

INVESTIGATIONS INTO THE DEPTH AND RATE OF WEATHERING ON WITWATERSRAND GOLD TAILINGS DAM SURFACES AS KEY INFORMATION FOR LONG-TERM ARD RISK ASSESSMENTS¹

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Abstract. The generation of Acid Mine Drainage (AMD) from tailings dams of the Witwatersrand goldfields in South Africa, is arguably one of the main strategic environmental issues facing the gold mining industry and the South African government. Fine grained, sulfidic, tailings impoundments generate AMD through the diffusive ingress of oxygen into unsaturated pore spaces. Golder Associates conducted research on behalf of the Water Research Commission of South Africa to determine what the likely rate and depth of oxidation is within typical tailings impoundments of the Witwatersrand goldfields. Investigations consisted of a combination of literature research, field work and sulfide-oxidation modeling. The results of these investigations suggest that the bulk of a typical Witwatersrand tailings facility will acidify after hundreds to thousands of years of sulfide oxidation. Acidic drainage, however, may be generated within the wall and toe paddock sections during the operational phase or shortly after decommissioning and closure. The production of acidic drainage, therefore, can be mitigated during the operational phase.

Additional Key Words: Oxygen diffusion, tailings seepage, acidification, neutralization.

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Introduction and Scope of Investigation

The oxidation of sulfide minerals, commonly associated with Witwatersrand gold mine residue deposits and the consequent release of acidic drainage, is arguably one of the main strategic environmental issues facing the gold mining industry and the South African government.

During the operational phase, seepage from the fine residue deposits (FRDs) is typically saline and neutral (even alkaline). During the decommissioning and closure phases of the FRDs, Acid Mine Drainage (AMD) can take place.

International research shows that oxygen ingress into FRDs is due to the O₂-concentration gradient that exists between the depth of active oxidation and the surface of the tailings impoundment. The O₂-concentrations gradient is controlled by the rate that sulfide oxidation consumes gaseous O₂. Because O₂ is being consumed at depth, the concentrations of gaseous O₂ decrease with depth resulting in an oxidised outer cap, Mayer et al., 2000. Enough acidity could be produced in this oxidised cap to consume all Neutralisation Potential (NP) in the rest of the FRDs, resulting in AMD from the waste facility. However, Mayer et al., 2000 also found that this oxidation rate decreases over time to significantly lower in-situ rates.

Golder Associates Africa carried out research, under contract for the Water Research Commission of South Africa, to determine the depth and rate of weathering on typical Witwatersrand gold FRD surfaces as key information for long-term AMD risk assessment.

Approach and Methodology

The approach followed in the research was to compare the total mass of acidity that will likely be generated in the oxidised outer surface of typical Witwatersrand FRD over time, to the total Neutralisation Potential of the rest of the FRD. The time to total Neutralisation Potential consumption was then used to evaluate the potential for long-term AMD risks of FRDs as well as to evaluate mitigation strategies.

The research methodology included a literature review of typical parameters such as dimensions, construction methods, mineralogy, climate, material properties and geochemistry of Witwatersrand FRDs. The literature study also included a study of the processes and parameters that drive and effect AMD processes in fine mine residue deposits. Analytical and probabilistic modelling, including the use of specialist models such as PYROX, PHREEQC and an Engineering spreadsheet model developed for the project were used to model ranges of likely consumption of available Neutralisation Potential (i.e. onset of acidic drainage conditions) for typical Witwatersrand FRDS .

Characteristics of Witwatersrand Fine Residue Deposits

Available data indicate that Witwatersrand fine residue materials do not vary greatly in terms of their physico-chemical characteristics. Witwatersrand FRDs are typically very large deposits. They are greater than 1km in length and breadth, and up to 60m high. Only two depositional

methods are recorded in the literature, i.e. paddock systems and cycloned tailings deposits Anglo American, 1995. Cyclone deposition occurs in a minority of cases in the Witwatersrand.

Witwatersrand gold tailings are constrained within a relatively narrow compositional range and are described as consisting of between 70% and 90% quartz, 10% to 30% phyllosilicates (mainly sericite) and 1% to 2% consisting of minerals such as uraninite, monazite, chromite and rutile Rösner et al., 2001; Förstner and Wittmann 1976. Rösner et al., 2001 found that besides quartz, the minerals found in tailings included muscovite, chlorite and pyrophyllite.

Minerals that contribute to the Acid Potential (AP) are present in relatively low concentrations (<1%). Acid Potential is, however, generally higher than the Neutralization Potential (NP). In the Witwatersrand FRDs, Acid Potential is predominantly ascribed to pyrite, as it is the most dominant sulfide mineral, with other sulfides occurring less frequently. Pyrite may occur in both euhedral (less reactive) or framboidal (more reactive) forms in the Witwatersrand reefs, due to the presence of biogenic carbonaceous material in the Witwatersrand reefs, along with the presence of hydrothermally precipitated pyrite grains.

Neutralization Potential is ascribed primarily to carbonates of Ca and Mg. Silicates can become important neutralizing minerals in a tailings deposit after all soluble carbonate phases are depleted. Because the dissolution of silicate minerals is kinetically limited, the NP of the silicate minerals depends on their respective dissolution rates Usher et al., 2001.

Acid Base Accounting (ABA) analyses conducted by PHD, 2002, on various Witwatersrand gold tailings samples showed AP: NP ratios from 1:0.5 to 1:2.78. Measured S concentrations varied between 0.17% and 0.45 % S, thus placing the tailings within the uncertain range, with its potential to generate AMD Sobek et al., 1978.

Some particle size fractionation may occur by hydraulic separation, thus resulting in differential permeabilities within these deposits. This effect is enhanced in cycloned deposits, where the wall sections are constructed with thickener underflow. In general, the grain size distribution is described as >75% of the material with particle sizes of <75µm Rösner et al., 2001; SRK, 1988 and a D₁₀ particle size of between about 0.02mm and 0.1mm Anglo American, 1995. Porosity in Witwatersrand FRDs is generally considered to vary between 31% and 34%, or expressed as void ratios, between 0.46 and 0.51 Wates, 2004 (pers. comm.), PHD, 2002.

The moisture content within an FRD or between different FRDs may be highly variable. Factors determining moisture content include climatic conditions and physical properties of the FRD on a macro and micro scale. Fine-grained layers, which may form capillary breaks, are important in FRDs, as they cause local variation in moisture contents and can significantly affect the flow of gases. Such layers were discussed by Nicholson et al., 1995 and were also observed in the field during this study. Measurements by PHD, 2002, recorded moisture contents between 20 and 40% (volume).

The hydraulic conductivity in Witwatersrand gold FRDs has been found to be highly anisotropic (the ratio of horizontal permeability vs. vertical permeability is significantly different), largely due to compaction of the sediment column in a water-saturated environment. Hydraulic conductivity in the horizontal plane is considerably higher than in the vertical plane. The anisotropy is caused mostly by the stratification within the sediment and shrinkage cracks. Anisotropy coefficients of between 5 and 10 are considered normal, but values up to 200 have

been reported Rösner et al., 2001. The saturated hydraulic conductivities of Witwatersrand tailings have been found in selected studies to range between 10^{-6} and 10^{-8} ms^{-1} Bezuidenhout and Vermaak, 2000.

Mass transport processes affecting FRD AMD potential

Water and gas transport are the main processes affecting AMD and pollution mobilization at Witwatersrand gold FRDs. From a mass transport perspective, the rate at which pore water moves through the tailings material directly influences the rate at which dissolved salts (from AMD processes) in the pore water are mobilized from the FRDs into the receiving environment. Various geochemical processes along the flow path influence the chemical composition of the pore water. Gas transport processes limits the rate of sulfide oxidation in the tailings material which directly influences the rate at which typical AMD processes take place within the tailings deposit and hence its influence on the pore water chemical composition.

Pore water flow in an FRD is determined by a number of factors, such as climatic variation, material properties, type of deposit and water management strategy, and determines the moisture content within the tailings body at any one point in time and space. This moisture content influences the oxidation rate by determining the fraction of air-filled pore space available for air to move through. Both saturated and unsaturated flow processes occur within an FRD. In the post closure phase, unsaturated flow processes become progressively more dominant as the piezometric surface subsides.

Gas transport is generally described as occurring either by advective or diffusive flow. In tailings materials it has been shown that advective flow, including barometric pumping (movement of air due to changes in atmospheric pressure), does not contribute significantly to O_2 transport, due to the fine-grained and hence relatively impermeable nature of tailings material James, 1997. Where preferential pathways exist in the near surface environment, a limited amount of advective flow may occur however this is not expected to significantly affect the overall O_2 transport rate within an FRD.

Diffusion in tailings is driven by the concentration gradient between the tailings surface and the matrix, caused by the oxidation of pyrite Yanful, 1991. Oxidation is usually confined to a few metres below the surface as a result of O_2 transport limitations due to diffusion, with O_2 concentrations becoming progressively lower with depth.

Previous studies have showed that the limiting parameters controlling pyrite oxidation through diffusion are the diffusion coefficient and the kinetic rate constant Nicholson et al., 1989; David & Nicholson, 1995; Troeh et al., 1995. As the diffusion depth increases, O_2 diffusion becomes a more dominant parameter than the inherent pyrite reaction rate in determining O_2 flux.

Investigations into diffusion controlled pyrite oxidation in FRDs have led to the conclusion that determinations that are made on bulk tailings tend to overestimate reaction rates Nicholson et al., 1989. This is caused by local variation in moisture saturation levels within the tailings profile, due to the presence of more saturated fine-grained layers and less saturated coarser grained layers.

Mineral reaction kinetics focus mostly on the oxidation of pyrite. The shrinking core model has commonly been used to calculate oxidation rates of pyrite grains over time. An oxidised layer, which forms through reaction of pyrite has been measured in the laboratory and calibrated empirically to have a secondary diffusion coefficient in the region of approximately $7.8 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ PTI Environmental Services, 1995.

Bacteria such as *Thiobacillus ferrooxidans* play an important part in determining oxidation rates of pyrite; however, in a diffusion controlled environment this has a more limited effect. It has been determined that under conditions of less than approximately 8% O_2 these bacteria do not contribute significantly to pyrite oxidation reactions Pugh et al., 1984; Perkins et al., 1995.

Modelling

A conceptual model was developed to represent the generation of acidity from pyrite oxidation within a typical Witwatersrand FRD. Figure 1 provides a schematic representation of the conceptual model. According to the model, the FRD will undergo oxidation through vertical ingress of oxygen by diffusion through the tailings profile. A large store of un-reacted material, with associated NP occurs below the oxidation zone and potentially provides a means to delay the onset of acidic seepage from the base of the FRD for an extended period.

Use was made of the dual diffusion PYROX model to develop envelopes for oxidation rates of typical Witwatersrand FRDs. Making use of the PYROX modelling results, a spreadsheet engineering type model was developed to model the likelihood of acidification from an FRD due to the time related consumption of neutralisation capacity.

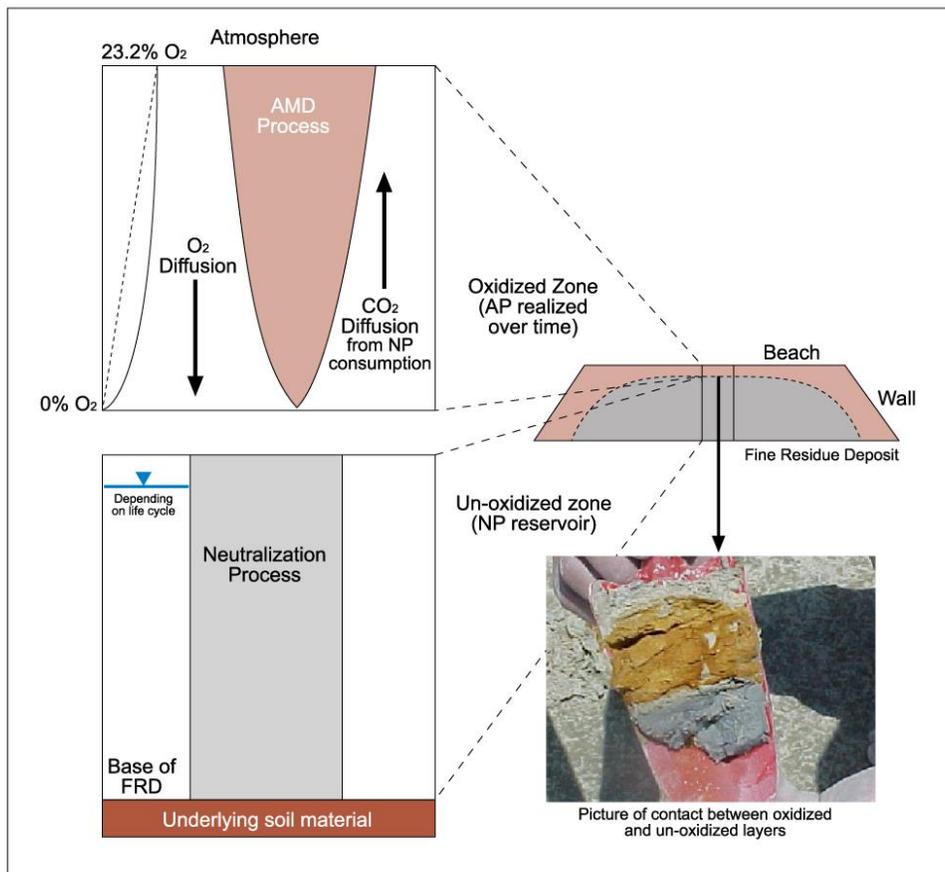


Figure 1: Schematic representation of the conceptual model

PYROX modelling

The PYROX model developed at the University of Waterloo is based on a double diffusion model developed by Davis and Ritchie, 1986 and has been used in international tailings investigations Wunderly et al., 1996. The model was used to evaluate pyrite oxidation rates within the known compositional range (physical, geochemical and climatic) of Witwatersrand tailings. The model was also used to perform sensitivity analyses for particle size, porosity, moisture and sulphur content on the control of pyrite oxidation rates over a 200 year period.

Typical to conservative values of sulphur content, particle size, saturation, porosity, temperature, secondary diffusion coefficient and the thickness of the tailings profile were used as a baseline for determining sensitivities of other parameters. Figure 2 provides time series graphs of modelled pyrite oxidation rates (expressed as $\text{kg SO}_4^{-2}/5$ year period) for various particle size distributions, porosity values, saturation and S contents.

The following inferences can be drawn from these graphs:

- Initial oxidation rates are lower for coarser material sizes (>5mm) compared to finer (<1mm) material and is expected to be related to the relative larger reactive surface area of the finer grained material. However, as oxidation coatings start developing on reactive surfaces, the diffusion of O_2 through the oxidation coatings become more rate controlling compared to the diffusion of O_2 through the pore space, resulting in long-term rates that follow a decay curve to oxidation levels that are similar for both fine and coarse grained material.
- Higher porosity values lead to higher initial oxidation rates. However, formation of secondary oxidation coatings on the reactive surface areas results in similar decay curves of pyrite oxidation, suggesting relative insensitivity of long-term oxidation rates to porosity.
- Higher moisture contents results in lower oxidation rates, due to restricted flow paths for gaseous O_2 diffusion. Again, long-term rates decrease to levels that are similar for all modelled moisture contents.
- Sulfide concentration (as Pyrite) was modeled over a range of between 0.1% and 4% of the bulk density of the tailings material. An upper bound of 6.6% pyrite was assumed for a Witwatersrand FRD, on the basis of maximum pyrite concentrations in the Au reef material mined. The lower bound relevant to AMD is considered to be 0.25% pyrite, as lower sulfide concentrations are not considered to pose a significant risk of acidification Lawrence & Day 1997. Lower S contents results in lower oxidation rates, however, because of the lower O_2 concentration gradient, this results in deeper oxidation fronts than high S tailings for variable sulfide concentrations. Oxidation rates decrease over time following a typical decay curve, however, greater variation in long-term rates for the different S contents are observed, relative to other variables this parameter was thus

shown to be the most sensitive parameter to consider in modeling long-term oxidation rates.

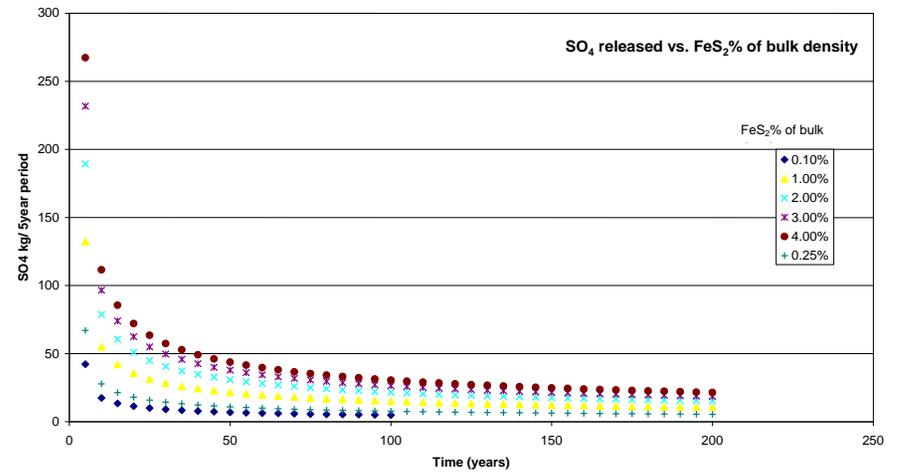
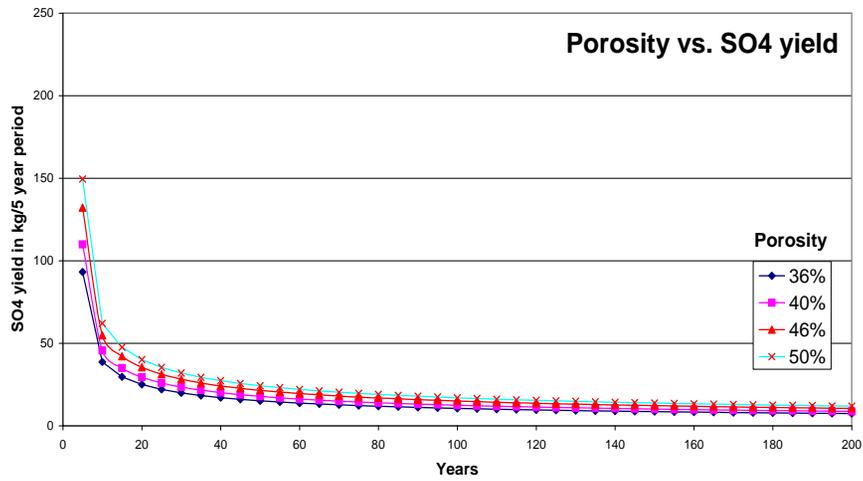
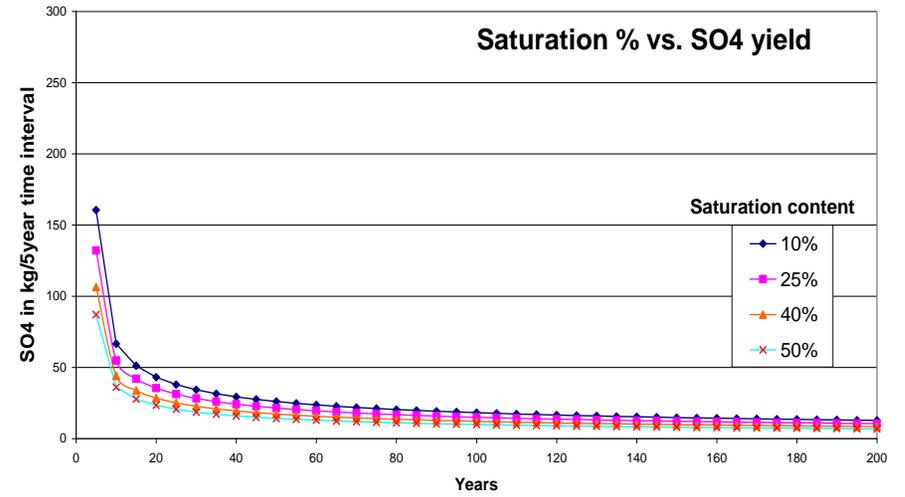
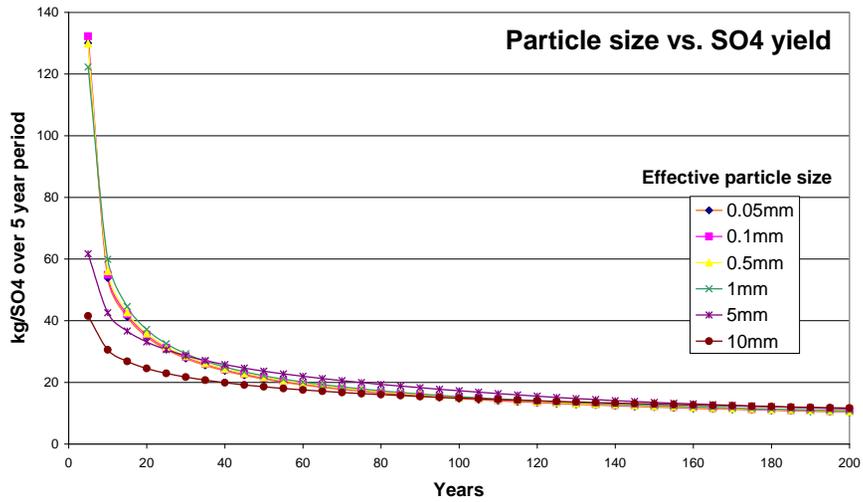


Figure 2: Results of PYROX sensitivity modeling of typical Witwatersrand tailings (incl. sensitivity analysis)sis

- It was found that within the ranges of these parameters within a Witwatersrand FRD, the absolute values of pyrite oxidation rates vary as expected. It was also, however, found that the rate of change of the oxidation rate over time fell within a relatively narrow range of fitted functions, especially when viewed over the long term. These findings were used in the construct of the engineering model as a function of static diffusion rates at initial compositions.

Probabilistic modelling using the TAP model

A probabilistic based spreadsheet model was developed to compare time related production of acidity of a typical Witwatersrand FRD versus the neutralisation of this acidity from the inherent base potential of the tailings material. This model is referred to as the Tailings Acidification Prediction (TAP) model and is aimed at developing an understanding of the likely time spans for the consumption of neutralisation capacity, and the change over to acidic drainage for a typical Witwatersrand FRD. The model consists of two components, i.e. an oxygen diffusion component to predict trends in pyrite oxidation rates and a neutralisation component to predict the rate of consumption of NP in the FRD.

The initial oxidation rate is calculated using the equation by Nicholson et al., 1989 [1]. The diffusion coefficient is calculated using the equation of Millington and Shearer (1971) ([2].

$$C_z = C_0 \times 10^{-z(k/D_{eff})^{1/2}} \quad [1]$$

C_0 = Start O_2 concentration at top of profile in $kg.m^{-3}$

z = Depth at which concentration is calculated in m

k = Kinetic rate constant for pyrite oxidation in s^{-1}

D_{eff} = Effective diffusion co-efficient in $m^2.s^{-1}$

C^z = Concentration of O_2 at depth z .

$$D_{eff} = (1 - S)^2 \cdot [\varepsilon(1 - S)]^2 \quad [2]$$

D_{eff} is the diffusion coefficient in the waste

S is the degree of saturation of pore space as a fraction

ε is the porosity of the material

It is important that ABA analyses should take note of the reaction pH, as the determined available NP is normally related to a strong acid at low pH, which will provide an artificially high available NP in neutral solutions. Reaction stoichiometries are entered into the model for the applicable pH range. The default values represent the worst case stoichiometric scenario that may occur within a tailings environment. Complete oxidation occurs, thus 3.75mol of O_2 is consumed per mol of pyrite. The assumption that Fe^{3+} will be formed during pyrite oxidation is also made, thus implying that 4mol of H^+ are produced per mol of pyrite oxidised. It is further assumed that NP is only consumed at an efficiency of 50%. This is due to the fact that HCO_3^- is

the stable carbonate ion at neutral pH. Since dissolved HCO_3^- is likely to be flushed from the system, before it is able to react with acidity, it is assumed that the formation of CO_3^{2-} and subsequent decomposition CO_2 will not occur.

The depth of a tailings profile is a significant input parameter, in that it is assumed that NP is available throughout the tailings profile, according to the average composition. Acidity on the other hand is only produced in the oxidised zone.

Modelling results

The TAP model was used to evaluate the sensitivity of the various input parameters of the model. The results of the sensitivity analyses are provided in Table 1.

Although all parameters have a significant influence on the rate of acidification of an FRD, the most sensitive parameters are clearly the S content and depth of the tailings profile. The impact of the S concentration is somewhat deceptive, as it is a measure which has a direct influence on the AP: NP ratio in the material:

- For example, increasing the S concentration (AP) in the material, but maintaining the same AP: NP ratio will result in an FRD taking longer to acidify due to the fact that O_2 diffusion flux will not increase proportionally to an increase in S concentration.
- Increasing the S concentration (AP) but maintaining the same NP concentration, i.e. increasing the AP: NP ratio will result in tailings acidifying more rapidly due to an increased rate of acid production depleting available NP at a faster rate.

Table 1: Modelled times to acidification for various parameters, as modelled within the constraints of Witwatersrand FRDs

	Minimum time to acidification (years)	Probable time to acidification (years)	Maximum time to acidification (years)
Porosity (46%)			
36%	4800	>10 000	>10 000
40%	3200	7400	>10 000
46%	2000	4000	7000
50%	1400	2800	4600
Moisture saturation (25%)			
10%	900	1800	2800
25%	2000	4000	7000
40%	5000	>10 000	>10 000
50%	>10 000	>10 000	>10 000
Depth of tailings profile (40m)			
5m	32	40	47
10m	119	174	224
20m	476	830	1213
30m	1095	2129	3359
40m	2000	4000	7000
50m	3200	7100	>10 000
60m	4600	>10 000	>10 000
Pyrite concentration (1%)	At constant AP:NP ratio		
0.1%	16	19	20
0.25%	194	302	407
0.5%	1638	3361	5508

1%	>10 000	>10 000	>10 000
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x Bold values relate to average values for Witwatersrand FRDs used for modelling purposes.

The TAP model output is presented for typical input values, as defined in Table 1 and illustrated in Fig. 3. The proportion of the available pyrite that is oxidised over the modelling period of 10 000 years can be deduced by the discrepancy between AP realised and total AP throughout the tailings profile. Figure 3 also shows how NP is reacted with the released acidity according to the given stoichiometry and a graph of the remaining NP is plotted on this basis vs. time. The time to acidification is equal to the point at which the NP value reaches zero. Lines of maximum, average and minimum acid generation rate and the associated consumption rate of NP are given to define the possible range of decay curves that could be observed according to fitting of PYROX data.

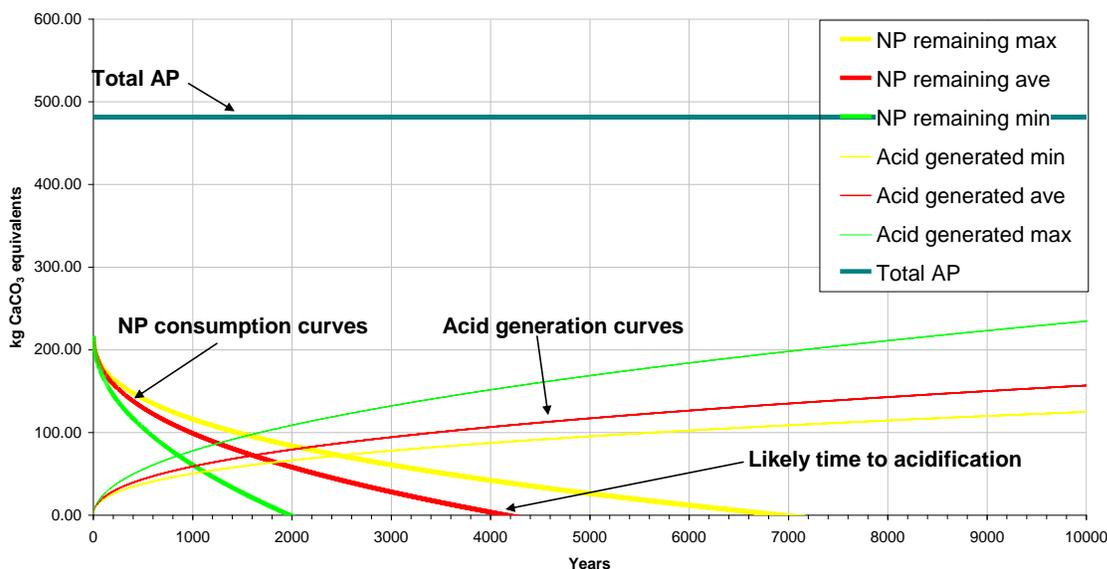


Figure 3: Cumulative curves for consumption of neutralisation capacity and acid generation in typical Witwatersrand FRDs.

Implications for the Mining Industry

Modelling and field observations indicate that weathering in the top of the tailings profile in a typical Witwatersrand FRD is rapid for approximately 10 years after decommissioning and that an oxidised horizon of up to 5m deep will form during this time. As discussed above, variations in physico-chemical parameters can modify these figures to form shallower or deeper profiles. Modelling results show that pyrite oxidation is only likely to consume available NP after approximately 1000 years in tailings of typical composition over the beach area of the FRD. Wall sections are most susceptible to AMD and will result in acidification much sooner than the beach areas due to the relatively lower height of air exposure and higher oxidation rates.

Exceptions were found to occur for FRDs with low sulfide concentrations and for facilities of low height, where acidification occurred at an earlier stage. These findings are ascribed to the

deeper penetration of O₂ in a low sulfide scenario, where O₂ consumption occurs more slowly, but there is a relatively large proportion of oxidised tailings relative to un-oxidised tailings in a shallow profile, thus eliminating the reservoir of unaffected NP below the oxidised zone.

The findings of this research therefore suggest that for typical Witwatersrand Goldfields FRDs, that the neutralisation capacity/reservoir in the FRDs below the oxidised outer zone is significant and could buffer acid produced in the outer oxidised zone on the leach areas for >1000 years. The research provided a screening tool (TAP model) and assessment methodology to assess the likelihood of seepage acidification from Witwatersrand FRDs and was also useful for evaluating a number of mitigation measures. The research report and model is available from the Water Research Commission of South Africa for a nominal fee (www.wrc.org.za).

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