The Evolution of Tailings Seepage Chemistry at One of Australia's Largest and Longest Operating Mines

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ABSTRACT

The Mount Isa mines have been active for more than 90 years and have played a significant role in the economy of Queensland. Process tailings from both copper and lead-zinc production streams are deposited in a large valley fill tailings storage facility which currently has an approximate surface area of 14 km². The site is a world class sedimentary deposit with climatic conditions typified by prolonged dry seasons with relatively short, high intensity wet seasons. This paper discusses the mineralogical and geochemical characterization of the tailings, and the assessment of the potential for metal leaching, to aid mine closure planning.

Historical tailings at Mount Isa are generally non-acid forming based on conventional acid base accounting, mineralogical examination, and kinetic testing. However, modern tailings are characterized by higher sulfide and associated trace element content, and lower carbonate mineral content than historical tailings. Changes in tailings composition are likely attributable to changes in ore and mineral processing methods, and time since tailings deposition. Strategies are currently being developed for the placement of a suitable cover system to reduce surface water – tailings interaction following decommissioning of the TSF. This study demonstrates the benefit of integrating detailed geochemical characterization into closure and rehabilitation planning.

Keywords: Mount Isa Mine, tailings, seepage

INTRODUCTION

Mount Isa is located in North-West Queensland, Australia, approximately 360 km south from the Gulf of Carpentaria and 960 km west of the Coral Sea (20° 43' S, 139° 29' E) (Figure 1). Mineralization in the Mount Isa region was first discovered in 1923 and subsequently a large number of mining leases were established with ore extracted through numerous shallow open pit operations. In 1924, Mount Isa Mines was incorporated, consolidating a number of the major leases and initiating the development of an underground mining operation. Today, ore is sourced from both underground and open pit mines. Copper ore is mined and processed separately from lead and zinc ore. The ore is concentrated by grinding and flotation, followed by smelting. Typically 30% of the combined copper and lead-zinc tailings stream is utilized as hydraulic fill in underground mine operations and the remaining portion is sent to a tailings storage facility (TSF).
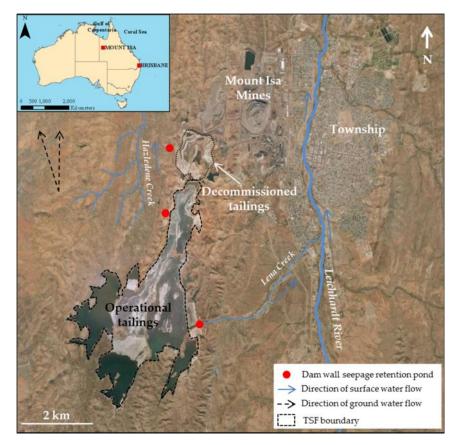

Figure 1 Site locality

The TSF is approximately 14 km^2 in surface area. The area is contained by three visible dam walls, with a further two dam walls buried under a waste rock dump to the north. Toe seepage emanates from the base of the dam walls at rates of less than 1 to 5 L/s and is captured in retention ponds prior to being pumped back to the TSF. It is characterized by neutral pH (7.3 to 8.2) and elevated sulfate (3100 to 8400 mg/L SO$_4$), with concentrations of arsenic, boron, cadmium, cobalt, copper, lead, manganese, and zinc slightly elevated in comparison to Australian National Water Quality Guidelines for 95% aquatic ecosystem protection (ANZECC, 2000) and regional surface and ground waters.

The TSF is composed of decommissioned cells to the north, and operational cells to the south (Figure 1). Decommissioned cells were operational from the early 1950s to the mid-1960s. Tailings within the decommissioned cells (hereon referred to as 'decommissioned tailings') have been exposed to approximately 40 years of weathering and exposure. Since decommissioning of the northern cells, tailings have been actively deposited within the operational cells ('operational tailings'). Tailings deposition is cycled through the different operational cells so that the operational tailings sampled by this study had been exposed for less than two years.

This paper discusses the mineralogical and geochemical characterization of the tailings, and the potential release of metals and acidity due to long-term mineralogical weathering.

Climate and Geology

The climate of Mount Isa is semi-arid and subtropical, characterized by average annual pan evaporation (3100 mm) exceeding average annual rainfall (430 mm), and warm to hot temperatures (19 to 32 °C mean monthly minimum and maximum) (BoM, 2012). On average, 75% of annual rainfall occurs between the months of December and March ('wet season') with the intervening months of April to November typified as the 'dry season'.

The Mount Isa Mines' ore deposits are located within the Mount Isa Inlier; an area of more than 50,000 km^2 of Early and Middle Proterozoic rocks host to economically significant quantities of lead-zinc, copper, uranium, cobalt, and gold (Blake *et al.*, 1990). Mount Isa shares some geological similarities to other well-known shale-hosted massive sulfide deposits, such as the Red Dog and Lik deposits in Alaska (Kelley *et al.*, 1995; Kelley and Taylor, 1997), and McArthur River and Broken Hill deposits in Australia (Huston *et al.*, 2006).

A distinct feature of Mount Isa is the occurrence of spatially separate lead-zinc and copper ore bodies. Both occur within the Urquhart Shale, a unit of metamorphosed pyritic and dolomitic shale approximately 1000 m thick at the mine. The main economic sulfides within lead-zinc ore are galena (PbS) and sphalerite (ZnS) with minor amounts of silver occurring in close association with galena. Copper ore principally occurs as chalcopyrite ($CuFeS_2$). The main gangue sulfide is pyrite (FeS_2). The main carbonates are dolomite ($CaMg(CO_3)_2$) and calcite ($CaCO_3$), with ankerite ($Ca(Fe,Mg)(CO_3)_2$) and siderite ($FeCO_3$) also present. Other gangue minerals include quartz, chlorite, K-feldspar, muscovite, biotite, albite, rutile, and carbonaceous material (Heinrich *et al.*, 1995; Chapman, 2004).

METHODOLOGY

Sample Collection

Decommissioned and operational tailings were sampled to less than 1 m depth with the assistance of an excavator, and to depth using a truck-mounted drill rig equipped with a push-in piston sampler. Drill cores were sub-sampled at 1 m intervals from 0 to 3 m below the tailings surface, and at 3 m intervals to bedrock (typically 27.5 m below surface). A total of 20 samples from four locations within decommissioned tailings, and 30 samples from 10 locations within operational tailings were collected.

The selection of sample sites at Mount Isa was ultimately constrained by accessibility. The unconsolidated nature of operational tailings restricted sampling to areas accessible by internal roads that run the length of the TSF. Analytical data were interpreted with supporting information

from secondary sources including aerial photography, ore geology and mineral processing publications, concentrator records, annual Xstrata sustainability reports, and personal communication with long-term mine staff.

Mineralogy

The mineral composition of tailings samples was determined using X-ray diffraction, optical microscopy, Scanning Electron Microscopy coupled with Energy Dispersive Spectroscopy (SEM-EDS), and Mineral Liberation Analysis (MLA). All analyses were undertaken at the University of Queensland.

Static Testing

Tailings were analyzed for acid base accounting (ABA) (AMIRA International, 2002) and elemental composition by four-acid digestion and ICP-MS analysis. Maximum Potential Acidity (MPA) was calculated using sulfide sulfur corrected for non-acid generating forms of sulfide, specifically sphalerite and galena using zinc and lead concentrations, respectively. Acid Neutralization Capacity (ANC) was measured using the Sobek method modified to oxidize and hydrolyze dissolved ferrous iron during the titration step (AMIRA International, 2002). Analyses were undertaken at the University of Queensland and a commercial laboratory (ALS) in Brisbane, Australia. Quality checking and assurance consisted of duplicate, certified reference material, and method blank inclusion in each sample batch.

Column Leaching

Three decommissioned and three operational tailings column samples were extracted from in-situ, near-surface (0 to 1 m depth) deposited tailings. The samples were collected intact with minimal disturbance to stratigraphy by inserting the column tubing housed within a soil corer directly into the tailings profile. The columns were 125 mm in length and 70 mm in internal diameter. The tailings were leached with deionized water supplied under constant head pressure using a Mariotte-type design. A flow rate of 2 to 5 mL/hr was targeted as this allowed the accumulation of leachate sufficient in volume to be collected at least once every two days. The columns were subject to four phases (Figure 2):

a) Saturated leach 1. Deionized water was slowly introduced from the column base over the course of one to two weeks to exclude air from pore spaces while minimizing loss of pore structure from increasing pressure, and then leached under saturated conditions until electrical conductivity showed little variation between collected samples (typically two to three months).

b) Suction and aeration. Drying was induced within the columns by applying suction (-15 to -60 kPa). The concept was to empty pores of water and increase oxygen ingress. Compressed air was fed directly into the tailings via an inlet on the side of each column for a period of one week. This was to encourage oxidation reactions and to hasten the mineral weathering process.

c) Saturated leaching 2. Deionized water was re-introduced in order to mobilize any soluble constituents released as a result of the suction and aeration step.

d) End of leach characterization. A non-reactive tracer test (KCl) was conducted to estimate solute transport parameters. Chloride break-through curve data were modelled using a non-equilibrium dual porosity, Mobile-Immobile Model within Hydrus-1D (Version 4.14) (Simunek *et al.* 2008). Further method detail is provided by Forsyth *et al.* (2013).

Collected leachate was analyzed for pH, electrical conductivity, oxidation-reduction potential, sulfate, chloride, major cations by ICP-AES, and trace elements by ICP-MS. Element release rates (mg/kg/day) were calculated for leach phases 1, 2a, and 2b as shown in Figure 2.
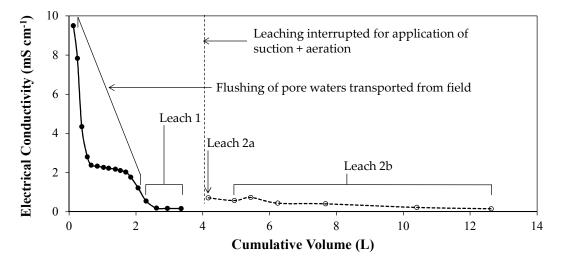

Figure 2 Column leach phases

RESULTS AND DISCUSSION

Tailings Characterization

Mineralogy

Bulk mineralogy was similar between decommissioned and operational tailings. The main minerals were quartz, K-feldspar, chlorite, muscovite, dolomite, calcite, and pyrite. Quantitative XRD showed that operational tailings contained more pyrite (12 to 15 wt. %) and less dolomite (6 to 16 wt. %) than decommissioned tailings (1 to 2 wt. % and 11 to 18 wt. %, respectively). Pyrrhotite, chalcopyrite, galena, sphalerite, ankerite, and siderite were observed in low abundance (less than 2 wt. %) in both tailings types.

Secondary minerals within decommissioned tailings included goethite and gypsum in sub-surface samples, and gypsum, hexahydrite, jarosite, melanterite/rozenite, chalcanthite, and poitevinite in surface samples. SEM-EDS analysis on decommissioned tailings indicated the presence of poorly crystalline iron (oxy)hydroxides, iron oxyhydroxysulfates, and a calcium-iron-silica gel of variable chemical composition. Secondary mineralization in operational tailings was restricted to gypsum.

Acid Base Accounting

ABA classifications were defined using ANC/MPA ratios (Figure 3), with samples forming four groups reflective of their origin within the TSF:

a) Group 1: decommissioned tailings, 0 to 1 m depth below surface. Samples within this group were depleted in ANC (geometric mean 110 kg H_2SO_4/t) and MPA (20 kg H_2SO_4/t) as a result of over 40 years of mineral weathering and exposure.

b) Group 2: decommissioned tailings, 1 to 27.5 m depth below surface. Samples within this group contained high ANC (320 kg H_2SO_4/t) and low MPA (120 kg H_2SO_4/t), and were classified as non-acid forming (NAF) or uncertain.

c) Group 3: operational tailings, 0 to 12 m depth below surface. Samples within this group typically contained less ANC (160 kg H_2SO_4/t) and more MPA (200 kg H_2SO_4/t) than operational tailings below 12 m depth, and were classified as potentially acid forming (PAF) or uncertain.

d) Group 4: operational tailings, 12 to 27.5 m depth below surface. Samples within this group were characterized by ANC (240 kg H_2SO_4/t) intermediate to that of group 2 and 3, and low MPA (120 kg H_2SO_4/t), and were classified as uncertain.

No samples were acidic. Paste pH ranged from 6.0 to 8.0, with group 3 samples at the lower end of the range (pH 6.0 to 7.4).
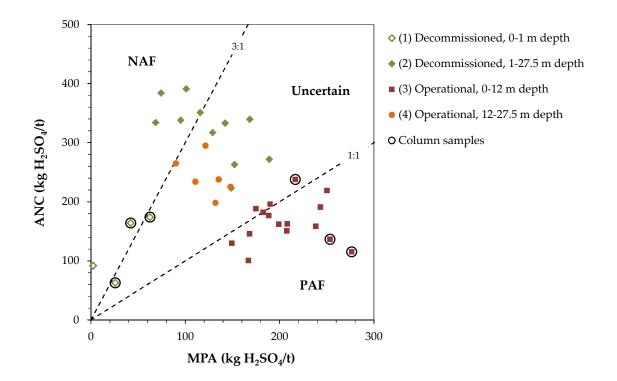

Figure 3 Acid base account plot for decommissioned and operational tailings

Trace Element Analysis

A comparison of decommissioned and operational tailings using a two-sample t-test indicated that operational tailings from 0 to 12 m depth contained significantly (p ≤ 0.01) greater concentrations of trace elements commonly associated with mineral sulfides, including arsenic, copper, lead, molybdenum, nickel, and zinc (Table 1). Slightly elevated concentrations of some metals were observed in near-surface decommissioned tailings, including copper, molybdenum, nickel, and zinc (Table 1). These metals are likely to occur in association with efflorescent sulfate salts.

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Table 1   Element concentrations by four-acid digestion summarized as the geometric mean with minimum and maximum values in parentheses

Tailings Group	As mg/kg	Cu mg/kg	Pb mg/kg	Mo mg/kg	Ni mg/kg	Zn mg/kg
Decommissioned 0-1 m depth (n = 7)	480 (200, 790)	1900 (1000, 4600)	4200 (3200, 5700)	4 (2, 100)	22 (15, 32)	5500 (4400, 6800)
Decommissioned 1-27.5 m depth (n = 11)	330 (210, 460)	960 (790, 2000)	2800 (1800, 6100)	1 (1, 2)	11 (8, 14)	3700 (2200, 5900)
Operational 0-12 m depth (n = 21)	810 (610, 1400)	2200 (1300, 6100)	5400 (2400, 8500)	26 (3, 220)	26 (19, 35)	8600 (3400,20000)
Operational 12-27.5 m depth (n = 6)	540 (440, 800)	1200 (690, 1500)	4000 (2600, 6600)	2 (2, 3)	16 (12, 22)	4800 (4300, 7100)

Note: All values are reported to two significant figures.

Changes in Mine Operation Influencing Tailings Composition

In comparison to historical tailings, the modern tailings stream at Mount Isa, represented by operational tailings from 0 to 12 m depth, is characterized by higher sulfide and associated trace element content, and less carbonate. Information from secondary sources suggests changes in tailings composition are likely attributable to changes in ore source and processing.

Decommissioned tailings were primarily derived from the processing of shallow copper ore bodies (200, 400, 500 and 600 mRL) which are predominantly dolomitic in composition (Wilde *et al.*, 2006), and lead-zinc ore from the Mount Isa underground mine. In contrast, operational tailings were derived from the processing of deeper, more siliceous copper ore bodies (1100, 1900, 3000 and 3500 mRL), and lead-zinc ore from George Fisher underground and Handlebar Hill and Black Star open pit mines.

The introduction of ore preconcentration and ultra-fine grinding technologies such as the IsaMill allowed the processing of lower grade ores (Gao *et al.*, 2002), which contain more pyrite, and resulted in a finer, higher sulfide tailings stream.

Mineral Reaction Rates

Column Leaching

Leachate pH remained near-neutral throughout the experiment with little difference between the ranges of decommissioned (pH 7.5 to 8.3) and operational tailings (pH 7.3 to 8.4). Major ions were dominated by calcium and sulfate, with leachate from two of the three decommissioned tailings remaining supersaturated with respect to gypsum throughout the experiment.

The columns were characterized by long pore water residence times (15 to 53 hours) and high water retention with 84 to 98% of pores remaining water-filled after the application of -60 kPa pressure. However, this small change in water content in operational tailings was sufficient to increase sulfate, and calcium and magnesium release rates (indicative of sulfide mineral oxidation and acid neutralization, respectively), and release rates for certain trace elements, including manganese, cobalt, and zinc (for example, operational tailings column 2, Table 2). The increase in element release rates from operational tailings was accompanied by a 0.4 to 0.7 unit decrease in leachate pH.

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Saturated hydraulic conductivity (Ks) under constant head increased during the experiment (Table 2). In two columns, this was attributed to the formation of fine vertical cracks observed after the application of suction. In other columns, it was attributed to the slow dissolution of soluble phases (particularly gypsum) resulting in more pores contributing to water flow.

Sulfate, calcium, and magnesium release rates were used to calculate rates of acid generation and neutralization, whereby:

$$\text{Sulfide oxidation rate (kg } H_2SO_4\text{/t/day)} = \text{SO}_4 \text{ production rate (kg/t/day)} \times \frac{98.08}{96.06}$$

(1)

$$\text{ANC consumption rate (kg } H_2SO_4\text{/t/day)} = \text{sulfide oxidation rate (kg } H_2SO_4\text{/t/day)} \times \frac{[\text{Ca (mol)}+\text{Mg(mol)}]}{\text{SO}_4\text{(mol)}}$$

(2)

Leachate solutions saturated and/or supersaturated with respect to gypsum were excluded from the calculations.

A comparison of the calculated rates (Table 3) indicated that ANC would likely be consumed before the acid-generating sulfides in operational tailings 1 and 2 under all test conditions, which supports a PAF classification. Conversely, ANC was unlikely to be consumed before the acid-generating sulfides in decommissioned tailings, which supports a NAF classification. Operational tailings 3 were classified as uncertain using static ABA. Kinetic testing suggests this sample has the potential to become acid generating if conditions were to remain unsaturated and highly oxidizing.

CONCLUSIONS

Modern tailings at Mount Isa are characterized by higher sulfide and associated trace element content, and lower carbonate mineral content than historical tailings. Changes in tailings composition are likely attributable to changes in ore and mineral processing methods, and time since tailings deposition.

Strategies are currently being developed for placement of a suitable cover system to reduce surface water – tailings interaction following decommissioning of the TSF. It is anticipated that covering the surface tailings would potentially limit the accumulation of efflorescent sulfate salts on the tailings surface, which if allowed to accumulate, would be subject to dissolution and transportation during wet season rain events. Infiltrated water that passes through the oxidation zone is expected to be readily neutralized by ANC in the tailings below. Modification of the final tailings layer prior to deposition to remove or pacify pyrite could also potentially be considered as an alternative to a suitable cover system.

Table 2 Element release rates from operational tailings column 2

Leach Phase[a]	Water Content cm³/cm³	pH	SO_4 mg/kg/day	Ca mg/kg/day	Mg mg/kg/day	Mn mg/kg/day	Co mg/kg/day	Zn mg/kg/day
1	53 (100%)	8.1	18	10	0.33	0.0074	0.00006	0.0093
2a	49 (92%)	7.7	450	160	40	1.1	0.059	2.0
2b	53 (100%)	7.9	77	31	5.8	0.14	0.006	0.14

Note: (a) Refer to Figure 2.

Table 3 Sulfide oxidation and ANC consumption rates measured from column leaching

Column	Leach Phase[a]	Mean K_s cm/day	Acid Generation MPA kg H_2SO_4/t	Sulfide Oxidation Rate kg H_2SO_4/t/day	Depletion Time in Lab years	Neutralization ANC kg H_2SO_4/t	ANC Consumption Rate kg H_2SO_4/t/day	Depletion Time in Lab years	Class.
Decommissioned 1	1	1.2	71	0.17	1.1	174	0.10	4.9	NAF
	2a	3.5	71	0.54	0.4	174	0.34	1.4	NAF
	2b	0.9	71	0.13	1.5	174	0.09	5.4	NAF
Operational 1	1	0.3	293	n.a.[b]	n.a.[b]	115	n.a.[b]	n.a.[b]	n.a.[b]
	2a	0.4	293	0.32	2.5	115	0.26	1.2	PAF
	2b	12	293	0.10	7.9	115	0.09	3.4	PAF
Operational 2	1	0.2	276	0.03	25	137	0.03	13	PAF
	2a	62	276	0.46	1.6	137	0.45	0.8	PAF
	2b	120	276	0.09	8.7	137	0.09	4.2	PAF
Operational 3	1	1.6	231	0.07	9.4	238	0.04	15	NAF
	2a	83	231	0.23	2.8	238	0.55	1.2	PAF
	2b	120	231	0.20	3.2	238	0.15	4.3	NAF

Notes: (a) Refer to Figure 2.   (b) Rate indeterminate due to gypsum saturation.

ACKNOWLEDGEMENTS

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