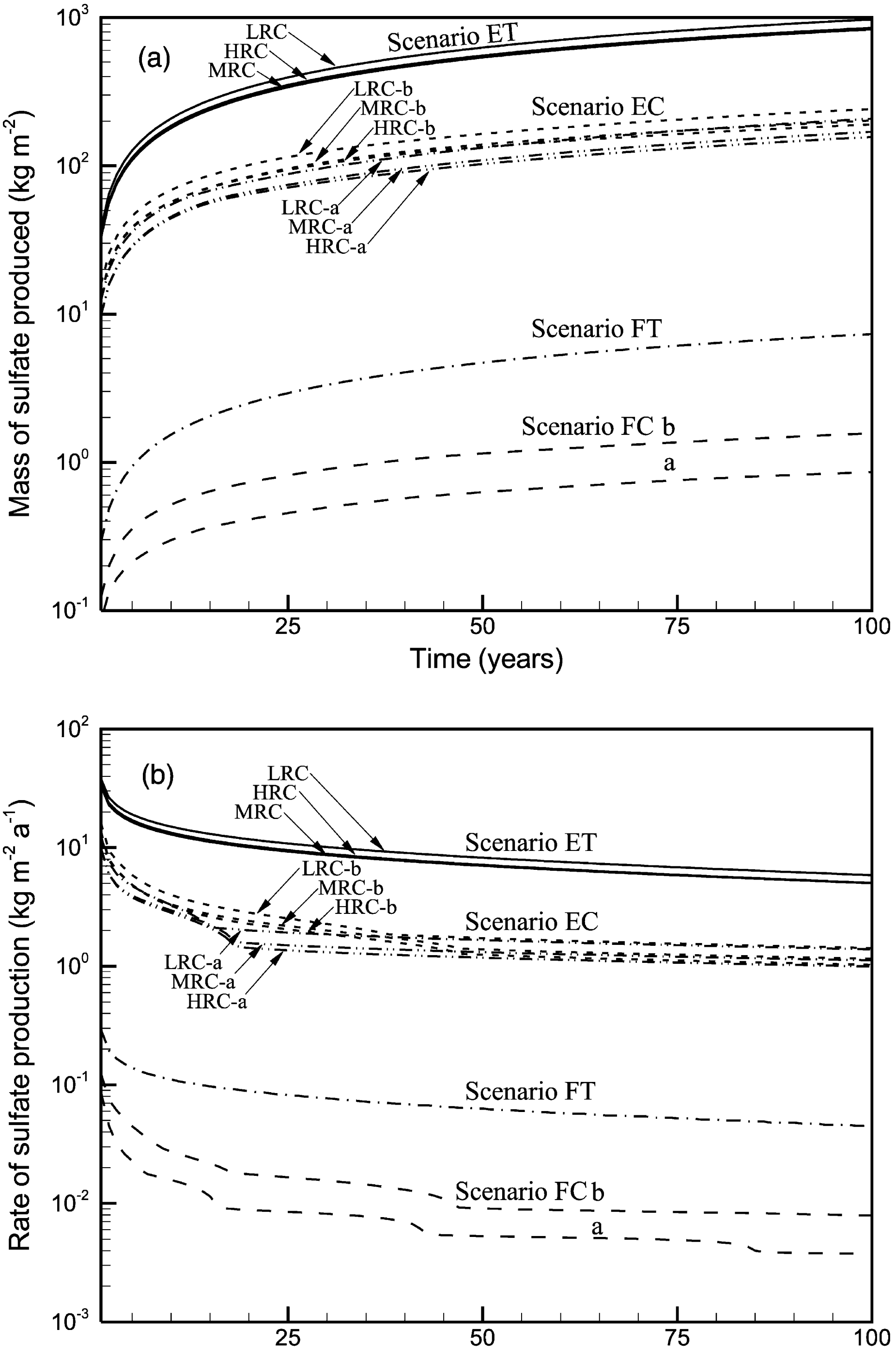

Fig. 2. Cumulative (a) and yearly total (b) mass of sulfate produced for a 100-year simulation time. (LRC = low rainfall condition; MRC = medium rainfall condition; HRC = high rainfall condition; CFT with a sulfide mineral fraction of $a = 0.005$ and $b = 0.015$).

(water cover), and a factor of 530 to 1129 (99.8%) less for Scenario FC. Similarly, the sulfate production rates are highest for Scenario ET (5.0 to 5.9 kg m^{-2} a^{-1} at 100 years considering all three rainfall scenarios), and lowest for Scenario FC (0.004 to 0.008 kg m^{-2} a^{-1} at 100 years for 0.5 and 1.5% pyrrhotite content in the CFT). In general, production rates are highest at the start of the simulation and decline exponentially with time.

The effect of low, medium, and high rainfall conditions on the generation of sulfate are represented by three curves for Scenario ET and two sets of three curves each for Scenario EC in Figs. 2(a) and (b). Generally, the rates of sulfate production for each scenario are similar, independent of the rainfall condition, indicating that the variations in precipitation do not affect the sulfide oxidation rate as much as the cover type. For Scenario EC, lesser rates of sulfate production were predicted for higher precipitation conditions. On average, relative to the medium rainfall condition, the low rainfall condition results in 21% greater production, and the high rainfall condition results in 7% lower production after 100 years. Similarly for Scenario ET, the low rainfall condition results in 17% greater production relative to the medium rainfall condition after 100 years. However, the high rainfall condition for Scenario ET results in a slightly greater rate of sulfate production (2% greater after 100 years) when compared to the medium rainfall condition. This is because the data set for the medium rainfall condition is characterized by water saturations which are slightly greater in the upper 25 cm of the vertical profile on average over a one-year time period, in comparison to the high rainfall condition, despite the lower yearly recharge rate. This is likely due to differences in annual recharge distributions for the two rainfall conditions. This anomaly disappears when the water saturations are averaged over a greater depth, as is confirmed by the model results for Scenario EC.

The stepwise reduction of the sulfate production rates for Scenario FC, as shown in Fig. 2(b), is due to the spatial discretization in the numerical model, which can only approximately describe the advance of the sharp oxidation front. Nevertheless, the trend of the evolution is described correctly.

Vertical profiles of the sulfate production rate at 20-year time intervals for Scenarios ET and EC are plotted in Fig. 3. For Scenario ET, the bulk of sulfide mineral oxidation takes place within the top 1.25 m of the vertical profile, while the bulk sulfide mineral oxidation for Scenario EC takes place between 1.0 and 1.25 m depth. This depth range for Scenario EC corresponds to the top 0.25 m of tailings underlying the CFT cover. The differences between these scenarios can be explained by the low sulfide content of the CFT cover. For Scenario EC, sulfide minerals are depleted within the CFT cover (0.5% FeS), or are nearly depleted (1.5% FeS) after a simulation time of 20 years, allowing the oxidation front to migrate further down. The deeper location of the oxidation front and higher water saturations at this depth (see Fig. 4) lead to the significant decrease in sulfide oxidation rates in comparison to Scenario ET. These plots confirm that variances in precipitation do not significantly affect the spatial distribution in production rates along the vertical profile.

Fig. 5 presents vertical profiles of sulfate production for Scenarios FT and FC. Most of the reaction products of Scenario FT are produced within only the top 2 cm of the fully saturated tailings, and all the products of Scenario FC are produced within the top 5–6 cm of the fully saturated CFT cover. Under fully saturated conditions and assuming the advective influx of oxygen is negligible, the primary factor limiting sulfide oxidation to the very top of the profile is the low solubility and low diffusion coefficient of oxygen in water. Sulfide oxidation occurs at a greater depth for Scenario FC, albeit at a much lower rate, due to the low sulfide mineral content in the CFT which is rapidly consumed allowing oxygen to be transported further downwards by diffusion relative to Scenario FT.

For the fully-saturated scenarios it was assumed that recharge into the tailings material, and thus the advective influx of oxygen into the tailings impoundment, are negligible. For greater recharge rates, advective oxygen transport may become more significant than oxygen transport by diffusion, leading to higher sulfide mineral oxidation rates. To test the validity of this assumption, the maximum advective flux of dissolved oxygen that could be realized under unit gradient conditions and oxygen-saturated pond water was calculated. The calculation is based on the saturated hydraulic conductivity of 3.3×10^{-7} m s^{-1} (0.029 m day^{-1}) for the tailings (from Table 1), and the solubility of

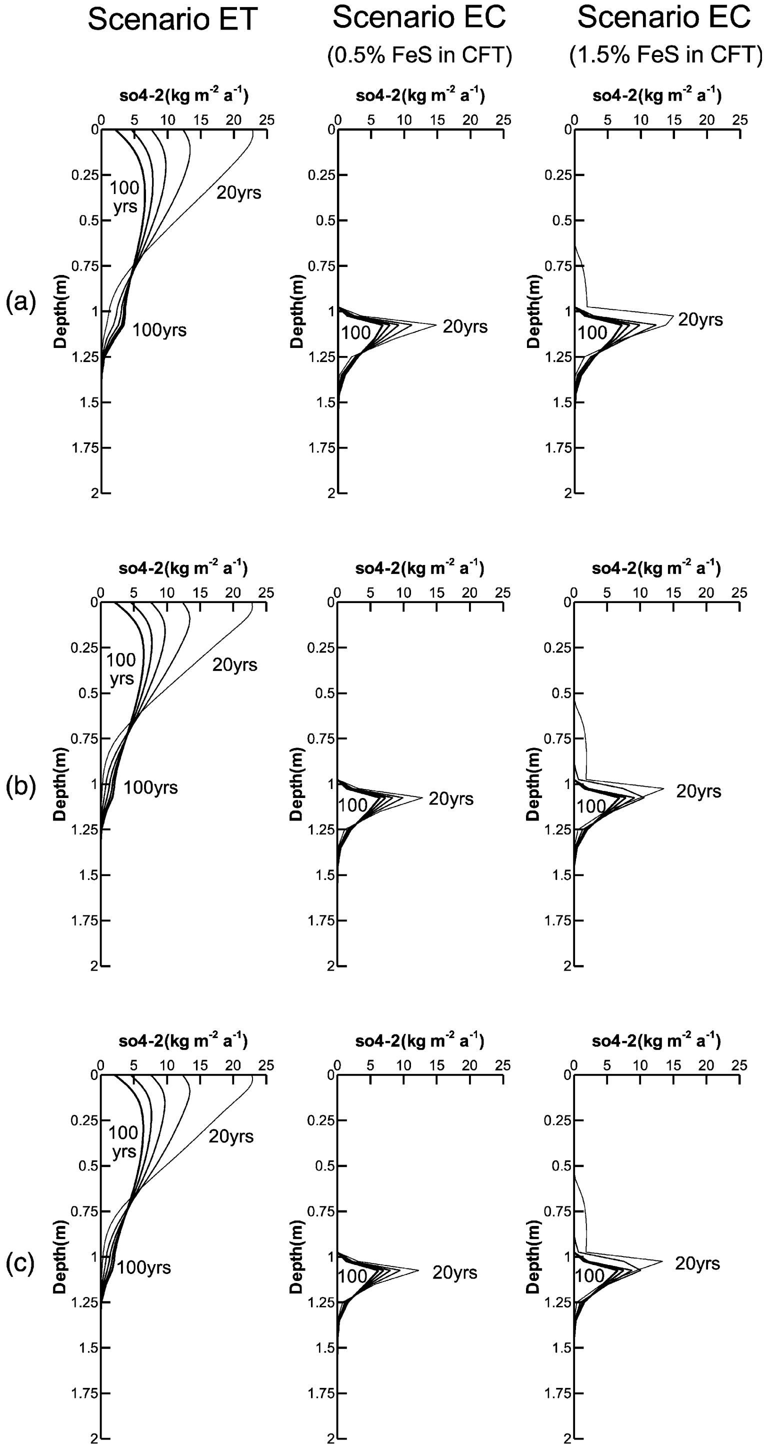

Fig. 3. Sulfate and iron production rates (kg m^{-2} a^{-1}) with depth at 20-year intervals for Scenarios ET and EC. (a) Low rainfall condition. (b) Medium rainfall condition. (c) High rainfall condition.

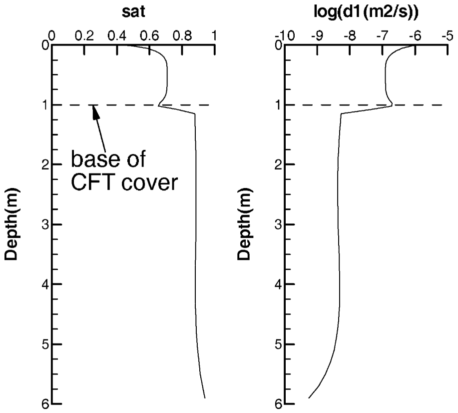

Fig. 4. Water saturation and oxygen diffusion coefficient with depth on day one of every year for Scenario EC using the low rain fall condition.

oxygen in water at 25 °C ($8.6\ \mathrm{mg\ l^{-1}}$). The maximum advective flux was determined to be $0.09\ \mathrm{kg\ m^{-2}\ a^{-1}}$, which is equivalent to the initial calculated diffusive flux for Scenario FT ($0.09\ \mathrm{kg\ m^{-2}\ a^{-1}}$), but is significantly larger than the minimum diffusive flux of $0.001\ \mathrm{kg\ m^{-2}\ a^{-1}}$ after 100 years for Scenario FC. This implies that the calculations for the water-FT may slightly underestimate actual sulfide oxidation rates. However, the unit-gradient assumption is highly conservative and water flow will likely be more restricted in reality. The presence of an organic rich layer on the tailings surface was not considered in the simulations. Such a layer may lead to additional oxygen consumption and even smaller sulfate production rates than predicted due to the consumption of oxygen before its ingress into the tailings material.

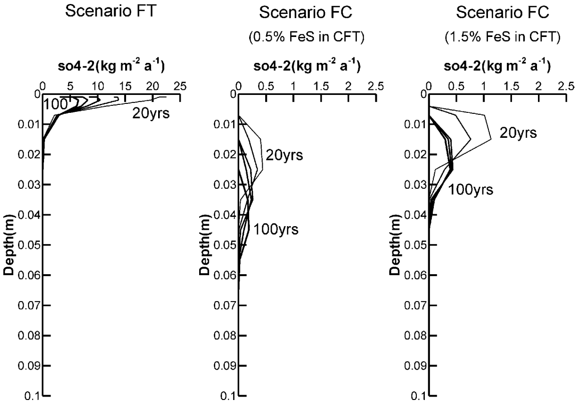

Fig. 5. Sulfate and iron production rates ($\mathrm{kg\ m^{-2}\ a^{-1}}$) with depth at 20-year intervals for Scenarios FT and FC.

Vertical profiles of normalized oxygen concentrations and normalized fractions of sulfide mineral consumed at 20-year intervals are presented in Figs. 6 and 7 for all four scenarios. The profiles of normalized sulfide mineral fractions consumed indicate the reaction front position. For the scenarios subject to directly incident rainfall (Scenarios ET and EC), the results in Fig. 6 indicate that, for the most part, variances in precipitation do not greatly affect the spatial distribution in oxygen and sulfide minerals reacted (see Fig. 6). Therefore, only the simulations using the low rainfall condition are presented in Fig. 7, which compares the results for the 'open ground' and 'flooded' covers.

The profiles for Scenario ET (Fig. 7) indicate that oxygen is transported to a depth of 1.25 m, corresponding to the maximum depth to which sulfide mineral is reacted. The reaction front position for Scenario ET extends from the top of the ET indicating that the sulfide minerals along the profile have not been fully oxidized. However, the sulfide mineral particles at the top of the profile are almost fully consumed after 100 years.

The profiles for Scenario EC (Fig. 7) indicate that oxygen is transported to a depth of approximately 1.35 m. The oxygen concentration declines slowly through the CFT cover and rapidly through the top of the tailings. The depth to which oxygen is transported corresponds to the maximum depth at which sulfide mineral is reacted. At 100 years, the reaction front starts at or near the base of the CFT cover, indicating that the sulfide mineral content in the CFT material has been fully or mostly consumed. The model results indicate that the reaction front reaches the base of the cover at 1 m depth within the first year (not shown). Depending on the rainfall condition, it can take 14–18 years for the cover to become completely oxidized if the CFT material has a 0.5% sulfide mineral content, and it can take 37 to 46 years if the CFT material has a 1.5% sulfide mineral content. When the cover becomes fully oxidized, oxygen is transported through the CFT cover and is consumed within the top 0.35 m of tailings.

Oxygen is transported to a depth of 2.0 and 5.5 cm for Scenario FT and FC, respectively, these depths corresponding to the maximum depth at

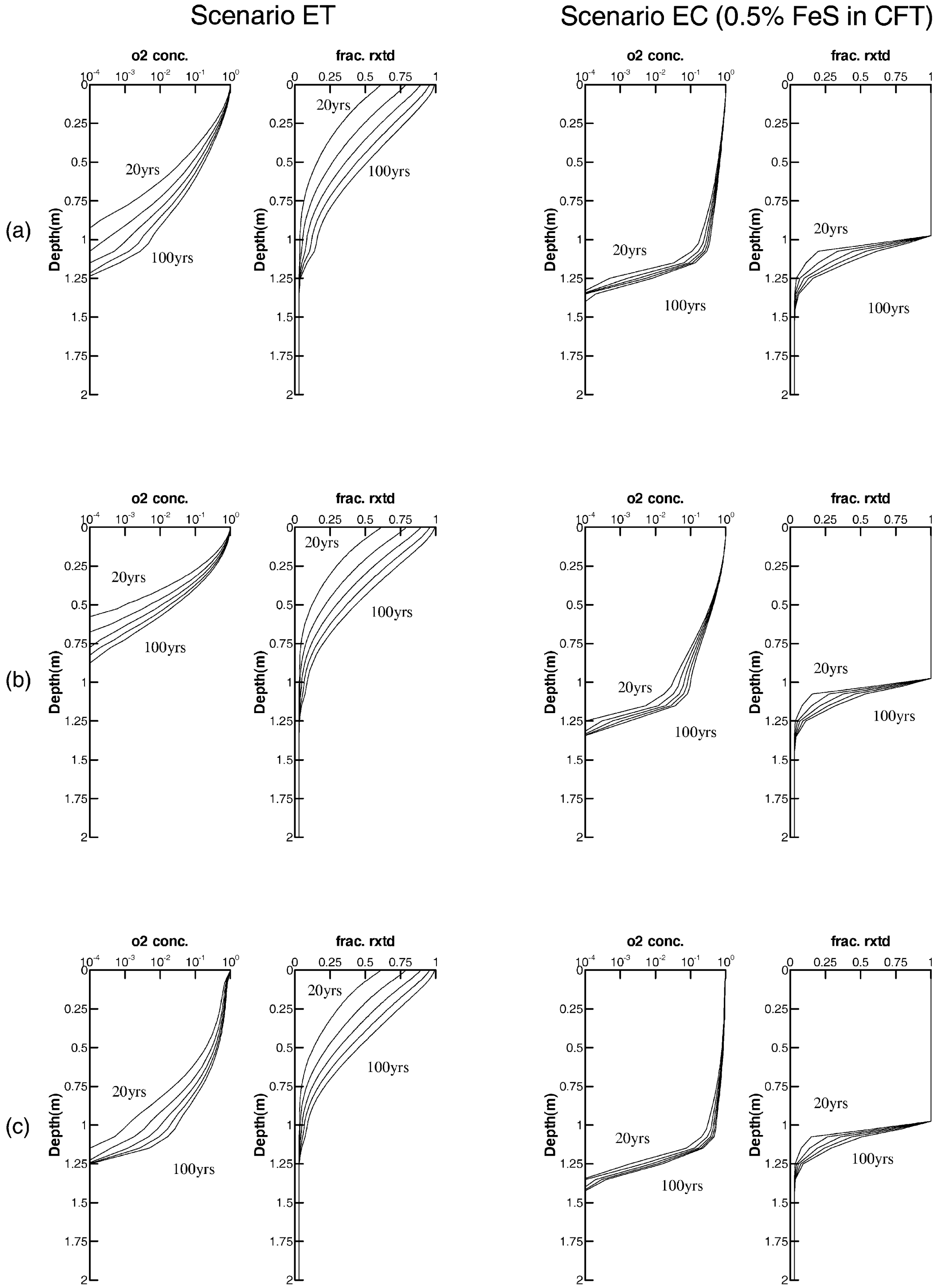

Fig. 6. Profiles of normalized oxygen concentration and fraction of sulfide mineral consumed at 20-year time intervals for Scenarios ET and EC. (a) Low rainfall condition. (b) Medium rainfall condition. (c) High rainfall condition.

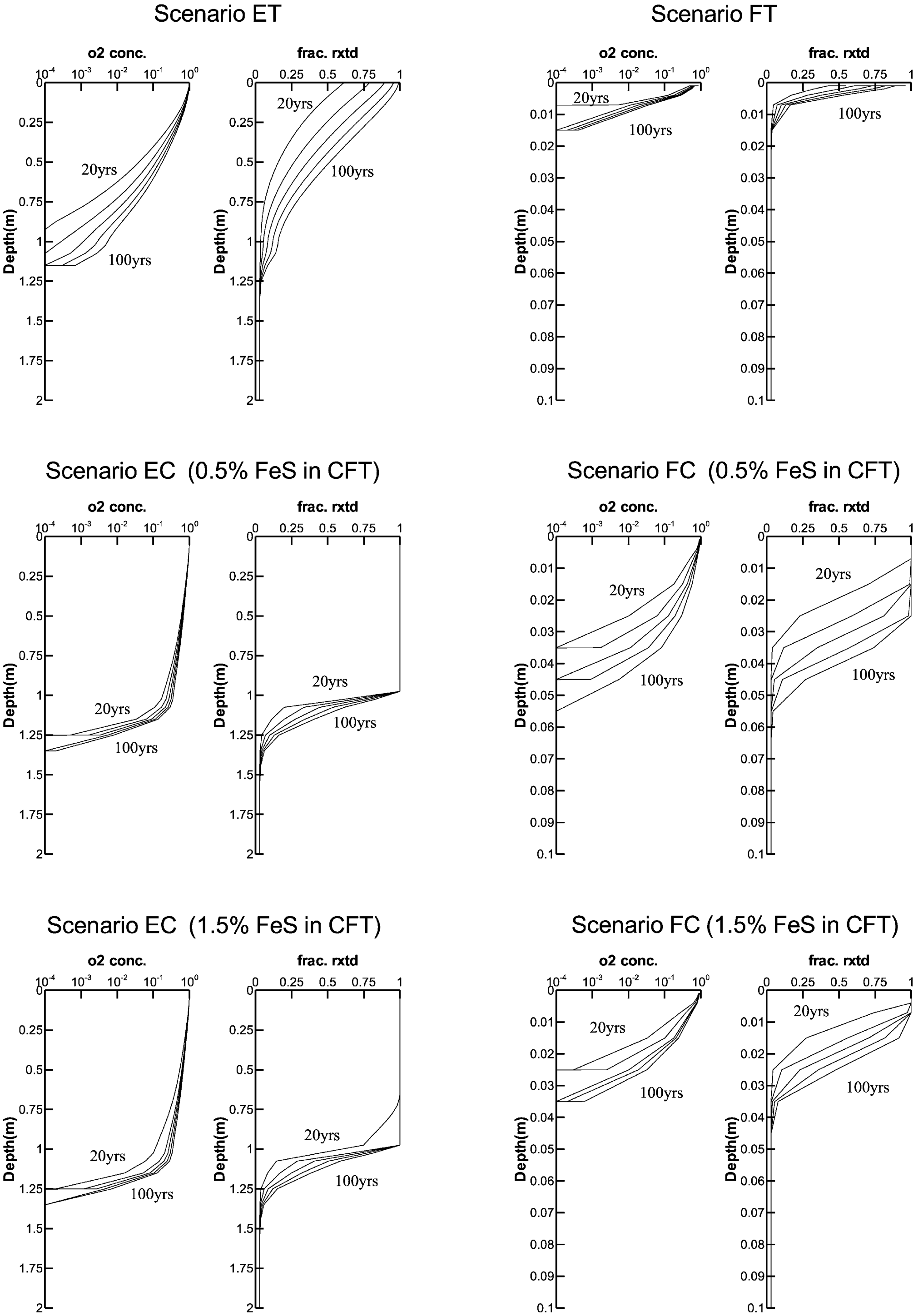

Fig. 7. Profiles of normalized oxygen concentration and fraction of sulfide mineral consumed at 20-year time intervals. Plot for Scenario ET and EC are based on the low rainfall condition.

which sulfide mineral is reacted (Fig. 7). The reaction front for Scenario FT extends from the top of the tailings indicating that the sulfide minerals at the surface of the saturated tailings have not yet oxidized fully, however, at this location they are almost fully oxidized at 100 years. In the case of Scenario FC, the reaction front position moves progressively downward through the saturated CFT as the upper zone of the CFT cover becomes fully oxidized. It takes approximately 80 years for the top 3 cm of the cover to become completely oxidized if the CFT material has a 0.5% sulfide mineral content, and it takes over 100 years if the CFT material has a 1.5% sulfide mineral content. The reaction front for Scenario FC does not reach the tailings underlying the CFT cover. Complete oxidation of the top part of the CFT cover allows oxygen to travel slightly further through the saturated material relative to Scenario FT.

For the simulations, which involve dynamically changing moisture contents over time (Scenarios ET and EC), the relative error in total oxygen mass in relation to the mass of oxygen consumed due to sulfide oxidation has been computed. The relative error is due to the neglect of oxygen mass conservation when moisture contents are updated, and is computed both yearly and cumulative from the start time. The maximum computed yearly and cumulative relative errors are in the range of 1.5–3.2 and 0.02–0.9%, respectively, and occur at the end time of 100 years. Generally, the error in total oxygen mass increases with time due to the decrease in oxygen consumption with time. These low error values suggest that the process of air displacement can be neglected and that PYROX can approximate well the effect of dynamically changing moisture contents on the rate of sulfide oxidation.

5. Discussion and conclusions

The objective of tailings covers is to minimize the sulfide oxidation rate and to reduce the generation of AMD, primarily in this instance, by limiting the ingress of atmospheric oxygen into the tailing. Limiting the water flux through the tailings may enhance the performance of such a cover. The work described here has specifically evaluated the performance of desulfurized tailings as a cover material, by comparing the potential effectiveness of various types of cover configurations in reducing the rate of sulfide oxidation in the underlying tailings. The cover scenarios considered here utilize either a cover of CFT waste, a water cover, or a combination of these cover types. In addition, the effect of climatic variability on the rate of sulfide mineral oxidation in tailings with an open ground surface was evaluated.

The modelling results over a simulation time frame of 100 years indicate substantial differences in the effectiveness of the three cover types investigated. The sulfide oxidation zone extends to depths greater than 1 m after 100 years for the scenarios subject to directly incident rainfall (Scenarios ET and EC). In contrast, the depth of penetration of oxygen does not exceed 0.06 m over 100 years for the scenarios that involve a permanent water cover (Scenarios FT and FC). For the case of a CFT cover overlain by a permanent water cover (Scenario FC), the underlying tailings remain unoxidized after closure, even after a simulation time of 100 years.

A CFT cover overlain by a permanent water cover (Scenario FC) is the most effective method for minimizing the sulfide oxidation rate (oxidation reduced by 99.8%) compared to ET. Somewhat less effective is a water cover alone (Scenario FT, oxidation reduced by 99.1%), followed by a CFT cover alone (Scenario EC, oxidation reduced by 75–82%), regardless of the rainfall condition and considering CFT material with a sulfide mineral content ranging from 0.5 to 1.5%. For the scenarios subject to directly incident rainfall, (Scenario ET and EC), even though the low annual rainfall condition resulted in the highest sulfide oxidation rate relative to the other rainfall conditions, variability in precipitation does not have as significant an effect on the sulfide oxidation rate as does the cover type. There was less than 10% variation in the performance of the CFT cover (Scenario EC), with respect to the extent of sulfide oxidation, over the likely range of climatic conditions at the mine site.

The comparison of the different cover types has some practical implications for the construction of mine tailings covers, which should be generally applicable based on the site conditions. A simple water cover (Scenario FT) appears to be the best solution, if a water-covered tailings impoundment is technically and economically feasible. Whilst the use of a combined cover (Scenario FC) only slightly improves (in absolute terms) the performance, it may also be useful in maintaining the water cover (i.e. reduce infiltration into the tailings) if the material layer has a lower permeability relative to the underlying tailings. The use of a CFT cover in the case of FT would thus be primarily for hydrologic reasons (minimization of water influx), not for geochemical reasons (minimization of sulfide oxidation rates).

A secondary benefit of the CFT layer would be to shield the tailings from re-suspension by wave action. This can be a very important consideration in areas, such as at the Renison mine, where strong winds can produce substantial turbulence through several metres of water. Re-entrainment of tailings would lead to direct exposure to oxygen in the water column and hence accelerate the rate of oxidation of the tailings particles.

If the maintenance of a water-covered tailings impoundment is not feasible as a result of engineering/geotechnical constraints on stability of the dam wall, a fine-grained, nearly saturated, cover comprised of CFT can still substantially limit sulfide oxidation in comparison to the uncovered scenario. For a cover of this type both the permeability and the sulfide mineral content of the material should be as low as possible.

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