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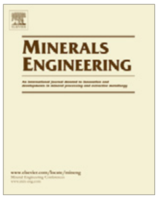

Coupling oxidation to transient drying during multilayer deposition of thickened gold tailings

Rachel Bryan [a], Paul Simms [b,*], Rens Verburg [c]

[a] KGS consultants, 560 Squier Place, Thunder Bay, Ontario, Canada P7B 6M2
[b] Department of Civil and Environmental Engineering, Carleton University, Ottawa, Ontario, Canada K1S N6A
[c] Golder Associates, 18300 NE Union Hill Road, Suite 200, Redmond, Washington 98052, USA

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ABSTRACT

Dewatering tailings prior to deposition is an attractive alternative to conventional practice, as it minimizes the use of embankments and therefore reduces the risk of catastrophic failure associated with dammed slurry impoundments. One of the potential detriments is the absence of a water cover and the consequent increase in oxygen ingress and generation of acid drainage. This paper investigates the amount of oxidation that occurs in shallow layers of desiccating gold tailings during deposition. Oxidation is modelled by coupling transient predictions of unsaturated flow to predictions of oxygen diffusive transport and consumption. A first-order rate law is adopted to model oxidation. Coupling is accomplished through the dependency of oxygen diffusivity on degree of saturation. Experiments and numerical predictions agree within the order of the measurement error, despite the relative simplicity of the oxygen consumption model. Though these tailings crack, the cracks do not influence the drying behaviour, nor do they significantly influence the horizontal distribution of oxidation. The modelling method is applied to the case of a fresh layer of tailings deposited over a deep stack. This analysis shows that some oxidation will occur during deposition of a fresh layer of thickened tailings, and the rate of drying and oxidation is strongly influenced by the underlying previously-deposited tailings. It is also illustrated how drying time to achieve a given density, and consequently oxidation, can be limited by hydraulically decoupling a fresh layer from the underlying stack using a capillary break.

1. Introduction

Surface disposal of thickened or dewatered high density tailings is an increasingly utilized alternative to conventional disposal of slurried tailings in dammed impoundments. Tailings can be dewatered to the point where they are non-segregating and exhibit a yield stress upon deposition, and therefore can form gently sloped self-supporting stacks that do not necessarily require containment by dams or dykes (Robinsky, 1999). These characteristics make thickened tailings an attractive alternative to conventional slurry deposition, due to the significant risk and consequences of dam failure (ICOLD, 2001) associated with the latter. Other advantages include ease of closure due to earlier trafficability afforded by the higher density at deposition, and the economic and environmental benefits of recycling the recovered water. Potential disadvantages include greater risk of acid generation due to the absence of a water cover, the potential for remobilization of the stack during seismic events (Li et al., 2009), and challenges in managing deposition geometry (Henriquez and Simms, 2009; Shuttleworth et al., 2005).

Many practitioners believe that the homogeneous nature of thickened tailings makes them less likely to produce acid drainage than conventional tailings (Martin et al., 2006; Meggyes and Debreczeni, 2006). This has some justification, as under steady-state conditions a relatively high capillary fringe would be maintained. In addition, infiltration, and therefore the quantity of potentially acidic leachate, is reduced by the natural slope of the surface, the physical barrier created by precipitated salts, and the low hydraulic conductivity (Verburg, 2001; Newman, 2003).

However, oxidation has been reported at thickened tailings sites. At the Bulyanhulu mine in Tanzania, visible oxidation is reported in disturbed areas of the tailings storage facility (Shuttleworth et al., 2005). The tailings facility at Kidd Creek Mine near Timmins, ON, has also undergone significant oxidation at the surface (Al and Blowes, 1995). Therefore, it is clear that thickened tailings stacks are susceptible to some degree of oxidation. The goal of this paper is to show how the degree of oxidation *during deposition* can be evaluated by modelling transient unsaturated flow during drying of the tailings, and coupling the predicted degree of saturation profiles to oxygen diffusion and consumption rates. This

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* Corresponding author.
E-mail address: paul_simms@carleton.ca (P. Simms).

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modelling approach incorporates several simplifying assumptions with respect to sulfide oxidation, such as employing a first-order rate law, ignoring any potential feedback mechanism from other geochemical processes, and ignoring the influence of cracks on oxygen transport. The modelling method is tested on three laboratory column tests simulating the sequential deposition of two layers of thickened tailings. The method is then employed to analyze a hypothetical deposition scheme to illustrate how deposition planning can be optimized to minimize oxidation.

2. Background

2.1. Sulfide oxidation

The chemistry of sulfur oxidation in tailings impoundments is described extensively in the literature (e.g., Blowes et al., 2003). Many ore bodies are found in sulfidic deposits, which contain high percentages of pyrite, pyrrhotite, and other sulfide minerals. Underground, these minerals are stable in the anaerobic, reducing environment. When the waste materials are discharged to a surface storage facility, weathering of the sulfide minerals can lead to acidic, heavy-metal-laden runoff, commonly known as acid rock drainage (ARD). Since pyrite is a ubiquitous sulfide and the primary sulfide mineral in the tailings used in these experiments, its behaviour will be the focus of further discussion. However, this should not disinterest the reader in the behaviour of other sulfide minerals, such as arsenopyrite (e.g., Komnitsas et al., 1995).

Due to the relatively short time a fresh layer is left to dry before placement of the next layer (weeks not months), we focus on circumneutral pyrite oxidation. The sum of the various reactions, including initial oxidation of pyrite by oxygen, oxidation of ferrous iron to ferric iron, and its subsequent precipitation, can be represented by (Garrels and Thompson, 1960):

$$FeS_2 + \frac{15}{4}O_2 + \frac{3}{2}H_2O \rightarrow Fe(OH)_3 + 2SO_4^{2-} + 4H^+ \tag{1}$$

At circumneutral pH, oxidation is driven by oxygen availability; ferric iron does not contribute as an oxidant due to its insolubility at pH > 4. Also, under circumneutral conditions, the role of bacteria that can accelerate sulfide oxidation, such as *Acidithiobacillus ferrooxidans*, is minimal.

The pH of the tailings pore water will be maintained at a relatively neutral value for a certain time, depending on the quantity of neutralizing minerals. The most effective neutralizing minerals are the carbonates, which buffer in the circumneutral pH range depending on the mineral (e.g., calcite [$CaCO_3$] buffers at pH 6.5–7; siderite [$FeCO_3$] buffers at pH 5.5–6). A mineral such as siderite may tend to form in the very early stages of oxidation, when calcite is dissolving and Fe^{2+} ions are available from pyrite oxidation. Once calcite is consumed and the pH continues to fall, siderite would then start to dissolve around pH 6, providing some secondary buffering capacity. In the lower pH range, hydroxides such as $Al(OH)_3$ (pH 4.0–4.3) and $Fe(OH)_3$ (pH 2.5–3.5) and various aluminosilicate minerals offer some buffering capacity (White and Brantley, 1995; Jurjovec et al., 2002).

2.1.1. Rate of pyrite oxidation

The principal factors affecting the rate of pyrite oxidation under circumneutral pH conditions are the specific surface area and reactivity of the grains, oxygen concentration, and temperature. Nicholson et al. (1988) showed that the rate of pyrite oxidation increased linearly with decreasing grain size and consequently with increasing specific surface area.

In terms of oxygen concentration, the reaction kinetics for pyrite oxidation can be described generally by the following equation:

$$r = \frac{-d[FeS_2]}{dt} = -\frac{4}{15}\frac{d[O_2]}{dt} = k[O_2]^a \tag{2}$$

where r is the reaction rate (mol/day), k is the reaction rate constant (day^{-1}), $[O_2]$ is the dissolved oxygen concentration (mol/L), and a is the reaction order with respect to oxygen. The value of a can be approximated by zero-order at high oxygen concentrations, and first-order at low, diffusion-controlled oxygen concentrations. Experimental conditions with intermediate oxygen concentrations take on fractional values for a (Nicholson et al., 1988).

Williamson and Rimstidt (1994) compiled the rate data available in the literature and determined an abiotic rate law for pyrite applicable over four orders of magnitude in DO concentration and a pH range from 2 to 10. The rate law is given as:

$$r_{py} = 10^{-8.19(\pm 0.10)}\frac{m_{DO}^{0.50(\pm 0.04)}}{m_{H^+}^{0.11(\pm 0.01)}} \tag{3}$$

where r_{py} is the rate of pyrite destruction (mol/m^2/s) and m is the species concentration (mol/kg). Their analysis confirmed a reaction order of $a = 0.5$ with respect to DO. Although the rate is pH-dependent, the small reaction order with respect to H^+ ($a = 0.11$) indicates that pH has a minor effect. The H^+ concentration must change by nine orders of magnitude to effect a single order of magnitude change in the reaction rate.

Jerz and Rimstidt (2004) derived a similar rate equation describing the oxidation of pyrite by oxygen. The Jerz and Rimstidt equation incorporates a time-dependent factor, which accounts for the slowing of the reaction rate over time due to the build-up of reaction products on the pyrite surface:

$$r_{O_2} = \frac{dn}{dt} = \frac{10^{-6.6}P_{O_2}^{0.5}}{t^{0.5}} \tag{4}$$

where r_{O_2} is the rate of oxygen consumption (mol/m^2/s), P_{O_2} is the partial pressure of oxygen (atm), and t is the time (s).

2.2. Mass transfer through the pore space

Both advection and diffusion in the gas and liquid phase can contribute to oxygen movement in mining waste. Advective transport is a concern when dealing with waste rock dumps (e.g., Molson et al., 2005) but it is generally agreed that advection does not contribute significantly to gas transport in tailings piles because of the fine grain size and low permeability (Jaynes et al., 1984; Elberling and Nicholson, 1996). This leaves diffusion as the primary mechanism for oxygen transport from the tailings surface to the depth where oxidation takes place.

The diffusion coefficient of oxygen in water is 1.8×10^{-9} m^2/s, whereas the diffusion coefficient of oxygen in air is 2.0×10^{-5} m^2/s, 10,000 times the rate of diffusion in water (Elberling, 1996). Therefore, the saturation profile in tailings strongly controls the rate of oxygen diffusion and consequently the extent of sulfide oxidation. Aachib et al. (2004) proposed the following semi-empirical equation for calculating the effective diffusion coefficient of oxygen in unsaturated porous media:

$$D_{eff} = \frac{1}{n^2}\left(D_a^0\theta_a^p + HD_w^0\theta_w^p\right) \tag{5}$$

where D_{eff} is the effective diffusion coefficient of oxygen (m^2/s), n is the porosity, D_a^0 is the free diffusion coefficient of oxygen in air at room temperature, θ_a is the volumetric air content of the soil, p is a fitting parameter which accounts for tortuosity (for simplified models, Aachib et al. (2004) concluded that a fixed value of $p = 3.4$ can be adopted without much loss of accuracy), H is the dimensionless Henry's equilibrium constant ($H = 0.03$ for oxygen at 20 °C), D_w^0 is the free diffusion coefficient of oxygen in water at