

Waste Rock Dump Geochemical Evolution: Matching Lab Data, Models and Predictions with Reality

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ABSTRACT

The prediction of how waste materials will evolve geochemically within waste rock storage facilities (WRSF) has been the subject of many hundreds of thousands of hours’ research by geoscientists globally. Despite the quantity of research there are two significant areas of uncertainty that remain as partially resolved issues: (a) the majority of the research carried out has been at the laboratory not site scale (b) many predictions made using computer modelling have not, or are not normally able to be validated from site data.

OKC has had the opportunity to lead two large scale WRSF drilling programs at different sites in Western Australia where 12 historical waste dumps of around 10-30 years in age have been subject to detailed intrusive investigation. This has included approximately 2,000 m of sonic drilling, the recovery and detailed analysis of over 2000 samples of core material and the installation of over 150 sensors at depths between 5-140 m within the waste which have provided over 5 million points of monitoring data.

The extensive data gathered as part of the assessment has allowed OKC to determine the geochemical, hydrological and geophysical evolution of the waste rock facilities as a result of 10-30 years of exposure. This valuable site data has been used to both back test the results of predictive models made for the waste facilities, and to optimise predictive models for future scenarios. In addition the data has allowed the development of scaled up site specific kinetic leach columns.

Keywords: Kinetic testing, geochemical evolution, scale parameters, intrinsic oxidation rate

INTRODUCTION

The prediction of how waste materials will evolve geochemically within WRSFs has been the subject of many hundreds of thousands of hours of research by geoscientists globally. Despite the quantity of research there are two significant areas of uncertainty that remain as partially resolved issues: (a) the majority of the research carried out has been at the laboratory not site scale (b) many predictions made using computer modelling have not, or are not normally able to be validated from site data.

As part of a large scale investigation to improve the understanding of waste rock geochemistry at macro scale OKC has had the opportunity to lead two large scale WRSF drilling programs at different sites in Western Australia. 12 historical WRSFs of around 10-30 years in age have been subject to detailed intrusive investigation. This has included approximately 2,000 m of sonic drilling, the recovery and detailed analysis of over 2,000 samples of core material and the installation of over 150 sensors at depths between 5-140 m within the waste which have provided over 5 million points of monitoring data.

This paper focuses specifically on research carried out on sites within a semi arid climate and waste rock containing sulfidic black shale material taken from mines within the Pilbara in Western Australia.

LABORATORY TESTING AND THE ISSUE OF SCALABILITY

Laboratory testing methods have been developed and refined over many years for geochemistry assessment and AMD prediction which has resulted in a broad set of standardised testing methods being generally accepted internationally by the mine-geochemistry industry. This has been possible by and large because laboratory experiments are completed in a controlled environment with high degree of precision and accuracy. However a common factor and limitation to all laboratory assessments is that the scale at which these experiments are carried out.

Kinetic testing methods offer a good example of the issue of trying to determine how laboratory data should be used to extrapolate from the laboratory (micro) to site (macro) scale. Common kinetic tests such as humidity cells and leach columns utilise a few kilograms of material that has been crushed to a few millimetres in size that are then exposed to wetting and drying cycles with very high leaching rates. The concept of these kinetic tests is that by using small grain sizes and high leaching ratios the weathering processes (sulfide oxidation rates and carbonate dissolution) can be accelerated allowing the assessment of sulfide oxidation rates, acidity generation, neutralisation reactions, and leachate geochemistry to be assessed within a reasonable timeframe. It has been proven that these tests achieve the aim of simulating accelerated weathering. However, the precise method (and value) of how to utilise these results in a scaled up situation, for example in the extrapolation of data to predictions of field conditions such as oxidation rates or seepage quality has not yet been conclusively determined.

Table 1 outlines some scale factors that require consideration when using kinetic test data for “predictions” of field conditions, which will have a significant influence on the ability to predict field geochemical conditions and processes from laboratory data.

Table 1 Scale variables for laboratory kinetic tests compared to field conditions for medium to large size waste rock storage facility (>1 million tonnes)

Scale parameter	Laboratory test conditions	Typical Australian field conditions	Challenges extrapolating from lab to field
Grain size	PSD = 100% <6mm	PSD = 40% <6mm	Oxidation rates faster and dissolution of silicates/carbonates higher for smaller grain sizes. Lab tests may over estimate both sulfide oxidation rates and acidity buffering processes
Geochemical system	Open	Variable from open to closed	Open systems discharge acidity closed systems store acidity, lab tests are not reflective of field drainage geochemistry
Oxygen consumption measurement	Estimate from sulfate release	Estimate from in situ measurement	Using sulfate produced from leaching tests may not provide accurate prediction of oxygen consumption
Mass of material	1-2 kg	>1 Mt	Heterogeneity effects not accounted for in lab tests, bulk geochemistry of material in the field may not be well represented by material tested in the lab
Air flow (Oxygen supply)	Diffusion dominated unrestricted	Advection dominated potentially restricted	Lab tests assumed unlimited oxygen supply, field conditions may vary, generally significantly lower than lab conditions
Temperature	20-30 degrees	0-100+ degrees	Due to effect of thermal properties by total mass of material, field temperatures may be significantly higher than lab, this can effect geochemical reaction rates
Liquid : solid ratio	8:1 per year	0.001: 1 per year	Lab leachate is more dilute then field conditions due to high L:S ratio. No restriction on sulfide oxidation rates based on H ₂ O supply in lab, field reaction rates may be H ₂ O supply limited.

REACTION KINETICS IN CLOSED AND OPEN SYSTEMS: THE SUPPLY OF O₂ AND H₂O AS CRITICAL FACTORS

At the most basic level sulfide oxidation reaction kinetics are controlled by the relative supply of O₂ and H₂O. The commonly stated (simplified) reaction for iron sulfide oxidation in this case pyrite, to form sulphuric acid H₂SO₄ and ferrihydrite Fe(OH)₃ is (from Lottermoser, 2010):



It is important to note that this reaction represents the interaction of O₂ with FeS₂ (pyrite) with H₂O thought to be acting as a catalyst. It is clear then that if either water or oxygen are not present then the reaction will not proceed, the kinetics of the reaction will therefore be determined by the relative supply of both. Lottermoser (2010) states that “*there is little consensus in the literature on the precise reaction mechanisms describing the oxidation of pyrite*”. Therefore it is not clear how the varying

supply of O₂ and H₂O will impact the pyrite oxidation reaction kinetics. It is however widely accepted that “the transport of oxygen to the oxidation sites is considered the rate limiting process in dumps” Lottermoser (2010). For example from equation (1) 7/2 moles H₂O are required for oxidation of 1 mole of FeS₂. However, in field conditions reactions take place in pore spaces of unsaturated waste rock where the pyrite mass is disseminated (at grades of a few percent) through the host rock. As a result this molar ratio of H₂O to Pyrite on a per kg rock mass basis will not be sufficient because at such low water contents the pore waters may not physically be in contact with sulfide mineral surfaces. The supply of O₂ and H₂O is in turn determined by the nature of the geochemical system as open or closed. In an open system there is a potentially unlimited supply of O₂ and H₂O into the system and a means for reaction products to exit the system. These systems are represented by laboratory free draining leach columns. A closed system in contrast is represented by either a restriction of supply of oxygen or water or a restriction on the means for reaction products to exit the system. With respect to field conditions the internal zones of WRSFs can vary between open and closed system conditions, and can often fluctuate between conditions on a seasonal basis.

Supply of H₂O, O₂ and the intrinsic oxidation rate (IOR)

The liquid to solid ratio (L:S) is a convenient way to express the relative supply of H₂O into the system, this simply reflects the weight for weight balance of water against the mass of the porous solid through which the liquid is passing. Free draining leach columns have very high L:S ratios in general, an AMIRA (IWRI and EGi, 2002) column has an annual L:S ratio of approximately 8:1 for example. In contrast WRSFs typically have low L:S ratios and annual ratios in the Pilbara are estimated to be around 0.001:1 (assuming a 40 m high WRSF has 400 mm of net percolation per annum).

Oxygen supply is dominated by the degree of saturation of the pore space within the material, and the process of air movement which can be driven by advective or diffusive processes. Within free draining leach columns the supply of oxygen is driven by diffusion as no advective forcing is applied. In WRSFs the supply of oxygen can be driven by advection or diffusion depending on site conditions. The work of Brown *et al.* (2014) indicates that convection of oxygen accounts for 90% of the oxygen transport into a WRSF and diffusion accounts for 10%. This is close to an order of magnitude difference between the two oxygen ingress processes.

The IOR is the oxidation rate where oxygen is freely available (Bennett *et al.*, 1995). It has been reported that based on free draining leach column tests that the IOR ranged from 1.6×10^{-8} to 3.3×10^{-6} kg O₂/m³/sec (EGi, 2001) for samples of pyritic shale materials (Mt. McRae shale) sourced from the Pilbara, Western Australia. Levay and Co (2013) report an acidity generation rate of ~1.5 kg H₂SO₄/t/week for a Mt. McRae shale sample having 3.2 wt.% S, which is equivalent to an IOR of 2.5×10^{-6} kg O₂/m³/sec and comparable to the EGi (2001) work.

Because an IOR of 2.5×10^{-6} kg O₂/m³/sec has been calculated from a free draining leach column then it is assumed that given an unrestricted oxygen supply that this is the minimum oxygen requirement for the pyrite oxidation reaction not to be rate limited. On the same basis the amount of H₂O required for the reaction to not be rate limited can also be calculated. Based on reaction (1) and the production of 1.5 kg H₂SO₄/t/week requires a minimum of approximately 0.5 kg t/week of H₂O, which can be converted into an L/S ratio of 0.026 per year. Clearly given the L:S ratio is 8:1 in

column tests then there is a significant excess of H₂O for pyrite oxidation not to be rate limited, in fact there is approximately 300 times excess H₂O. It should be noted that the use of such excess H₂O in these tests will inevitably result in significant dilution of leachates, that is to say leachate strengths are unlikely to be representative of field conditions. For materials with low sulfide contents this is likely to be of particular concern as these tests will tend to significantly underestimate the concentration of contaminants of AMD leachates in the field

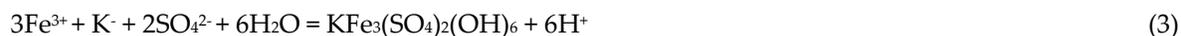
It is widely accepted in the industry that laboratory column leach tests and similar field based lysimeter trials provide elevated reaction rates compared to the field (Miller *et al.*, 2003; Andrina *et al.*, 2012). For example the calculated IOR for waste rock was an order of magnitude lower in trial dump experiments compared to a 500 tonne trial and lab columns in the work of Andrina *et al.*, 2012. Most importantly, Andrina *et al.* (2012) notes that the trial dump was often well oxygenated with oxygen content often at 20%, which is comparable to atmospheric concentrations. Based on these studies when oxygen is generally freely available it may be proposed that the laboratory based IOR can be reduced by an order of magnitude due to scaling effects, which, for the purpose of this report is referred to as Laboratory to Field Conversion Factor (LFCF). However, as demonstrated by Table 1 there are many scaling effects that may impact the determination of field IOR rates, in addition the method of estimating IOR from sulfate production in these tests is subject to a degree of error that may be significant depending on the geochemistry of the situation.

WEATHERING PRODUCTS OF PYRITE OXIDATION: GEOCHEMICAL CONSIDERATIONS

Although a simple summary equation to represent the oxidation of pyrite (Equation 1), the weathering of metal sulfides in the environment do not often progress as shown in the direct production of H₂SO₄ and Fe(OH)₃. This is because interaction of the H₂SO₄, Fe^{x+}, H⁺ or SO₄²⁻ with H₂O and O₂ results in the formation of many different compounds which are often meta-stable in the surface environment. Further interaction with H₂O and O₂ can occur depending on the pH/Eh of the geochemical environment. This simplified model of pyrite oxidation does not tell the whole story. Sulfide weathering has the potential to release all the acid 'potential' by the precipitation of hydroxides and oxides such as goethite or ferrihydrite:



This process does not store H⁺ as stored metal acidity and releases maximum acidity. Hudson and Edwards *et al.* (1999) point out that even low concentrations of SO₄²⁻ in solution can suppress the formation of Fe hydroxides, and instead favours the formation of oxyhydroxysulfates. This alternative is the storage of acidity in secondary salts, which are only stable in oxidising acidic pH conditions. For example in the formation of jarosite:

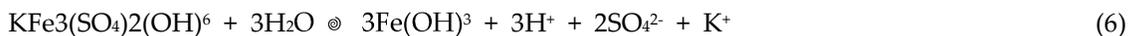


Often there is incomplete oxidation of the ferrous (Fe²⁺) iron to ferric (Fe³⁺) iron and ferrous salts such as melanterite, FeSO₄ (Equation 4) can form, which following any subsequent wetting can release the stored ferrous acidity (Equation 5). These acid salts are highly soluble.





However, if oxidation to ferric iron is complete yet the hydrolysis is incomplete, jarosite type secondary minerals can form. Jarosite type minerals form at pH values below 3.5 and release only 2H⁺ per Fe³⁺ incorporated into jarosite, not the associated 3H⁺ ions associated with complete Fe hydrolysis. Thus jarosite type minerals store acidity that can be released once pH increases (Equation 6). Jarosite is stable at pH values < 4 and above pH 4.7 it is soluble, dissolving slowly (Li et al., 2007), which has long term implications for the rebound of pH to circum-neutral conditions after sulphide exhaustion and/or for the treatment of AMD impacted waters.



The acid load associated with the dissolution of ferrous salts such as melanterite will occur immediately upon wetting. The precipitation and dissolution of these sulfate bearing minerals will exert a significant control on the sulfate produced in leachates and therefore the ability to accurately predict IOR rates from laboratory leaching experiments.

In semi-arid environments the precipitation of these secondary sulfate minerals as a result of low L:S ratios may therefore be a significant factor in determining how much acidity is released from the WRSF.

OPEN AND CLOSED GEOCHEMICAL SYSTEMS

Table 2 outlines the importance of consideration of the geochemical system being studied as open or closed. Laboratory methods are open systems and will generally have unlimited oxygen supply and very high L:S ratios so oxidation reaction rates are not rate limited. In addition these conditions will limit the potential storage of acidity as secondary sulfate minerals, which will also result in high output of dissolved phase oxidation reaction products. Field conditions in comparison represent a variable state system which may have a seasonal aspect as the system ranges from open to a closed state. It should be noted that contrary to widespread belief oxygen supply can still represent an unlimited condition in WRSFs as a result of advective air movements in coarser zones of waste. Whereas in semi-arid environments the supply of H₂O is likely to be the limiting factor with respect to formation of an open system due to low L:S ratios.

Table 2: Open and closed systems

Scenario	System	Oxygen supply	H ₂ O supply	Storage of acidity as meta stable minerals	Output of Fe ²⁺ , Fe ³⁺ , H ⁺ and SO ₄ ²⁻ in leachate
AMIRA Leach column	Open	Diffusion controlled potentially unrestricted	Unrestricted. Annual LS ratio 8:1	Low	High due to high flushing rates (L:S ration 8:1)
Waste rock prior to "wetting up"	Closed	Advection controlled potentially unrestricted	Restricted: depth variable dependence,	Very high	Low due to very low flushing rates
Waste rock "wetted up"	Open	Advection controlled potentially unrestricted	Limited to net percolation rate, Annual LS ratio of 0.001:1 common	Variable	Variable due to seasonality of flushing rates. Annual L:S ratio of 0.001:1 common

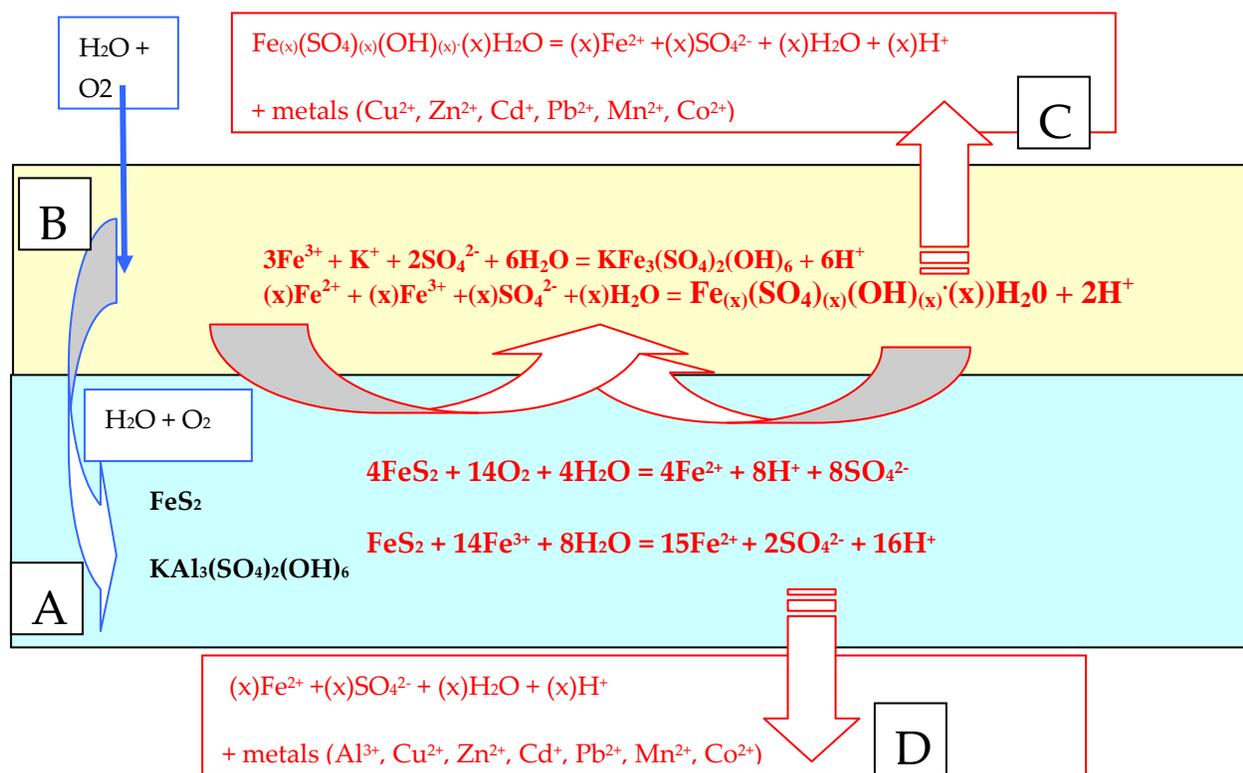


Figure 1 Simplified geochemical model of sulfide oxidation showing open and closed system pathways

Figure 1 shows the simplified geochemical model for initial stages of sulfide oxidation in an open system

- Box A indicates sulfide minerals that are undergoing oxidation in the presence of aluminium and potassium bearing silicate minerals and in contact with an unrestricted supply of oxygen and water resulting in production of Fe²⁺, Fe³⁺, H⁺ and SO₄²⁻
- Box B indicates production of secondary oxyhydroxysulfates on mineral surfaces within pore spaces from dissolved phase species released from Box A. This process is likely to dominate in a closed system where L:S ratios are low and precipitation of these minerals exceeds losses from leachate discharges
- Box C indicates a situation where the precipitated oxyhydroxysulfates dissolve to form Fe²⁺, Fe³⁺, H⁺ and SO₄²⁻ and H₂O. This situation is representative of an open system where L:S ratios are high enough to dissolve any secondary minerals present that are soluble.
- Box D indicates the situation where the soluble products from Box A are directly discharged from the system. This is an open situation common to free draining laboratory kinetic tests where L:S ratios (flushing rates) are high. In this scenario secondary minerals (Box B) do not have much chance to form due to high dissolution rates caused by high L:S ratios.

CASE STUDY: INTRUSIVE WRSF INVESTIGATION

The presented study includes 12 WRSFs at various mine sites in the Pilbara, Western Australia. The mine sites are made up of multiple WRSFs which have been constructed by various techniques including end dumping. A proportion of the dumps contain pyritic black shale with some incorporating encapsulation techniques. OKC completed a drilling programme that resulted in 2,000 m of sonic drilling, the recovery and detailed analysis of over 2000 samples of core material the installation of over 150 instruments within 12 WRSFs up to a depth of 100 m. The WRSF monitoring systems were equipped with instrumentation to measure *in situ* moisture, oxygen pore-gas concentrations, pore-water pressure and *in situ* temperature within the WRSFs.

The Pilbara region consists of a climate classified as arid-tropical. Summers last from October to April and mild winters occur from May to September. Sporadic and intense thunderstorms are typical for the region from January to March, and tropical cyclones can result in daily rainfall amounts of up to 200mm over a 24hr period.

Presentation and analysis of the data collected from the instrumentation to date is beyond the scope of this paper and is reported in a separate paper published in Pearce and Barteaux 2014. A summary of the results is provided below which is important for understanding of field conditions within the WRSFs that are driving geochemical conditions:

- Wetting up of the WRSF's is not occurring uniformly throughout the waste. Generally the coarse cobble zones did not match the wetted-up model as moisture contents were lower than expected, whereas the finer textured materials had moisture contents closer to the wetted up model scenario.

- Based on responses of installed instrumentation the internal movement of water is considered to be governed by preferential flow paths predominantly within zones of coarser waste rock particle sizes. Fast responses to rainfall events are noted in these zones through the middle of the WRSFs, which can be tracked to basal zones of the WRSFs.
- The results of moisture content analysis indicates that water content varied between the borehole locations with the majority of results ranging from 3-15% by volume. If the moisture content is assumed to be on the upper end of the estimate at 10% on a mass basis this represents a current static L:S ratio of 0.1.
- Oxygen sensors installed indicate the WRSF's have high internal air flow rates and connectivity which is providing ample supply of oxygen to the whole waste rock profile with oxygen levels of between 16-20% being recorded through the whole profile of the waste in many WRSFs. In general oxygen concentrations increase with depth indicating convective supply source.
- Elevated internal dump temperatures (30-40 °C), high (and connected) internal air flows and supply of H₂O within preferential zones are postulated to be driving evaporative drying of pore waters which results in the transport and re distribution of water in the gaseous phase within the waste mass throughout much of the profile. This mechanism is considered to comprise a significant means of transfer of H₂O within the WRDs from zones of preferential flow to zones that do not receive much direct net percolation

Given the above observations it may be inferred that oxidation reactions within the WRDs studied may not be O₂ limited, but are more likely to be limited by the supply and movement of H₂O.

Geochemical analysis of drill core supports this conjecture as there is a significant presence of secondary sulfate minerals which are indicative of precipitation of sulfate bearing minerals as a result of stationary pore fluids (and therefore a very low L:S ratios). The geochemical system can therefore be best thought of as semi closed in that oxygen and H₂O (possibly supplied from internal evaporation rather than matrix pore water flow) can enter the waste but very limited leachate leaves the system. This results in a buildup of secondary sulfate minerals. Sulfur speciation results from analysis of black shale samples suggest a high degree of oxidation of sulfides for a large proportion of samples tested. From older WRSFs (30 yrs+) approximately 1500 samples were analysed from 49 boreholes, the average and median percentages of total sulfur that was in the form of sulfate was 80% and 72% respectively. The average proportion of total sulfur present as sulfate was 80%. At higher grades of >1% sulfur the sulfate ratio is slightly lower, around 60% average. It is assumed based on understanding of the mineralogy of the black shale that the material when deposited comprised sulfide as pyrite and little sulfate minerals. It is assumed therefore that the majority of sulfate present as measured represents secondary sulfate minerals that are the result of oxidation of the pyrite.

If it is assumed that the oxidation reactions have been occurring for approximately 30 years and no loss of sulfate from the system has occurred then IOR rates are determined as followed:

- IOR field = 3%S @ 60% oxidation over 30yrs = 0.04 kg H₂SO₄/t/week = 7.5⁻⁸ kg O₂/m³/sec
- IOR field = 9%S @ 60% oxidation over 30yrs = 0.1 kg H₂SO₄/t/week = 1.9⁻⁷ kg O₂/m³/sec

The field IOR rate calculated is considered a lower bound estimate as reaction rates may have declined with time as a result of fresh sulfide surfaces being smothered by secondary minerals. Laboratory calculated values lie in the range 4.9E-7 to 2.9E-6 kg O₂/m³/sec. In general the results of the case study assessment broadly agree with Andrina *et al.* (2012) where an order of magnitude difference in IOR was observed between field and lab IOR rates. It is interesting to note that even with the very low L:S ratios observed in the WRSFs that oxidation reactions have proceeded to 60% or greater completion. If it is assumed based on field monitoring data that oxygen is not the limiting factor, the main causes of the lower observed field rates are likely to be:

- The impact of restricted water supply and time taken for “wetting up” to occur
- Formation of secondary sulfate mineral coatings around sulfide grains as a result of low matrix pore water flow and leachate “flushing”
- Larger particle sizes than laboratory tests

CONCLUSIONS

The use of laboratory leach column data for field estimates of IOR and seepage quality requires careful consideration as scaling factors will considerably impact the validity of the results. For WRDs in semi arid climates such as the Pilbara the low rainfall environment creates conditions of very low L:S ratios and geochemical systems within WRDs may remain in a quasi closed state for many years. High levels of oxygen ingress are indicated to occur as a result of high internal WRSF air temperatures and low waste rock saturation levels which means that sulfide oxidation reactions may become limited to a greater extent by net percolation rates than oxygen supply. It is noted that even with very low WRSF L:S ratios (0.1 or less), oxidation reactions have proceeded to 60% or greater completion. Storage ratios of acidity as secondary sulfate minerals are high during the period that the WRD remains as a quasi closed system. As a result future predictions of seepage quality must therefore consider the impact that future dissolution of these minerals will have if the WRD approaches a “wetted up” state. Laboratory leach columns in comparison to field conditions are characterised by high L:S ratios and low levels of stored acidity. When extrapolating laboratory results to field conditions in semi arid environments therefore it must be considered that leach columns data may overestimate IOR (by an order of magnitude), but likely significantly underestimate potential seepage quality compared to field conditions.

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