AN INVESTIGATION INTO THE DEPTH AND RATE OF WEATHERING ON GOLD TAILINGS DAM SURFACES AS KEY INFORMATION FOR LONG-TERM AMD RISK ASSESSMENTS

N Bezuidenhout • PDS Rousseau

WRC Report No. 1347/1/05

Water Research Commission

AN INVESTIGATION INTO THE DEPTH AND RATE OF WEATHERING ON GOLD TAILINGS DAM SURFACES AS KEY INFORMATION FOR LONG-TERM AMD RISK ASSESSMENTS

Report to the
WATER RESEARCH COMMISSION

by

Nico Bezuidenhout and Pierre D.S. Rousseau
Golder Associates (Africa) Pty Ltd

WRC Report No: 1347/1/05
ISBN No: 1-77005-327-1

NOVEMBER 2005

Obtainable from:

Water Research Commission
Private Bag X03
Gezina
0031

ISBN No: 1-77005-327-1

Printed in the Republic of South Africa

ACKNOWLEDGEMENTS

The research in this report emanated from a project funded by the Water Research Commission, entitled:

AN INVESTIGATION INTO THE DEPTH AND RATE OF WEATHERING ON GOLD TAILINGS DAM SURFACES AS KEY INFORMATION FOR LONG-TERM AMD RISK ASSESSMENTS

The Steering committee responsible for this project included the following persons:

Mr HM du Plessis	Water Research Commission (Chairman)
Mr N Bezuidenhout	Golder Associates Africa (Pty) Ltd
Mr JA Wates	Golder Associates Africa (Pty) Ltd
Mr W Pulles	Pulles, Howard and de Lange Inc.
Dr B Zhao	Pulles, Howard and de Lange Inc.
Mr N Lesufi	Chamber of Mines of SA
Mr L LaBuschagne	Department of Minerals and Energy
Mr P Mthunywa	Department of Minerals and Energy

The following contributions are also gratefully acknowledged:

- AngloGold is kindly thanked for their assistance in providing data and assisting with the arrangements to conduct the fieldwork for this research. We would especially like to thank Mr Ulrich Sibilski and Ms Simone Barnard in this regard.

- The kind assistance and advice of our international Golder Associates colleagues in the United States is also acknowledged, specifically Dr Rens Verburg, Dr Ken de Vos and Ms Cheryl Ross.

EXECUTIVE SUMMARY

BACKGROUND AND MOTIVATION

The oxidation of sulphide minerals in gold Fine Residue Deposits (FRDs) of the Witwatersrand and its potential effect on water resources is arguably one of the major regional strategic environmental issues for the gold mining industry and the South African government.

Seepage from these FRDs tends to be alkaline to neutral during the operational phase, but can become acidic in the post operational phase as Acid Mine Drainage (AMD) processes become significant when the piezometric surface subsides.

International research and field observations have shown that an oxidised outer cap forms on FRDs, below which limited oxidation takes place. Enough acidity may be produced in this outer cap to consume all available neutralisation potential within an FRD body, however, as oxidation slows, the risk associated with AMD decreases.

The key question is thus how the oxidised zone will develop over time and how much acidity will be generated relative to available neutralisation potential in the operational phase of an FRD. This will determine the likelihood that a particular FRD will start generating acidic seepage.

OBJECTIVES

A top down approach was used to evaluate practical methods for evaluating the acidification risk associated with Witwatersrand FRD's. The study was guided by three study objectives, including:

1. To understand the relationship between the depth and rate of weathering and the potential mass of acidity at gold FRDs through empirical analysis.

2. Develop procedures to conduct rapid accurate AMD risk assessments of gold FRDs.

3. Evaluate mitigation or risk reduction measures by making use of the AMD risk assessment procedure and provide industry guidelines.

SUMMARY OF FINDINGS

Characteristics of FRD's

Acid Mine Drainage (AMD) originates due to the combined effects of flow or transport processes, and geochemical processes. Transport processes include transport of liquids and gases through the mineral matrix of a tailings deposit and geochemical processes include all mineral reactions and interactions between gases, liquids and solids within a deposit.

The parameters relating to each of these processes are determined by the physico-chemical nature of the tailings material in question. It was the purpose of this study to discuss the transport and geochemical processes as they pertain specifically to gold FRDs of the Witwatersrand, as these deposits are relatively well constrained in character and composition.

The nature of FRD varies throughout its lifecycle and depends amongst other factors on the method by which it was constructed, its mineralogy, climate, particle size distributions local topography and geology, age, dimensions and geochemical character. Available data indicate that the tailings materials of the Witwatersrand do not vary greatly in terms of their physico-chemical characteristics. Only two depostitional methods were recorded in the literature, i.e. paddock systems and cycloned tailings deposits.

Witwatersrand FRDs are typically very large deposits. They are typically greater than 1km in length and breadth and up to 60m high. Mineralogy is dominated by quartz (70% and 90%), phylosilicates (mainly sericite) (10% to 30%) and minor amounts of primary minerals (1% to 2%), including pyrite and calcite, which are key minerals in controlling AMD.

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. The effect is expected to be relatively limited in Witwatersrand tailings, as the particle size distribution tends to fall within a relatively narrow range, when compared to tailings from some other mineral deposits.

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. Existing measurements on the study site indicated that moisture tends to vary between 20 and 40% by volume in the material. Along with porosity, the moisture content plays a major role in determining the diffusion coefficient within a waste body, as it has been determined that the diffusion coefficient through waste is proportional to the air filled porosity. This is largely a result of the fact that the diffusion coefficient of oxygen in water is four orders of magnitude slower than the diffusion coefficient of oxygen through air.

TRANSPORT PROCESSES

Water and gas transport are the main processes affecting AMD and pollution mobilisation at gold FRDs. These processes are expected to be rate limiting in that flow processes determine the rate at which contaminants are mobilised from the tailings material into the receiving environment, whereas gas transport processes limit the rate at which AMD processes take place within the tailings deposit.

The flow of water determines how much of the contaminant load is mobilised and also determines how fast and in which direction it will move. Additionally, it also determines the moisture content within the tailings body at any one position in time and space and thus influences the oxidation rate by determining the fraction of air-filled pore space available for air to move through.

In fine tailings material, where oxygen diffusion is the major gas transport mechanism related to pyrite oxidation, gas transport rates are essential in determining the rate at which sulphides can be oxidised. This amounts to a direct measurement of the rate at which contaminants (acidity and toxic elements) are generated.

The rate of contaminant generation, along with the rate of contaminant transport will determine the concentration and volume of seepage from the FRD's and thus the environmental impact.

Water flow processes

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).

During the operational phase, seepage from an FRD in the vertical direction is largely dependant on the permeability of the material at the base of the FRD and the presence of subsurface drains. Where the base material is less permeable in the vertical direction than the tailings material, seepage will move horizontally towards the walls of the FRD. The results of this movement are observed in toe seepage. When tailings material is less permeable than the underlying material, seepage will tend to move downward, through the foundation into the local groundwater system.

Both saturated and unsaturated flow processes occur within an FRD, especially during the operational phase. In the post closure phase, unsaturated flow processes become progressively more dominant as the piezometric surface subsides. Future recharge then occurs

under unsaturated conditions. Previous modelling studies found that recharge would occur at a rate of between 1 and 6% of MAP after decommissioning.

Flow under saturated conditions can be represented by a simple form of Darcy's equation and is applicable below the piezometric surface within an FRD. This zone extends from the edges of the pool along the hydraulic gradient towards the edges of the FRD. If the piezometric surface intersects the wall of the FRD, toe seepage occurs. Morin and Hutt (1997) indicated that flow in the walls of an FRD is likely to be more rapid than near the pool. This is because of the coarser grain sizes and the shorter flow path length. Anisotropy can also be expected to play a part. In the post closure phase, it has been shown that the piezometric surface subsides at approximately 0.5m per year, which is an indication of the saturated hydraulic conductivity. The saturated hydraulic conductivities of Witwatersrand tailings have been determined to be between 10^{-6} and 10^{-8} ms^{-1}.

Unsaturated flow also obeys Darcy's law, however, it is more complex to calculate, because it is dependent on the degree of moisture saturation which typically varies over time. This is because large pores drain more quickly than small pores and water filled pores conduct water more easily than air filled pores. Empirically, unsaturated hydraulic conductivity is a non-linear function of volumetric water content and is also dependent on hydraulic head. It is commonly expressed as a function of soil suction ($K(\theta)$), pore water pressure ($K(u_w)$) and/or effective pore radius ($K(R)$). The effect of hysteresis has resulted in the preferential use of the moisture retention (soil suction) curve, as the differences between wetting and drying curves are less than for pore water pressure. A large number of empirical models have been developed to make use of these parameters.

Gas transport processes

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 oxygen transport, due to the fine-grained and hence relatively impermeable nature of tailings material. 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 oxygen transport rate within an FRD. Throughout the literature surveyed, diffusion was recognised as the major mechanism for oxygen transport through an FRD, and also serves as the basis upon which oxidation models for fine tailings are based.

Diffusion in tailings is driven by the concentration gradient between the tailings surface and the matrix, caused by weathering reactions that consume oxygen, e.g. the oxidation of pyrite. Oxidation is usually confined to a few metres below the surface as a result of oxygen transport limitations due to diffusion, with oxygen concentrations becoming progressively lower with depth.

Oxygen is the most significant gas in determining reactions in tailings material. The ability to determine the oxygen diffusion rate through an FRD over time will thus enable long-term prediction of AMD potential. Along with knowledge of water flow, calculation and/or modelling of pyrite oxidation rates provides the tools for determining pollutant concentrations within and around a tailings deposit over time.

Gas diffusion is mathematically described by Fick's law. Previous studies have showed that the limiting parameters controlling pyrite oxidation through diffusion are the diffusion coefficient and the kinetic rate constant. As the diffusion depth increases, oxygen diffusion becomes a more dominant parameter than the inherent pyrite reaction rate in determining oxygen flux.

Investigations into diffusion controlled pyrite oxidation in FRDs internationally has led to the conclusion that determinations that are made on bulk tailings tend to overestimate reaction rates. 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.

Geochemical processes

Mineral reactions

Witwatersrand gold tailings are constrained within a relatively narrow compositional range. Minerals that contribute to the Acid Potential (AP) are present in relatively low concentrations, however, AP is generally higher than the Neutralisation Potential (NP).

Acid Potential is predominantly ascribed to pyrite, as it is the most dominant sulphide mineral, with other sulphides occurring less frequently. Pyrite may react along different evolutionary paths, depending on the pH and redox conditions in which the reaction takes place.

Neutralisation Potential is ascribed primarily to carbonates of Ca and Mg. Silicates may also contribute significantly to the NP in a tailings deposit, depending on their inherent neutralisation potential and reactivity. Some silicate minerals are capable of neutralising acidity, especially where reaction rates are slow.

The formation of secondary minerals was also shown to be of major importance as the reaction stoichiometries of neutralisation reactions are critically dependent on mineral reactions that occur. Formation of ferrihydrite after pyrite oxidation, for example, results in double the requirement in neutralisation potential to prevent the reaction solution from acidifying.

Mineral reaction kinetics

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 7.8×10^{-12} m^2s^{-1}

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% oxygen these bacteria do not contribute significantly to pyrite oxidation reactions.

FIELDWORK

Fieldwork for this research concentrated on supplementing field data that were collected for Anglogold Limited by Pulles, Howard and de Lange in 2002. The approach that was followed, was to generate additional data and to discover relevant field related variables that would be of interest in constructing an oxygen diffusion based model for assessing acidification risk. Parameters that were determined in the field and laboratory included sample descriptions, particle sizes, X-Ray diffraction mineralogical analyses (XRD) and shake flask tests. Additional parameters, such as ABA analyses, moisture contents, particle size distributions, porosities and sulphur concentrations were obtained from the literature.

Observations and data.

Surface features were found to be abundant. Shallow mud cracks, the growth of plants and changes in particle size distributions were observed across the surface of the deposit. The presence of mud cracks and plants is expected to result in some variation in permeability across the surface of the FRD through macropore spaces. In the case of particle size distributions, variation is expected between conductivities in the wall and beach sections of the deposit.

Subsurface features included the presence of a leached zone directly below the surface, a hardpan horizon below this and an oxidation front, where iron stained sediments contacted grey, apparently unoxidised tailings.

The leached horizon is expected to be the result of rainfall leaching of oxidised products. The hardpan horizon is a result of mobilised iron from pyrite oxidation forming secondary minerals. Evidence of variable oxygen concentrations and preferential flow paths was noted by the presence of mottling and accentuated staining respectively.

The phenomenon of the oxidation front was researched and it was found that this transition is related to the presence of oxidised Fe and that it does not necessarily define the point of zero oxygen concentration. Redox conditions were found to be a function of the Fe^{2+}/Fe^{3+} concentration. The rapid transition, colour transition that is normally observed, along with occasional evidence of pyrite formation in cracks below the transition zone, indicates that the colour transition is likely to represent the position of the transition from oxic to anoxic conditions.

The presence of moist, fine-grained layers was observed. These layers are known to retard oxygen diffusion and were found to occur at different levels within an oxidation profile. Few moist layers were found in the top two meters of the tailings profiles investigated, probably as a result of desiccation during the long preceding dry season.

A number of yellow stained strata were observed in tailings auger cuttings that were collected. These were ascribed to syn-depositional oxidation. Due to rapid oxidation of near surface pyrite grains, such layers form rapidly during any drying cycle experienced across an FRD.

A significant finding that was observed from hydrometer particle size distributions, was that a significant proportion of analyses indicated that vertical variability in particle sizes is at least as significant as horizontal variability. This is important with respect to hydraulic conductivity and moisture retention, two key parameters in modelling impacts from tailings dams.

MODELLING

A conceptual model was developed to represent the generation of acidity from pyrite oxidisation within a typical Witwatersrand FRD. According to the model, wall sections of a tailings dam will undergo rapid acidification, as a result of horizontal influx of oxygen across the profile. The bulk of an FRD will, however, undergo oxidation through vertical ingress of oxygen by diffusion through the tailings profile. A large store of unreacted 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 FRD's. 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 AMD process. The purpose of the probabilistic spreadsheet model (Tailings Acidification Prediction or TAP model) that was constructed as part of this project, is to make use of this concept to predict the likely rate of acidification of seepage at the base of an 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.

Acidification of seepage is considered to be a major impact, relative to neutral drainage, as a result primarily of the potential for metal leaching to the receiving environment, but also as a result of acidity being harmful in itself.

Actual impacts will be a function of flow processes, coupled with the rate of contaminant generation within an FRD and the sensitivity of the receiving environment. Receptor impacts, however, will have to be evaluated on a case-by-case basis.

PYROX modelling

The PYROX model developed at the University of Waterloo is based on a double diffusion model developed by Davis and Ritchy (1986) and has been used extensively in international tailings investigations. The model was used to perform sensitivity analyses on relevant variables on the control of pyrite oxidation rates. Parameters were tested within the known compositional range of Witwatersrand tailings, in order to assist in developing a simplified empirical model, which could be used for assessing the likely time span for generation of acidic seepage from the base of a Witwatersrand gold FRD.

Typical to conservative values of sulphur content, particle size, moisture content, porosity, temperature, secondary diffusion coefficient and the thickness of the tailings profile were used as a baseline for determining sensitivities of other parameters.

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 to construct the TAP model as a function of static diffusion rates at initial compositions.

The TAP model was constructed from the point of view that the impact due acidification of seepage from the base of an FRD is dependent on the time span over which it is expected to occur. Consideration of the likely lifespan of suggested mitigation measures is a major consideration in this respect, as well as the likelihood of major impacts occurring after an extended period of weathering, when contaminant generation rates are likely to be much lower.

The model makes use of humidity cell secondary oxidation rates, porosity, the degree of moisture saturation, pyrite concentrations, the depth of a tailings profile and the NP:AP ratio of the tailings to predict a time to acidification of seepage.

Sensitivity analyses that were run for the model, indicated that most FRDs are likely to take more than 1000 years before acidic seepage will be generated from their bases. Exceptions were found to occur for FRDs with low sulphide concentrations and for facilities of low height. These findings are ascribed to the deeper penetration of oxygen in a low sulphide scenario, where oxygen consumption occurs more slowly and the relatively large proportion of oxidised tailings relative to unoxidised tailings in a shallow profile.

DISCUSSION OF RESULTS AND 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.

Considering the particle size distributions of tailings, it is likely that engineered surface covers will not significantly reduce oxidation rates on an FRD, except in the short period after decommissioning. The value of a cover may be realised in maintaining higher moisture levels and in reducing recharge through an FRD, however.

PROPOSED ASSESSMENT AND DESIGN CRITERIA FOR MITIGATION OF ACIDIFICATION IN WITWATERSRAND FINE RESIDUE DEPOSITS.

Field measurements

Field measurements need to concentrate on providing calibration data for the TAP model. Relevant measurements, which will assist in this regard include:

- Directly determined oxygen flux measurements, which can be checked against modelled values. These measurements should be made regularly to determine temporal variations.

- Moisture contents, which are important for determining diffusion coefficients and can be determined with geophysical methodologies. Measurements should be ongoing to determine temporal variations.

- Porosity.

- Pyrite concentrations in representative samples below the oxidised zone.

- NP measurements from samples above the piezometric surface, but below the zone where acid reactions have already occurred.

Sampling should be conducted such that oxidation reactions are prevented, through exclusion of air and if possible the inclusion of inert gases in samples.

Potential mitigation options

A number of potentially promising mitigation options were identified on the basis of the conceptual model. The various options need to be subjected to technical and financial feasibility assessments before they can be considered as guidelines, however. A list of identified measures is included below:

- Alkaline addition – Addition of limestone, especially in the wall section, will decrease the AP:NP ratio and will reduce the likelihood of acidification of seepage due to the acid-base balance.

- Depyritisation – This technique has been documented in the literature, especially with regard to cycloned tailings. Pyrite can be separated and included in a lower risk environment elsewhere or can be reprocessed for a value added product.

- Engineered covers – These are commonly used in the industry and are designed to reduce oxygen ingress through maintaining high moisture levels and to reduce recharge and consequent seepage through the store and release principle.

- Fly ash – Fly ash is alkaline and fine grained, as well as having pozzolanic properties in many cases. Mitigation with this material may therefore be possible, however, caution needs to be exercised due to the commonly high leachable metal concentrations in fly ashes.

- Bottom liner – Where seepage is generated, a bottom liner can be used to collect affected seepage for treatment. This will be especially of relevance where acidic seepage will be generated at an early stage in the wall section of an FRD.

The suggested measures need to be considered in the context of available opportunities throughout the lifecycle of an FRD, for instance, a liner must be installed prior to construction for maximum efficiency, whereas a cover can be constructed only after decommissioning of the facility.

CONCLUSIONS AND RECOMMENDATIONS

The research has shown that the depth and rate of weathering in Witwatersrand FRDs is highly dependent on physico-chemical properties, however, it was shown that facilities of typical composition and morphology should take more than 1000 years, before acidic seepage is released to the receiving environment below the base of the facility.

The objectives for the study were partially achieved, in so far as it was found that accurate modelling of oxidation rates is relatively complex and data intensive. Providing a general model is therefore difficult due to the various site-specific variations in parameters affecting AP and NP realization on an FRD. The recommended mitigation measures need to be subjected to technical and financial feasibility assessments before they can be accepted as best practice measures.

It is thus recommended that further work should be conducted in evaluating these mitigation measures. Other recommendations include:

- Verification of findings by application of other models.

- Collection of field data to develop a database against which calibration can take place and which can serve as a record. Parameters to be determined include:

- Pyrite concentrations
- Intrinsic pyrite oxidation rates (humidity cells)
- Empirical determination of diffusion coefficients
- Moisture profile determinations
- Depth dependent oxygen concentration profile determinations for time line calibration
- Oxygen flux determinations for oxidation rate verification.

TABLE OF CONTENTS

SECTION **PAGE**

1 INTRODUCTION AND SCOPE OF INVESTIGATION 1
1.1 Background to project 1
1.2 Project objectives and deliverables 1
1.3 Research methodology 2
1.4 Field work and investigation 3
1.5 Analytical, numerical modelling and sensitivity analyses 3
1.6 Recommendations and procedures 4
2 LITERATURE REVIEW OF FACTORS THAT GIVE RISE TO ACID MINE DRAINAGE IN SULPHIDIC GOLD FRDS IN SOUTH AFRICA 5
2.1 Characteristics of Witwatersrand gold FRDs 5
2.1.1 Construction methods 6
2.1.2 Particle size distributions 7
2.1.3 Moisture content in Witwatersrand tailings 9
2.1.4 Porosity 9
2.1.5 Geochemistry of Witwatersrand gold FRDs 9
2.2 Transport processes 10
2.2.1 Water flow within a Witwatersrand gold FRD. 10
2.2.2 Gas transport processes 19
2.3 Geochemical processes 27
2.3.1 Mineral reactions 27
2.3.2 Mineral reaction kinetics 30
2.3.3 Secondary mineral formation 32
3 FIELDWORK 33
3.1 Objectives 33
3.2 Methodology 34
3.3 Observations and data 34
3.3.1 Surface features 34
3.3.2 Subsurface features 36
3.3.3 Laboratory analyses 38
3.4 Discussion 41
4 MODELLING 42
4.1 Modelling objectives 42
4.2 Conceptual model 42
4.3 PYROX modelling 46
4.3.1 Sensitivity analysis 47
4.4 Probabilistic modelling 52
4.4.1 Model construction 53
4.4.2 Modelling results 54
5 DISCUSSION OF RESULTS AND IMPLICATIONS FOR THE MINING INDUSTRY 60
5.1 Development of the oxidised zone and the associated impact on the risk of acidification 60
5.2 Pyrite oxidation in the wall section of an FRD and associated risk of acidification 61
6 PROPOSED ASSESSMENT AND DESIGN CRITERIA FOR MITIGATION OF ACIDIFICATION IN WITWATERSRAND FRDS 62
6.1 Field measurements 62
6.1.1 Measurement of modelling parameters in tailings 62
6.1.2 Sampling 65
6.2 Long term monitoring of FRDs with a view to prediction of AMD impacts 66
6.2.1 Field measurements 66

6.3 Mitigation of AMD impacts from Witwatersrand gold FRDs....67
6.3.1 Mitigation options....67
6.3.2 Lifecycle considerations....69
7 CONCLUSION....72
8 RECOMMENDATIONS....73
9 REFERENCES....75

LIST OF APPENDICES

Appendix A Analytical data and sample descriptions
Appendix B CD containing the report as a printable pdf file and a copy of the TAP excel spreadsheet model

LIST OF TABLES

Table 2.3.2(a) Pyrite reaction rates for different species at different oxygen concentrations (after perkins et al. (1995)....31
Table 2.3.2(b) Sulphide oxidation rates measured with the oxygen consumption method by Tibble & Nicholson (1997)....31
Table 4.4.2(a) Times to acidification for various parameters, as modelled within the constraints of Witwatersrand FRDs, using the spreadsheet model developed for this study....57

LIST OF FIGURES

Figure 2.1.1(a) Stratification visible within a wall cross-section of an FRD.... 7
Figure 2.1.1(b) Particle size distributions associated with various types of tailings, after (Anglo American (1995)....8
Figure 2.2.1(a) Hydrological model of an FRD with K_{sat} base > K_{sat} tailings.... 13
Figure 2.2.1(b) Hydrological model of an FRD with K_{sat} base < K_{sat} tailings.... 13
Figure 2.2.1(c) Hydrological model of an FRD post closure 13
Figure 2.2.1(d A surface oxidised zone and near-surface preferential pathways visible through iron staining (Bezuidenhout and Vermaak (2002)..... 16
Figure 2.2.1(e) A conceptual graph of seepage rates from an FRD pre- and post-decommissioning.... 18
Figure 2.2.2(a) A contact zone between oxidised and unoxidised tailings.... 20
Figure 2.2.2(b) A schematic diagram depicting the oxidation profile during the operational phase of an FRD.... 26
Figure 2.2.2(c) A conceptual diagram, depicting the total oxygen flux (pyrite mass oxidised) within an FRD as a function of its lifecycle.... 27
Figure 3(a) Sampling positions at the main study site.... 33
Figure 3.3.1(a) Mud cracks hosting a grass community on an FRD, as well as other plant mass.... 35
Figure 3.3.3(a) Particle size analyses of all sub-samples collected at the study site...40
Figure 3.3.3(b) Particle size variations between moist layers in tailings matrix for samples 2, 5 and 6.... 40
Figure 4.2(a) A conceptual model of oxidation and neutralisation processes within an FRD.... 43
Figure 4.2(b) A conceptual representation of average seepage water quality evolution during the lifecycle of a sulphidic FRD.... 44
Figure 4.3.1(a) Sensitivity analysis to particle size differences within a tailings body..49

Figure 4.3.1(b) Reaction rates modelled over a 200 year period, showing sensitivity to porosity..... 50
Figure 4.3.1(c) Sensitivity analysis of reaction rates vs. moisture saturation in percent 51
Figure 4.3.1(d) Sensitivity analysis for sulphur concentration..... 52
Figure 4.4.1(a) An example of the input screen for the Tailings Acid Probability (TAP) spreadsheet model..... 54
Figure 4.4.2(a) An example of the acid generation output of the spreadsheet model accompanying this report..... 58
Figure 4.4.2(b) An example of the spreadsheet model output. Acidification occurs when NP= 0 kg $CaCO_3$ equivalents..... 58
Figure 4.4.2(c) A set of graphs depicting the relationship between the height of an FRD of typical composition and the time to acidification.....59

Glossary and abbreviations

Acidification	Acidification in the context of this study is considered to be the point at which acidic seepage is generated at any particular point in a tailings profile after consumption of NP or on a larger scale, the point at which acidic seepage flows from the base of the dump into the subsurface geology.
ABA	Acid-Base accounting
AMD - Acid Mine Drainage	Acidic drainage from broken rock related to mining, due to the oxidation of sulphide minerals and subsequent release of sulphuric acid.
AP	Acid potential. This is the potential for the formation of H_2SO_4, as calculated from sulphide species in a sample.
ARD - Acid Rock Drainage	ARD is synonymous with AMD in the mining milieu, but is considered to be a more general term.
FRD - Fine mine residue deposit	A deposit of fine residues from mineral processing. This is synonymous with a "tailings dam", "tailings deposit" or "slimes dam", but is the preferred modern usage where fine grained materials make up the body of the deposit.
MAP	Mean Annual Precipitation.
NP	Neutralising Potential. This is the capacity for a sample to neutralise acidity through buffering reactions and is expressed in $CaCO_3$ equivalents, commonly as kg/t.
XRD	X-ray diffraction spectrometry.

1 INTRODUCTION AND SCOPE OF INVESTIGATION

1.1 Background to project

The oxidation of sulphide minerals, commonly associated with gold mine tailings in South Africa 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 of fine mine residue deposits (FRDs), seepage is typically saline and neutral (even alkaline). During the decommissioning and closure phases of the FRDs, Acid Mine Drainage (AMD) processes take place. Once started, AMD processes are persistent and worsen the salinity impacts associated with the operational phase, since metal mobilisation continues for long periods after decommissioning.

International research shows that oxygen ingress into FRDs as a result of consumption by pyrite oxidation, decreases with depth in an FRD resulting in an oxidised outer cap (Mayer et al. (2000). Enough acidity could be produced in this oxidised cap to consume all Neutralisation Potential in the rest of the FRD, resulting in Acid Mine Drainage (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.

The inherent store of neutralisation capacity of an FRD should be seen as a non-renewable resource and is the most significant and important defence barrier to long-term generation of AMD. Because pyrite oxidation is kinetically driven, the longer it has to oxidise the more acidity it will produce.

It therefore seems that the key question that needs to be answered in assessing the AMD potential of an FRD is how deep the oxidised zone will develop over time and hence how much acidity will be generated in this zone compared to the total neutralisation potential of the FRD. Understanding the depth and rate of weathering, and the mass of acidity generated, will thus provide first order input data for an AMD risk assessment for an FRD. By comparing the total mass of acidity that will likely be generated in the oxidised outer surface of the FRD (developed through this research) to the total Neutralisation Potential of the rest of the FRD will provide an estimate of the potential for long term AMD.

This report documents the works that have been carried out by Golder Associates Africa under contract with the Water Research Commission into the depth and rate of weathering on gold FRD surfaces as key information for long-term Acid Mine Drainage (AMD) risk assessment.

1.2 Project objectives and deliverables

There are basically two approaches that can be used to assess the potential for complete consumption of Neutralisation Potential in gold FRDs, including a top down or bottom up

approach. Bottom up approaches will typically include complex geochemical and mass-flow transfer modelling requiring large data inputs. This type of approach is typically expensive, complex and difficult to calibrate. This proposal follows a top down approach by making use of probabilistic analysis and risk methodology to assess the potential for AMD from gold FRDs.

The objectives of the research documented in this report were:

1. To understand the relationship between the depth and rate of weathering and the potential mass of acidity at gold FRDs through empirical analysis.

2. Develop procedures to conduct rapid accurate AMD risk assessments of gold FRDs.

3. Evaluate mitigation or risk reduction measures by making use of the AMD risk assessment procedure and provide industry guidelines.

The objectives in this research were aimed at providing an assessment methodology or guideline and risk assessment tool to evaluate the long-term AMD potential of typical Witwatersrand sulphidic gold FRDs. The approach included both empirical and analytical works and made use of relevant and current international research on this topic.

The deliverable of this research can be summarised as follows:

1. A technical research report consisting of the following components.

   - Documenting the latest knowledge regarding AMD processes on sulphidic gold FRDs.
   - Analytical and numerical modelling and sensitivity analyses of key processes driving and effecting AMD processes on sulphide gold FRDs.
   - Procedures and guidelines for rapid accurate assessment of the AMD potential of gold FRDs.

2. A technical paper documenting the findings and a presentation to authorities and industry representatives.

3. Capacity building through the University of Western Cape resulting in the completion of a project as part of an honours degree.

1.3 Research methodology

The research methodology followed in this investigation can be summarised by the following four steps, which form the basic structure of the report.

1. Parameters and processes, which give rise to Acid Mine Drainage at gold FRDs.

2. Field observations and analyses to confirm phenomena discussed in the literature.

3. Sensitivity analysis of relevant parameters in tailings oxidation.

4. Model construction and risk assessment for acidification of Witwatersrand gold FRDs.

Conceptual modelling and literature surveys were used to develop an understanding of process and parameters that drive and effect AMD processes on gold FRDs. The literature study included a survey of international research and is documented in Section 2 of this report. The aim was to understand and document the latest knowledge regarding processes and parameters effecting AMD on gold FRDs.

1.4 Field work and investigation

Based on the findings of the conceptual modelling and literature survey, field investigations were conducted to evaluate the practicalities of measuring the various processes and parameters affecting AMD in gold FRDs. Various FRDs were visited to evaluate the potential for screening type measurements.

AngloGold commissioned Pulles, Howard and de Lange during 2002 to sample gold tailings for geochemical parameters. The data generated included results for static geochemical tests, mineralogy and physical properties. One of these FRDs was re-sampled by the authors to gather additional information.

1.5 Analytical, numerical modelling and sensitivity analyses

In order to evaluate the inter-relationships of the key processes that determine the AMD potential of FRDs, various analytical and numerical models were constructed to simulate the various AMD processes. Use was primarily made of O_2-diff, a steady state oxygen diffusion model developed by Golder Associates Africa, and PYROX, a dual diffusion finite difference pyrite oxidation model developed by the University of Waterloo. These models were used to conduct sensitivity analyses on parameters affecting AMD processes and to develop an Excel based spreadsheet model that can be used for screening analyses. The spreadsheet model is intended to be a tool to determine the timespan involved in the consumption of Neutralisation Potential (NP) within a tailings profile, due to acid generated from pyrite oxidation. As it has been shown that Acid Potential (AP) is generally higher than NP in samples of Witwatersrand tailings, it is always theoretically likely that acidification will occur at some point in time. It is, however, the objective of this model to determine what this time frame is with a view to improving the understanding of associated risk, to assist with decision making on management options for closure.

It must be remembered that this research is aimed only at evaluating whether an FRD will turn acidic or not within the foreseeable future. A timeframe is defined within approximately

one order of magnitude. This means that in the interim neutral saline drainage can still take place from an FRD, which has been classified as having a low potential for AMD.

1.6 Recommendations and procedures

This component of the research focused on documenting the critical parameters that need to be measured in order to conduct simplistic AMD risk assessments using the accompanying spreadsheet model as a screening tool. This component also provides guidelines on how to take these measurements and how to conduct the screening analyses of AMD potential of South African sulphidic FRDs. Guidelines are also provided to assist the reader in interpreting the results from the screening model.

2 LITERATURE REVIEW OF FACTORS THAT GIVE RISE TO ACID MINE DRAINAGE IN SULPHIDIC GOLD FRDS IN SOUTH AFRICA

Acid Mine Drainage originates due to the combined effects of two processes, namely flow or transport process and geochemical processes. Transport processes include transport of liquids and gases through the mineral matrix of a tailings deposit and geochemical processes include all mineral reactions and interactions between gases, liquids and solids within a deposit.

The parameters relating to each of these processes are determined by the physico-chemical nature of the tailings material in question. It is the purpose of this section to discuss the transport and geochemical processes as they pertain especially to gold FRDs of the Witwatersrand, as these deposits are relatively well constrained in character and have compositions that fall within well defined boundaries, based on a detailed understanding of the resource mineralogy of the source material.

The nature of an FRD varies throughout its lifecycle and depends amongst other factors on the method by which it was constructed, its mineralogy, climate, particle size distributions local topography and geology, as well as other factors. Available data, however, indicate that the tailings materials of the Witwatersrand do not vary greatly in terms of their physico-chemical characteristics. Only two depostitional methods were recorded in the literature, i.e. paddock systems and cycloned tailings deposits (Anglo American (1995).

2.1 Characteristics of Witwatersrand gold FRDs

Witwatersrand FRDs are typically very large deposits, due to the very large volumes of material that are mined and the high proportion of waste relative to recoverable products of value. They are typically greater than 1km in length and breadth and up to approximately 60m high.

The tailings are typically of a relatively uniform composition, commonly consisting of between 70% and 90% quartz, 10% to 30% phylosilicates (mainly sericite) and 1% to 2% primary minerals such as uraninite, monazite, chromite and rutile (Rösner et al. (2001) after Wittman (1976). In the study by Zhao (1998), other associated minerals were also shown to be common in the mineralised zones of the Witwatersrand , e.g. titanite (sphene), albite and calcite. These were, however, not reported quantitatively. Various sulphides were also shown to be present in previous studies reviewed by Zhao (1998), e.g. pyrrhotite, sphalerite, chalcopyrite and galena, however, these are minor phases and are less important with respect to their acid generation potential, when compared to pyrite.

These FRDs have mostly been deposited by the open-ended paddock method, with some deposits having been constructed from cycloned materials (Anglo American (1995). The deposits have a stratified morphology, caused by hydraulic separation of coarser and finer materials between depositional events. Some variation also occurs between the wall and

beach sections as a result of differential settling of particulates over the FRD profile, although the significance of this effect is likely to be limited in paddock deposits of the Witwatersrand due to the relatively uniform particle size distribution of the Witwatersrand gold tailings in general (Anglo American (1995) and Bezuidenhout and Vermaak (2002).

2.1.1 Construction methods

FRDs are constructed, using various methods throughout the world. All methods fall within essentially three classification classes (MacG. Robertson (1987):

- No embankment facilities, including submarine and lacustrine deposition, commonly applied in parts of the world where conditions and regulations permit subaerial deposition.

- Central discharge methods, which make use of tailings viscosity to shape the deposit from a central point, also eliminating the need for an embankment system.

- Embankment retained deposits are constructed, using upstream, centreline and/or downstream construction methods. These methods are defined according to the direction in which successive lifts in the wall are constructed relative to the point of discharge.

Among these construction methods, the Witwatersrand FRDs classify within the embankment retained deposits. Paddocked systems fall mostly within the upstream construction methods due to successive lifts being constructed in an upstream direction to the previous ones. Alternating upstream and downstream depositional methods have also been used (Anglo American (1995). Cycloning falls within the centreline construction method according to MacG Robertson (1987) as the deposition point moves essentially vertically as the size of the deposit grows.

The paddock system makes use of pipes to fill constructed paddocks (day paddocks) in a ring dyke with slurry during the day. The coarser material is allowed to settle until the late afternoon, whereafter the remaining fluid is allowed to drain through a breach to a larger night paddock within the body of the FRD. Here material that is still suspended can settle over night. Finally, clarified water is allowed to decant through the penstock system back to the plant for reprocessing (Anglo American (1995) and Rösner et al. (2001).

Distinctive layers of between 5cm and 10cm thickness, visible by variations in colour and texture, are commonly observed. Sharp contacts between layers are visible (see Figure 2.1.1(a).

Cyclone deposition occurs in a minority of cases in the Witwatersrand. This method makes use of thickened tailings underflow from a cyclone system to construct a ring dyke, with the cyclone overflow being directed into the impoundment. The method is dependent on

particle size distributions, as sufficient coarse material is necessary to achieve a good separation by cycloning (MacG. Robertson (1987).

2.1.2 Particle size distributions

Particle size distributions within FRDs have been described in a number of studies. Yanful (1991) describes the particle size distributions as silty sands or sandy silts, with a proportion of clay sized material. The material is generally fine grained as is required to liberate all mineral grains for metallurgical processing. The grain size distribution is described as >75% of the material with particle sizes of <75μm (Rösner et al. (2001) after SRK (1988).

The gold tailings of the Witwatersrand are described as being relatively uniformly graded and thus do not possess great variability in their saturated conductivity values (Anglo American (1995). In general, four types of tailings are recognised, i.e. FC/FF, CC/CF, FC/CF and CC/FF (see Figure 2.1.1(b) after Anglo American (1995), with C representing Coarse and F representing Fine. FC for example represents a fine coarse fraction and FF represents a fine fraction. Gold tailings are recognised as an FC/CF type of tailings.

Figure 2.1.1(a): Stratification visible within a wall cross-section of an FRD.

The well-graded tailings in the FC/CF group tend to form a relatively flat beach, of 0.5% and result in a relatively uniform permeability as well. A typical curve for this type of distribution

has a D_{10} particle size of 0.03mm (Anglo American (1995). This distribution falls within a set of curves, describing tailings of various types, with D_{10} particle sizes ranging between 0.003mm and 0.04mm, with FC/CF tailings tending to fall between about 0.02mm and 0.1mm. An envelope containing all tailings materials is defined with an upper D_{10} bound of 0.25mm.

Available particle size distributions for this study were all obtained from paddocked tailings and conformed with literature descriptions. No data was available for cycloned tailings. It is, however, stated that cycloning is feasible in gold tailings even with 90% of particles <75μm. Dewatering of underflow tailings to 8%-10% water allows rapid consolidation and high rates of rise on cycloned tailings deposits (Anglo American (1995).

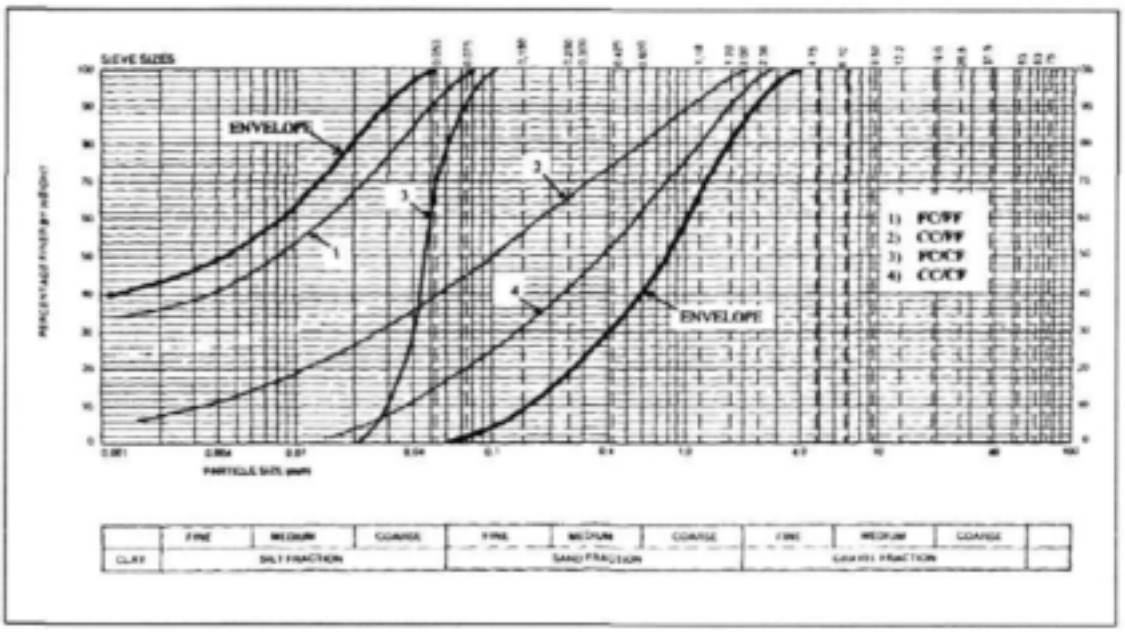

Figure 2.1.1(b): Particle size distributions associated with various types of tailings, after (Anglo American (1995).

Particle size differences between the wall and beach sections are likely to be more pronounced in cycloned tailings. It can thus be expected that the differences in permeabilities to both gases and liquids between wall and beach sections will be more pronounced in these deposits than would be the case for paddocked tailings.

Morin and Hutt (1997) indicated that a degree of mineral separation, due to density separation, can occur in certain FRDs where suitable conditions exist. No data were available to confirm or negate this effect in Witwatersrand gold FRDs. Stratification due to density separation was also observed with rotational pumping of material between paddocks, a common phenomenon in Witwatersrand gold tailings (see Figure 2.1.1(a).

The presence of fine-grained strata within tailings deposits has also been noted. These features are of major significance, as they are capable of retarding the flow of water due to the formation of capillary breaks in the tailings material and in consequence will also retard

gas flow due to the presence of these water saturated layers. Such layers were discussed by Nicholson et al. (1995) and were also observed in the field during this study.

2.1.3 Moisture content in Witwatersrand tailings

The moisture content of tailings material is variable and is a function of climatic variation and the lifecycle of an FRD. Rösner et al. (2001) after Funke et al (1990) distinguish tailings material derived from gold processing plants and tailings derived from gold and uranium processing plants. The solids to water ratio of the wet slimes material is approximately 1:1 for the wet gold slimes material and 1:4.5 for the material derived from combined gold and uranium recovery.

The moisture concentration may be affected by a number of factors, including climatic factors, like ambient temperature, Mean Annual Precipitation (MAP) and evaporation rate. Other factors affecting moisture content include physical factors and mineralogical factors. The moisture content in any specific portion of an FRD, especially below the zone where evaporation takes place is largely governed by the particle size distribution.

Moisture contents measured in the laboratory for the upper portion of closed FRDs by (Pulles, Howard and Delange (2002) varied between 8.5 and 20.5% by weight.

As mentioned in Section 2.1.2, fine-grained layers exist within FRDs, which may lead to the formation of water-saturated layers where capillary breaks form. Variations in moisture content over small distances may therefore be significant and can consequently significantly affect the flow of water and gases through the tailings profile.

2.1.4 Porosity

Porosity has been shown to be an important factor in determining gas diffusion rates and is a fundamental input parameter in modelling the oxidation rate of pyrite in a fine grained matrix. This is because the diffusion coefficient of oxygen through the material is defined primarily by the material porosity and the moisture content (David and Nicholson (1995), Wunderly et al. (1995) and Nicholson et al. (1989). The diffusion coefficient for oxygen through air is greater by four orders of magnitude than the diffusion coefficient for oxygen in water. Along with tortuosity causing increased path lengths, this phenomenon determines the effective diffusion coefficient through tailings material.

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 (pers. comm.)).

2.1.5 Geochemistry of Witwatersrand gold FRDs

The mineral composition of Witwatersrand reefs are described as consisting of between 70% and 90% quartz, 10% to 30% phylosilicates (mainly sericite) and 1% to 2% primary minerals such as uraninite, monazite, chromite and rutile (Rösner et al. (2001) after Förstner and

Wittmann (1976). More detailed analyses from Feather and Koen (1975) showed quartz having abundances of 88% and 89%, muscovite of 4.4% and 3.0% and pyrite of 6.6% and 3.2% respectively for the Vaal Reef and the Ventersdorp Contact Reef. Minor proportions <1% of other minerals were found to be present. Rösner *et al.* (2001) found that besides quartz, the minerals found in tailings included muscovite, chlorite and pyrophyllite. The secondary mineral, jarosite, which is formed after pyrite oxidation, was also detected in minor amounts and where sufficient calcium was present gypsum was also found to occur.

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.

2.2 Transport processes

Water and gas transport are the main processes affecting AMD and pollution mobilisation at gold FRDs. These processes are expected to be rate limiting in that flow processes determine the rate at which contaminants are mobilised from the tailings material into the receiving environment, whereas gas transport processes limit the rate at which AMD processes take place within the tailings deposit.

The flow of water determines how much of the contaminant load is mobilised and also determines how fast and in which direction it will move. Additionally, it also determines the moisture content within the tailings body at any one position in time and space and thus influences the oxidation rate by determining the fraction of air-filled pore space available for air to move through.

In fine tailings material, where oxygen diffusion is the major gas transport mechanism related to pyrite oxidation, gas transport rates are essential in determining the rate at which sulphides can be oxidised. This amounts to a direct measurement of the rate at which contaminants (acidity and toxic elements) are generated.

The rate of contaminant generation, along with the rate of contaminant transport will determine the concentration and volume and thus the environmental impact of any pollution that does occur.

2.2.1 Water flow within a Witwatersrand gold FRD.

Water flow is commonly described by two different models, i.e. saturated flow and unsaturated flow. In saturated flow models, a basic form of Darcy's law is commonly used to describe the flow rate of water in a porous material. In unsaturated flow models, the situation becomes far more complex, as hydraulic conductivity becomes a function of the moisture content of the material. This is because water tends to flow in saturated pore spaces and does not easily flow through dry, air-filled, pore spaces.

It is essential to be able to calculate or measure the water balance associated with an FRD. This implies that fluid fluxes can be determined, as well as the moisture content of the tailings body itself. These are the main variables, other than chemistry, that will directly and indirectly determine the environmental impact that will be associated with a particular tailings deposit. To construct such a water balance, water inputs from rainfall and process water need to be quantified on a temporal basis. Losses due to seepage and evaporation/evapotranspiration also need to be quantified. The inherent moisture content of tailings will be a function of their physico-chemical makeup. Ideally a good understanding of such a water balance enables one to determine how much water flows through an FRD to the groundwater table or to the foot of the FRD, as well as to determine what the moisture content of the tailings deposit is over time.

The seepage volume from an FRD will also relate to contaminant concentrations in seepage water, as a dynamic balance will exist between seepage rates and contaminant generation by mineral weathering. Along with spatial distribution of contaminants, these factors will determine the extent of environmental impacts.

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). Deeper cracking due to faulting or seismicity may also be significant (Zhao pers. com. (2004).

A list of characteristics of elevated FRDs is provided by de Vos et al. (1999). Water is described as moving only vertically, with horizontal outflows of water not being observed, largely due to the large horizontal extent of FRDs, relative to their height. The effects of anisotropy are thus eliminated.

During the operational phase, seepage from an FRD in the vertical direction is largely dependent on the permeability of the material at the base of the FRD and the presence of subsurface drains. Where the base material is less permeable in the vertical direction than the tailings material, seepage will move horizontally towards the walls of the FRD (see Figures 2.2.1(a) and 2.2.1(b). The results of this movement are observed in toe seepage. When tailings material is less permeable than the underlying material, seepage will tend to move downward, through the foundation into the local groundwater system. Interception drains at the base of an FRD will also improve vertical movement of water. The anisotropy of the system however favours movement of water in the horizontal direction, thus making it difficult to drain water from the system from below. For faster drainage of the water from an FRD (Rösner et al. (2001) suggested the use of vertical drains.

A conceptual diagram of the post closure hydrology is presented in Figure 2.2.1(c). A receding piezometric surface is indicated. The level of this surface and rate at which it recedes may vary considerably, depending on local conditions, i.e. tailings permeability, permeability of the base and climate.

Saturated flow processes

Saturated flow processes are described by Darcy's law. The flow rate is dependent on the hydraulic conductivity of the porous material, the cross-sectional area of the "aquifer" or water carrying body and the head of water. The head is defined as the hydraulic gradient, which is the difference in height of the piezometric surface divided by the horizontal distance between two measuring points within the flow path.

The equation for Darcian flow is expressed as $Q=KIA$, where Q is the flow rate (m^3s^{-1}), K is the hydraulic conductivity (ms^{-1}), I is the head (hydraulic gradient), which is dimensionless and A is the cross-sectional area (m^2) of the aquifer.

Although there are deviations from Darcy's law under field conditions due to non-linearity of hydraulic conductivity, a single value of K describes saturated flow processes accurately enough for application to engineering problems. Non-linearity is observed, where turbulent flow may occur, or where significant electric forces cause interaction between water and solid materials.

Permeability or hydraulic conductivity is a function of the conducting material's particle size distribution and porosity and physico-chemical properties of the fluid involved, such as viscosity, density, dielectric constant and temperature.

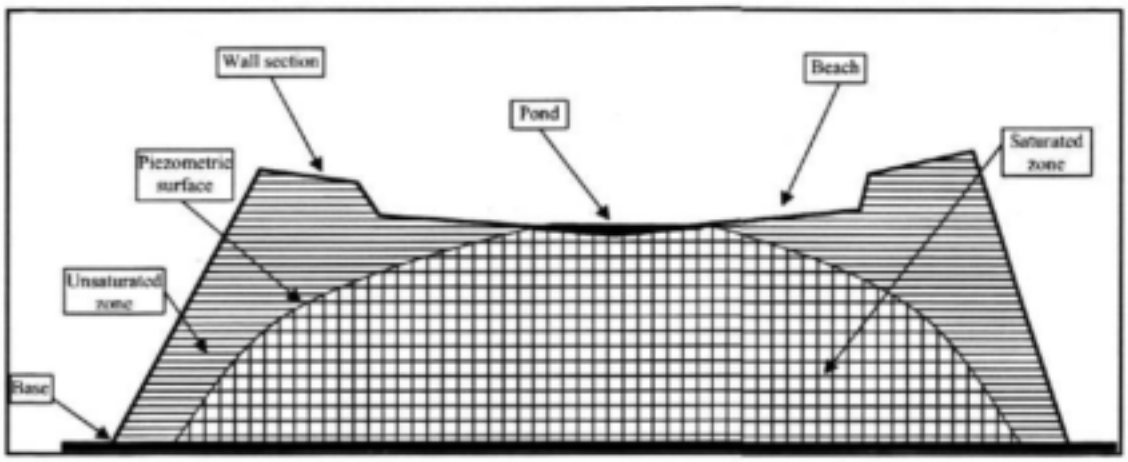

Figure 2.2.1(a): Hydrological model of an FRD with K_{sat} base > K_{sat} tailings.

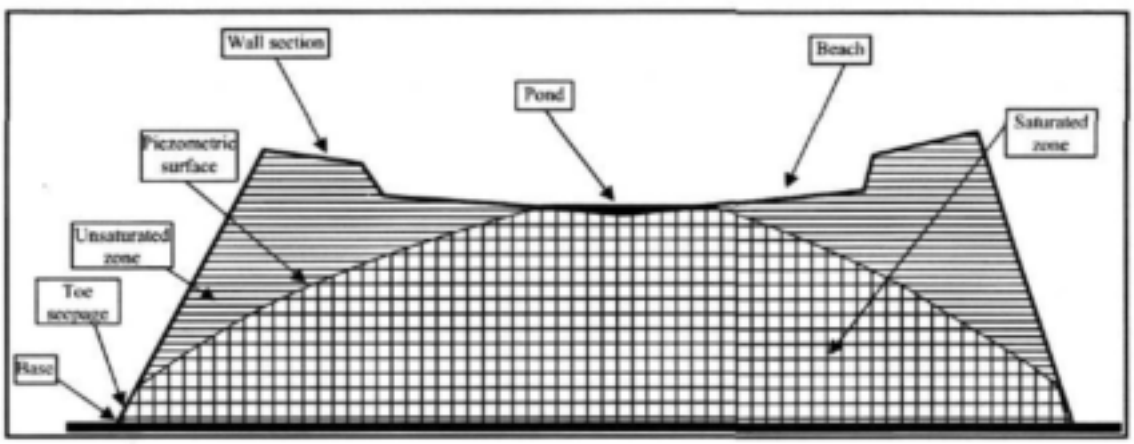

Figure 2.2.1(b): Hydrological model of an FRD with K_{sat} base < K_{sat} tailings.

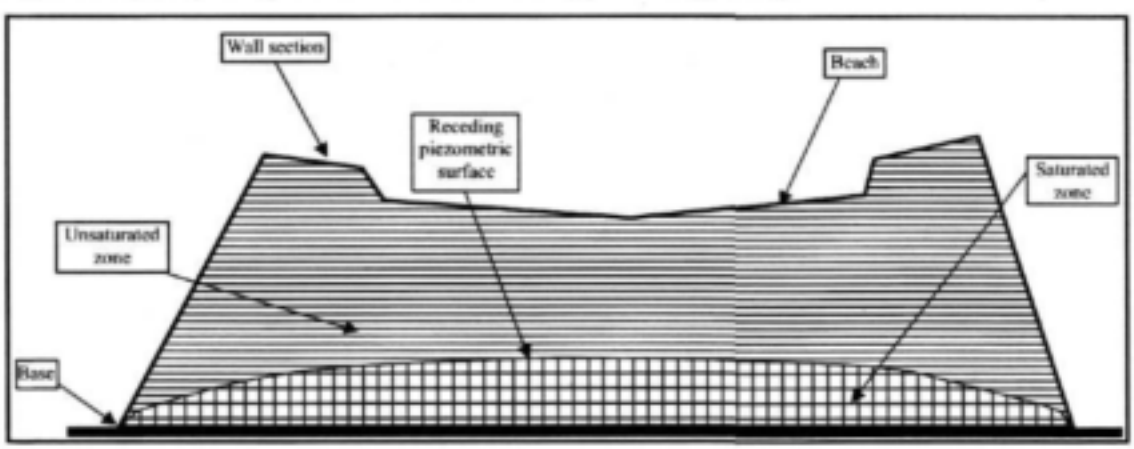

Figure 2.2.1(c): Hydrological model of an FRD post closure.

Commonly D_{10}[1], other characteristic particle sizes or combinations of characteristic particle sizes between D_5 and D_{50} are used to calibrate empirical models (van Schalkwyk and Vermaak (2000). Where particle size distributions are used to calibrate an empirical model, porosity can be considered to be a function of the particle size distribution if it is assumed that a sedimentary body is made up of spherical particles, which are packed in a random order. Typical values of hydraulic conductivity, determined for tailings materials from the Witwatersrand vary between 10^{-6} and 10^{-8} ms^{-1} (Anglo American (1995).

Water movement in the saturated zone is controlled by three factors, i.e. diffusion, advection and dispersion:

- Diffusion is the movement of molecules within a fluid, due to a concentration gradient. It is an artifact of elastic collisions between fluid molecules. Diffusion flux is described by Fick's law, which states $F=D_d(dC/dx)$. F is the flux in mol m^{-2} s^{-1}, D_d is the diffusion coefficient (m^2s^{-1}) and dC/dx is the concentration gradient (mol/m^2).

- Advection is the movement of solutes due to movement of the fluid body through the matrix and is related to Darcy's law.

- Dispersion is the result of mechanical and diffusive forces. Mechanical dispersion occurs due to the mixing of a plume with the water through which it is moving and diffusive dispersion occurs due to the concentration gradient between the plume and surrounding fluids. Dilution occurs in the direction of flow, as well as a spreading of the plume in the direction normal to flow. The sum of mechanical and diffusive dispersion is known as hydrodynamic dispersion.

FRDs display saturated flow characteristics during their operational phase throughout a volume defined by the zone below their piezometric surface. This zone extends below the surface from the edges of the return water pond at the top of the FRD along the hydraulic gradient towards the base and toe of the dump (see Figures 2.2.1(a) and 2.2.1(b) above). Chemical reactions due to acidification are limited in this zone of the FRD and pore water qualities are thus expected to approximate those of penstock return water (Bezuidenhout and Vermaak (2002).

After tailings deposition ceases, accumulated water in the pond slowly seeps into the tailings body or evaporates to the atmosphere. Once the surface dries, the saturated zone slowly recedes below the surface at a rate of >0.3m/year (Stuparyk et al. (1995) or approximately 0.5m per year (Rösner et al. 1997). Morin & Hutt (1997) indicated that the rate of water level decrease is likely to be faster in the wall sections than in the beach sections of the FRD due to the shorter flow path and larger grain sizes in this zone.

---

[1] 10% of the material analysed has a smaller particle size than the D_{10} particle size.

Unsaturated flow processes

Unsaturated flow processes are critical in determining the pollution potential within an FRD, as they are the main mechanism responsible for the transport of contaminants through the system. Within the saturated zone, it is normal for porewater to contain excess lime from metallurgical processes, thus reducing or eliminating AMD impacts from initial pore waters in the saturated zone. It can thus be deduced that AMD impacts on groundwater beneath FRDs are primarily due to recharge from rainwater over unsaturated tailings material or from process water moving through partially saturated zones in the beach and wall sections of the FRD where pyrite oxidation can occur.

Partially saturated zones will begin to generate acidity as soon as air comes into contact with sulphide minerals in the material. Any further water that moves through pore spaces, which contain pyrite weathering residues, will act as a reaction and transport medium for these residues.

Unsaturated flow is complex and is influenced by a number of factors in the field, e.g. mineralogy, particle size distribution, biological activity and formation of preferential flow paths. Two types of preferential flow paths exist in soil materials, which can also be recognised in tailings deposits.

The first type consists of "macropores", e.g. animal burrows, root channels, cracks and fissures caused by swelling clays, partings on bedding planes and features associated with soil structure that cause short circuiting of flow, as well as disturbance through geological activity.

The second type of preferential flow is through the soil matrix as a result of microscopic variations in particle distributions within the matrix that cause water to flow preferentially between some grains and to avoid other flow paths (van Schalkwyk & Vermaak (2000). Field observations indicated that biological activity on FRDs is limited, although some vegetation was noted. This is likely to be due to the low fertility of quartz-rich tailings and the aggressive chemistry of tailings material, which may contain cyanide residues and is also likely to be acidic near the top of the profile. This is where oxidation of pyrite occurs soon after deposition and also occurs most rapidly due to the short diffusion distance to the oxidising pyrite grains. An example of tailings displaying preferential flow paths due to aggregation or jointing is provided in Figure 2.2.1(d).

Figure 2.2.1(d): A surface oxidised zone and near-surface preferential pathways visible through iron staining (Bezuidenhout and Vermaak (2002).

Unsaturated flow estimations do not consider preferential flow paths associated with macropores, but can be calculated. Parameters required for such calculations include: porosity, the soil characteristic curve (soil suction as a function of water content), the saturated hydraulic conductivity and the residual water content (water remaining after a soil has been allowed to drain under gravity). Modelling unsaturated flow in general is considered to be challenging, as a result of the presence of macropores and layering (van Schalkwyk & Vermaak (2000).

Both these features are evident in FRDs to some extent. Vegetation and possibly burrowing are likely to result in macropore formation in the near surface environment. The presence of thin, water-saturated layers was for instance noted in material collected during the fieldwork programme and has been referred to in other studies (Nicholson et al. (1995). Capillary breaks may be formed where small particle size material is placed over larger particle size material, e.g. silt sized material over sand sized material. Water is then held within the matrix of finer material due to capillary forces and is thus prevented from draining to the sub-surface. This results in higher moisture contents in the finer material (Yanful and St. Arnaud (1991).

Similarly to saturated flow processes, unsaturated flow processes follow basic Darcian principles. This means that flow takes place in the direction of decreasing hydraulic potential, the flow rate is proportional to the hydraulic gradient and is also affected by the geometric properties of the pore channels. The relationships are complicated by variable moisture content, soil suction and hysteresis.

Variation in hydraulic conductivity is the main difference between saturated and unsaturated flow. This occurs due to large pores draining faster than smaller pores. Air saturated pores conduct water poorly. Water is thus forced to flow through smaller pores and bypasses the larger pores, causing an increased tortuosity in the flow path and consequently a lower hydraulic conductivity. This factor has been shown to cause a decrease in hydraulic conductivity in unsaturated sands, relative to saturated sands (van Schalkwyk and Vermaak (2000).

In the unsaturated zone the moisture saturation and local water flow path are controlled by a number of factors. Grain size distribution, along with mineralogy will control the tendencies with regard to capillary flow. The capillary zone above the piezometric surface can be subdivided into three zones, i.e. the capillary fringe, the capillary zone and the discontinuous zone (van Schalkwyk & Vermaak (2000) and Rösner et al (1997) after Martin & Koerner (1984a). The capillary fringe is a saturated zone above the piezometric surface, which exists at negative pore pressure. The capillary zone is a partially saturated zone, with smaller pores filled with water and larger pores filled with air. In the discontinuous zone, water is adsorbed onto the surfaces of hygroscopic minerals, especially clays, and requires external force to remove it, e.g. evaporation. Depending on the particle size distribution, a capillary head of variable height will exist above the piezometric surface. Yanful (1991) after Lambe & Whitman (1969) used an empirical relationship to determine the capillary head, i.e. h_c=-0.01(-21.9-85.12(logD_{10})), where h_c is the capillary head and D_{10} is the effective particle size defined as the largest grain size of the finest 10% of the material measured.

Unsaturated hydraulic conductivity is a non-linear function of volumetric water content and is also dependent on the hydraulic head. It is commonly expressed as a function of soil suction (K(θ), pore-water pressure (K(u_w) and/or effective pore radius (K(R).

A large number of empirical methods for determining unsaturated hydraulic conductivity have been developed. The equations derived from these studies commonly make use of particle size distributions and characteristic curves (pore pressure and water retention curves) of porous materials to determine their hydraulic conductivities.

Various authors, as described by Van Schalkwyk and Vermaak (2000) showed that hysteresis (variation in characteristic curves between wetting and drying cycles) has variable significance with respect to its effects in determinations of the hydraulic conductivity of material. It was found that variation due to hysteresis is least for the moisture retention curve (max 300%), compared to the pore pressure curve (max 20 000%), thus leading to the

conclusion that K is best-determined using moisture retention curves, i.e. $K(\theta)$ is more reliable than $K(u_w)$.

Van Genuchten and Wierenga (1977) used Tritium to show that zones of immobile water exist in an unsaturated soil profile. Immobile water masses ranged between 6% and 45% of the total water mass. This indirectly indicates that water follows preferential flow paths in such systems. It was also shown that diffusion occurs between the mobile and immobile water masses and that the mobility of water is inversely related to the flow velocity of water through the pores, thus indicating that isolated pathways may still interact with the flux moving through preferential pathways.

Detailed discussions of unsaturated flow can be found in other Water Research Commission reports and in other geotechnical, geohydrological and fluid dynamics literature. Van Schalkwyk and Vermaak (2000) provide a detailed discussion of saturated and unsaturated flow.

Lifecycle effects on water flow

Both saturated and unsaturated flow processes occur within FRDs and are a function of the lifecycle and morphology of the system, i.e. the stage of development and the spatial position within the FRD. A conceptual time dependent graph, demonstrating the change in seepage rates from an FRD from the operational phase to the closure phase is given in Figure 2.2.1(e).

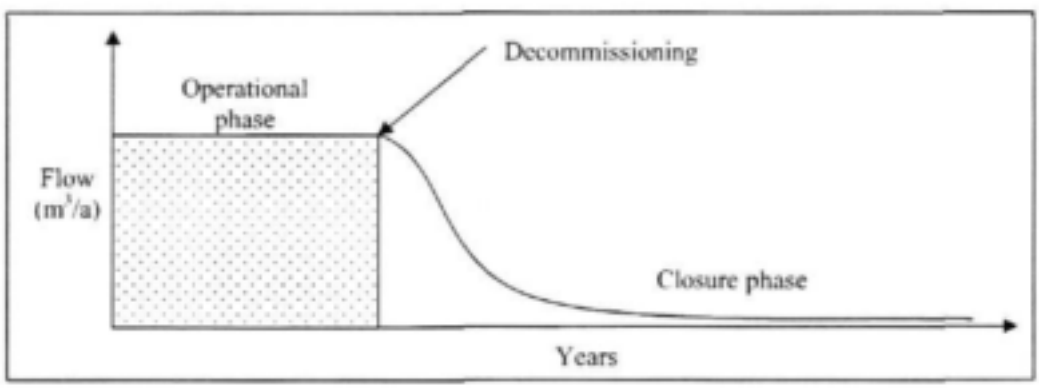

Figure 2.2.1(e): A conceptual graph of seepage rates from an FRD pre- and post-decommissioning.

This graph represents the water flux during the operational phase of an FRD and the subsidence of the piezometric surface after deposition of tailings ceases. As the saturated zone moves lower, the seepage rate decreases due to the decrease in head. Processes such as capillary action and recharge become more important in the water balance during this time. The piezometric surface moves downward at approximately 0.3m to 0.5m per year shortly after closure and progressively slows as the water surface approaches the base of the deposit under a lower head. In the long term the piezometric surface will vary around a

dynamic equilibrium position within the deposit, the level of which will depend on recharge from rainfall and changes in mineralogy and morphology, as well as the hydraulic conductivity of the base materials. Post closure modelling by Bezuidenhout and Vermaak (2002) found that seepage would be generated at a rate of 1% to 6% of Mean Annual Precipitation after the phreatic surface subsides.

In the post closure phase, the unsaturated zone expands into the zone below the pond as air progressively fills some of the void space, previously occupied by process water. In the long term, this zone extends through most of the deposit up to the point where rainfall recharge is able to maintain a dynamic equilibrium or the deposit is completely unsaturated.

Variations in flow in different types of deposits

Variations in the flow regimes of FRDs constructed by different methods may be expected. The small differences between particle sizes of tailings materials of the beach and wall sections on Witwatersrand paddock deposits indicate that variations should not be very large.

Where cycloning achieves preferential separation of coarser grained material, a more pronounced difference in permeabilities may be expected between wall and beach sections of a deposit. Morin & Hutt (1997) noted that cycloned tailings manifested significant particle size differences and even mineral separation tendencies between wall and beach sections of a deposit. It is likely that variations will not be very large in the Witwatersrand paddock deposits, however, due to the relatively uniform particle size distributions, as mentioned in Section 2.1.2.

The effects of particle size variations will cause a steepening of the piezometric surface where they do occur. This will cause the piezometric surface to be convex towards the exterior of a deposit as depicted in Figures 2.2.1(a) and 2.2.1(b). The degree to which this phenomenon will be observed will depend on variations in the permeability of the tailings material, as well as the base of the deposit. Where permeability increases are observed, it is likely that higher oxidation rates will be observed in the wall of the FRD than would otherwise be the case, as the material will have a lower level of moisture saturation and the unsaturated zone is likely to penetrate deeper into the tailings deposit.

2.2.2 Gas transport processes

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 oxygen transport, due to the fine-grained and hence relatively impermeable nature of tailings material. Where preferential pathways exist in the near surface environment, a limited amount of advective flow may occur (James (1997), however, this is not expected to affect the overall oxygen transport rate significantly within an FRD. Throughout the literature

surveyed, diffusion was recognised as the major mechanism for oxygen transport through an FRD, and also serves as the basis upon which tailings oxidation models are based.

Diffusion in tailings is driven by the concentration gradient between the tailings surface and the matrix, caused by weathering reactions that consume oxygen, e.g. the oxidation of pyrite (Yanful (1991). Oxidation is usually confined to a few metres below the surface as a result of oxygen transport limitations due to diffusion (Rösner et al. (2001), with oxygen concentrations becoming progressively lower with depth. Figure 2.2.2(a) shows a profile through fine tailings, which demonstrates a rapid transition between oxidised and unoxidised tailings, related to the movement of an oxidation front.

Figure 2.2.2(a): A contact zone between oxidised and unoxidised tailings.

Oxygen is the most significant gas in determining reactions in tailings material. This is because it is the rate limiting reactant of pyrite oxidation reactions in diffusion-controlled systems, a fact that has been recognised throughout the literature referenced in this study. The ability to determine the oxygen diffusion rate through an FRD over time will thus enable long-term prediction of AMD potential. Along with knowledge of water flow, calculation and/or modelling of pyrite oxidation rates provides the tools for determining pollutant concentrations within and around a tailings deposit over time. The challenge lies in accurate determination of these processes.

Gas diffusion is mathematically described by Fick's first and second laws, which are formulated as follows:

Fick's first law:

$$J = -D\frac{\delta C}{\delta Z} \tag{1}$$

Fick's second law:

$$\frac{\delta C}{\delta t} = D\frac{\delta^2 C}{\delta Z^2} \tag{2}$$

J = flux of oxygen (mol/m²s)
D = diffusion coefficient (m²/s)
C = concentration (mol/m³)
Z = depth in the tailings (m)

It can be seen by inspection of the first law formulation that the gas diffusion rate is dependent on the diffusion coefficient and the gas concentration gradient through the material in question.

Schafer et al. (1997) added a term r_s for oxygen consumption by pyrite in the following equation:

$$\frac{\delta}{\delta t}(aC_g) = \frac{\delta J_g}{\delta Z} + r_s = 0 \tag{3}$$

J_g = Gas flux (mol/m²s)
a = Volumetric air content (m³)
Z= Diffusion distance (m)
r_s = Gas consumption term (mol s^{-1})

The solution for steady state one-dimensional flow for this equation was provided after Jury et al. (1991):

$$C_g(z) = C_o + r_g\frac{r_g}{D_s'}\left(\frac{z^2}{2} + Lz\right) \tag{4}$$

C_g (z) is the concentration at depth z (mol m^{-3})
C_o is the atmospheric gas concentration at depth z=0 (mol m^{-3})
r_g is the gas consumption term (mol s^{-1})
D_g is the diffusion coefficient of the gas in soil (m² s^{-1})
Z is the depth (m)
L is the depth at the depth of zero gas flux (m)

Nicholson et al. (1989) developed the following equation to determine steady state oxygen concentration vs. depth according to Fick's law. This is also the equation used to develop the O_2-diff model on which part of the accompanying spreadsheet model is based.

$$C_z = C_0 \times 10^{-z(k/D_{eff})^{\frac{1}{2}}} \quad (5)$$

C_0 = Start oxygen 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 coefficient in m^2s^{-1}
C_z = Concentration of oxygen at depth Z.

From these equations, it can be seen that the important rate limiting parameters are the diffusion coefficient and the kinetic rate constant. As the diffusion depth increases, oxygen diffusion becomes a more dominant parameter than the inherent reaction rate in determining oxygen flux.

The diffusion coefficient within tailings is highly dependent on the pore space characteristics of the material. Oxygen diffusion into an FRD occurs mostly through gas-filled pore space as opposed to liquid filled pore space (David & Nicholson (1995) and Yanful (1991). This is mostly due to the difference between the diffusion coefficient of oxygen in water ($2x10^{-5}cm^2/s$) vs. the diffusion coefficient of oxygen through air ($0.178cm^2/s$). By implication diffusion rates are therefore affected by both the porosity of the material, as well as the moisture content of the material.

David & Nicholson (1995) determined the oxygen concentration gradient within tailings by using near surface measurements (the top 35cm) to calibrate the value of D_{eff}. To calculate the effective diffusion coefficient under different degrees of saturation, the following equation was used:

$$D_{eff} = -\pi D_a^0 (1-S)^{\alpha} + \tau \frac{D_w^0}{H} \quad (6)$$

Where D_a^0 is the coefficient of diffusion for oxygen in air and D_w^0 is the diffusion coefficient for oxygen in water as discussed above, S is the degree of water saturation, H is the modified Henry's constant (H=26.32 at 10°C) and τ and α are fitting parameters, which were determined by non-linear regression of laboratory column data (after van Genuchten (1987). The values for τ and α were calcualted at 0.27 and 3.27 respectively.

Stuparyk et al. (1995) used a modified version of a semi-empirical equation after Reardon and Moddle (1985) to determine the oxygen diffusion coefficient. Yanful (1991) calculated the Effective diffusion coefficient using empirical methods and also performed measurements. Troeh et al. (1982) and Millington & Shearer (1971) also developed equations to determine D_{eff}. The equations are all primarily functions of gas filled porosity.

Numerous studies have been conducted, which make use of this knowledge to quantify the sulphide oxidation rates within tailings. David & Nicholson (1995) performed calculations on pyrrhotite oxidation, using a gas concentration oxygen flux method.

This method was found to be sensitive to changes in the permeability of the tailings profile due to variation in the tailings properties, such as alternating coarse and fine-grained layers. The problem was treated similarly to the application of Darcy's law for hydraulic conductivity, using the harmonic mean value of the diffusion coefficient for calculations (after Freeze & Cherry (1979). It was found that rates were overestimated using this method due to the difficulty of accurately measuring the water saturation of the tailings material on a local scale.

Nicholson et al. (1995) noted that oxygen diffusion will be retarded by finer grained layers relative to coarser grained layers due to the higher moisture saturation in the fine-grained material. This is likely to occur where capillary breaks form, as discussed under unsaturated flow processes in Section 2.2.1.

Porosities may be variable in aggregated materials, such as aggregated soils. This phenomenon may also be seen in tailings, as illustrated with the formation of mud cracks and iron staining along preferential pathways within tailings below the surface (See Figure 2.2.1(d). Porosities between aggregates will be higher than within matrix material.

Nicholson et al. (1995) measured oxidation rates between the walls and centre sections of an FRD and found that gas diffusivities varied by two orders of magnitude. This was attributed to the reduction in sulphide content and the lower permeability of the finer material at the pool area, relative to the beach area. It should be noted, however, that similar tendencies will not necessarily occur in Witwatersrand FRDs as mineralogy and particle size distribution differences are likely to be significant between the study sites discussed by Nicholson et al.(1995) and Witwatersrand FRDs. David and Nicholson (1995) described fine grained layers within an FRD profile, which had higher moisture contents than layers above and below. Low gas flow measurements were attributed to the effects of these layers on the diffusion coefficient of bulk tailings (see Sections 2.1.2 and 2.1.3).

Søgaard Andersen et al. (2001) showed that during carbonate reaction with pyrite oxidation products from marine sediments, a gas volume reduction occurs due to the 1:2 molar ratio of CO_2 released relative to O_2 consumption. This mechanism may play a part in gas transport through tailings material, however, no measurements of this phenomenon are known.

Methods used for assessment of oxidation rates in FRDs

Nicholson et al. (1995) describe a method by which oxygen consumption within the tailings profile can be measured directly. It entails the measurement of oxygen concentration change in the headspace of a thin walled Aluminium cylinder inserted into the tailings. A cap with sensor equipment is placed on the open cylinder and the change in oxygen

concentration over time is measured. It was found that oxygen consumption rates varied between 250mol O_2 $m^{-2}yr^{-1}$ in the areas that were located above the water table to 0.1mol O_2 $m^{-2}yr^{-1}$ closer to the FRD pond area. It was also found that oxygen consumption rates and sulphate production rates were stoichiometric, according to pyrite oxidation equations, except at lower oxidation rates, where it is believed incomplete oxidation resulted in lower sulphate concentrations. Nicholson et al. (2000) and Bussiére and Aubertin (1999) used the oxygen consumption method for studies of liming requirements and cover performance respectively.

Timms and Bennet (2000) discuss a device that also performs headspace measurements to calculate oxygen consumption. This device is more sophisticated and can be used on the surface of an FRD due to a pressure equilibration mechanism that is included on the device, to prevent inflow of gas from the outside of the cylinder to the inside as a result of the formation of pressure gradients. This device was constructed from an old 44 gallon drum and was driven into the tailings to a depth of 1cm with a 3cm headspace for measurements.

The method used by David and Nicholson (1995) and. de Vos (pers. Comm.) to measure oxygen concentration changes entails measurement of the oxygen concentration profile within the tailings to calibrate an oxidation model. This method uses a thin stainless steel tube that is driven progressively deeper into the tailings to determine the oxygen concentrations at different depths. A depth dependent oxygen profile is thus measured. This method is useful, in that it is capable of defining the pseudo-steady state oxygen concentration profile within an FRD. Oxygen diffusion models, such as the model by Davis & Ritchie (1986) or van Niekerk (1997) use the oxygen concentration profile to calibrate the momentary pyrite oxidation rate as well as the model time axis, which is not possible when using headspace measurements.

David and Nicholson (1995) concluded that measurements of the oxygen concentration gradient cause overestimation of the oxygen flux. Two reasons were given for this:

- The oxygen gradient or flux method determines a "snapshot" value, which if determined in the dry season will be higher than the average value.

- Thin fine-grained layers within an FRD, which have higher moisture contents than average, control oxygen flux due to their low permeability. Measurements of average moisture contents thus do not represent the field conditions adequately.

David and Nicholson (1995) also used a geochemical mass balance approach to determine historical weathering in the same FRD on which oxygen measurements were performed. Weathered pyrite mass was calculated by integrating the dissolved sulphate and iron masses across the weathered profile in 5cm core sections. It was found that secondary minerals play an important part in these determinations and will tend to cause an underestimation of weathering products due to precipitation reactions. It was found that areas affected by high oxidation rates would be better represented by sulphate concentrations, as sulphide

oxidation was likely to be the major source of sulphate under these conditions. Iron was found to be a better estimator at lower concentrations; however, it was acknowledged that mineral precipitation was likely to cause lower concentrations to be recorded than were actually released by pyrite oxidation. The calculated oxidation rates obtained from the oxygen concentration method and the geochemical assessment method varied by two orders of magnitude due to the effects of under- or overestimation.

Changes in gas transport during the lifecycle of an FRD

As discussed above, oxygen transport will be the rate-limiting factor in pyrite oxidation reactions in FRDs, especially once the near surface layer of the tailings body has been oxidised. It was also shown by various authors that the diffusion rate of oxygen is determined by the air-filled porosity of a tailings material as a result of the fact that oxygen diffusion through air is four orders of magnitude faster than through water.

This observation indicates that oxidation will occur primarily in the unsaturated zone of a tailings deposit throughout its life. This implies the wall and beach sections during the operational phase, as unsaturated conditions dominate in these zones. During the operational phase, the pond section of an FRD will be primarily a saturated zone, with desiccation being unlikely during this period. Tailings that occur below the piezometric surface can thus be expected to remain essentially unoxidised during the construction phase of the FRD.

Materials that occur above the piezometric surface are at least partially exposed to oxygen, with the possible exception of a part of the capillary zone. These materials will thus begin to oxidise during deposition and will contribute to acid generation at an early stage in the life of the tailings deposit.

Observations and models show that oxidation rates tend to be rapid where the diffusion distance to a pyrite grain is short. It is thus likely that a significant proportion of pyrite will be oxidised in the near-surface unsaturated zone as successive layers are allowed to dry prior to deposition of the next layer. Yellow and orange staining observed in Witwatersrand FRDs at different levels within their stratigraphy attests to this.

The vertical exposure of pyritic material on the outside of an FRD wall implies that oxidation will occur from the onset of the depositional process over the entire height of the profile. The depth of the oxidation front will define the affected zone. This effect will be further pronounced, if the wall material is significantly coarser than the beach materials within the deposit. Due to the high AP:NP relationship generally observed in Witwatersrand tailings, this will result in acid seepage from the wall of a dump early on in its lifecycle. Vertical flow of process water from paddock areas may have some mitigating effect in terms of maintaining higher saturation levels and providing alkalinity during the depositional phase. The downslope, unsaturated, parts of the wall section will, however, be exposed to weathering.

After decommissioning, as the piezometric surface subsides, unsaturated pore space will increase. This will immediately provide additional pathways for diffusion of oxygen into the tailings profile. Observations, modelling and literature, however, indicate that oxidation of pyrite in the body of the FRD is only likely to be rapid for the first few years after deposition ceases. This is because pyrite that occurs at the surface of the FRD will be rapidly oxidised, due to the short diffusion distance and lower moisture saturation in the evaporative zone. After this initial period, pyrite oxidation rates will drop rapidly behind a progressively more slowly moving oxidation front. The rate of progress of this oxidation front will be dependent on a number of factors, including the diffusion distance, the pyrite concentration the moisture content, particle size and porosity of the tailings material. Factors such as temperature will

also play a role. A schematic diagram of the zones affected by oxidation is depicted in Figure 2.2.2(b) and a conceptual graph, depicting the likely evolution of pyrite oxidation rates, as discussed above, within an FRD is provided in Figure 2.2.2(c).

Figure 2.2.2(b) shows the morphology of an FRD in cross-section. The outer parts of the wall section and the beach zone are oxidised near the surface. Below the oxidised zone, however, an unsaturated zone exists, which is essentially unoxidised.

Figure 2.2.2(c) depicts a conceptual oxygen flux evolution curve over the lifecycle of an FRD. During the operational phase, oxygen flux remains essentially constant due to the continued addition of new material. After decommissioning, a slight increase in oxygen consumption is expected as the centre of the FRD becomes unsaturated and allows oxygen ingress to occur. After a few years, the upper zone of the FRD will have been depleted of pyrite and pyrite armouring will occur throughout the oxidised zone, causing oxidation rates to fall progressively over time. Due to the lower oxidation rate, pyrite consumption will decrease and the rate of change in the oxidation rate will thus also decrease.

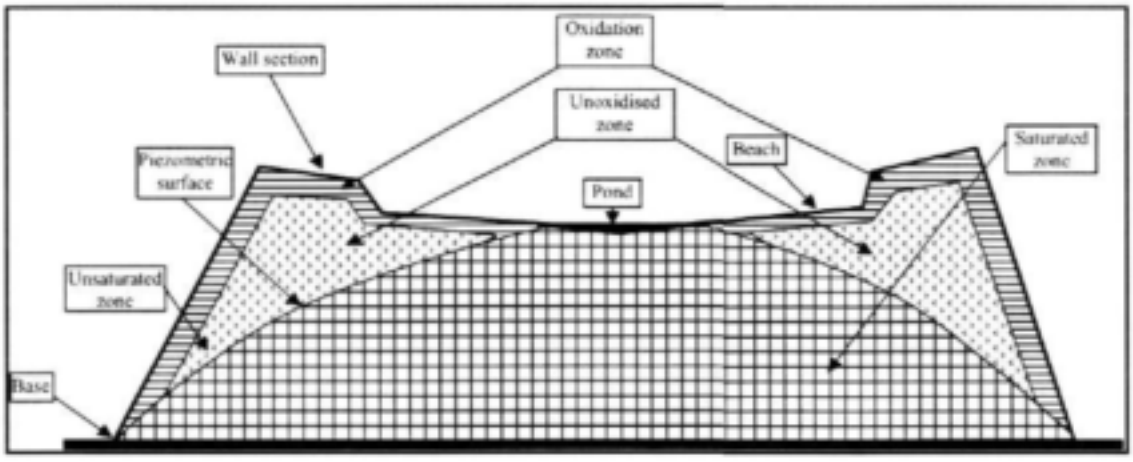

Figure 2.2.2(b): A schematic diagram depicting the oxidation profile during the operational phase of an FRD.

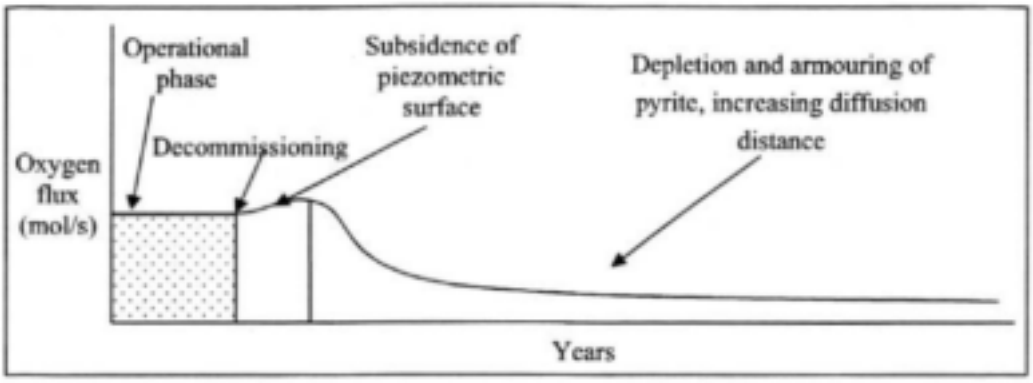

Figure 2.2.2(c): A conceptual diagram, depicting the total oxygen flux (pyrite mass oxidised) within an FRD as a function of its lifecycle.

Effects of deposit type on oxygen diffusion

Oxygen diffusion in an FRD may be affected to some extent by the deposition methods used as a result of temporal and physical considerations.

Variations in particle sizes or mineralogy due to variable degrees of density separation between paddock type deposits and cycloned deposits have the potential to influence oxygen flux rates into these deposits. This will be manifested if significant mineral separation occurs, especially with respect to pyrite concentrations or if significant particle size separation occurs, thus affecting hydraulic conductivities and diffusion coefficients.

Variable moisture content resulting from differences in deposition mechanisms or permeability of the materials may play a role in controlling oxygen flux. For example, rapid consolidation of cycloned wall materials is likely to reduce the periodicity and significance of oxidised layers within a tailings deposit due to the short waiting periods required between depositional events. Variations in moisture saturation levels, especially in the wall sections, are also likely to occur as a result of the differences between depositional methods.

2.3 Geochemical processes

2.3.1 Mineral reactions

The geochemical makeup of tailings material is the determining factor with regard to mobilisation of pollutants. As was mentioned in Section 2.1.1, the mineralogy of the Witwatersrand gold tailings is constrained within a relatively narrow compositional range, consisting predominantly of quartz and other minerals of low reactivity. A minor proportion of the tailings consist of minerals which will contribute to AP. Some of the silicate minerals that are likely to react in an acidic environment are likely to contribute to NP in the system. Furthermore, the AP is generally higher than the NP within these tailings, thus implying that all Witwatersrand tailings will at some stage become acidic, once all NP has been consumed.

Mineral reactions in the Witwatersrand tailings material are dominated by the oxidation of pyrite, with other sulphides not being present in significant quantities (Rösner et al. (2001).

The reactions of pyrite under various conditions are documented by Perkins et al. (1995) as follows:

$$FeS_2 + \frac{7}{2}O_2 + H_2O \Leftrightarrow Fe^{2+} + 2H^+ + 2SO_4^{2-} \quad (1)$$

$$2Fe^{2+} + \tfrac{1}{2}O_2 + 2H^+ \Leftrightarrow 2Fe^{3+} + H_2O \quad (2)$$

$$Fe^{3+} + 3H_2O \Leftrightarrow Fe(OH)_3 + 3H^+ \quad (3)$$

$$FeS_2 + \tfrac{15}{4}O_2 + \tfrac{7}{2}H_2O \Leftrightarrow Fe(OH)_3 + 2H_2SO_4 \quad (4)$$

Reaction (1) occurs in environments of intermediate oxidation potentials, where the sulphate ion is the dominant species and Fe^{2+} is the dominant Fe ion, rather than Fe^{3+}. At higher oxidation potentials Fe^{2+} oxidises to Fe^{3+}, as depicted by equation (2). Where the pH of the solution is high enough, Fe precipitates as $Fe(OH)_3$ as depicted by equation (3). The sum of all these reactions is given in equation (4).

Minerals are classified into five groups based on their dissolution kinetics in a normal weathering environment Perkins et al. (1995) after Sverdrup and Warfvinge (1988). The most rapidly dissolving group (with rate constants between 10^{-5} to 10^{-6} mol/m^{2}s) are the carbonate minerals (i.e. aragonite, calcite, dolomite, magnesite) and brucite. The silicates are split into four weathering groups:

- Fast, with rate constants between 10^{-9} to $10^{-10.5}$ mol/m^{2}s (i.e. anorthite, nepheline, jadeite, leucite, spodumene, pyroxenes such as forsterite, diopside, wollastonite, hedenbergite, bronzite, amphiboles such as hornblende and glaucophane, chlorites, garnets and epidotes).

- Intermediate, with rate constants between $10^{-10.5}$ to 10^{-12} mol/m^{2}s (i.e. pyroxenes such as enstatite, hypersthene, and augite, amphiboles such as tremolite, actinolite and anthophyllite, and serpentines such as chrysotile and talc).

- Slow, with rate constants between 10^{-12} to 10^{-13} mol/m^{2}s (i.e. feldspars such as albite, orthoclase, and plagioclase, clay minerals like gibbsite and kaolinite, and micas such as muscovite and biotite).

- Non-reactive or inert minerals have the smallest rate constants (10^{-14} to 10^{-16} mol m^{-2}s^{-1}). Quartz, rutile and zircon are included in this group.

From these results, it can be seen that the mineralogy of the Witwatersrand is relatively stable, except for the presence of pyrite, which will rapidly react with oxygen. Acid Base Accounting (ABA) and mineralogical analyses of these tailings have shown that AP exceeds NP (Rösner et al. (2001), Pulles, Howard and de Lange (2002) and Section 3.3.3 of this report).

Neutralisation reactions in Witwatersrand tailings will be limited by the abundance of quartz in the system. Quartz is classified as an unreactive mineral (Perkins et al.(1995). Perkins et al. (1995), however, also demonstrated that silicates, e.g. Mg-chlorite could provide significant neutralisation potential. The case study in question demonstrated that Mg in solution was derived from Mg-chlorite in the relevant study sample, rather than dolomite which was also present at 5% of the abundance of the Mg-chlorite. In situations where residence times are long, such as in FRDs, such silicate minerals may provide significant neutralisation potential where they make up a significant portion of the material composition. Usher et al. (2001) demonstrated that some silicate minerals potentially have a higher buffer capacity than calcite. In these cases reaction rates will be the limiting factors in determining whether NP will be realised in buffering acidic porewater solutions.

In the study by Rösner et al. (2001), muscovite and chlorite were identified as significant constituents of gold tailings based on field data. These minerals may thus be capable of neutralisating some of the acid produced by pyrite oxidation. It is, however, not stated whether the chlorite is Fe or Mg-rich. Considering the geological environment in the Witwatersrand, is likely that the chlorite will be Fe rich and that neutralisation ability will thus be limited. This is because Fe-rich chlorite will not provide alkalinity to the same extent as Mg-chlorite. Muscovite will on the other hand react slowly, as indicated by the reactivity classification in Perkins et al. (1995). Of the other minerals identified in the Witwatersrand tailings material, jarosite forms as a secondary mineral after pyrite oxidation and is known to release acidity on decomposition. Pyrophyllite, a simple aluminosilicate mineral, is not expected to be reactive and will not possess significant buffering capacity, as it does not contain alkali cations.

A general rule, which should always be considered and is illustrated in Perkins et al. (1995) is that the extent to which minerals will neutralise acids cannot be predicted without knowing what secondary minerals will form. Minerals which are used as examples, include siderite and ferrihydrite, formation after neutralisation with calcite. Siderite formation increases the amount of calcite required for neutralisation by 50% due to release of H^+ during siderite precipitation. Similarly, formation of ferrihydrite after calcite reaction doubles the amount of calcite required to affect neutralisation of pyrite oxidation products due to release of H^+ by hydrolysis of Fe^{3+}.

In this review, surface area effects of reactions will not be considered in detail. It is, however, important to note that mineral reactive surface area is an important parameter in determining the reaction rates of minerals. The exposed surface area of a pyrite grain can be affected by factors such as morphology and secondary mineral precipitation. Framboidal pyrite is for instance more reactive than euhedral pyrite due primarily to a larger effective

surface area, but also because of the presence of more reactive sites on the mineral surface.

Determination of these influences are complicated, as surface area measurements are not necessarily correlated with mineral abundance and are not necessarily represented by the Brunauer-Emmett-Teller (BET) method for surface area determinations (Perkins et al. (1995). The formation of secondary mineral layers on oxidising pyrite surfaces can impede the transport of oxygen to the pyrite surface and transport of reaction products away from the reactive surface. These considerations, along with small-scale heterogeneities in the morphology of a tailings deposit, make it difficult to determine mineral reaction rates within a tailings deposit, as experienced by Nicholson et al (1995). It is thus likely that mineral reaction rates will be better handled by empirical means than by theoretical calculations.

2.3.2 Mineral reaction kinetics

The kinetics of pyrite oxidation are largely controlled by oxygen diffusion within an FRD, as was discussed in the section on gas transport above. The model most commonly used is the double diffusion model, making use of the shrinking core model for pyrite consumption, combined with diffusion through the pore spaces of a tailings body (Davis and Ritchie (1986). This model has been practically implemented in the Pyrox model Wunderly et al. (1996) and the Generic simulation model for Opencast mine water systems van Niekerk (1997).

The pyrite reaction rate is calculated as being controlled by diffusion through a layer of reacted material, which grows as pyrite is oxidised. Oxygen is required to diffuse through the oxidised layer to reach the unreacted core, thus leading to progressively slower reaction rates. In the Pyrox model, the diffusion coefficient (D_2) through the reacted, oxidised, core has been calibrated to approximate 7.8×10^{-12} m^2s^{-1}, using empirical and laboratory tests (PTI Environmental Services (1995).

It is, however, also important to consider the presence of bacteria such as *Thiobacillus ferrooxidans*, which can increase the reaction rate of pyrite by two to three orders of magnitude.

Pugh et al. (1984) found that *Thiobacillus ferrooxidans* was inhibited in environments where oxygen was present at less than 8%. The results of their study, as provided in Perkins et al. (1995) are given in Table 2.3.2(a) below. Other interactions of pyrite in solution, besides bacterial activity, that are mentioned in the summary by Perkins et al. (1995) and other sources, include indirect oxidation of pyrite by ferric iron and electrochemical effects, neither of which are definitively quantified with respect to their contribution to pyrite oxidation rates.

Rate reactions and observations, as provided by Perkins et al. (1995) after Nicholson et al. (1988 and 1990) are given below. These experiments were done in the range (0.6-20% O_2), pyrite particle size (54-215 µm), temperature (25-60°C) and pH (6.7-8.5)

- Reaction rate varied inversely with grain size diameter to the first order, but approached inverse square rates with increasing time.
- Activation energy (described as pseudo activation energy) was determined at 88kJ/mol.
- The shrinking core model provided a surface rate constant of 3.07×10^{-6} mh^{-1} ± 46% and a diffusion coefficient for oxygen of 1.08×10^{-12} m^2h^{-1}.

TABLE 2.3.2(A): PYRITE REACTION RATES FOR DIFFERENT SPECIES AT DIFFERENT OXYGEN CONCENTRATIONS (AFTER PERKINS ET AL. (1995).

Oxygen %	*Slope	*r^2	*k	Relative rate
Massive Pyrite				
0	-0.006	0.32	-0.013	1.0
8	-0.018	0.98	0.087	10.0
20	0.048	0.97	0.111	12.1
Frambodial Pyrite				
0	0.007	0.26	0.017	3.0
8	0.054	0.97	0.125	13.8
20	0.069	0.96	0.158	17.1
Marcasite				
0	0.031	0.34	0.070	8.3
8	0.094	0.85	0.215	22.8
20	0.094	0.77	0.215	22.8

***Slope** refers to the slope of the fitted curve to rate data for each experiment, r^2 refers to the correlation coefficient for the same curve, **k** is the theoretical first order rate constant, as determined from the kinetic data and **Relative rate** is a measure to compare different rates of oxidation under different conditions.

Tibble & Nicholson (1997) applied the oxygen consumption method to determine the oxygen consumption rates on various tailings deposits. The results obtained for pyritic tailings material are given in Table 2.3.2(b) below.

TABLE 2.3.2(B): SULPHIDE OXIDATION RATES MEASURED WITH THE OXYGEN CONSUMPTION METHOD BY TIBBLE & NICHOLSON (1997).

Scenario investigated (Sulphide mineral)	Characteristic variable	Oxygen consumption as O_2 (mol m^{-2} a^{-1})		Max. Potential Equivalent $CaCO_3$ kg $m^{-2}a^{-1}$ (Fe:Ca = 1:1)
		Range	Average	
Exposure Time (25% pyrite)	Fresh (<2years	2-67	33	4
	2-4 years	5-1617	273	29
	5-6 years	1-518	192	21
	Approx. 10 years	17-328	81	9
Simple cover (8% pyrite)	Exposed	82-105	105	11
	Covered	<1-33	10	1
Engineered Cover (6% pyrite)	Exposed	228-836	517	55
	Covered	<1-30	7	1

2.3.3 Secondary mineral formation

The formation of secondary minerals is an important factor in determining the ion balance within a pore water solution, as secondary mineral formation may influence the acidity of the porewater solution by buffering, dissolution or precipitation. In sulphidic systems, this commonly occurs with minerals containing iron. An example of this can be seen where Fe^{3+} forms as a secondary step in pyrite oxidation to form Ferrihydrite in a sufficiently alkaline solution. This results in a concomitant release of acidity according to the equation:

$$Fe^{3+} + 3H_2O \Leftrightarrow Fe(OH)_3 + 3H^+$$

Similarly, after neutralisation of acidity by the alkalinity in $FeCO_3$, the Fe^{2+} component can oxidise to Fe^{3+}, resulting in a zero nett change in the proton balance (Perkins et al. (1995). Mayer et al. (2000) showed buffering between secondary minerals and acid solutions.

Jarosite ($KFe_3(SO_4)_2(OH)_6$) and its aluminous analogues are minerals that commonly form in Acid Mine Drainage (AMD) environments. These minerals are referred to as acid minerals, as they capture acidity, which is only released on decomposition of the mineral. Acidity can be stored in the system for an extended period in this form.

3 FIELDWORK

3.1 Objectives

The objectives of the fieldwork conducted as part of this research were aimed at constraining the physico-chemical parameters within FRDs that are likely to affect the depth and rate of weathering. Sampling was not focussed on an array of Witwatersrand FRDs, but rather focussed on one FRD where historical geochemical and physical data were available. Such an FRD was kindly provided by AngloGold and all geochemical data generated for AngloGold on this FRD complex was made available for the research. Initial physico-chemical data were generated by Pulles, Howard and de Lange for AngloGold during 2002.

Fieldwork for this study focussed on confirming previously reported parameters, as well as collection of specifically relevant data pertaining to physico-chemical characteristics identified as relevant in the literature.

Sampling positions for both studies are indicated in Figure 3(a) and laboratory results are included in Appendix A.

Figure 3(a): Sampling positions at the main study site.

3.2 Methodology

For this study pyrite oxidation history is investigated primarily as a function of oxygen diffusion, as opposed to reconstruction of the oxidation history from geochemical composition of pore waters. As oxygen diffusion is the primary driver of pyrite oxidation in an FRD and geochemical analyses are expensive and are not likely to be more reliable than oxygen flux determinations, it was decided to follow this approach. Additionally, it was decided that a conservative approach would be followed, which would not permit underestimation of oxidation rates. David and Nicholson (1995) indicated that oxidation profile measurements were likely to cause overestimation of oxidation rates as fine grained, higher moisture content layers are not taken into account when measuring moisture contents. Geochemical assessments on the other hand would be more inclined to cause underestimation of oxidation rates as mineral solubility constraints play a significant role in pore water concentrations.

The disadvantage of using the oxygen flux method, is that it provides only a snapshot of the oxidation conditions within a tailings body and thus requires numerous readings over time to provide an accurate assessment.

Fieldwork was aimed at determining what physico-chemical phenomena are likely to impact on oxidation rates within an FRD, as well as to constrain the boundaries of the oxidised zone if possible. During July 2003, ten sites were sampled with a bucket auger within the residue deposit that was studied, with hole depths varying between 3.2m and 6.80m. Most holes were approximately 5m deep. Seventeen sub-samples were collected from the ten holes that were made. A number of photos were also taken to illustrate the features observed, some of which are included in this report. Particle size analyses, X-Ray Diffraction (XRD) analyses and shake flask tests were performed on selected samples. Descriptions were also recorded for all samples with regard to colour trends and any other noteworthy characteristics. Precise colours were not recorded, as it was decided that this would not add value, soil scientists having found that colour is not a reliable indicator of conditions within a soil profile (pers. comm. C. Viljoen). Colours were rather described as they might have a bearing on differences in oxidation state within a profile.

3.3 Observations and data

3.3.1 Surface features

A number of surface features were observed, which are likely to influence the weathering processes and hence AMD production on gold FRDs.

Plant growth

Plant growth was significant in certain parts of the FRD, particularly near the penstock where reeds grew in dense clumps. Some grass also occurred, mostly near the centre of the

deposit. The grass was mostly concentrated in mud cracks (see Figure 3.3.1(a), probably due to higher nutrient and moisture contents in the fine-grained filling material within the cracks. Bushes also grew in significant numbers over virtually the whole surface of the FRD. It is likely that some macropore formation would occur as a result of this plant growth, with resultant channelling of oxygen and water near the surface of the FRD. The introduction of organic content is also likely to consume oxygen in the sediment profile to some extent through decomposition processes.

Figure 3.3.1(a): Mud cracks hosting a grass community on an FRD, as well as other plant mass.

Mud cracks

Mud cracks were present in significant numbers. Cross sections were excavated through the cracks, which were found to be approximately 10 to 20cm deep from visual inspection. These features can be expected to increase oxygen ingress to some extent in the near surface where they occur, however, the limited space occupied by these features, compared to the volume of tailings is likely to limit their influence.

Surface texture

Surface texture varied between different parts of the deposit. Wall materials appeared slightly coarser and materials near the centre of the deposit finer (see Figure 3.3.3(a). Areas which clearly held standing water at some stage near the centre of the FRD also displayed significant signs of clay deposition in the form of a cracked, laminated, surface skin.

3.3.2 Subsurface features

Within the tailings body, a number of features, relevant to the pyrite weathering process were observed:

Leached horizon

In all tailings profiles, an apparent leached horizon, approximately 5 to 10cm thick, occurred directly below the surface. This is likely due to the rapid oxidation of pyrite near the surface of the deposit and rainfall infiltration washing weathering products deeper into the tailings body over time. Leaching is likely to be promoted by the availability of surplus acid after depletion of NP near the surface. Rainfall is also likely to move through this zone on a regular basis, prior to being evaporated again during drier periods, thus rinsing soluble salts into the zone below.

Hardpan horizon

An iron stained (hardpan) zone occurs below the leached zone, with iron tending to be more enriched with depth, until the "unoxidised" zone is reached. Some evidence of preferential flow paths was observed, where jointing was highlighted by planes stained with iron precipitates (see Figure 2.2.1(d).

A tendency towards mottling was also observed, which is an indication of differential oxygen concentrations within the sediment mass caused by variable moisture contents due to variable permeability. Mottling was especially evident in the coarser fractions. It is expected that this is due to a more dynamic moisture profile in these materials, i.e. moisture contents are more variable, as transport occurs more easily.

Position of the oxidation front

A distinct transition from iron-stained to grey or mottled conditions was observed. The transition is usually rapid (of the order of a few millimeters or centimetres depending on particle size) and distinct. This zone depicts the transition between the redox stability of Fe^{2+} and Fe^{3+}. From the literature and modelling excercises, it is evident that this does not necessarily represent the oxidation front per se.

Yellow to grey transitions are commonly observed in soil profiles, associated with the alteration of Fe^{3+} to Fe^{2+}. Holmes (2000) summarised the relationship for lake sediments, insofar as redox potential is related to pH and O_2 concentration. It was found that redox decreased by 58 mV per 1 pH unit increase; reduction in O_2 saturation from 100% to 10% decreased redox by 30 mV and the transition from Fe^{2+} to Fe^{3+} was found to occur at 200mV. As a relationship exists between redox, pH and O_2 concentration, this could be applied in determining oxygen concentration gradients in fresh tailings.

Theoretical discussions of these reactions (Appelo & Postma (1993) and van Niekerk (1997), however, indicate that redox is reflected in the Fe^{2+}/Fe^{3+} ratio by the equation $Eh=E^0+(RT/nF)\ln\{(Fe^{3+}/Fe^{2+})\} = 0.77+0.059\log\{(Fe^{3+})/(Fe^{2+})\}$. There is thus not a single point of transition and it can therefore not be assumed that the oxygen concentration can be directly measured by using the depth of iron staining within the tailings mass. The rapid transitions observed in the field, however, indicate that the colour changes observed are likely to occur within a well-constrained redox envelope.

Secondary sulphide formation

Occasional signs of sulphide formation exist within the unoxidised zone, indicating the dynamic nature of the system in question and suggesting that the anoxic zone is closely related to the zone of colour change. Black streaking within joints is indicative of the formation of greigite or framboidal pyrite within the tailings.

Exposing a sample of the material to oxygen highlights the reactive nature of the fine sulphide grains within the tailings. After two days of exposure, a distinct yellowing of the sediment is observed in the outer parts of the sample. It is possible that secondary sulphide formation thus plays a role in the redox dynamics of an FRD.

Moist layers, fine-grained layers

Within the tailings body, moisture rich layers, with unpredictable spacing exist. Three samples of such moist layers were recovered from auger samples and submitted for analysis. The samples were 2A, 5C and 6B.

Particle size distribution graphs for sub-samples from sample sites 2, 5 and 6 are shown in Figures 3.3.3(b) below and indicate that the moist layers show distinctly finer grain sizes than the matrix in which they occur.

These layers were probably deposited during periods of stagnancy in the settlement pond. The relatively high moisture content of these layers indicates the formation of capillary breaks within the tailings material.

In some samples, up to three moist layers were observed within a 5 metre auger profile, however, these layers were generally not observed in the top two metres of the deposit. This

is likely to be due to evaporative effects having been significant in the upper part of the deposit, especially during the dry season when the sampling was done.

The absence of moist layers is thus not necessarily due to the absence of fine-grained layers. Fine layers will be difficult to detect and separate if they are not moist, as moisture provides a means of cohesion and distinction.

The presence of layering has been recorded in the literature, along with its effects on the permeability within tailings (Rösner et al. (2001). A feature of this layering is seen in the presence of capillary breaks, which are responsible for the distinct, fine-grained, moist layers discussed above. Significant layering effects can, however, also be observed where cross sections of dumps can be observed, e.g. where erosion has taken place on the side of a dump, or where old dumps are being reclaimed (see Figure 2.1.1(a).

Syndepositional oxidation of pyrite

Yellow stained layers in cross sections of FRDs are an indication of some syndepositional oxidation of the pyrite mass. Modelling indicates that pyrite oxidation rates are high in near surface tailings, with total oxidation of the top meter of pyrite within a few months in relatively dry tailings. It is thus likely that partially saturated tailings (beach and wall material) will undergo a significant amount of oxidation in the time between depositional events, with the reaction rate being determined by the pyrite concentration and oxygen diffusion through the system.

3.3.3 Laboratory analyses

Acid Base Accounting (ABA) analyses.

ABA analyses indicated that the AP of the tailings material was higher than the NP in six of the nine samples analysed, thus indicating a long term probability of becoming acidic. The highest AP:NP ratio recorded was 1:0.5 and the lowest was 1:2.78. Measured sulphur concentrations varied between 0.17% and 0.45 % sulphur, thus placing the tailings within the uncertain range, with respect to their potential to cause Acid Mine Drainage, when compared to standard criteria.

Particle size analyses by hydrometer method for selected samples.

Particle size analyses were performed on all sub-samples collected. Inspection of the analyses indicates that the intra sample variation is higher than that between the beach and pond areas of the FRD. This is confirmation of a relatively uniform grading between the beach and pond areas of the FRD (Anglo American (1995). The major proportion of the tailings material falls within the silt size fraction, with sand and clay making up minor proportions, sand being more dominant than clay. One sample (2B) contains predominantly sand size particles. This is, however, an exception. The results for the particle size analyses are

graphically presented in Figures 3.3.3(a) and 3.3.3(b) below, with Figure 3.3.3(b) illustrating the effect of particle size on moisture content.

X-ray diffraction analyses for mineral composition.

X-ray diffraction (XRD) analyses were performed on all samples, to ascertain their mineral composition. It was found that quartz made up between 55% and 84% of sample composition, with chlorite and mica being the other significant minerals within the system. Minor fractions of other silicate minerals were also found to be present, generally making up less than 10% of the total composition. These analyses agree with those quoted by Rösner et al. (2001). Some of the minerals, e.g. plagioclase, chlorite and amphibole, are likely to provide NP to the system. No pyrite was detected and calcite was found to be present in amounts of 1% or less. Goethite was found to be present in some samples, with a maximum of 2% in sample 7B.

It is likely that pyrite was present even though it was not detected due to XRD being insensitive at the concentrations present. ABA analyses show that there is sufficient acidity available in the material for pyrite to be present. It is not certain to what extent available pyrite may already have oxidised in the samples collected. In samples collected within the oxidised zone, it is especially likely that some pyrite would already have reacted with oxygen. Modelling indicates that the oxidation front can penetrate relatively deeply into a tailings deposit, thus suggesting that partial oxidation of pyrite may have occurred, even in samples collected 5m deep, depending on the physico-chemical environment.

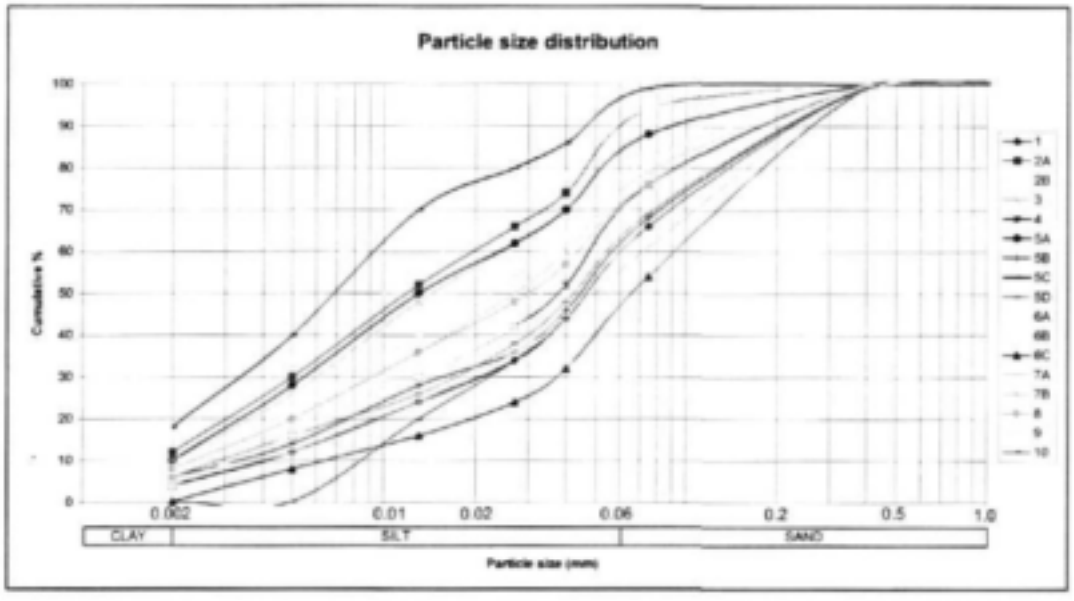

Figure 3.3.3(a): Particle size analyses of all sub-samples collected at the study site.

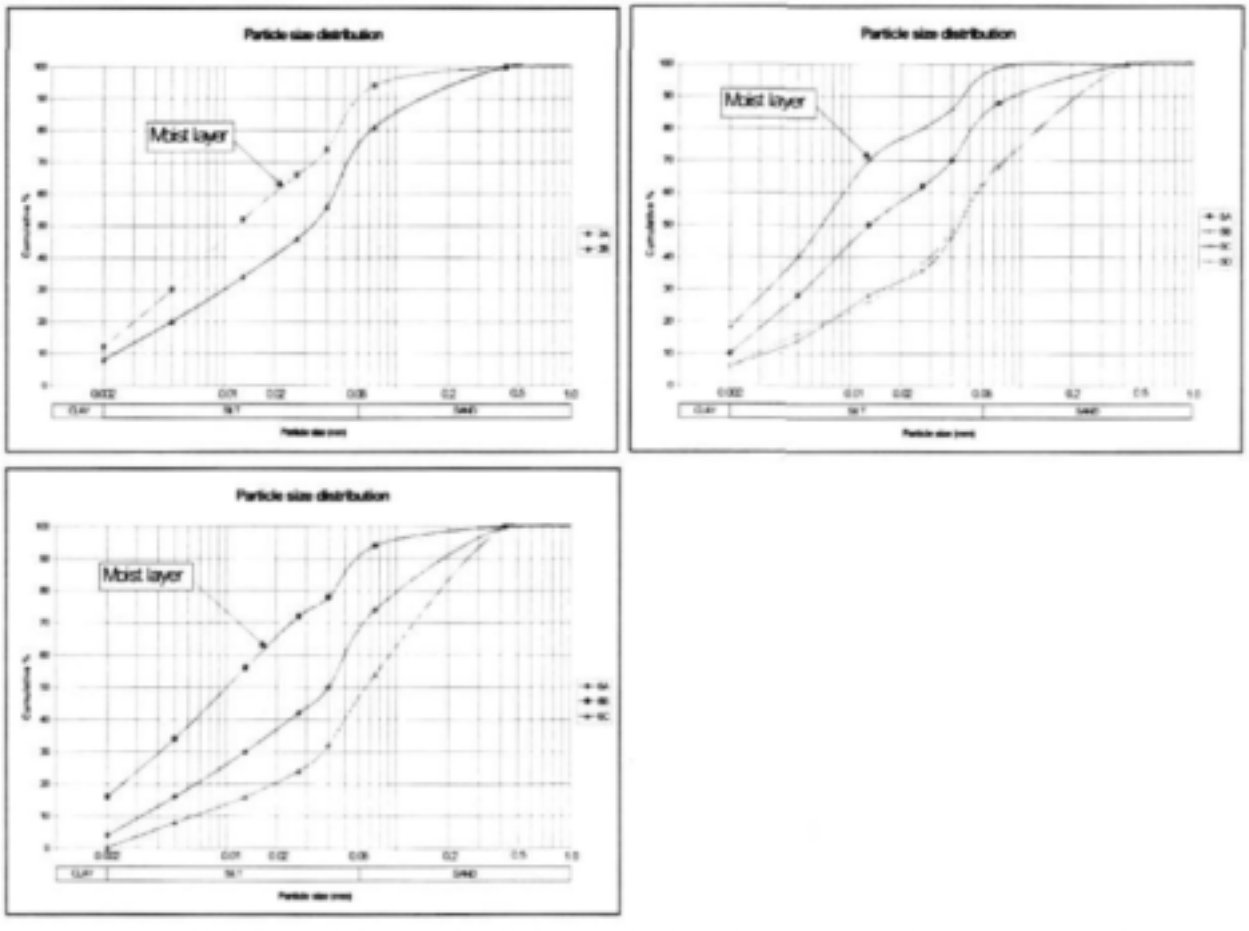

Figure 3.3.3(b): Particle size variations between moist layers in tailings matrix for samples 2, 5 and 6.

3.4 Discussion

Surface features on a tailings deposit are likely to have a limited impact on the overall chemical processes within a tailings deposit. The reason for this is that the extent of these features is only of the order of a few centimetres in almost all cases. The short diffusion distance within this upper zone will cause rapid oxidation to occur in this zone irrespective of the presence of additional pathways.

With regard to subsurface features, field results show that tailings are relatively heterogeneous materials. Variations within a tailings body occur both in the vertical and in the horizontal direction. Mineral compositions vary to some extent, albeit within a limited compositional range.

As observed previously (Anglo American (1995), the particle size distributions within a Witwatersrand gold FRD do not show significant gradation in the horizontal direction, due to the generally narrow range of particle size distributions. In the results for this study, it can be seen that size grading can occur as much in the vertical as in the horizontal direction.

Data show that AP:NP ratios vary significantly throughout an FRD (Pulles, Howard and de Lange (2002), and that there are no specific trends related to location within an FRD. It was also found in this study that AP:NP ratios varied significantly between samples tested. It is uncertain what the contributing factors to these variations are, however, the initial mineralogy, evolutionary history and degree of weathering are all likely to play a part in the observed effects.

Particle size and variations in moisture content imply that the gas diffusion coefficient is likely to vary considerably through the tailings mass. The presence of water saturated fine-grained layers, which are likely to be discontinuous, will be especially important in this respect. Infiltration and evaporation of moisture, to and from these layers respectively, will play an important part in determining oxidation rates, as this is the most dynamic and also the most important factor determining oxygen diffusion into the tailings mass (see Section 2.2.2 above).

4 MODELLING

4.1 Modelling objectives

The objectives of this study were to determine the risk of acidification in gold FRDs, resulting from diffusive oxidation of pyrite. It has been recognised previously that the wall sections of an FRD will be at a higher risk of acidification, as they will be exposed to the atmosphere, across the entire stratigraphic profile, additionally, the larger particle size, lower level of moisture saturation and the possibility of enrichment of pyrite by density separation is also present. Acidification of seepage water from the wall section of an FRD is thus likely to be rapid within a few metres of the surface (<10 years) and more severe than the remainder of the dump, but will cover a much smaller area. It is in fact likely that the major proportion of this risk will be realised during the operational life of the tailings facility. Targeted remediation of such areas is thus likely to be more cost effective than bulk treatment of the tailings mass.

The bulk of an FRD is located beyond the reach of horizontal oxygen flux and is only impacted by vertical diffusion of oxygen through the sedimentary profile. The risk of long-term acidification of seepage from the majority of the tailings material thus primarily depends on the likelihood of acidity consuming the neutralisation potential available in the vertical flow path through an FRD.

The objectives of modelling acidification risk from an FRD were thus recognised as the ability to define the rate of release of acidity from pyrite oxidation and to be able to calculate the stoichiometry and rate of consumption of neutralisation potential by the acidity produced.

4.2 Conceptual model

The conceptual model is based on the morphology of an FRD and the geochemical processes associated with AMD generation and neutralisation. It focuses on the factors that will affect oxygen diffusion and geochemical reactions within an FRD, such as described in Section 4.1.

The model evaluates the time within which it is probable that acidification will occur. This is based on the assumption that AP>NP, as is generally the case in Witwatersrand tailings. Where the AP:NP ratio is less than 0.5 it is unlikely that acidification will occur in any event, however, where AP:NP is greater than 0.5, the possibility exists that acidity will consume all available NP. This is due to the assumption that neutralisation is affected by a 50% efficiency of the $CaCO_3$ neutralisation reaction at neutral pH. This is a valid assumption, as HCO_3^- is the dominant species of carbonate at neutral pH, thus preventing CO_3^{2-} from contributing to neutralisation of acidity in a neutral solution.

Considering these details, an oxidation model of an FRD was conceived, which includes the following zones:

- An upper oxidised zone, which is dominated by pyrite oxidation and is bounded by an oxidation front and the atmosphere.

- The oxidation front is a point in the FRD where the O_2 concentration becomes zero as a result of consumption by pyrite oxidation. It is likely that a relationship exists between the depth of the oxidation front and the colour transition between oxidised and reduced iron within an FRD, although these are not necessarily equivalent points, as discussed in Section 3.3.2.

- The acid neutralisation front is a zone in which NP is consumed by acidity that is generated in the oxidised zone and subsequently seeps through the FRD. This is based on the assumption that rainfall recharge will move more rapidly than the oxidation front.

- The acidification front is a horizon within the FRD that is defined by the point at which all acidity in solution is neutralised and does therefore not consume any further NP.

- The zone between the acidification front and the base of the FRD does not take part in either oxidation or neutralisation reactions and is essentially inert, until encroached upon by acidity or oxygen from above.

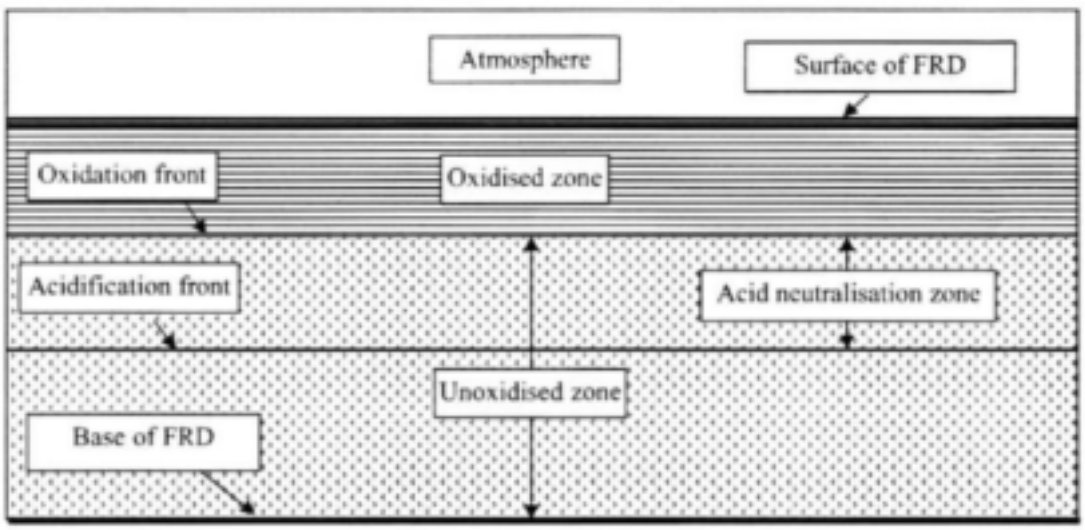

Figure 4.2(a): A conceptual model of oxidation and neutralisation processes within an FRD.

This model is the basis of the accompanying assessment tool that was developed on spreadsheet. The calculations are based on a diffusion model for oxidation reactions, coupled with stoichiometric mineral reaction equations for neutralisation of acidity.

The chemical evolution that is expected to occur over time, using this model is depicted in Figure 4.2(b) below. The chemical evolution of seepage qualities is divided into five stages, representative of the lifecycle of an FRD. The graph is not to scale, as post-decommissioning stages may each last for thousands of years, whereas the operational phase lasts at most a few decades.

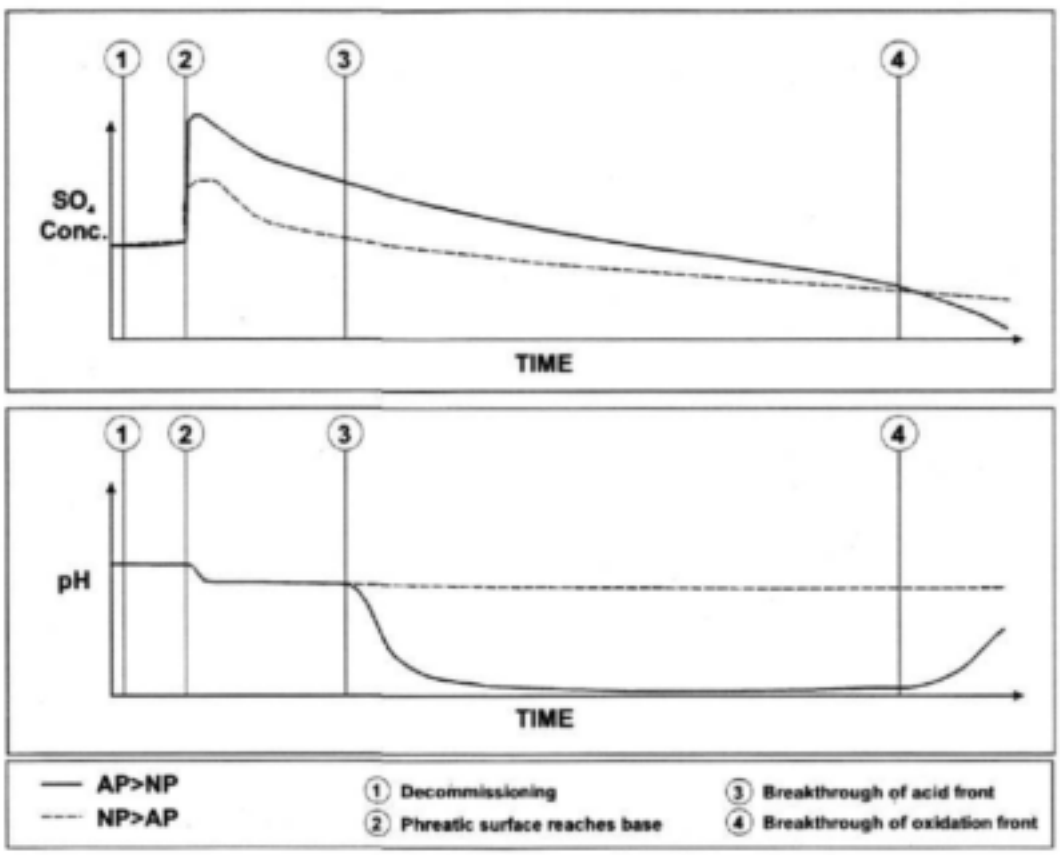

Figure 4.2(b): A conceptual representation of average seepage water quality evolution during the lifecycle of a sulphidic FRD.

In Figure 4.2(b), the time period leading up to decommissioning (point number 1) is the operational period. During this period pyrite oxidation and associated sulphate generation can be attributed to the unsaturated zone between the pond and the wall of the FRD being recharged by process water. Oxidation will take place primarily in the upper part of each layer of material that is deposited where unsaturated zones develop. Continued addition of alkaline process water ensures that sufficient alkalinity is added to the system to prevent acidification of seepage during this period.

After decommissioning of an FRD (point number 1), as the phreatic surface recedes, an increased volume of tailings comes into contact with oxygen and process water is no longer added to buffer acidity that is produced from pyrite oxidation. Seepage water qualities at the base of the deposit are likely to remain those of the process water, until such time as the remaining process water in the pore space, migrates to the base of the tailings deposit (point 2).

Depending on the AP:NP ratio in the material, neutralisation of acidity may take place for a limited period, or throughout the lifecycle of a tailings deposit where sufficient NP is present. In tailings with a high AP:NP ratio, acidification of the oxidation zone will occur rapidly, thus increasing oxidation rates due to bacterial action or Fe^{3+} oxidation. However, both bacterial action and oxidation of Fe^{2+} to Fe^{3+} will be limited by oxygen supply by diffusion as the oxygen diffusion distance becomes deeper. Where AP:NP ratios are low, buffering will prevent acidification from accelerating the oxidation rates, until such time as all NP is consumed in the reactive zone. This is because acid conditions are the most suitable for activity of bacteria such as *Thiobacillus ferrooxidans* and Fe^{3+} is only mobile under acidic conditions.

Where available AP>NP, the acidic front will break through at the base of the FRD at point 3, the time frame of which in practice will be determined by the NP:AP ratio of the tailings material and other physico-chemical parameters. Theoretical modelling showed that this is likely to be of the order of 1000 years or more in typical Witwatersrand tailings. The model, however, needs to be calibrated with applicable field data to confirm this observation. Where available NP>AP, acidification will never occur, as all acidity that is produced will be neutralised by NP. Sulphate concentrations will be controlled by recharge rates and pyrite oxidation rates during this period, unless secondary sulphate minerals should become saturated at any stage.

It can be expected that a large metal load will be released once the acid front reaches the base of the FRD. This is because most metals would be mobilised and transported from their original positions upon acidification by percolating pore water. These metals will then again be immobilised as precipitates where pH increases occur lower down in the sedimentary profile due to neutralisation of acidity by NP in the matrix. This process will continue until such time as the entire profile has been acidified, a process which is likely to take thousands of years in the Witwatersrand tailings environment. At this stage, the concentrated load of metal ions scavenged from higher in the tailings profile will become available to the environment below the FRD over a relatively short time span, depending on transport and kinetic rates. The potential for significant environmental impacts thus exists at this stage.

Once acidification has occurred at point 3, the acid load will continue to be generated, although reporting at progressively lower concentrations due to lower pyrite oxidation rates resulting from the longer oxygen diffusion distance, and a constant average precipitation recharge rate. The entire tailings profile will oxidise in this way, until the oxidation front reaches the base of the tailings deposit at point 4. Once this point is reached, all pyrite grains

are at least partially oxidised and a substantial proportion of the available pyrite has been consumed.

As the pyrite remnants are oxidised to completion, porewaters dissolve progressively less sulphate from the deposit, until such time as pyrite is completely exhausted. The reduction in sulphate concentrations and likely carbonate buffering from rainwater will cause porewater pH to begin to rise again towards more neutral values over time or in the case of neutral drainage, salinity will decrease.

Where acidification of a tailings profile does not occur, reaction times are likely to be longer, due to slower reaction rates. Impacts are also likely to be less significant, as metals will not easily be transported through the sediment profile in neutral porewaters and acidification of the base of the FRD will not occur.

Porewater sulphate concentrations that occur will depend on the interactions between recharge water and pyrite oxidation products, as well as the possible formation of secondary minerals. Where recharge rates are high and oxidation rates are very low, impacts due to salinity and acidity may be limited. It is, however, not the purpose of this study to quantify these factors, as sophisticated water flow, geochemical and oxygen diffusion modelling, as well as substantial field data are required to obtain accurate results. Modelling, based on oxidation rates and available NP is, however, able to provide an indication of the rate at which acidification may occur. This is also the basis on which the accompanying model was constructed.

4.3 PYROX modelling

Sensitivity analyses were conducted, using the PYROX program (Wunderly et al. (1996).

The PYROX model (Wunderly et al. (1995) is designed on the basis of a double diffusion model, i.e. diffusion is modelled through air-filled pore spaces within a porous sulphide-containing medium, as well as through the oxidised coating on individual sulphide grains. The model makes use of a shrinking core component to simulate oxidation of pyrite grains and a finite element component to simulate the transport of oxygen. Contaminant loads are calculated for specified time steps, as well as remaining oxygen and sulphide concentrations at various levels. It is possible to use the model with either pyrite or pyrrhotite as sulphide mineral.

The program is designed to predict acid waste loads due to diffusion controlled sulphide oxidation in mining waste. This program has been widely used for investigations of sulphidic wastes internationally and has thus been verified to some degree. It thus provides a defensible foundation upon which to test and develop specific modelling applications, as was done for this project. It is, however, cautioned that the model should be calibrated with field data to obtain accurate results for prediction of oxidation rates. Considering its wide

use. PYROX was considered an ideal tool for development of a simplified model for inclusion in this study.

4.3.1 Sensitivity analysis

Sensitivity analyses were performed over a simulation period of 200 years. It was found that under field conditions, considering the physico-chemical constraints of the Witwatersrand FRDs, reaction rates tend to be defined within a relatively narrow range of exponential decay curves of the form provided in equation (7).

$$y = Cx^{-z} \tag{7}$$

y is the mass of pyrite oxidised
C is the initial reaction rate
x is the time in years
z is a fitting parameter determined empirically

The range of values for z were found to vary between -5 and -7, across the physico-chemical range which may realistically represent Witwatersrand tailings. It is thus clear that the initial reaction rate is critical in determining the pyrite oxidation history within an FRD, but that the rate at which pyrite reacts over time will tend to behave similarly thereafter, with respect to the rate of change of oxidation of pyrite.

For the PYROX sensitivity analysis, relevant parameters were fixed in the model at values that were considered to be typical to conservative. Sensitivity to different parameters was tested by varying one parameter at a time, whilst maintaining the others at their chosen values. Due to limitations in the PYROX model, it was not possible to test the full range of values that were of interest, specifically with respect to particle size and moisture contents. The input values used are given below.

- Sulphur content 1% of bulk density
- Particle size 1mm
- Moisture content 25% of porosity (11.5 % of total volume, which is considered to be conservative)
- Porosity 46%
- Temperature 18°C
- D2 = 7.8×10^{-12} m^2/s (where D2 is the diffusion coefficient of the oxidised layer on an oxidising pyrite particle)
- Thickness of FRD 40m.

Sensitivity analyses of a similar nature were conducted by Singh & Doulati Ardejani (2003) to demonstrate contaminant generation trends in backfilled open cut spoils. They used a modified version of a fluid dynamics package called PHOENICS to develop a model for oxidation of pyrite and transport of oxidation products within backfilled opencast coalmine spoils.

Particle size

Particle size within the Witwatersrand tailings all grades within the fine sand to silt fraction, with remnant sand dumps being scarce. The sensitivity analysis related to particle size was thus run in PYROX within this range, to determine the sensitivity of the model to particle size. In Figure 4.3.1(a), it can be seen that particle sizes of less than 1mm tend to react similarly and that a negligible effect is noted due to the particle size variation. Only where particles become significantly larger, can significant changes in reaction rates be seen. It can be seen in Figure 4.3.1(a) that oxidation rates are reduced for larger particles of 5mm and 10mm radius, such as would be found in coarse waste, however, in the finer fraction with radii of 0.05mm to 1mm the rates remain virtually identical. It can thus also be inferred that rates will be comparable at smaller particle radii. The effects noted are likely to be related to the thickness of the oxidised coating on a pyrite grains becoming progressively less significant as the grains become smaller, as well as diffusion through the matrix becoming rate limiting.

Due to divergence of the calculations in the PYROX model under certain combinations of parameters, it is not possible to cover the full range of particle sizes below 1mm radius. It should, however, be considered that variation in the pyrite reaction rate due to particle size changes is related to the exposed surface area of the mineral and the thickness of the oxidised layer on the outside of the particle, which regulates the diffusion of oxygen to the pyrite core. Variation due to particle size would thus be seen in the modelling range, if it were a sensitive parameter within the range of interest in Witwatersrand FRDs (see Figure 4.3.1(a).

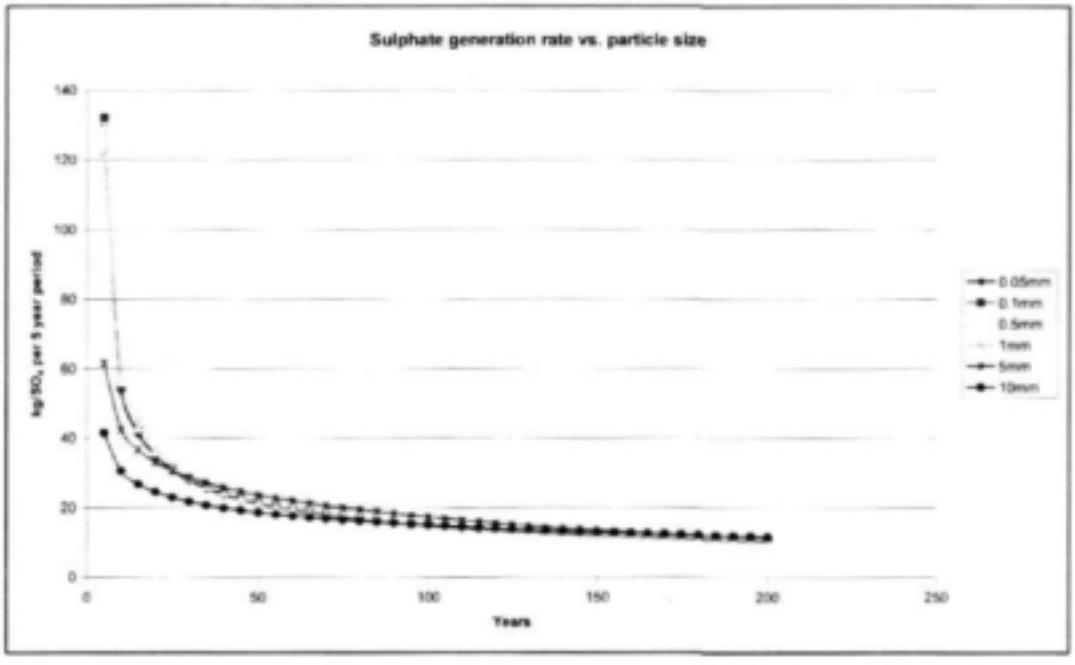

Figure 4.3.1(a): Sensitivity analysis to particle size differences within a tailings body.

Porosity

Porosity is a parameter that has been recognised as an important factor in determining the diffusion coefficient within a tailings body, specifically the air filled porosity, which is directly related to the diffusion coefficient of the tailings (see **Section 2.2.2**). Porosity values were varied in a range between 36% and 50%, to define the envelope in which tailings may possibly fall. The change in porosity resulted in a contaminant load variation of 60% between the lower end and the upper end of the porosity range, thus confirming that porosity is a significant factor in determining the diffusion and hence pyrite reaction rate within an FRD. It should, however, be noted that the rate of change of the reaction rate is the same for both graphs and that the same "decay curve" is thus applicable. It is thus realistic to model both scenarios based on the initial reaction rate and assuming that they will reduce at the rate of $Cx^{-0.567}$. The sensitivity analyses for porosity are represented in Figure 4.3.1(b).

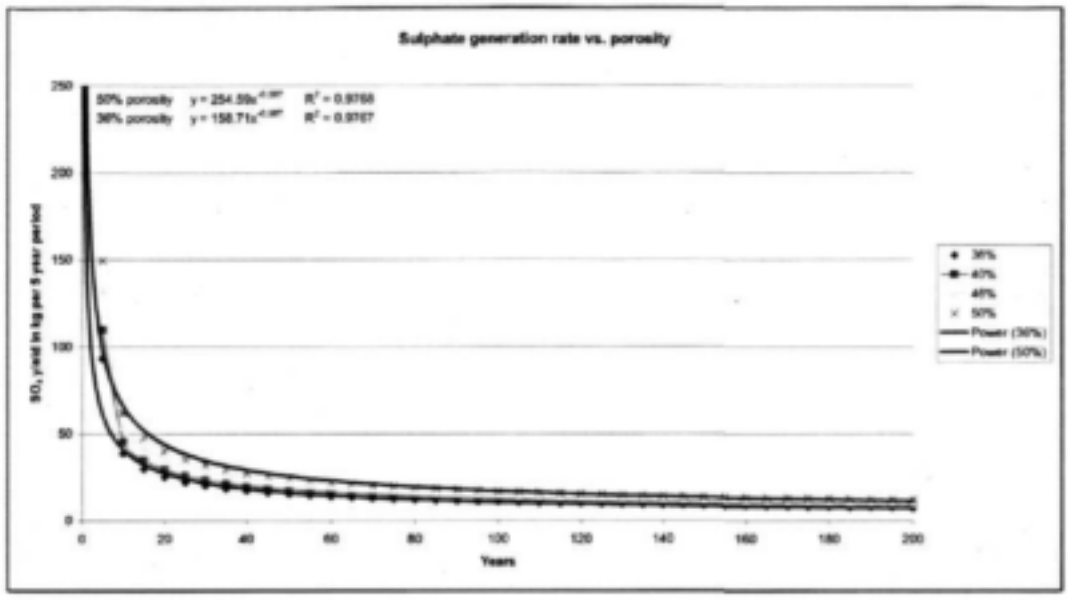

Figure 4.3.1(b): Reaction rates modelled over a 200 year period, showing sensitivity to porosity.

Moisture content

The moisture content was varied between 10% and 50% of porosity. At the small particle sizes and deep tailings profile that was modelled, PYROX was found to be unstable and could not calculate rates at higher moisture contents. The values that were modelled were, however, found to reduce, as expected with increasing moisture content. The oxidation rate (sulphate generation rate) was found to be 46% lower in the 50% saturated tailings than in the 10% saturated tailings, which is a direct result of the lower diffusion coefficient of the more saturated material. Similarly to the other parameters, it can be seen, however, that the rate of change of the "decay curve" remains constant and that the equation $y=Cx^{-587}$ remains a good representation. The sensitivity analysis for moisture content is included in Figure 4.3.1(c).

Sulphur concentration

Sulphur concentration was modelled over a range of between 0.1% and 4% of the bulk density of the tailings material. This translates into approximately 0.2wt% and 8wt% pyrite respectively and approximates the sulphur wt% for a dry tailings material. An upper bound of 6.6% pyrite was assumed for a Witwatersrand FRD, on the basis of maximum pyrite concentrations in the gold reef material mined. The lower bound relevant to AMD is considered to be 0.25% pyrite, as lower sulphur concentrations are not considered to pose a significant risk of acidification. Lawrence & Day (1997) indicated that sulphur concentrations of less than 0.3% have limited potential to cause AMD in most cases. These constraints thus indicate that the likely spectrum of possible pyrite concentrations has been considered in the sensitivity analysis. The results obtained for the sensitivity analysis are presented in Figure 4.3.1(d) below.

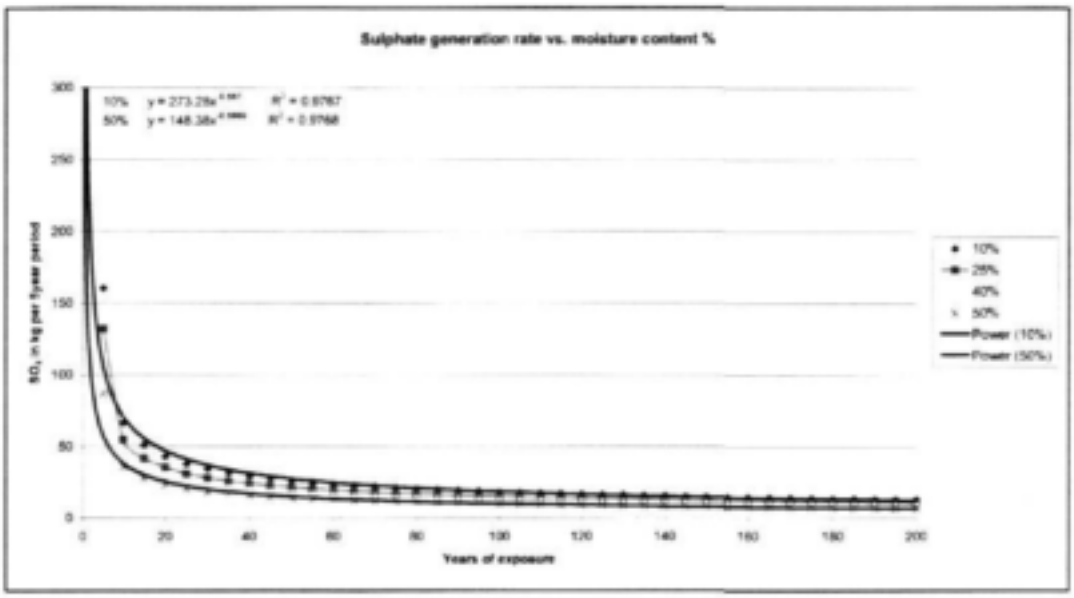

Figure 4.3.1(c): Sensitivity analysis of reaction rates vs. moisture saturation in percent.

It can be noted that the best-fit equation is different for the 0.1% sulphur model when compared to the other curves generated thus far. A z value of –0.642 instead of –0.587 is obtained. This is likely to be related to the fact that the entire pyrite source was depleted after 100 years, thus making the number of significant points less than those recorded for the other pyrite concentrations measured, as well as the final point falling off the line. This also illustrates how the oxygen concentration gradient is much shallower and the oxidation profile deeper at low pyrite concentrations, due to the low rate of consumption, thus enabling the oxidation front to progress rapidly through a tailings body.

It can be noted that the curves do not fit the data perfectly and that the R^2 values are generally around 0.97. It can be seen by inspection, however, that the curves tend to be slightly too shallow in the early time steps, which would cause an overestimation of the oxidation rate if the initial oxidation rate is used as the starting point for the curve of form $y=Cx^z$. This implies, however, that the results that would be obtained by applying this equation to calculate the rate of consumption would be conservative and that the qualitative nature of the result would remain the same.

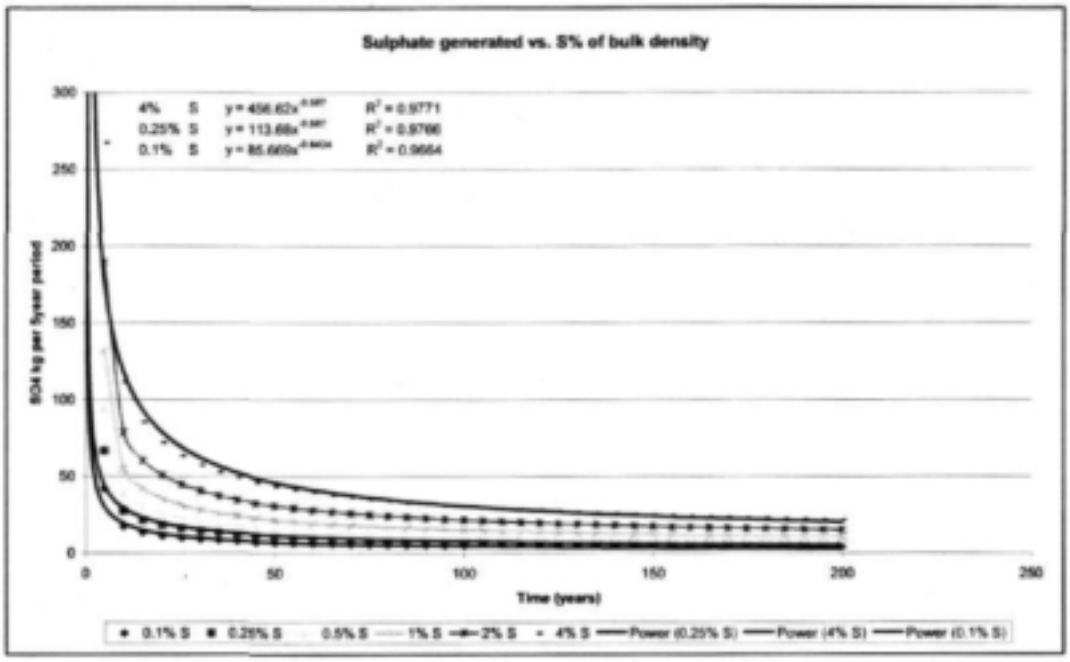

Figure 4.3.1(d): Sensitivity analysis for sulphur concentration.

4.4 Probabilistic modelling

A probabilistic spreadsheet model was developed that is aimed at providing insight into the following aspects:

- The potential for acidification needs to be assessed with reference to a time span of relevance to geological and environmental processes. Where risks may possibly only be realised after many centuries or even millennia, is it reasonable to expect that a mine operator can plan for such an eventuality.

- The life expectancy of specific mitigation measures should be evaluated prior to implementation as no discernable benefit will be obtained from rehabilitation practices, which have a lower life expectancy than the timeframe in which an impact will eventually be realised. This includes possible erosion losses and decomposition and weathering of materials.

- Risks that may realistically materialise in the foreseeable future need to be considered, as they are also likely to cause the highest impact. This is especially true for diffusive pyrite oxidation, where initial reaction rates are the highest. It may be assumed that impacts that will only realise after millennia will be less significant, as processes occurring over this length of time are highly unlikely to cause major impacts as processes will be slow and contaminant concentrations can be expected to be low.

4.4.1 Model construction

The (Tailings Acidification Prediction (TAP) spreadsheet model that was developed for this study functions on the basis of probability, insofar as it predicts the rate at which NP will be depleted in an FRD within a range of possible weathering and oxidation trends. The ranges of probabilities were defined as possible variations in the oxidation history in a typical Witwatersrand FRD, based on the sensitivity analyses conducted with PYROX. It can be seen by inspection of the graphs in Section 4.3 that a value of z=-0.587 (see equation (7)) is generally applicable for the rate of decay in the reaction rate such as discussed in Section 4.3.1. Slight deviations from this rule occur at extreme values of some parameters.

The initial oxidation rate is calculated using equation (5) in Section 2.2.2 by Nicholson et al. (1989). A variant of the model has previously been used in other studies (Bezuidenhout and Vermaak (2002). This rate is taken as the baseline to which the above mentioned 'decay rate' is applied. The diffusion coefficient is calculated using the equation of Millington and Shearer (1971) ((8) below).

$$D_{eff} = (1-S)^2 \cdot [\varepsilon(1-S)]^2 \qquad (8)$$

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

Required data inputs for the model include:

- Humidity cell oxidation rates to define the inherent kinetic rate of the material in question.
- Porosity of tailings.
- Degree of moisture saturation of tailings.
- Pyrite concentration in tailings.
- Depth of the tailings profile.
- NP:AP ratios from ABA analyses.

In this model, the common parameters have the same significance as those used in the PYROX model. Kinetic rates are, however, defined by the use of humidity cells, rather than using theoretical calculations of the shrinking core method.

Acid Base Accounting (ABA) analyses are used to define the AP:NP ratio. 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. 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 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.

The depth of a tailings profile is significant, 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. The model is also based on this hypothesis.

Default values are provided for other parameters in the model, as obtained from the literature. An example of an input screen is provided in Figure 4.4.1(a)

Steady-state Oxygen Ingress model

Constants and formulae	Symbol				
Diffusion coefficient (waste)	De	1.21E-06	m^2/s		Calculation cells
Oxygen concentration at waste interface	Cw	2.8E-01	kgO_2/m^3		Physical constants
Total oxygen in waste profile	MO_2	5.12E+00	kg/m^3		Empirical constants
Oxygen flux at waste interface	Fc	1.77E-08	kgO_2/m^2s		DATA INPUT CELLS
H^+ produced as $CaCO_3$		0.92905038	kg $CaCO_3$/year		
S concentration		0.27	wt% S		
Average specific gravity of minerals		2.66	t/m³		
Dry Bulk Density		1.44	t/m³		
Total mineral mass		1437345.00	g/m³		
Pyrite mass		7186.73	g/m³		
Molar pyrite concentration		59.90	mol/m³		
Load H+		2156.03	mols/m² dump area		
Load H+ as $CaCO_3$		239.60	kg/$CaCO_3$/m³		
NP as $CaCO_3$		107.82	kg/$CaCO_3$/m³		
Real NP:AP reaction ratio		0.23			
Effective available $CaCO_3$		53.91	kg$CaCO_3$ eq/m³		
Reaction rate for constant pyrite oxidation	K	3.31E-09	1/s		
Pyrite density		5.00	g/cm³		
Quartz density		2.65	g/cm³		
Oxygen concentration (air)	Co	0.28	kg/m³		
Diffusion coefficient in air phase	Da	1.80E-05	m²/s		
Depth intervals	i	0.05	m		
Molar ratio of H+ per mol pyrite oxidised		4.00			
Molar ratio of O_2 to pyrite		3.75			
H+:$CaCO_3$ molar stoichiometry		1.00	mol/mol		
Data entry for model calculations					
Oxidation Rate (Humidity Cells)		50	gFeS_2/t/week	0.083349309	mols FeS_2/t/week	3.30688E-05	mol pyr/mol pyr/sec
Porosity of waste		0.46			
Degree of saturation of waste	S	0.75	m^3/m^3 porosity		
Pyrite concentration		0.50	wt% FeS_2		
Height of tailings (m)		40.00	m		
ABA NP:AP molar ratio from lab results		0.46	kg $CaCO_3$ ratio		

Figure 4.4.1(a): An example of the input screen for the Tailings Acid Probability (TAP) spreadsheet model.

4.4.2 Modelling results

The model was tested to determine the sensitivities and constraints on the NP consumption rate over time. To facilitate comparison, tailings variables were chosen to coincide with those used for PYROX modelling as far as possible and were otherwise estimated from available

values in the literature. A summary table is given below, for times to NP depletion under various sensitivity scenarios, such as were compiled for PYROX. For this analysis the sulphur concentration was reduced to 0.53% (1% pyrite), as it did not affect the stability of the model and more closely resembled average field concentrations that were available. Humidity cell rates were estimated at 50g FeS_2/ton/week on the basis of available information for other types of FRDs. The depth of the tailings profile was estimated at 40 meters.

In Table 4.4.2(a) default values are indicated behind each heading in brackets and the closest value to the default value is highlighted under each subheading in bold. It can be seen that the model is sensitive to all the parameters that were considered. Although all parameters have a significant influence on the rate of acidification of an FRD, the most sensitive parameters are clearly the sulphur content and the height of the FRD.

The impact of the sulphur 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 sulphur 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 oxygen diffusion flux will not increase proportionally to an increase in sulphur concentration.

- Increasing the sulphur 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.

In Figures 4.4.2(a) and 4.4.2(b), the output is presented for typical input values, as defined in Table 4.4.2(a). In Figure 4.4.2(a) it can be seen that the total AP is given, as well as the amount of acidity that will theoretically be released by sulphur oxidation over time. The proportion of the available pyrite that is oxidised over the modelling period of 10 000 years can be deduced by the discrepancy between oxidised pyrite and total pyrite throughout the tailings profile. In Figure 4.4.2(b), an example of actual model output, Neutralisation Potential 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. Theoretical sensitivity curves of the time to acidification of a tailings profile of average composition, relative to its thickness, are shown in Figure 4.4.2(c).

Unfortunately no humidity cell rates were available for Witwatersrand tailings, thus requiring the estimation of kinetic rates on the basis of experience with other tailings materials. Conservative values, based on literature and other types of tailings were used. These rates will have a significant effect on the oxygen flux rate and hence the pyrite oxidation rate in an FRD. A database of humidity cell rates should thus be developed to define the

boundaries of oxidation rates of Witwatersrand tailings. A comprehensive discussion on humidity cell tests is included in the work by Usher et al. (2003).

Table 4.4.2(a): Times to acidification for various parameters, as modelled within the constraints of Witwatersrand FRDs, using the spreadsheet model developed for this study.

	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
Humidity cell rates (50g FeS_2/ton/week)			
10g FeS_2/ton/week	>10 000	>10 000	>10 000
25g FeS_2/ton/week	4000	9500	>10 000
50g FeS_2/ton/week	**2700**	**4000**	**7000**
100g FeS_2/ton/week	1000	2000	3000
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
Sulphur concentration (0.53%)	***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

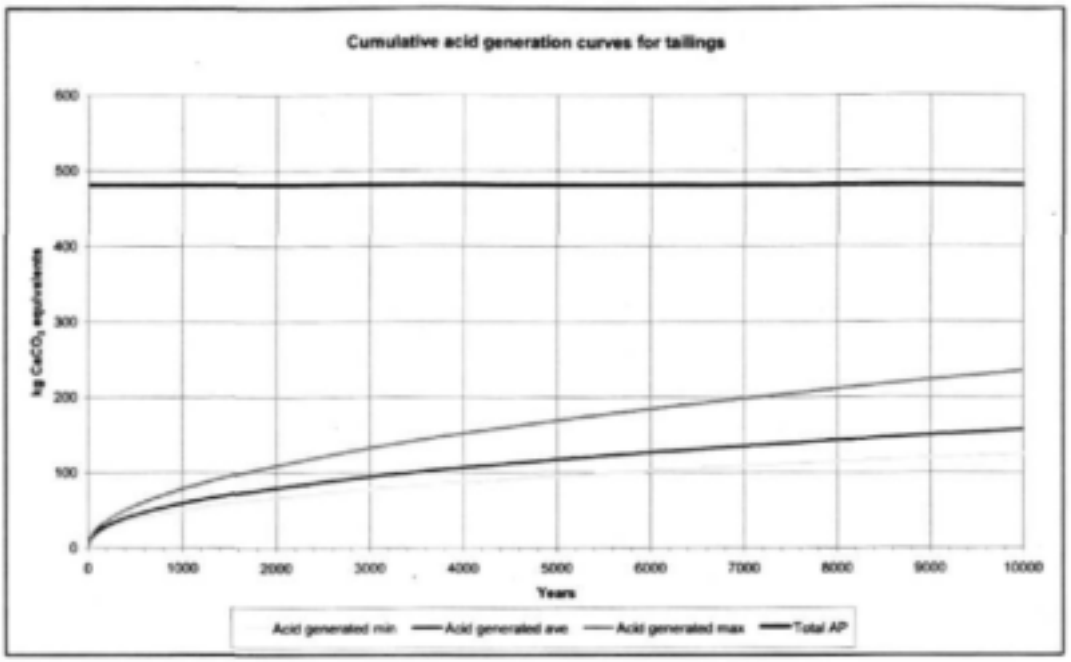

Figure 4.4.2(a): An example of the acid generation output of the spreadsheet model accompanying this report.

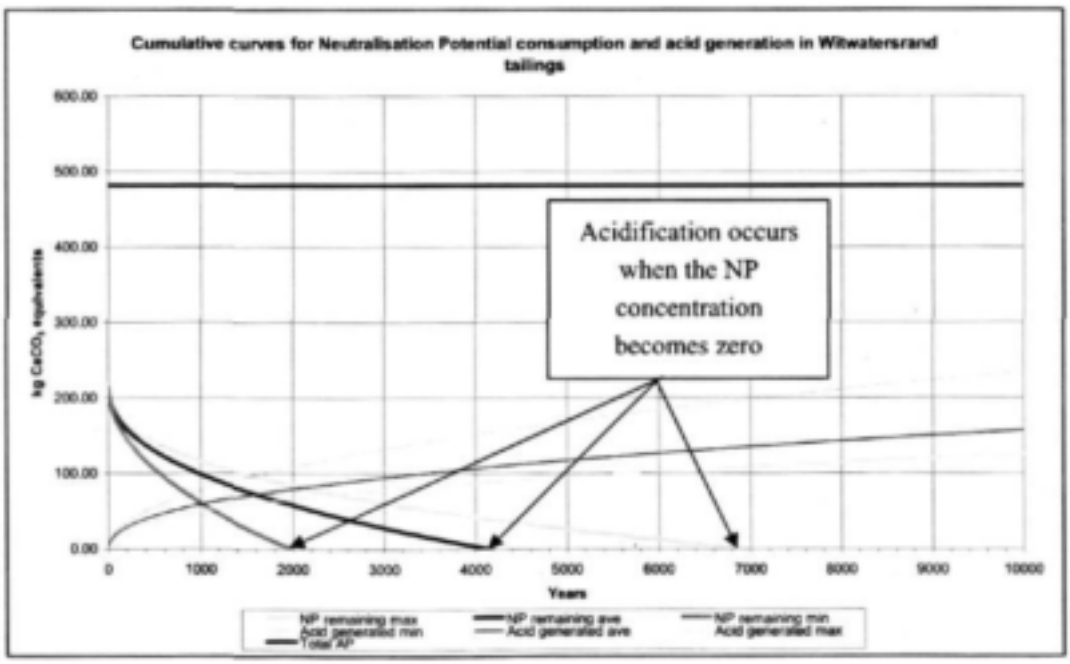

Figure 4.4.2(b): An example of the spreadsheet model output. Acidification occurs when NP= 0 kg $CaCO_3$ equivalents.

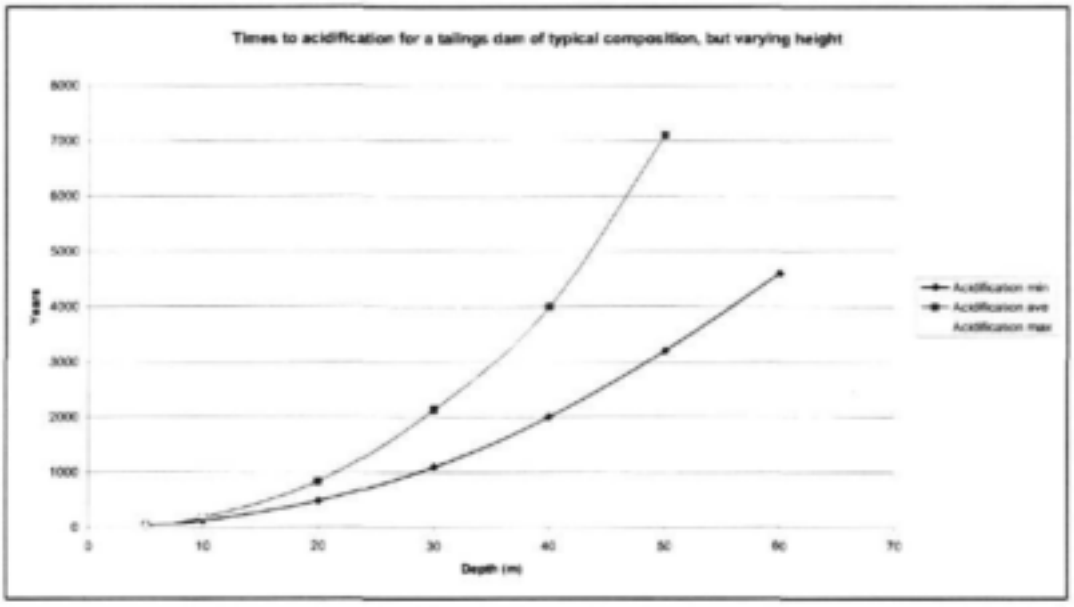

Figure 4.4.2(c): A set of graphs depicting the relationship between the height of an FRD of typical composition and the time to acidification.

5 DISCUSSION OF RESULTS AND IMPLICATIONS FOR THE MINING INDUSTRY

The implications for the mining industry from this research revolve around time issues, including:

- The potential for FRDs to undergo AMD processes to such an extent that the long term seepage quality turns acidic. Although impacts on the receiving water environment will occur through neutral seepage, acidic seepage is perceived to be much worse, due to mobilisation of metals under acidic conditions.

- The time frame for FRDs to generate an acidic long term seepage.

- Altering the potential for acidification by implementation of selected mitigation options.

Research has shown that these probabilities and consequences are different for different tailings deposits, as well as being different between the wall and beach sections of an FRD.

5.1 Development of the oxidised zone and the associated impact on the risk of acidification

Oxidation of pyrite in the upper strata of an FRD is caused by the diffusive flux of oxygen. The driver for this oxygen flux is a concentration gradient between the atmosphere and pore spaces in the tailings due to the continuous consumption of oxygen through pyrite oxidation and bacterial action. The generally low organic content of Witwatersrand FRDs dictates that the major portion of oxygen is likely to be consumed by pyrite oxidation rather than bacterial action, unless pyrite concentrations are very low.

Model calculations indicated that the upper two metres of tailings are oxidised very rapidly (<10 years), as was also demonstrated by Mayer et al. (2000). After this initial period reaction rates decline to much lower values, as longer diffusion distances and armouring of pyrite grains slow oxidation processes over time.

Due to the stoichiometric ratio of oxygen to pyrite, the depth and gradient of the oxygen concentration profile is significantly influenced by the pyrite concentration and reaction rate. Where pyrite concentrations are high, oxygen is consumed rapidly in the upper parts of the FRD and a shallow oxidation profile will form. Where low pyrite concentrations are present, oxygen can diffuse much deeper into the tailings profile, but the overall rate of contaminant generation is much lower due to the longer average diffusion path to available pyrite grains.

At equivalent AP:NP ratios, the generation of acidity throughout the tailings profile in low sulphide tailings will mean that a greater proportion of the available acidity is realised more quickly relative to high sulphide tailings. This will thus result in more rapid consumption of available NP and therefore earlier acidification of seepage from the base of the tailings deposit.

Field observations and the literature indicate that the oxidised zone in Witwatersrand FRDs extends only a few meters (up to 5m) below the surface. Theoretical calculations indicate that the oxidation front may extend somewhat deeper, but this is not observed on a macro scale when considering colour changes through the FRD. It should, however, be remembered that colour changes are dependent on oxidation of iron with subsequent precipitation of iron hydroxy complexes and thus do not necessarily define the zone of pyrite oxidation per se. Changes in moisture contents, bacterial activity and the presence of organic material will all serve to moderate calculated values, especially where bulk assessments are done, such as have been applied in this study.

It should, however, be noted that the rate of oxidation expected from a typical Witwatersrand FRD, as was depicted in Table 4.4.2(a), is only likely to cause acidification after a minimum time of the order of >1000 years. Variation around this norm will depend on the physico-chemical variations within the tailings, such as were discussed in Section 4.4.

Observations and modelling results bring one to the conclusion that the major proportion of acid is generated in the near surface of an FRD due to the factors discussed above. It would thus be advantageous to reduce the potential for acid generation in the surface of an FRD, rather than to apply mitigation measures aimed at the entire profile. Furthermore, the fine grain sizes generally present in tailings material are likely to result in a limited benefit from the use of soil covers designed to act as oxygen barriers, especially in tailings deposits that have been decommissioned for more than 5 to 10 years. The greatest advantages that are likely to emanate from the use of engineered covers will be in their potential to maintain a higher moisture content and/or to act as store and release systems to prevent percolation of water through the tailings profile. These properties will result in a much lower oxygen diffusion coefficient and will prevent contaminant transport.

5.2 Pyrite oxidation in the wall section of an FRD and associated risk of acidification

The outer wall of an FRD is already exposed to the atmosphere along its entire vertical extent during the operational phase. For this reason, acidification is likely to occur soon after deposition of the material and will be dictated by the AP:NP ratio and redox conditions within the affected zone, as discussed in Section 2.3 above.

This oxidation zone will extend horizontally to approximately the same depth as the surface oxidised zone extends vertically throughout the remaining portion of the FRD. Higher horizontal diffusivities, lower moisture contents and larger particle sizes may result in a deeper oxidation zone forming in the wall section than is found in vertical tailings oxidation profiles. The fact that this zone is located on the perimeter of an FRD makes it significant, as it may make up a significant proportion of the tailings mass, depending on the ratio of the perimeter length to the surface area covered by the FRD.

To prevent acidification of an FRD, it is thus of primary importance to mitigate the potential impacts from the wall sections of the FRD, before concerning oneself with the remainder of the system where a vertical oxidation profile is dominant. Various mitigatory measures are discussed in Section 6.

6 PROPOSED ASSESSMENT AND DESIGN CRITERIA FOR MITIGATION OF ACIDIFICATION IN WITWATERSRAND FRDS

After consideration of the criteria that have been assessed during this study and the perspectives gleaned from the fieldwork and modelling, it is possible to formulate assessment and design criteria to apply to Witwatersrand gold FRDs for the mitigation of AMD impacts.

The point of departure in the methodology for this study is that a rehabilitation solution should be found, which is a functional, sustainable means to control acidification of groundwater due to pyrite oxidation within gold FRDs on the Witwatersrand. Some of these arguments are indeed also valid for other FRDs. Although it is likely that an FRD with a relatively high AP:NP ratio will become acidic over time, it is important to keep a perspective on the time frame attached to this probability, as well as the likely impacts that will be noted.

6.1 Field measurements

A number of field measurements will be useful in constraining the timeframe and depth of weathering in an FRD. In this study a gas diffusion approach has been followed and the relevant criteria are thus based on the premise that a similar approach will be used. Both physical and chemical parameters need to be quantified satisfactorily to determine the applicable constraints.

6.1.1 Measurement of modelling parameters in tailings

It has been shown in the preceding sections that the reaction rate of pyrite is largely dependent on the physico-chemical conditions within a body of tailings material. Relevant parameters include porosity, moisture contents and particle size.

For a first order assessment, it is not practical to conduct a large number of measurements, as the cost in terms of time and money would be excessive. It should be borne in mind that estimates should be as realistic as possible, but that one should err on the side of caution where uncertainties exist.

Oxygen flux measurements

The oxygen concentration profile needs to be evaluated, with a view to calibrating the diffusion model against field data. The reason for this is that the model functions on the basis of differential concentrations across the tailings profile. Measuring of oxygen flux alone would not enable the concentration gradient to be determined and would thus leave greater

doubt as to the long-term changes that may be expected. Any doubt with respect to the diffusion coefficient of the tailings material, the pyrite concentration or kinetic rates would result in excessive uncertainty. Factors such as the period of exposure of the tailings material would also be critical for the determination.

Where the oxygen concentration gradient is known, it is possible to define a curve, which should approximately coincide with the modelled curve in the spreadsheet model or PYROX and which will therefore coincide with the history of the tailings deposit with respect to pyrite oxidation. Uncertainties in moisture contents, pyrite concentration or kinetic rates will be visible if the curve shapes differ significantly.

The oxygen concentration gradient can be measured by sampling pore gases in an FRD at different depths with a thin tube. This method was used by de Vos (pers. comm.) and David and Nicholson (1995). A similar technique has been developed by ANSTO of Australia, who use a multiple port oxygen analyser.

Where the pyrite concentrations and reaction rates are well known and other physical parameters are known, the model can be calibrated by using oxygen flux measurements. It is important to know the time of exposure of the tailings material in this situation, however.

To measure the oxygen flux, the methods used by Tibble and Nicholson (1997) or Timms and Bennet (2000) can be used. These methods both entail measurement of changes in oxygen concentration in the headspace of a cylinder in response to diffusion as a result of consumption by pyrite oxidation reactions.

Laboratory humidity cell measurements may also be used as a source of oxidation rates. Knowledge of diffusion coefficients of the material in question will permit calculation of oxygen flux rates in diffusion dominated systems. These measurements will be prone to overestimation of oxidation rates, as no account is taken of the dependence of kinetic rates on oxygen concentrations.

Moisture content

For an initial assessment of the moisture concentration, a bulk sample should be collected at a depth of at least 3m below the tailings surface. This is to ensure that evaporative losses do not cause unnecessary underestimation of the average moisture contents of the tailings material. Samples should also not be collected too low in the sediment profile, as it is possible that moisture contents that are too high will be determined due to the influence of the receding piezometric surface or a capillary zone. This is especially pertinent within the first few years after decommissioning. Field collected samples can be oven dried to determine their moisture contents.

For higher order assessments, detailed knowledge about porosity and moisture saturation will be required at high resolution for accurate modelling. This will be difficult and expensive to determine without the use of high resolution down-the-hole geophysical methods.

Where long term monitoring of moisture contents is possible, installation of time domain reflectometers can be used. This will not enable high-resolution measurements to be made economically, but will be useful for understanding the long-term changes in the average moisture contents within a tailings profile.

Porosity

The porosity of tailings material can be measured directly by collecting undisturbed core samples and measuring the mass to volume ratio and subtracting the mass of water, as determined through moisture content analysis. Knowledge of the mineralogy will enable an approximate average specific gravity to be calculated so that the porosity can be calculated on the basis of the difference between average Specific Gravity and bulk density. Results from in situ geophysical density determinations can also be used, if they can be shown to be of acceptable accuracy.

Pyrite concentrations

It should be borne in mind that pyrite will begin to react with oxygen as soon as it is exposed. Measurement of pyrite concentrations within a tailings profile should thus occur in tailings that have not been exposed to oxygen. This is important, as pyrite concentrations will be underestimated and AP:NP ratios may be overestimated if samples are collected above the oxidation front. It will thus be advantageous to measure pyrite concentrations in material that is obtained from as deep within the tailings profile as possible.

Due to the fine grain size and the low likelihood of intermittent exposure to oxygen, material from within the pool section of an FRD will be least affected by oxidation. It is, however, necessary to collect samples from different parts of an FRD for an assessment and these considerations should be kept in mind.

In Witwatersrand tailings the pyrite concentrations are generally relatively low when compared to other tailings materials. This is advantageous with respect to acid generation potential, but makes it difficult to measure pyrite concentrations accurately. The X-ray diffraction (XRD) method is not very sensitive to minerals at low concentrations and pyrite is commonly not detected, even though it is present in sufficient quantities to generate significant acidity. Direct measurement of pyrite by this method is thus not feasible in most situations.

It is thus recommended that sulphide sulphur should be measured to determine its availability within the tailings material (Usher et al. (2003). This is more likely to provide a representative concentration of pyrite than the XRD method. It is also recommended that sulphate sulphur

should be determined to ensure that any sulphate that is present is not included as potentially oxidisable pyrite. As the Witwatersrand tailings are known to contain pyrite as the dominant sulphide mineral, it is acceptable to assume that sulphide sulphur can be attributed to pyrite. This provides a measure of the potentially oxidisable iron within the system as well.

Neutralisation Potential

The Neutralisation Potential of a sample within an FRD may be affected by two possible features of the system.

- Firstly, where oxidation of pyrite has already occurred, it is likely that a portion of the NP will have reacted and that a depleted value will be obtained if measured from a sample.

- Secondly, the area below the phreatic surface will contain porewaters laden with alkaline process water from the operational phase. If a sample is collected from this part of the FRD, then it is likely that NP will be overestimated.

Considering these points, it is advisable to collect a sample in which NP is to be determined below the oxidised zone, but above the phreatic surface to obtain a representative value for the NP in the FRD.

In conclusion, the criteria for accurate sampling of the various parameters in an FRD require that samples be collected as deeply as possible within the tailings body, but at least a few metres (±6m capillary fringe according to J.W. Wates (pers. com.) above the phreatic surface. This part of an FRD is likely to be the most representative of fresh tailings material, especially in the beach section of an FRD. The same considerations are also relevant when sampling wall material.

The Neutralisation Potential of tailings material can be measured by using the standard ABA method of Sobek et al. (1978) or refined methods, considering mineral buffering effects, can also be used for better results (Usher et al. (2003). It will, however, also be of benefit to determine the pH dependence of NP, as NP that only becomes effective at low pH is not effective in preventing pollution. The lower bound of carbonate alkalinity at approximately pH4.5 may be considered an acceptable point of reference. This value may be directly included in model calculations.

6.1.2 Sampling

Sampling needs to be conducted in such a way that all required parameters can be measured from a single bulk sample at a particular sampling point for calibration of the model. Heterogeneity within an FRD still needs to be considered, thus requiring that a representative number of samples should be taken to represent such spatial variation.

Sampling equipment needs to consider the needs of the study at hand. Where possible, a mechanical coring or augering device that does not expose the sample excessively to air should be used. This will prevent oxygen from easily coming into contact with pyrite in the sample and will also prevent loss of moisture by evaporation. The sample should be stored in an airtight container, with as little air as possible being included in the sample. A sealable cylinder or an airtight plastic bag may be used. For accurate results, samples should be stored under an inert gas, as the fine pyrite found in tailings material will react very rapidly with any available oxygen.

It is important to store and transport samples under refrigerated conditions and to have them analysed as soon as possible. It should also be stipulated that samples should be dried in an oxygen free atmosphere prior to analysis, as increased temperatures and exposure to the atmosphere are likely to cause a large proportion of available pyrite to oxidise during sample preparation.

It is not recommended that a hand auger be used to collect fresh samples if it is possible to use suitable mechanical sampling equipment. The reason for this is that a hand auger cannot be comfortably used at depths deeper than 5 meters and becomes inefficient and potentially dangerous to use at depths deeper than 7 to 8 meters due to top-heaviness of the long string of the rods and the associated difficulty in handling the equipment.

6.2 Long term monitoring of FRDs with a view to prediction of AMD impacts

6.2.1 Field measurements

The long-term fluctuations in the oxidation rates in FRDs are not well known. Fluctuations will occur primarily as a function of the depositional processes in an FRD or due to climatic factors, especially in the post-decommissioning phase.

It is well known that variations in moisture content do occur in conjunction with the seasonal fluctuations in rainfall (David and Nicholson (1995). In the Witwatersrand, these fluctuations can be extreme, with almost all rainfall occurring during the spring and summer months. It can, however, be expected that moisture fluctuations will occur primarily in the upper part of a tailings profile, where evaporative processes readily occur.

Measurement of moisture contents and long term oxygen flux rates will thus be advantageous in calibrating long-term models.

Oxygen measurements

Oxygen measurements can be conducted using the methods described by Tibble and Nicholson (1997) or Timms and Bennet (2000). These methods require a cylinder to be placed into or over the tailings profile respectively for direct measurements of tailings oxidation rates. The apparatus are relatively simple in principle and will provide a database of oxidation

rates, which can be compared with modelling results to enable prediction of long-term trends.

Moisture contents

Field measurements of moisture contents are useful, as the moisture concentration is the most variable physical parameter determining AMD generation and transport in an FRD. The construction and instrumentation of field lysimeters will thus enable long term monitoring of moisture trends in a tailings profile. Various instruments are available for measuring moisture content, with time domain reflectometry being one of the favoured methods in FRDs. Any reliable method may be used, however. Records of moisture contents will be invaluable in calibrating time dependant unsaturated flow models and oxygen diffusion models that include water flux and moisture content as inputs.

6.3 Mitigation of AMD impacts from Witwatersrand gold FRDs

Mitigation options that are considered, take cognisance of the morphological and geochemical observations that have been recorded for Witwatersrand gold FRDs.

Typical modelling results show that acidification in the short to medium term is primarily a concern in the wall section of an FRD, as vertical diffusion of oxygen is only likely to cause acidification on the order of 1000 years but horizontal diffusion will lead to immediate acidification of wall materials. This is compounded by the low piezometric surface in the wall section when compared to beach and pond areas and indicates that acidification will already occur during the operating phase.

Various mitigation options that have been used in the past and which could be considered for use in the future are discussed below, along with insights gained from modelling.

6.3.1 Mitigation options

Alkaline addition

Addition of Neutralisation Potential is a technique that has been used in a number of cases for mitigation of AMD. This technique usually involves the addition of crushed limestone or dolomite to acid generating materials to increase the NP:AP ratio sufficiently to eliminate the potential for AMD. In these cases leachates are saline, rather than acidic and metal impacts are reduced to much lower levels than is the case under acidic conditions.

Leachate volumes are not impacted, but acid generation rates are likely to be reduced to some extent due to lower activity of pyrite oxidising bacterial and the removal of Fe^{3+} in precipitates at the higher pH values. At high concentrations of calcium, which is present in both dolomite and calcite, gypsum saturation and precipitation is likely to occur. Sulphate

loads emanating from the system are thus likely to be reduced and seepage quality will also be maintained at a higher pH.

Carbonate rock may be included as a base material, a surface material (rock cladding) or as an addition to the tailings stream during deposition. The latter method is likely to be the most effective, as neutralisation material is in close contact with acidic porewater and will thus have a direct impact on porewater chemistry. Where neutralising material is placed as a surface layer, the rate of leaching cannot be controlled and effectiveness can therefore not be guaranteed. Where neutralising material is placed as a basal layer, porewaters will not be neutralised until they leach from the base of the deposit. This will cause metals to accumulate at the base of the deposit and bacterial and Fe^{3+} oxidation will continue unabated in the oxidised zone.

Depyritisation

Depyritisation has been used in the past as a mitigation option at various sites around the world. Techniques such as selective flotation are able to remove pyrite from a waste stream so that an inerted tailings stream can be created (Bussiére & Aubertin (1999). Recovered pyrite can be used for production of sulphuric acid or it can be disposed of in a less reactive zone of the affected FRD.

It is thus possible to build at least part of the FRD with unreactive material. Where this is applied to the wall of an FRD, AMD risk can be substantially reduced. The method is, however, not applicable to all tailings materials (Martin et al. (2002).

Engineered covers

Engineered covers that can be applied to tailings in the Witwatersrand are of essentially two types, i.e. store and release covers and barrier covers.

Store and release covers make use of the nett evaporative climate of the highveld region to prevent recharge. In these covers, capillary breaks are constructed with the purpose of holding water within the evaporative zone below the surface of the cover. This water is then released during drier periods. Very low recharge rates can be achieved using this method and leachate volumes are thus significantly reduced.

Barrier covers are designed to prevent the ingress of oxygen and are commonly constructed of a subsurface layer of fine material, such as compacted clay. If these layers are kept at a high moisture saturation they present an effective barrier to oxygen diffusion. Dessication cracking is, however, often a major concern with regard to the performance of these covers.

Combinations of store and release and barrier covers can be effective in reducing environmental impacts from acidic tailings, as the rate of waste load generation is reduced and waste that is generated is not transported to the surrounding environment.

Fly ash

Fly ash from power generation is a waste product that occurs in large quantities. In South Africa, fly ashes are commonly alkaline in nature. These materials are generally fine grained and in some instances have pozzolanic properties. Fly ashes, however, also have high metal concentrations and are potentially leachable, especially in acid or alkaline solutions.

Where appropriate, fly ash could potentially be used as a remediation material on FRDs. The addition of alkalinity and potentially lower permeability of cover materials due to pozzolanic activity will reduce the rate of acid generation in an FRD. It is important to note, however, that detailed risk assessments are required before applying such measures. It is also important to realise that fly ash should only be applied as a cover and not as an additive to tailings, as the risk of metal leaching by acidity should be avoided.

Bottom liner

Bottom lining of an FRD is a further option that can be considered, especially where it pertains to the wall section of the FRD in question. The placement of a suitably impermeable barrier with associated drainage infrastructure will enable leachate that is generated to be collected and treated as required. Subsurface impacts can thus be avoided.

6.3.2 Lifecycle considerations

Remediation options that may be applied or considered for application to FRDs are highly dependent on the stage in the lifecycle of the facility. Applicable options for consideration are the most numerous during the design phase and reduce progressively from the operational phase to the decommissioned phase. Applicable options are discussed below.

Design phase

During the design phase, all possible options are available. At this stage of an operation, a full feasibility study should be conducted where possible to include remediation options for AMD mitigation, considering the technologies mentioned above. Options that can be considered at this stage of development include the placement of a liner below the footprint of the FRD or at least part of the wall footprint. The most important part of the system that should be considered is the wall section, where the highest risk of acidification occurs and where acid generation is likely to have an impact, even during the operational phase.

Operational phase

Technologies that require treatment of the tailings stream can only be considered during the operational or design phases. These technologies include depyritisation and addition of alkalinity to the tailings stream. Where a portion of the FRD has already been created without

the technologies in question having been applied the effectiveness of the measures will be reduced proportionally.

This observation is especially true for the wall section, because of the exposure of the entire profile to oxygen. Where these technologies are applied to the entire tailings mass, the isolation of the subsurface layers from oxygen will result in a high effectiveness of these measures, even when they are implemented in the latter part of the facility's lifespan. Depyritisation of the upper few meters of tailings material would for instance prevent the initially high AMD generation rate that was modelled for this study from ever occurring. Where a large excess of alkalinity is added, this will also have a beneficial effect with regard to retarding the transport of reaction products through the tailings profile.

Decommissioned operations

Where decommissioned FRDs are concerned, the options are limited to surface applications. Here one can consider the construction of engineered covers, including the possibility of fly ash or alkaline addition. In the case of alkaline addition, rock cladding techniques that are currently applied can be altered to use dolomite or limestone to add NP to the system, whilst also controlling erosion by water and wind, although the efficacy of this measure will have to be investigated with regard to the rate of release of NP.

It should be noted that erosion control is important in preventing AMD processes as well. The removal of tailings material reduces the diffusion path length to fresh pyrite material and also contributes to the mobilisation of reaction products into the environment.

From the modelling and sensitivity analyses conducted in this study, it has been shown that application of mitigation measures to a decommissioned FRD should be applied as soon as possible after closure. In such an instance, a combined barrier cover, for the prevention of oxygen ingress and a store and release cover for prevention of precipitation recharge will significantly reduce the rate of contaminant generation and transport through an FRD.

In an FRD that has been exposed to the atmosphere for a few years a barrier cover is likely to have a limited effect on pyrite oxidation and thus acid generation rates. This is because the upper part of an FRD will be oxidised within 5 to 10 years and subsequent increases in the diffusion path length will cause oxygen flux rates to reduce substantially, due to the upper tailings profile acting as a cover. The fine grained nature of tailings and the presence of water saturated layers within a tailings profile are likely to result in future oxidation rates being only slightly reduced by applying cover materials. However, a store and release cover will still be beneficial, where recharge and associated contaminant transport rates can be reduced.

Prior to cladding with carbonate rock, it should be determined whether NP is likely to be leached to reactive sites in sufficient quantities before implementing such a strategy. As mentioned previously, fly ash has associated metal leaching risks and may have limited

efficacy in treating AMD, depending on its source and should thus also be carefully evaluated before application.

It is thus recommended that prior to application of any of the remediation strategies to a decommissioned FRD, they should be carefully evaluated before expenses are incurred in implementation, as they may have very little benefit. The age of the FRD is of great importance in this instance, as much of the acid generation risk will be realised within ten years of decommissioning.

Recharge control measures, such as store and release covers or impermeable liners are strategies that are always likely to provide some benefit, even after initial oxidation has occurred. The main benefit of a cover in this situation, however, would be to prevent rainfall recharge and thus to prevent contaminant transport to the receiving environment, rather than to prevent the generation of contaminants by pyrite oxidation.

7 CONCLUSION

In summary, the objectives of this research and a description of how they were achieved are provided below:

- *To understand the relationship between the depth and rate of weathering and the potential mass of acidity at gold FRDs through empirical analysis.* An in depth study of local and international literature on the weathering of pyritic tailings material was conducted and documented in Section 3 of this report. Based on this understanding and experience, conceptual engineering models were developed that will provide insight into the depth and rate of weathering in Witwatersrand tailings and their long-term AMD potential. Use was also made of the existing, internationally applied, PYROX numerical model to conduct detailed modelling of sensitive processes, and to be used for development of engineering models.

  An empirical spreadsheet model (engineering model), referred to as the Tailings Acidification Prediction (TAP) model has been developed, which provides a tool for determining the likely time span over which an FRD will produce acidic drainage. It is not intended that this tool be used for detailed risk assessment, but rather as a screening tool for determining what the likely risks of acidification are and what the time span is over which acidification will occur.

- *Develop procedures to conduct rapid accurate AMD risk assessments of gold FRDs.* Key parameters and drivers affecting the potential of an FRD to turn acidic were identified during the research. Guidelines for sampling and measurement are provided in Section 6 of this report. Key parameters include tailings geotechnical properties, morphology and height of an FRD, mineralogy and Acid-Base geochemistry of tailings material, oxygen flux rates and the FRD lifecycle.

  The objective of determining accurate methods proved to be challenging, as the complexity and variability of physical physico-chemical properties within an FRD requires that higher order assessments be conducted with considerable levels of specialist expertise and data intensity.

- *Evaluate mitigation or risk reduction measures by making use of the AMD risk assessment procedure and provide industry guidelines.* The conceptual model was used to evaluate various mitigation measures for AMD at Witwatersrand FRDs. Mitigation is specifically focussed on the significant parameters that determine the AMD potential of Witwatersrand FRDs. Mitigation measures for different lifecycle phases of the tailings were also assessed and evaluated. It should be cautioned, however, that the methodologies that have been suggested have not been verified in terms of technical and financial feasibility and as yet are therefore only indicative of promising methodologies that should be evaluated.

In conclusion, the research demonstrates that oxidation rates and the depth of weathering of an FRD can be modelled, provided key processes and parameters are recorded and known for a specific FRD. The simplified empirical model results suggest that beach areas on a typical Witwatersrand FRD will take >1000 years before acidic seepage is generated from the base of the deposit, whereas AMD is likely to be realised in the wall and paddock sections of an FRD early on in its lifecycle, possibly even during the operational phase.

Acidic seepage from the base of the wall will occur as a result of horizontal oxygen ingress into the tailings deposit and acidification in the paddock will occur as a result of slow deposition rates, frequent flushing effects and high oxidation rates, when compared to the body of the FRD.

The model should be seen as a screening tool and follows a conservative approach regarding rate equations and flow processes. The model can be used to evaluate a range of potential mitigation measures and can ultimately be used for cost-benefit optimisation studies.

NOTE: The application of this research and model should be done with caution. The results have not been verified extensively and will require a knowledgeable person to operate and apply the research and model sensibly. It is important to use realistic values in calibrating the model, as some parameters are sensitive to inaccuracy and can have a very significant influence on the outcome.

8 RECOMMENDATIONS

In the light of the findings of this research, it is recommended that the following should be considered:

- The potential measures that are provided as promising best practice techniques should be further evaluated to determine what their technical and financial feasibility status is for possible incorporation into formal guidelines.

- Further research should be conducted to verify the findings of this study. This research should include the application of other available models and especially data collection. Data collection should include:
  - Parameters required by the TAP model should be more reliably and comprehensively determined and collated. These include:
    - Pyrite concentrations
    - Intrinsic pyrite oxidation rates in specific tailings
    - Diffusion coefficient determinations
    - Moisture profile determinations
  - Depth dependent oxygen profiles within tailings for time line calibration
  - Oxygen flux measurements across the surfaces of FRDs for rate verification

9 REFERENCES

Andersen M.S., Larsen F., Postma, D. (2001) Pyrite oxidation in unsaturated aquifer sediments. Reaction stoichiometry and rate of oxidation. Environ. Sci. Tehnol., **35**, 4074-4079.

Anglo American 1995. A basic course on slimes dams (South African gold paddock dams) for metallurgists. Tailings unit, Geotechnical section, Civil engineering department, Anglo American Corporation of South Africa.

Appelo C.A., Postma D (1993) Geochemistry, groundwater and pollution. Balkema publishers. ISBN 9054101067.

Bezuidenhout N, Vermaak, J (2002) Water and Salt Balance Modelling combined with ARD Assessment of the FS North 6 FRD, Welkom. Internal Report Golder Associates.

Bussiére B., Aubertin M. (1999) Clean tailings as cover material for preventing Acid Mine Drainage: An in situ experiment. Mining and Environment conference proceedings. Sudbury Ontario. 19-28.

David D.J., Nicholson R.V. (1995) Field measurements for determining rates of sulphide oxidation. Sudbury. Mining and the Environment Conference proceedings. ISBN 0-660-16043-9, **1**, 201-210.

Davis G.B., Ritchie A.I.M. 1986 A model of oxidation in pyritic mine wastes, 1. Equations and approximate solution. Appl. Math. Modell. **10**, 314-322.

DeVos K.J. Personal communication. Tailings specialist. Mississauga office. Canada. Golder Associates.

DeVos, K.J., Ritchie, D.G., and Bocking, K.A. 1999. Practical considerations for covering sulphidic tailings deposits situated above the groundwater table. Proceedings Sudbury '99 Mining and the Environment. Sudbury Ontario. **1**, 291-300.

Feather C.E., Koen G.M. (1975) The mineralogy of the Witwatersrand reefs Minerals Sci. Engng., **7**, 189-224.

Förstner U., Wittmann G.T.W. (1976) Metal accumulation in acidic waters from gold mines in South Africa. Geoforum, **7**,44-49.

Freeze R.A., Cherry J.A. (1979). "Groundwater". Prentice Hall inc. Englewood Cliffs. New Jersey.

Funke J.W. (1990) The water requirements and pollution potential of South African gold and Uranium mines. Water Research Commission (Rep. No. KV9/90).

Holmes J.A. (2000) Water chemistry in lakes. BGY C56H3 Lecture 4 - Oct 2, 2000. The Environmental Information office. University of Toronto. http://www.utoronto.ca/env/eio.htm.

James A.R. (1997) The prediction of pollution loads from coarse sulphide-containing waste materials. Water Research Commission Report 559/1/97.

Jury W.A., Gardner W.R., Gardner W.H. (1991) Soil physics (Fifth edition). John Wiley & Sons.

Lambe T.W., Whitman R.V. (1979) Soil mechanics. John Wiley & Sons Inc. New York.

Lawrence R., Day S. (1997) Short course #2 on: Chemical prediction techniques for ARD. 4th International Conference on Acid Rock Drainage. Vancouver, B.C. Canada.

MacG. Robertson, A. (1987) The influence of depositional methods on the engineering properties of tailings deposits. International conference on mining and industrial waste management. Johannesburg. South Africa.

Martin J.P., Koerner R. (1984a) The influence of vadose zone conditions on groundwater pollution – Part1: Basic principles and static conditions. J. Hazard. Mat., **8**, 349-366.

Martin T.E., Davies M.P., Rice S., Higgs T., Lighthall P.C.(2002) Stewardship of tailings facilities. Mining Minerals and Sustainable Development. International institute for Environment and Development.

Mayer K.U., Blowes D.W., Frind E.O. (2000) Numeric modeling of Acid Mine Drainage generation and subsequent reactive transport. Proceedings from the fifth international conference on Acid Rock Drainage (ICARD 2000), 135-141.

Millington R.J, Shearer R.C. (1971) Diffusion in aggregated porous media. Soil Science., **111**, 372-378

Morin K.A., Hutt N.M. (1997) Environmental Geochemistry of Minesite Drainage. Practical Throry and Case Studies. ISBN 0-9682039-0-6.

Nicholson R.V., Gillham R.W., Reardon E.J. (1988) Pyrite oxidation in carbonate-buffered solution: 1. Experimental kinetics. Geochim. Cosmochim. *Acta*. **52**, 1077-1085.

Nicholson R.V., Gillham R.W., Cherry J.A., Reardon E.J. (1989) Reduction of acid generation in mine tailings through the use of moisture-retaining cover layers as oxygen barriers. Can. Geotech. J., **26**, 1-8.

Nicholson R.V., Gillham, R.W., Reardon E.J. (1990) Pyrite oxidation in carbonate-buffered solutions: 2. Rate control by oxide coatings. *Geochim. Cosmochim. Acta* **54**, 395-402.

Nicholson R.V., Elberling B., Williams G. (1995) A new oxygen consumption technique to provide rapid assessment of tailings material in the field and laboratory. Sudbury. Mining and the Environment Conference proceedings. ISBN 0-660-16043-9, **3**, 999-1006.

Nicholson R..V., Rinker M.J., Tibble P.A., Williams G., Wiseman M. (2000) An integrated approach to assess acid generation and metal release from sulphide tailings using oxygen consumption measurements, porewater chemistry and geochemical modeling. Proceedings from the fifth international conference on Acid Rock Drainage (ICARD 2000), 693-700.

Perkins E.H., Nesbitt H.W., Gunter W.D., St-Arnaud L.C., Mycroft J.R. (1995) Critical review of geochemical processes and geochemical models adaptable for prediction of acidic drainage from waste rock. MEND Project 1.42.1

PTI Environmental Services, 1995. Predicted Water Quality of the Batu Hijau Pit Lake, Sumbawa, Nusa Tenggara, Indonesia. Prepared for Newmont Mining Company, October 1995.

Pulles, Howard & de Lange. 2002 Geochemical sampling for AngloGold.

Pugh C.E., Hossner L.R., Dixon J.B. (1984) Oxidation rate of iron sulfides as affected by surface area, morphology, oxygen concentration, and autotrophic bacteria. *Soil Sci.***137**,309-314.

Reardon E.J. and Moddle P.M. (1985) Gas diffusion coefficient measurements on Uranium mill tailings: Implications to cover design. Uranium **2**, 111-131.

Rösner T., Boer R., Reyneke R., Aucamp P., Vermaak J. (2001) A preliminary assessment of pollution contained in the unsaturated and saturated zone beneath reclaimed gold – Mine residue deposits. WRC report No. 797/1/01.

Schafer W.M., Grady, T., Runnells D.D., Luckay C., Jones R. (1997) Control of tailings oxidation rate using spigotting techniques. Proceedings of the Fourth International Conference on Acid Rock Drainage, Vancouver., **3**, 1047-1057.

Singh R.N., Doulati Ardejani F. (2003) Evaluation of factors affecting pyrite oxidation and subsequent pollutant generation in backfilled open cut coal mines. 8th International Congress on Mine Water & the Environment, Johannesburg South Africa.

Sobek A.A., Schuller W.A., Freeman J.R., Smith R.M. (1978) Field and laboratory methods applicable to overburdens and minesoils. EPA-600/2-78-054. USEPA. Cincinnati. Ohio.

Søgaard Andersen M., Larsen F., Postma D. (2001) Pyrite oxidation in unsaturated aquifer sediments. Reaction stoichiometry and rate of oxidation. Environ. Sci. Technol., **35**, 4074-4079.

Steffen, Robertson and Kirsten (SRK) (1988) Research contribution of mine dumps to the mineral pollution load in the Vaal barrage. Water Research Commission (Rep. No. PT 3632/10)

Stuparyk R.A., Kipkie W.B., Kerr A.N., Blowes D.W. (1995) Production and evaluation of low sulphur tailings at INCO's Clarabelle Mill, Sudbury. Mining and the Environment Conference proceedings. ISBN 0-660-16043-9, **1**, 159-169.

Sverdrup, H.U. and Warfvinge, P. (1988) Weathering of primary silicate minerals in the natural soil environment in relation to a chemical weathering model. Water, Air, Soil Poll. **38**, 387-408.

Tibble P.A., Nicholson R.V. (1997) Oxygen consumption on sulphide tailings and tailings covers: Measured rates and applications. Proceedings of the Fourth International Conference on Acid Rock Drainage, Vancouver., **2**, 647-661

Timms G.P. and Bennet J.W. (2000) The desigh and use of novel devices for measuring Oxygen flux through covers on sulfidic waste rock. Proceedings from the fifth international conference on acid rock drainage (ICARD), **2**, 841-849.

Troeh F.R., Jabro J.D., Kirkham D. (1982) Gaseous diffusion equations of porous materials. Geoderma, **27**, 239-253

Usher B., Cruywagen L.M., de Necker E., Hodgson F.D.I. (2001) On-site and laboratory investigations of spoil in opencast collieries and the development of Acid-Base Accounting procedures. Water Research Commission Report.

van Genuchten, M. Th. and P.J. Wierenga. 1977. Mass Transfer Studies in Sorbing Porous Media. II. Experimental Evaluation with Tritium. Soil Sci. Soc. Amer. J. 41:272–278.

Van Genuchten R.(1987) Documented computer programs (Part 1). Source code for non-linear least squares analysis (CFIT). USDA-ARS. U.S. Salinity laboratory, Riverside California.

Van Niekerk A.M. (1997) Generic simulation model for opencast mine water systems. Water Research Commission. WRC report No. 528/1/97.

van Schalkwyk, A, Vermaak, J.J. 2000 The relationship between the geotechnical and hydrogeological properties of residual soils and rocks in the vadose zone. Water Research Commission. WRC report No. 701/1/00.

Viljoen C. Personal Communication. Soil scientist. Viljoen and Associates. Johannesburg.

Wunderly M.D., Blowes D.W., Frind E.O., Ptacek C.J. 1996 Sulfide mineral oxidation and subsequent reactive transport of oxidation products in mine tailings impoundments: A numerical model. Water Resources Research., **32 no 10**, 3173-3187.

Yanful E.K. (1991) Conference proceedings, Second International Conference on the abatement of Acidic Drainage Montréal., **1**, 461-487.

Yanful E.K., St-Arnaud L.C. (1991) Conference proceedings, Second International Conference on the abatement of Acidic Drainage Montréal., **1**, 487-504.

Zhao B. (1998) A mineralogical and geochemical study of alteration associated with the Ventersdorp Contact Reef in the Witwatersrand Basin South Africa. (unpub.) Ph.D thesis, University of the Witwatersrand, Johannesburg.

APPENDIX A

Analytical data and sample descriptions

Descriptions of field samples collected.

Site number	Site description	Sample number	Feature depth below surface	Description
1	Wall section near the entrance ramp to the tailings dam		3.7m	Transition to grey (unoxidised) colour noted at approximately 3.7m, with some mottling visible
		1	4.95m	Sampling point in grey material
2	Mid beach section of tailings dam		±0.05m	Leached surface layer
			1.03m	Light to darker yellow transition to yellow coloured material
			2.57m	Yellow to grey transition
			5.90m	Grey to yellow colour change
		2A	6.75m	Wet layer (wet clay like consistency) *This is the fourth distinct layer of water saturated material encountered, thought to be a capillary break zone*
		2B	6.80m	Layer below water saturated layer. *Sampled to determine possible particle size differences*
3	Lower beach section in the vicinity of the penstock.		±0.10m	Leached zone at the top of the profile, light grey coloured.
			2.39m	Yellow to grey transition
		3	2.44m to 2.59m	Sample position
4			1.65m to 1.70m	A moist gley horizon below a weathered profile, with some iron staining. Below the zone is a lower moisture content, yellow (oxidised) horizon
			2.23m	Transition from yellow to gleyed horizon
		4	3.50m	Sample collected in grey material
5	Middle beach section of tailings dam		0.165m	Light grey leached zone transition to yellow stained

				oxidised zone
			0.46m	Light yellow transition to darker yellow horizon
			0.75m	Mottling, with slight orange colour prominent
		5A	2.55m	Zone Mottled oxidised zone above transition to grey material
		5B	2.60	Sample of grey material, directly below the transition from yellow material
		5C	4.20m	Water saturated layer
		5D	4.25m	Material directly underlying moisture layer
6	Upper beach area, approximately 100m from the wall		±0.05m	Light grey coloured leached zone transition to y
			0.65m	Slight sign of gleying
			3.40m	Transition from mottley zone to grey zone
		6A	4.80m	Semi-saturated layer with grey material
		6B	5.05m	1 to 2cm thick water saturated layer with wet clay like consistency
		6C	5.10m	Material below wet layer, with a more sandy appearance
7	Wall section on the plant side of the tailings dam		±0.05m	Transition from light coloured leached zone below surface to light yellow coloured material
			1.9m	Light yellow material grades into orange and grey mottled material
			2.20m	Motley material transition to grey material
		7A	2.50	Transition zone from motley material to grey (unoxidised material)
			2.93m	Colour again becomes yellower

			4.69m	Colour becomes yellow again
		7B	5.35m to 5.80m	Sample from bottom of hole
8	Wall section		±0.05m	Light grey coloured leached zone transition to yellow oxidised material
			3.10m	Transition from yellow to grey material
		8	4.87m	Continuous grey material after transition at 3.10m
9	Middle to upper beach section		0.05m	Transition from Light grey leached zone to yellow oxidised tailings
			2.89	Dark orange staining, probably in bedding planes or fractures
			3.76m	Transition from yellow to grey. A distinct black band formed in a fracture or a bedding plane. It is expected that the black colour is caused by framboidal pyrite re-precipitation
		9	5.74m	End of hole. Material remains essentially homogeneously grey.
10	First batter road cutting, transverse hole drilled		0.4m	Bright yellow-orange mottling.
			1.15m	Transition from yellow to grey with some
		10	1.75m	Grey material with some mottling (material below transition)
			3.20m	End of hole. Grey with some mottling

HYDROMETER ANALYSIS

Sample number	63mm	53mm	37.5mm	26.5mm	19mm	13.2mm	4.75mm	2mm	0.425mm	0.075mm	0.04mm	0.025mm	0.013mm	0.005mm	0.002mm	D10 grain size	% Clay	% Silt	% Sand	% Gravel	Grading Modulus
1	100	100	100	100	100	100	100	100	100	66	44	34	24	12	4	0.004	4	53	43	0	0.34
2A	100	100	100	100	100	100	100	100	100	94	74	66	52	30	12	0.0015	12	73	15	0	0.06
2B	100	100	100	100	100	100	100	100	100	81	56	46	34	20	8	0.0015	8	62	30	0	0.19
3	100	100	100	100	100	100	100	100	100	88	70	62	48	26	10	0.0015	10	70	20	0	0.12
4	100	100	100	100	100	100	100	100	100	76	52	42	30	10	4	0.0022	4	62	34	0	0.24
5A	100	100	100	100	100	100	100	100	100	88	70	62	50	28	10	0.0017	10	70	20	0	0.12
5B	100	100	100	100	100	100	100	100	100	68	46	36	28	14	6	0.0017	6	53	41	0	0.32
5C	100	100	100	100	100	100	100	100	100	99	86	80	70	40	18	0.0015	18	75	7	0	0.01
5D	100	100	100	100	100	100	100	100	100	69	48	38	26	16	6	0.002	6	54	40	0	0.31
6A	100	100	100	100	100	100	100	100	100	74	50	42	30	16	4	0.01	4	60	36	0	0.26
6B	100	100	100	100	100	100	100	100	100	94	78	72	56	34	16	0.001	16	71	13	0	0.06
6C	100	100	100	100	100	100	100	100	100	54	32	24	16	8	0.2	0.002	0	44	55	0	0.46
7A	100	100	100	100	100	100	100	100	100	79	60	50	36	20	8	0.0015	8	63	29	0	0.21
7B	100	100	100	100	100	100	100	100	100	61	44	36	24	12	6	0.0015	6	48	46	0	0.39
8	100	100	100	100	100	100	100	100	100	76	57	48	36	20	8	0.0015	8	60	32	0	0.24
9	100	100	100	100	100	100	100	100	100	82	64	54	40	22	10	0.0015	10	64	26	0	0.18
10	100	100	100	100	100	100	100	100	100	66	44	34	20	0.2	0.2	0.002	0	56	43	0	0.34

Waterlab (Pty) Ltd / (Edms) Bpk
(Reg Nr/No 83/09165/07)

Management & Laboratory Services for Water and Wastewater
Environmental Monitoring and Impact Assessment

PO Box 11508 Hatfield 0028 (T) 012-349-1044/1066 (F) 012-349-1072
E-mail: waterlab@iafrica.co.za

WATERLAB

CERTIFICATE OF ANALYSIS

ATTENTION: Mr. P. Rousseau **ORDER NO : 000590** **DATE: 04/08/2003**
REPORT NO: 11533 **PROJECT NO: 159** **SAMPLES: 6028-6044**

GOLDER ASSOCIATES
X-RAY DIFFRACTION MINERALOGICAL ANALYSES

Mineral	Concentration (%)		
	Sample 1	Sample 2A	Sample 2B
Calcite	-	1	2
Hematite+/Goethite	1	-	-
Microcline	1	-	1
Plagioclase	2	1	1
Quartz	79	65	65
Mica	4	8	8
Chlorite	10	18	17
Amphibole	1	2	2
Pyrophyllite	3	6	5
Gypsum	-	-	-
Ilmenite/Smectite Interstratification	-	-	-

per.....
Waterlab (Pty) Ltd

Waterlab (Pty) Ltd / (Edms) Bpk
(Reg Nr/No 83/09165/07)

Management & Laboratory Services for Water and Wastewater
Environmental Monitoring and Impact Assessment

PO Box 11508 Hatfield 0028 (T) 012-349-1044/1066 (F) 012-349-1072
E-mail: waterlab@iafrica.co.za

WATERLAB

CERTIFICATE OF ANALYSIS

ATTENTION: Mr. P. Rousseau | **ORDER NO : 000590** | **DATE: 04/08/2003**
REPORT NO: 11533 | **PROJECT NO: 159** | **SAMPLES: 6028-6044**

GOLDER ASSOCIATES
X-RAY DIFFRACTION MINERALOGICAL ANALYSES

Mineral	Concentration (%)		
	Sample 3	Sample 4	Sample 5A
Calcite	Trace	1	Trace
Hematite+/Goethite	-	-	-
Microcline	-	1	1
Plagioclase	2	2	1
Quartz	70	65	66
Mica	6	5	7
Chlorite	15	19	17
Amphibole	2	3	-
Pyrophyllite	4	4	5
Gypsum	-	-	3
Ilmenite/Smectite Interstratification	-	-	-

per.....
Waterlab (Pty) Ltd

Waterlab (Pty) Ltd / (Edms) Bpk
(Reg Nr/No 83/09165/07)

Management & Laboratory Services for Water and Wastewater
Environmental Monitoring and Impact Assessment

PO Box 11508 Hatfield 0028 (T) 012-349-1044/1066 (F) 012-349-1072
E-mail: waterlab@iafrica.co.za

WATERLAB

CERTIFICATE OF ANALYSIS

ATTENTION: Mr. P. Rousseau | **ORDER NO : 000590** | **DATE: 04/08/2003**
REPORT NO: 11533 | **PROJECT NO: 159** | **SAMPLES: 6028-6044**

GOLDER ASSOCIATES
X-RAY DIFFRACTION MINERALOGICAL ANALYSES

Mineral	Concentration (%)		
	Sample 5B	Sample 5C	Sample 5D
Calcite	1	1	1
Hematite+/Goethite	-	-	-
Microcline	1	1	-
Plagioclase	1	2	1
Quartz	75	51	81
Mica	6	9	4
Chlorite	12	26	9
Amphibole	-	2	1
Pyrophyllite	3	8	3
Gypsum	-	-	-
Ilmenite/Smectite Interstratification	-	1	-

per.....
Waterlab (Pty) Ltd

Waterlab (Pty) Ltd / (Edms) Bpk
(Reg Nr/No 83/09165/07)

Management & Laboratory Services for Water and Wastewater
Environmental Monitoring and Impact Assessment

PO Box 11508 Hatfield 0028 (T) 012-349-1044/1066 (F) 012-349-1072
E-mail: waterlab@iafrica.co.za

WATERLAB

CERTIFICATE OF ANALYSIS

ATTENTION: Mr. P. Rousseau | **ORDER NO : 000590** | **DATE: 04/08/2003**
REPORT NO: 11533 | **PROJECT NO: 159** | **SAMPLES: 6028-6044**

GOLDER ASSOCIATES
X-RAY DIFFRACTION MINERALOGICAL ANALYSES

Mineral	Concentration (%)		
	Sample 6A	Sample 6B	Sample 6C
Calcite	1	1	-
Hematite+/Goethite	-	-	1
Microcline	-	1	-
Plagioclase	1	2	1
Quartz	78	52	78
Mica	4	8	5
Chlorite	12	28	11
Amphibole	2	1	-
Pyrophyllite	3	6	3
Gypsum	-	-	-
Ilmenite/Smectite Interstratification	-	-	-

per.....
Waterlab (Pty) Ltd

Waterlab (Pty) Ltd / (Edms) Bpk
(Reg Nr/No 83/09165/07)

Management & Laboratory Services for Water and Wastewater
Environmental Monitoring and Impact Assessment

PO Box 11508 Hatfield 0028 (T) 012-349-1044/1066 (F) 012-349-1072
E-mail: waterlab@iafrica.co.za

WATERLAB

CERTIFICATE OF ANALYSIS

ATTENTION: Mr. P. Rousseau | **ORDER NO : 000590** | **DATE: 04/08/2003**
REPORT NO: 11533 | **PROJECT NO: 159** | **SAMPLES: 6028-6044**

GOLDER ASSOCIATES
X-RAY DIFFRACTION MINERALOGICAL ANALYSES

Mineral	Concentration (%)		
	Sample 7A	Sample 7B	Sample 8
Calcite	1	Trace	Trace
Hematite+/Goethite	1	2	-
Microcline	1	Trace	Trace
Plagioclase	1	1	1
Quartz	63	76	70
Mica	7	4	7
Chlorite	14	12	14
Amphibole	4	-	1
Pyrophyllite	5	4	5
Gypsum	1	-	-
Ilmenite/Smectite Interstratification	2	-	2

per.....
Waterlab (Pty) Ltd

Waterlab (Pty) Ltd / (Edms) Bpk
(Reg Nr/No 83/09165/07)

Management & Laboratory Services for Water and Wastewater
Environmental Monitoring and Impact Assessment

PO Box 11508 Hatfield 0028 (T) 012-349-1044/1066 (F) 012-349-1072
E-mail: waterlab@iafrica.co.za

WATERLAB

CERTIFICATE OF ANALYSIS

ATTENTION: Mr. P. Rousseau | **ORDER NO : 000590** | **DATE: 04/08/2003**
REPORT NO: 11533 | **PROJECT NO: 159** | **SAMPLES: 6028-6044**

GOLDER ASSOCIATES
X-RAY DIFFRACTION MINERALOGICAL ANALYSES

Mineral	Concentration (%)		
	Sample 9	Sample 10	
Calcite	1	-	
Hematite+/Goethite	-	-	
Microcline	1	-	
Plagioclase	2	Trace	
Quartz	72	84	
Mica	6	3	
Chlorite	13	6	
Amphibole	1	-	
Pyrophyllite	4	3	
Gypsum	-	4	
Ilmenite/Smectite Interstratification	1	-	

per.....
Waterlab (Pty) Ltd

Waterlab (Pty) Ltd / (Edms) Bpk
(Reg Nr/No 83/09165/07)

Management & Laboratory Services for Water and Wastewater
Environmental Monitoring and Impact Assessment

PO Box 11508 Hatfield 0028 (T) 012-349-1044/1066 (F) 012-349-1072
E-mail: waterlab@iafrica.co.za / www.waterlab.co.za

WATERLAB

CERTIFICATE OF ANALYSIS

ATTENTION: Mr. P. Rousseau | **DATE: 19/08/2003** | **ORDER NO : 000590**
REPORT NO: 11553 | **PROJECT NO: 159** | **SAMPLES: 6028-6042**

GOLDER ASSOCIATES AFRICA (Pty) Ltd
ACID BASE ACCOUNTING : EPA-600 Modified Sobek

Sample	Paste pH	Total S (%)	Acid Generation Potential (AP) $CaCO_3$ (kg/t)	Neutralisation Potential (NP) $CaCO_3$ (kg/t)	Netto Neutralisation Potential (NNP) $CaCO_3$ (kg/t)	AP:NP Ratio	Rock Type
Sample 2B	8.26	0.315	9.84	7.50	-2.34	1 : 0.8	I
Sample 3	8.47	0.322	10.06	12.50	2.44	1 : 1.2	II
Sample 4	8.32	0.356	11.13	9.25	-1.88	1 : 0.8	I
Sample 5D	8.28	0.239	7.47	8.25	0.78	1 : 1.1	II
Sample 6B	8.37	0.173	5.41	13.00	7.59	1 : 2.4	II
Sample 6C	8.22	0.286	8.94	7.75	-1.19	1 : 0.9	I
Sample 7A	8.16	0.308	9.63	9.00	-0.62	1 : 0.9	I
Sample 7B	7.26	0.390	12.19	6.50	-5.69	1 : 0.5	I
Sample 8	8.28	0.365	11.41	8.75	-2.66	1 : 0.8	I

***Negative NP values are obtained when the volume of NaOH (0.1N) titrated (pH:8.3) is greater than the volume of HCl (0.1N) to reduce the pH of the sample to 1.5 – 2.0. Any negative NP values are corrected to 0.00.**

Please refer to Appendix (p.4) for a Terminology of terms and guidelines for rock classification

per.....
Waterlab (Pty) Ltd

APPENDIX : TERMINOLOGY AND ROCK CLASSIFICATION

TERMINOLOGY (SYNONYMS)

- Acid Generation Potential (AP) ; *Syn*: Maximum Potential Acidity (MPA)
  Method: Total S(%) (Leco Analyzer) x 31.25

- Neutralization Potential (NP) ; *Syn*: Gross Neutralization Potential (GNP) ; *Syn*: Acid Neutralization Capacity (ANC)
  Method: Fizz Test ; Acid-Base Titration (Sobek & Modified Sobek (Lawrence) Methods)

- Nett Neutralization Potential (NNP) ; *Syn*: Nett Acid Production Potential (NAPP)
  Calculation: NNP = NP – AP ; NAPP = ANC - MPA

ROCK CLASSIFICATION

TYPE I	Potentially Acid Forming	Total S(%) > 0.25% and AP:NP ratio 1:1 or less
TYPE II	Intermediate	Total S(%) > 0.25% and AP:NP ratio 1:3 or less
TYPE III	Non-Acid Forming	Total S(%) < 0.25% and AP:NP ratio 1:3 or greater

REFERENCES

Lawrence, R.W. & Wang, Y. 1997. ***Determination of Neutralization Potential in the Prediction of Acid Rock Drainage***. Proc. 4th International Conference on Acid Rock Drainage. Vancouver. BC. pp. 449 – 464.

Sobek, A.A., Schuller, W.A., Freeman, J.R. & Smith, R.M. 1978. ***Field and laboratory methods applicable to overburdens and minesoils***. EPA-600/2-78-054. USEPA. Cincinnati. Ohio.

Environment Australia. 1997. ***Managing Sulphidic Mine Wastes and Acid Drainage***.

APPENDIX B

CD containing the report as a printable pdf file and a copy of the TAP excel spreadsheet model

Other related WRC reports available:

Rehabilitation of contaminated gold tailings dam footprints

Hattingh RP; Lake J; Boer RH; Aucamp P; Viljoen C

An earlier preliminary assessment of pollution contained in the unsaturated and saturated zones beneath reclaimed gold-mine dumps, indicated that this pollution can have a significant negative environmental impact on the underlying vadose zone and subsequently the groundwater system. This study identified five alternative rehabilitation strategies to deal with this pollution and modelled their expected impact at three study sites. The strategies were found to represent mainly two opposing alternatives, *i.e.* those that result in the pollutant remaining within the confines of the residue deposit area, and those in which the pollutants are transported to the deeper aquifer, away from the source. The preferred management option would thus depend on whether containment or dilution and migration of pollutants would be the desired option. The problems associated with reclaimed mine-residue deposits were also assessed by evaluating the various risks that are associated with the source of contamination, the pathway it would follow and the receptor that would be adversely affected by the contaminated substance. Site-specific variation in physical characteristics was found to have a marked influence on the resultant risk assigned to individual aspects. The introduction of a risk assessment approach, a phased and chemically orientated approach to site investigation, and the selection of remediating strategies from a number of technologies, were identified as intrinsic components of the rehabilitation process. This would require that rehabilitation be conducted on a site-specific basis. The basic methodology for this approach is outlined in a companion document to the project report.

Report Number: 1001/1/03 **ISBN No: 1 77005 006 X**

TO ORDER: Contact **Publications** - Telephone No: 012 330 0340
Fax Number: 012 331 2565
E-mail: publications@wrc.org.za

Water Research Commission
Private Bag X03, Gezina, 0031, South Africa
Tel: +27 12 330 0340, Fax: +27 12 331 2565
Web: http: www.wrc.org.za