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Geochemical predictions and closure planning implications for reprocessing legacy tailings at the old tailings storage facility complex, South Deep gold mine, South Africa

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Abstract

Tailings storage facility (TSF) closure planning requires an understanding of long-term acid generation and metal leaching risks to inform closure options. SLR conducted a comprehensive geochemical characterisation of the South Deep old TSF complex, made up of 4 compartments (TSF 1–4), parts of which is currently being reclaimed. Assessment of the mature residual tailings and associated water chemistries was used to develop a kinetic model to predict the complex's geochemical behaviour and groundwater contaminant source terms. Water quality results indicate that metal exceedances predominantly occur within the reticulated water system and in boreholes and scavenger wells proximal to the Old TSF complex. Acid-base accounting and mineralogical results infer that the tailings are potentially acid-generating and pose a risk of metal leaching, as indicated by metal exceedances reported from static leaching tests which correlate with modelled source terms. A 20-week humidity cell experiment was conducted on composite samples that showed TSF1 and 2 (currently being reclaimed) have pH and alkalinity profiles indicating acidic conditions over the long term. Low carbonate molar ratios (CMRs) and feldspar molar ratios (FMRs) predict that secondary neutralisation capacity will not keep pace with in situ acid generation. The dormant TSF3 reported fluctuating pH and alkalinity profiles supporting a potential acid-generating classification. In contrast, the dormant TSF4 showed a circumneutral pH profile, suggesting acid generation is unlikely over the longer term. Their CMR and FMR values demonstrate sufficient secondary neutralisation potential, but it is likely to be nullified once the reprocessing begins. Chlorite and Talc were the main minerals controlling the complex's neutralisation potential. The study predicts that acidic conditions will persist throughout the complex, posing a risk of metal leaching that could affect local water resources. Therefore, a conservative approach should be taken into consideration when advising closure options to manage water quality impacts optimally, post-closure.

Keywords: *geochemical predictions, legacy tailings, kinetic humidity cells, secondary neutralisation potential, closure planning*

1 Introduction

As a member of the International Council on Mining and Metals, Gold Fields Ltd is required to conform to the Global Industry Standard on Tailings Management (GISTM). To meet with GISTM requirements and align with international best practices, an updated and comprehensive geochemical characterisation of the residual tailings materials being systematically reprocessed within the South Deep gold mine (SDGM) old tailings storage facility (TSF) complex was undertaken. This was recommended due to changes in mining practices since previous geochemical investigations and the introduction of the GISTM guidelines (Global Industry Standard on Tailings Management 2020). When pyritic tailings materials are exposed to atmospheric oxygen and precipitation, oxidation of residual sulphide minerals can lead to drainage water developing an acid mine

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drainage (AMD) character with an acid pH, elevated sulphate and dissolved metal concentrations over time (Heikkinen et al. 2009).

The SDGM is a deep-level mine that is actively mining at depths between 2,600 and 3,000 m below surface, with plans to extend to approximately 3,400 m. The mine is located within the magisterial districts of Westonaria and Vanderbijlpark, approximately 40 km southwest of Johannesburg, South Africa, and situated on the northwestern rim of the Witwatersrand Basin. The Witwatersrand Basin comprises a 6,000 m-thick sequence known as the Witwatersrand Supergroup (Andrew et al. 2019), which formed over a period of 360 million years (Ma), between 3,074 and 2,714 Ma, from a series of crustal plate movements. Pulses of sediment deposition within the sequence and its precursors were episodic, occurring between 3,086– 3,074 Ma (Dominion Group), 2,970–2,914 Ma (West Rand Group) and 2,894–2,714 Ma (Central Rand Group; Robb & Meyer 1995). The South Deep mining right area is underlain by outliers of the Karoo Supergroup shales and sandstones, followed by the Pretoria Group sediments and the Chuniespoort Group dolomites. The Chuniespoort Group in turn overlies the Klipriviersberg Group volcanic rocks, which are underlain by the Central Rand Group which hosts the gold-bearing conglomerates that are mined at SDGM (Catuneanu & Biddulph 2001). The SDGM Old TSF complex comprises the South Shaft (TSF 1 & 2) and Twin Shaft (TSF 3 and 4). The South Shaft TSFs were commissioned in 1968 while the Twin Shaft TSFs were commissioned in 1982, with both facilities consisting of two separate TSFs abutting one another. The Old TSF complex has been under care and maintenance since 2011 following the commissioning of the Doornpoort TSF. Reclamation activities of the South Shaft TSFs started with TSF1 in September 2019. The reclaimed tailings from the TSFs are combined with milled underground ore, passed through the plant and then pumped as a slurry to Doornpoort TSF. No reclamation activities have been undertaken at the Twin Shaft TSFs and it is anticipated that reprocessing will commence after depletion of TSF1 and 2 (SLR Consulting 2025).

The study aimed to predict rates of sulphide oxidation, and metal and salt releases from the Old TSF complex to determine the long-term acid-generating potential and metal leaching risks of the complex over several years and decades. This body of work will contribute to predicting the future geochemical behaviour of the complex and provide source term concentrations for use in a groundwater numerical model that simulates possible short-, medium- and long-term pollution plumes and informs closure planning of the facility.

2 Methodology

2.1 Tailings sampling

Two test pits were excavated on each TSF compartment (1–4), from which 3 discrete samples were obtained at depths of 0 to 1 m (surface), 1–2 m (subsurface) and 2–3 m (depth). This was done so that material from each geochemical horizon – namely, the oxidising, transition and reducing zones – could be obtained where feasible (i.e. at least in one pit on each TSF compartment). Selected discrete samples were constituted into 4 composites based on the highest pyrite composition determined during static testing for kinetic humidity cell (HC) leaching (Table 1). This was recommended, as the highest pyrite content is indicative of the worst-case scenario and is more representative for assessing the long-term rates of sulphide oxidation and acid generation in the tailings materials.

Table 1 Old tailings storage facility complex composite tailings samples

Composite ID	Sample ID	Pyrite composition (%)	Depth interval (m)	Proportion of composite (%)
	TSF1-002 Surface	0.44	0–1	33.30
TSF1	TSF1-001 Subsurface	0.91	1–2	33.30
	TSF1-002 Depth	0.61	2-3	33.40
	TSF2-002 Surface	0.19	0–1	33.30
TSF2	TSF2-002 Subsurface	1.15	1–2	33.40
	TSF2-001 Depth	1.14	2–3	33.30
	TSF3-002 Surface	-	0–1	33.30
TSF3	TSF3-001 Subsurface	0.38	1–2	33.30
	TSF3-002 Depth	0.43	2–3	33.40
	TSF4-002 Surface	-	0–1	33.30
TSF4	TSF4-002 Subsurface	0.46	1–2	33.40
	TSF4-001 Depth	0.63	2–3	33.30

Note: '-' = no value, 001 refers to test pit 1 and 002 refers to test pit 2 on a specific tailings storage facility

2.2 Laboratory testing

The Old TSF composite tailings samples underwent the following comprehensive geochemical analysis:

- pre- and post-HC X-ray diffraction (XRD) mineralogy
- pre- and post-HC acid-base accounting (ABA), paste pH and total sulphur
- 20-week kinetic HC testing
- weekly leachate analysis for pH, electrical conductivity, total dissolved salts, total alkalinity and major ions
- inductively coupled plasma mass spectrometry and optical emission spectroscopy analysis of > 40 metals.

3 Results and discussion

3.1 X-ray diffraction mineralogy

The mineralogical composition of the old TSF tailings composites before and after the 20-week kinetic leaching procedure is summarised in Table 2.

Table 2 Mineralogy of the old tailings storage facility complex composite tailings samples pre- and post-humidity cell leaching (weight %)

Mineral	Formula	TSF1	TSF2	TSF3	TSF4	TSF1	TSF2	TSF3	TSF4
		Pre-leaching				Post-leaching			
Quartz	SiO_2	91.20	93.10	90.60	93.50	94.90	90.90	89.70	93.10
Pyrite	FeS_2	0.90	0.60	0.50	0.50	0.80	0.60	0.30	0.60
Chlorite	(Mg, Fe)5Al(AlSi3O10)(OH)8	3.10	3.20	2.70	1.30	2.40	2.30	1.10	1.90
Talc	Mg3Si4O10(OH)2	0.90	-	1.00	0.80	-	-	-	-
Gypsum	$Ca(SO_4)(H_2O)2$	1.40	1.10	1.60	1.10	-	-	-	-
Muscovite	KAl2((OH)2AlSi3O10)	2.50	2.10	3.60	2.70	1.40	3.30	6.30	2.30
Pyrophyllite	$Al(Si_2O_5)(OH)$	-	-	-	-	0.50	2.90	2.60	2.20

Note: - = undetected

After completion of the 20-week HC testing, most composites showed lower proportions of chlorite, except for TSF4, which recorded an increase post-HC testing. The post-HC XRD results also showed complete depletion of talc and gypsum for all composite samples. The reduction or depletion of these minerals suggests that they may be controlling secondary neutralisation, with talc offering neutralisation potential (NP) over the short term (weeks to months), followed by the long-term (years to decades) NP offered by chlorite due to its slower dissolution kinetics. When comparing the pre- and post-HC XRD results, all composites retained elevated pyrite concentrations (> 0.3%) post-HC, suggesting they have the potential to generate acid in the longer term.

3.2 Acid-base accounting

The pre-HC paste pH values recorded for TSF1 and 2 (4.16 and 4.27, respectively) fall within a moderately acidic range, whereas the paste pH values for TSF3 and 4 (6.58 and 6.61, respectively) are within a circumneutral range. Conversely, the post-HC paste pH values recorded for TSF1 and 2 (5.26 and 5.71) increased to a slightly acidic range, whereas TSF3 and 4 remained within a circumneutral range, with TSF4 recording a minor increase (6.96) at completion of the HC testing. The increased paste pH values, post-HC testing, show that neutralisation has occurred over the short term due to the dissolution of talc and chlorite, as supported by the post-XRD analysis.

The old TSF composites also recorded NP ratio values < 1 as well as net NP values < -20 pre- and post-HC testing. These values are attributed to their elevated total sulphide (> 0.3) contents and larger acid potential (AP) versus NP values (Table 3). The significantly larger AP values are attributed to the lack of rapidly dissolving neutralising minerals like carbonates, which implies that the old TSF composites' NP is unlikely to keep pace with acid generation over the long term. Therefore, the ABA findings reinforce the Old TSF complex long-term acid generation risk profile.

Based on the pre- and post-HC ABA testing results, the following classifications were made for the Old TSF complex composites:

- TSF1 and 2 are acid-generating (AG) materials.
- TSF3 and 4 are potential acid-generating (PAG) materials.

Table 3 Old tailings storage facility complex composite tailings samples, acid-base accounting, pre- and post-humidity cell testing

Sample	Paste pH	Total S %	Sulphide acid potential kg/t CaCO3	Neutralisation potential kg/t CaCO3	Neutralisation potential ratio	Net neutralisation potential kg/t CaCO3	Classification
Non-potential acid-generating (non-PAG)	> 6	< 0.3			> 2	> 20	
Intermediate	3.5–5.5				1–2	-20 to 20	
PAG or AG	< 6	> 0.3			< 1	< -20	
Pre-humidity cell testing							
TSF1	4.16	1.21	38	-1.73	0.046	-40	AG
TSF2	4.27	1.26	39	-2.72	0.069	-42	AG
TSF3	6.58	0.94	29	0.75	0.025	-29	PAG
TSF4	6.61	1.05	33	0.99	0.030	-32	PAG
Post-humidity cell testing							
TSF1	5.71	0.83	26	-0.49	0.019	-26	AG
TSF2	5.26	1.05	32.80	-0.99	0.030	-33	AG
TSF3	6.34	0.67	20.80	-0.99	0.047	-21	PAG
TSF4	6.96	0.78	24.30	1.49	0.061	-23	PAG

3.3 Humidity cell experiment

Kinetic testing was conducted using HCs over a period of 20 weeks. Composites were selected to better understand the Old TSF complex tailings' potential for acid generation and available neutralisation capacity. This can inform mitigation strategies to protect water resources and address potential long-term acid generation and metal leaching from tailings materials by evaluating the reaction rates and depletion times of minerals present in tailings or mine waste. The key aspects relate to understanding the presence and availability of NP obtained from static ABA analyses, assessing the pH buffering capacity afforded by the minerals, and monitoring metal- and salt-leaching rates over time. For interpretation purposes, the last 3 weeks of the kinetic testing leachate data, namely weeks 18 to 20, were averaged and used to represent the final sequence of parameters in relation to their leachate generation profile. This can be regarded as an indication of the stabilised geochemical profiles of the Old TSF complex, which refers to the geochemical risks that are predicted to develop and persist over a period of several years to decades.. Refer to Table 4 for the approaches to the interpretation of kinetic test results.

Table 4 Approaches to the interpretation of kinetic results (Price 2009)

Humidity cell result interpretations
Acid generation
Sulphate production rate (mg/kg/week) = sulphate (mg/L) × mass of extraction fluid recovered/1,000
Neutralisation potential (NP) molar ratios
Carbonate molar ratio (CMR) = [(Ca (mg/L)/40.08) + (Mg (mg/L)/24.31)]/(SO_4 (mg/L)/96.06)
Feldspar molar ratio (FMR) = [(Ca (mg/L)/40.08) + (K (mg/L)/2*39.1) + Na mg/L)/(2*22.99)]/(SO_4(mg/L)/96.06)
Acid neutralisation and NP consumption
Carbonate ratio NP consumption (mg $CaCO_3$/kg/week) = CMR x theoretical NP consumption (mg/kg/week):
$2H^+ + SO_4^{2-} + (CazMg1_{-z})CO_3 \rightarrow zCa^{2+} + (1-z)Mg^{2+} + SO_4^{2-} + H_2CO_3^0$ or
$2H^+ + SO_4^{2-} + 2(Ca_zMg_{1-z})CO_3 \rightarrow 2zCa^{2+} + (2-2z)Mg^{2+} + SO_4^{2-} + 2HCO_3^-$
Theoretical NP consumption at pH = 6 (mg $CaCO_3$/kg/week) Sulphate production rate (mg SO_4/kg/week) × 100.09/96.06:
$2H^+ + SO_4^{2-} + CaCO_3 \rightarrow Ca^{2+} + SO_4^{2-} + H_2CO_3^0$
Remaining NP (% of original)
{[Initial NP (t $CaCO_3$/1,000 t) – (cumulative NP depletion rate (mg/kg)/1,000)]/initial NP (t/1,000 t)} × 100
FMR NP consumption (mg $CaCO_3$/kg/wk)
FMR theoretical NP consumption (mg/kg/week) based on:
$2H^+ + SO_4^{2-} + 6\ H_2O + CaAl_2Si_2O_8 \rightarrow Ca^{2+} + SO_4^{2-} + 2Al(OH)_3 + 2H_4SiO_4$
$2H^+ + SO_4^{2-} + 14H_2O + 2KAlSi_3O_8 \rightarrow 2K^+ + SO_4^{2-} + 2Al(OH)_3 + 6H_4SiO_4$
$2H^+ + SO_4^{2-} + 14H_2O + 2NaAlSi_3O_8 \rightarrow 2Na^+ + SO_4^{2-} + 2Al(OH)_3 + 6H_4SiO_4$

3.3.1 Assessment of pH and alkalinity leaching profiles

The temporal trends of the leachate pH and alkalinity profiles for the Old TSF complex composites are presented in Figures 1 and 2. TSF1 and 2 reported pH increases after the 'first flush'. Thereafter, both composites showed decreases in leachate pH, which stabilised within the ranges 2.47 to 3.29 for TSF1 and 2.73 to 3.28 for TSF2 over the remaining weeks of the HC test. TSF3 recorded an increase in pH after the first flush and reached a circumneutral level. Thereafter, the leachate pH gradually decreased from 6.49 to 4.83 and stabilised between 5.34 and 4.83 from week 12 onwards. TSF4 showed an increase in pH after the first flush, reaching a near-neutral level. The leachate pH stabilised earlier than the other composites and fluctuated between 6.60 and 6.41 over the remainder of the 20 weeks.

The stabilised pH trends recorded for TSF1, 2 and 3 infer that pH buffering is occurring within the HC leachates, which can be attributed to the precipitation of iron hydroxy-sulphate minerals like Schwertmannite (Espana et al. 2006; Botes & Steyn 2022) or Jarosite in the case of the leachates from TSF1 and 2, and aluminium hydroxy-sulphate minerals like Basaluminite (Sanchez-Espana et al. 2011) in the leachate from TSF3. The precipitation of these minerals in the HC leachates can release latent acidity in the form of H^+ ions (Taylor et al. 2005; Zipper et al. 2018), which contribute to the stabilised pH profiles (Uhlmann et al. 2004).

The mean stabilised pH results for weeks 18 to 20 for the old TSF composites indicated acidic conditions for TSF1 and 2 (2.51 and 2.77, respectively), moderately acidic conditions for TSF3 (4.93) and circumneutral conditions for TSF4 (6.36).

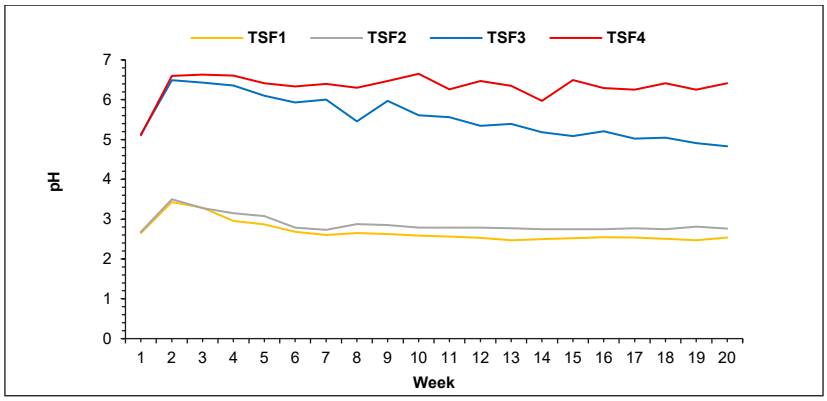

Figure 1 pH profile for old tailings storage facility complex tailings composites over a 20-week leaching experiment

TSF1 and 2 reported similar trends for their alkalinity profiles, which remained constant at a rate of 1.00 mg/kg/week throughout HC testing, with a mean long-term alkalinity of 0.750 mg/kg/week for both composites. TSF3 recorded an alkalinity profile ranging from 0.75 to 37 mg/kg/week, with a mean long-term alkalinity of 0.75 mg/kg/week. In comparison, TSF4 reported an alkalinity profile ranging between 1.50 and 36 mg/kg/week with a mean long-term alkalinity of 3.25 mg/kg/week.

The alkalinity profiles for TSF1 and 2 show that NP is negligible, which indicates that these composites have a high AG risk over the long term (years to decades) and is supported by the mean stabilised pH for these composites. However, the cumulative alkalinity profiles recorded for TSF3 and 4 show rapid increases during the first 4 weeks of the leaching experiment. Thereafter, the profiles increase gradually over the remainder of the 20 weeks.

The dissolution of talc is inferred to initially contribute to NP, followed by the slower dissolution of chlorite, which further contributes to NP over the long term. The alkalinity profiles for TSF3 indicate that it has the potential to generate acidity, which is supported by its moderately acidic mean long-term pH. However, the alkalinity profiles reported for TSF4 predict that net acid generation is unlikely, as supported by its circumneutral mean long-term pH in its current setting.

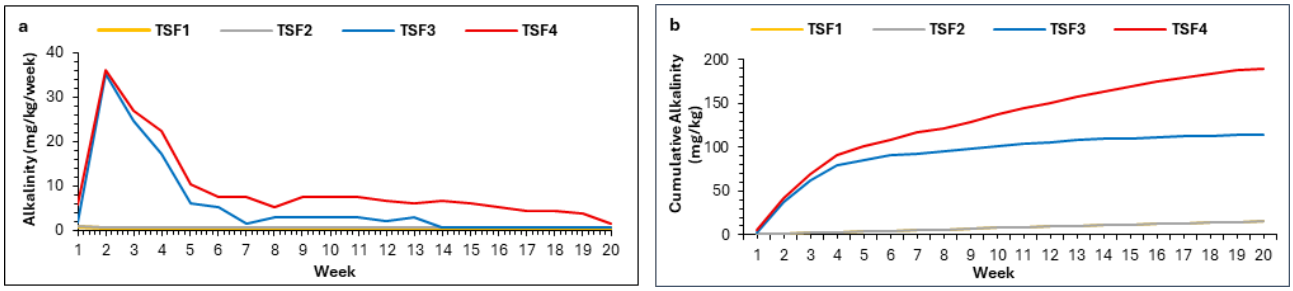

Figure 2 Alkalinity profile for old tailings storage facility complex composites over a 20-week leaching experiment. (a) Weekly alkalinity production; (b) Cumulative alkalinity

3.3.2 Sulphate production and neutralisation potential

The rates of sulphide oxidation and NP consumption are important parameters interpreted from HC leachate data. The HC tests determine the rate of sulphide mineral oxidation by the changes in sulphate concentrations analysed in the weekly leachate. It is also important to note that sulphate can be detected through the dissolution of sulphate-containing minerals like barite, anhydrite and gypsum, and this needs to be accounted for in any interpretation of sulphate generation. Sulphide oxidation is the major source of acid production in mine drainage, and the associated decrease in pH increases the solubility of metal ions, leading to their release.

When sulphide is oxidised, the dominant NP consumption reaction is acid neutralisation. When carbonates are present, the neutralisation reaction is pH-dependent (assuming no calcium and sulphate are lost to secondary mineral precipitation, indicated by the saturation indices for $CaSO_4$), and there are two main reactions (using pyrite as the sulphide mineral):

- At pH < 6.3, $FeS_2 + 15/4\ O_2 + 7/2\ H_2O + 2\ [Ca^{2+}, Mg^{2+}]\ CO_3 \rightarrow Fe(OH)_3 + 2SO_4^{2-} + 2H_2CO_3^0 + 2[Ca^{2+}, Mg^{2+}]$
- At pH 6.3 to < 10.3, $FeS_2 + 15/4\ O_2 + 7/2\ H_2O + 4\ [Ca^{2+}, Mg^{2+}]\ CO_3 \rightarrow Fe(OH)_3 + 2SO_4^{2-} + 4HCO_3^- + 4[Ca^{2+}, Mg^{2+}]$.

Therefore, at pH < 6.3, neutralisation will produce one mole of calcium/magnesium and sulphate, which gives a 1:1 carbonate molar ratio (CMR). Due to factors such as carbon dioxide exsolution and contributions from calcium and magnesium from non-carbonate minerals, this equation is theoretical. At pH values around 8, carbonic acid is not stable, and bicarbonate dominates. In such circumstances, carbonates are less efficient at neutralising acidity when a 2:1 calcium-to-magnesium ratio is achieved.

Weekly leachate concentrations of calcium, magnesium and sulphate are used to calculate the CMR (Table 4). The CMR indicates the actual rate of carbonate dissolution (NP consumption) and thus can be used to estimate the time to the onset of acid generation. The CMR values should be between 1 and 2 in carbonate-bearing rock that is actively and effectively neutralising acidity from sulphide oxidation (Morin & Hutt 1994, 2000). However, the CMR can fall below 1.0 when effective neutralisation falters as the rinse pH decreases to between 5 and 6. This is because the effective and fast-neutralising carbonate has been exhausted.

Where there is a low rate of in situ acid generation from sulphide oxidation, the predominant form of acidity is associated with the residual acidity of the deionised water (pH 5.5) introduced into the HCs. The carbonate minerals react with this water and yield large CMR values, significantly greater than 2. Furthermore, the CMR values do not accurately reflect the relative rates of sulphide oxidation and NP depletion when the sulphate concentration in the HC leachate is less than 20 mg/kg (Mattson 2005).

The mean results of the calculated CMR values for weeks 18 to 20 for the Old TSF composites are provided in Table 5. TSF1 and 2 reported CMR values < 1, indicating that NP will not keep pace with the rate of pyrite oxidation over the long term. Conversely, TSF3 and 4 reported CMR values slightly greater than 1, indicating that NP can keep pace with pyrite oxidation and suggesting that net acid generation is unlikely from these materials in the short term.

Table 5 Mean old tailings storage facility complex composites' carbonate molar ratio (CMR) results for weeks 18–20

Sample ID	Weeks 18 to 20 average CMR
TSF1	0.47
TSF2	0.66
TSF3	1.10
TSF4	1.06

3.3.3 Sulphide oxidation rates/sulphide production

The rate of sulphide oxidation in mine waste is estimated from the sulphate analyses of the HC leachate. To obtain sulphide oxidation rates, weekly sulphate concentrations are normalised to the mass of sample in the HC. Kinetic tests are used to evaluate whether the measured NP effectively buffers pH at the rate of acid generation from the oxidising sulphide minerals.

Weekly sulphate production rates and cumulative sulphate load of the Old TSF composites are shown in Figure 3. All composites recorded elevated sulphate production rates at the beginning of the leaching

experiment, which ranged between 6,330 and 10,427 mg/kg/week, with TSF3 and 4 reporting the highest rates. From week 2 onwards, the sulphate production rates significantly decreased, with TSF1 and 2 recording the highest sulphate production rates over the remainder of the 20 weeks.

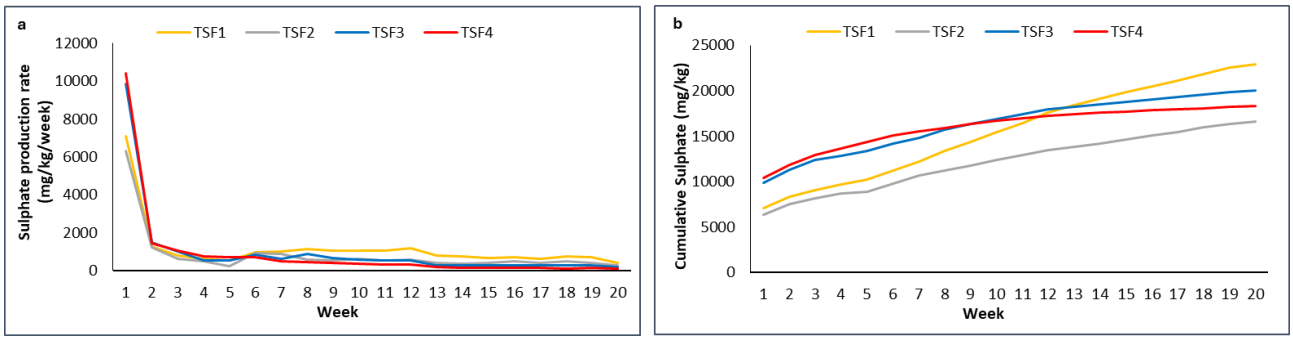

Figure 3 Old tailings storage facility complex composites' long-term sulphate production rate. (a) Weekly sulphate production rates; (b) Cumulative sulphate profiles

The continuously increasing cumulative sulphate loads recorded for TSF1 and 2 suggest that there is a source of residual sulphate in the composites, which is likely emanating from the oxidation of unreacted pyrite. This correlates with the acidic pH and negligible alkalinity profiles of these composites, further supporting their long-term AG potential. However, TSF3 and 4 recorded cumulative sulphate loads that started to stabilise midway through the leaching experiment. This suggests that most of the sulphate source in these composites is depleted or is in equilibrium with the NP.

The pre-HC XRD results showed that all TSF samples contained pyrite and gypsum, with TSF3 reporting the highest gypsum content. The elevated sulphate production rates initially reported for the Old TSF composites are likely due to gypsum dissolution, which is coupled with pyrite oxidation that generates acidic conditions and exacerbates mineral dissolution. The post-HC XRD analyses did not show the presence of gypsum in any of the Old TSF composites, which supports complete dissolution and contributions to sulphate production during the 20 weeks.

Furthermore, the decline in sulphate production observed for the Old TSF composites could be attributed to the encrustation of residual pyrite by precipitation of secondary minerals. This is well correlated to the shrinking core model in that the rate of oxygen diffusion towards the surface of residual pyrite is the rate limiting step (van Zweel et al. 2015). Thus a lag effect is introduced, which influences the oxidation kinetics of the unreacted pyrite and consequently leads to decreased sulphur production rates.

3.3.4 Non-carbonate (aluminosilicate) neutralisation potential

The presence of primary minerals and secondary phyllosilicates and aluminosilicates indicates that additional sources of NP may be available to offer pH buffering capacity. The method to assess the dissolution of these minerals involves calculating the FMR. Understanding the reactivity of silicate minerals is important when evaluating the long-term availability of NP in mine waste materials to ensure that silicate dissolution rates are sufficient to offset the acidity produced from sulphide oxidation.

These reactions are incongruent and typically release cations from the silicate mineral structure and generate residual secondary, and possibly tertiary, minerals that are stable under the geochemical weathering environment. When comparing the stoichiometric ratios of major ions such as Ca, Mg, Na, K and Si in the leachate from each HC, insight into the minerals controlling the NP for each of the Old TSF complex composites can be obtained.

Figure 4 shows the FMR trends for each of the Old TSF complex composites. At the beginning of the leaching experiment, all composites reported sufficient rates of silicate dissolution to counteract the acid generated from pyrite oxidation. However, from week 3 onwards, TSF1 and 2 recorded FMR values < 1, indicating that NP is restricted and supports a high AG risk over the long term. Conversely, TSF3 and 4 reported FMR values

that fluctuated between 0.72 and 1.23 over the remainder of the 20-week period, indicating that NP through silicate dissolution occurs, albeit at slower rates than if carbonate minerals dominated.

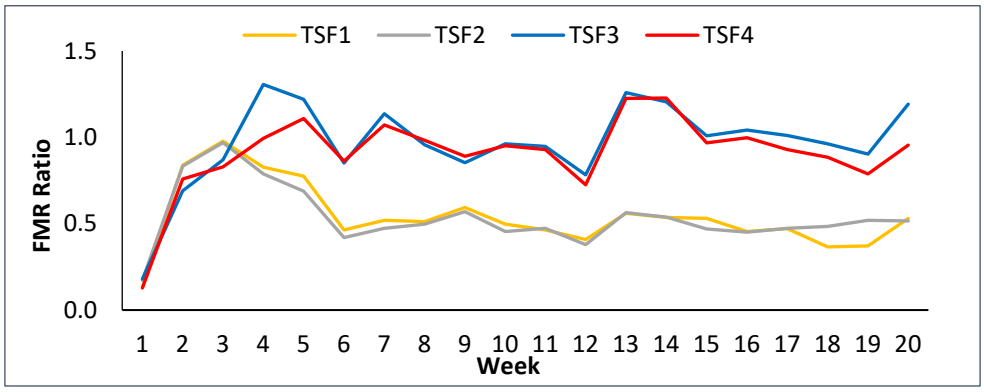

Figure 4 Feldspar molar ratio (FMR) ratios of the old tailings storage facility composites over the 20-week analysis period

Figure 5 shows the ratios of the major cations to silicon present in the HC leachate from each composite over the 20 weeks. All composites initially reported elevated Mg/Si ratios, which indicate that talc and chlorite are the main minerals controlling NP in the samples. TSF1 and 2 also show high Al/Si ratios at the beginning of the experiment, which is suggested as attributable to the dissolution of chlorite instead of muscovite due to negligible K/Si ratios recorded for the Old TSF composites. This suggests that muscovite plays a limited role in NP formation, likely due to its slow dissolution kinetics. The Ca/Si ratios for TSF1 and 2 are, on average, higher than the ratios for the other major ions from week 2 onwards, which is attributed to the dissolution of gypsum in the composites. However, the dissolution of this mineral does not imply that it is controlling NP.

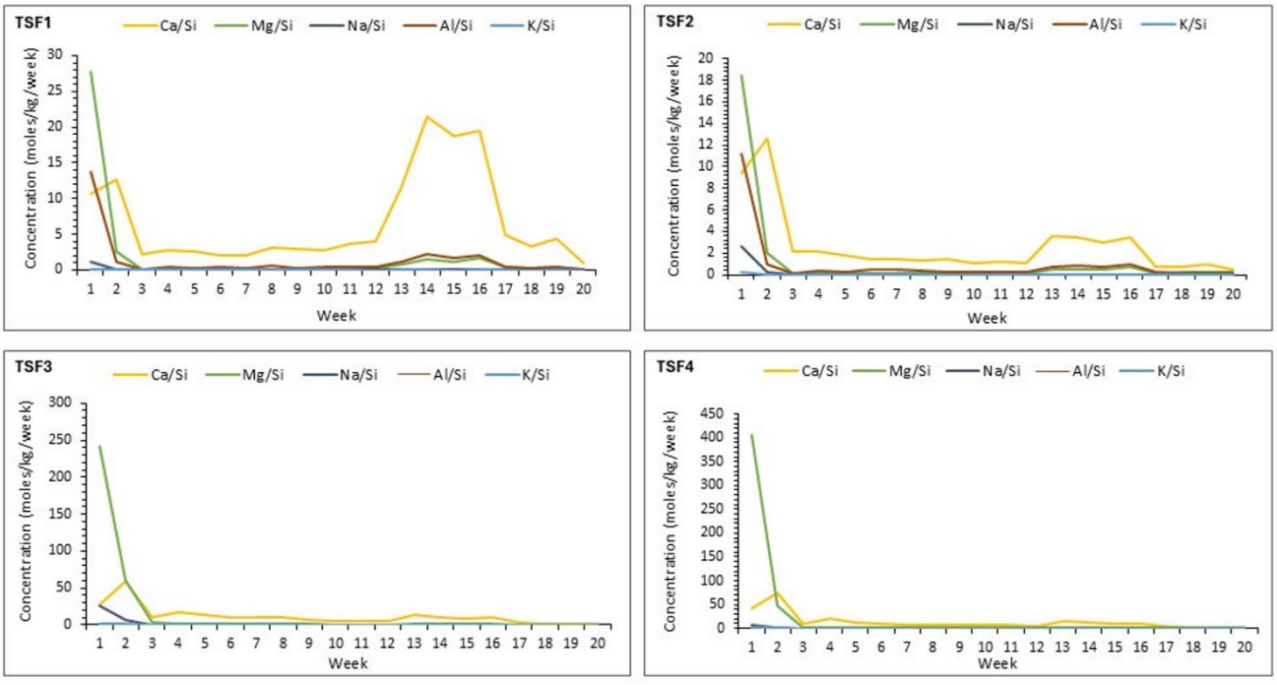

Figure 5 Major ion and silicon ratios present in the humidity cell leachates from TSF1, 2, 3, and 4 over the 20 weeks

3.3.5 Metal and metalloid leaching profiles

The HC analyte concentrations can be used to predict the leaching dynamics of the Old TSF composites and identify geochemical source terms that can impact groundwater and surface water resources over the long term. Figures 6 and 7 show the metal and metalloid leaching trends for the Old TSF complex composites, which could be potential constituents of concern.

Aluminium exceedances were persistent throughout the 20 weeks in TSF1 and 2 but limited to early stages in TSF3 and 4. However, aluminium mobility in groundwater is expected to be limited due to adsorption and secondary mineral precipitation. Arsenic exceedances were sustained in TSF1 and 2 before declining towards the end of the experiment, while TSF3 and 4 reported early-stage exceedances only. Manganese exceedances occurred throughout the HC experiment in TSF3 and 4, with decreasing concentrations over time, and in TSF2 for most of the period, including a temporary decline below screening thresholds mid-experiment, followed by renewed exceedances after week 15. TSF1 trended similarly but dipped below the exceedance threshold starting in week 15. Elevated lead exceedances were initially observed in TSF1 and 2, with TSF1 persisting above screening levels, while TSF2 declined below screening levels. TSF3 and 4 remained below lead screening thresholds throughout.

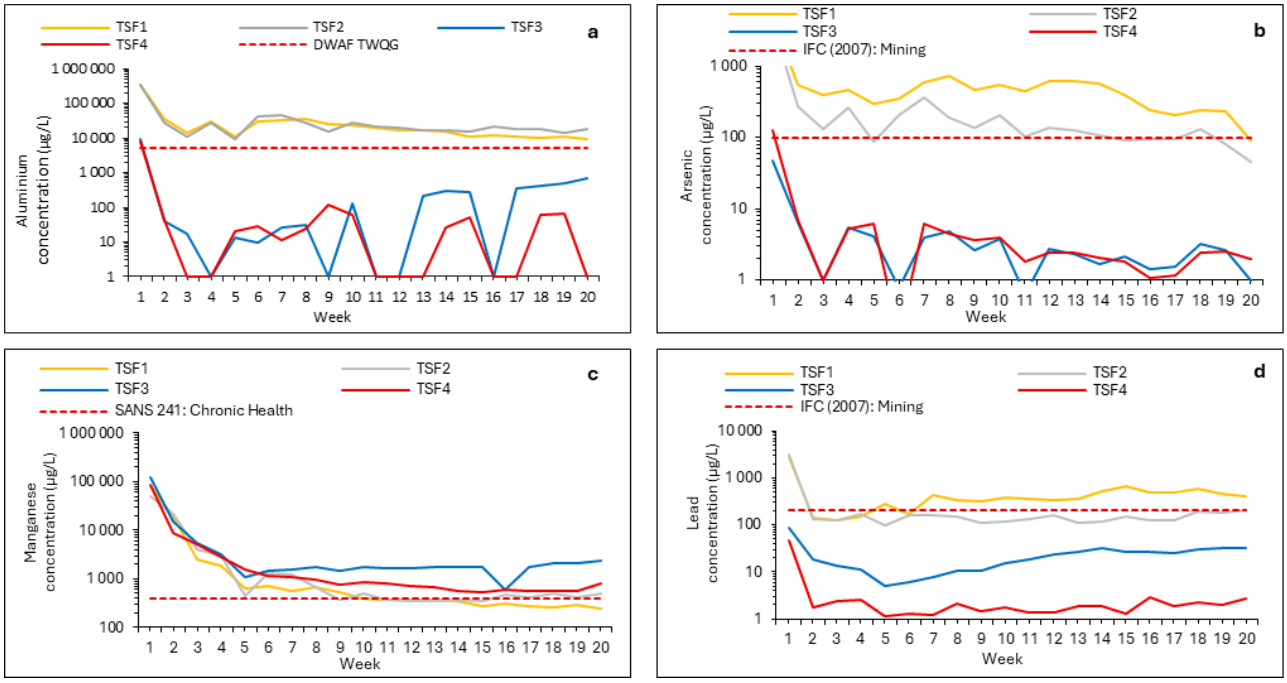

Figure 6 Old tailings storage facility (TSF) complex composites metal leaching profiles: (a) Aluminium; (b) Arsenic; (c); Manganese; (d) Lead. South African National Standards (SANS 241), Department of Water Affairs Target Water Quality Guidelines (DWAF TWQG); International Financing Corporation (IFC)

Uranium concentrations were consistently elevated in TSF1 and 2, exceeding screening thresholds throughout the leaching experiment. TSF3 and 4 initially recorded exceedances, but TSF4 concentrations declined below screening levels from week 5 onwards, while TSF3 exhibited an increase from week 5, resulting in sustained exceedances for the remainder of the period. Nickel concentrations exceeded screening thresholds in TSF1 and 2 throughout the experiment, despite a general downward trend. In contrast, TSF3 and 4 showed initial nickel exceedances followed by declines below screening thresholds, although TSF3 displayed late-stage variability with concentrations fluctuating near the threshold.

Cadmium exceedances were most significant in TSF1 and 2, with concentrations decreasing over time but remaining above screening thresholds for most of the experiment. By the final stage, TSF1 concentrations decreased below the threshold while TSF2 approached it. TSF3 and 4 also reported declining cadmium concentrations, with TSF3 showing a secondary increase from week 5 that resulted in exceedances beyond

the midpoint of the experiment. Cadmium concentrations reported for TSF4 remained below screening levels thereafter. Chromium exceedances were largely confined to TSF1 and 2 for most of the leaching period, with TSF1 consistently exhibiting higher concentrations. No chromium exceedances were observed in TSF3 or 4 over the 20-week duration.

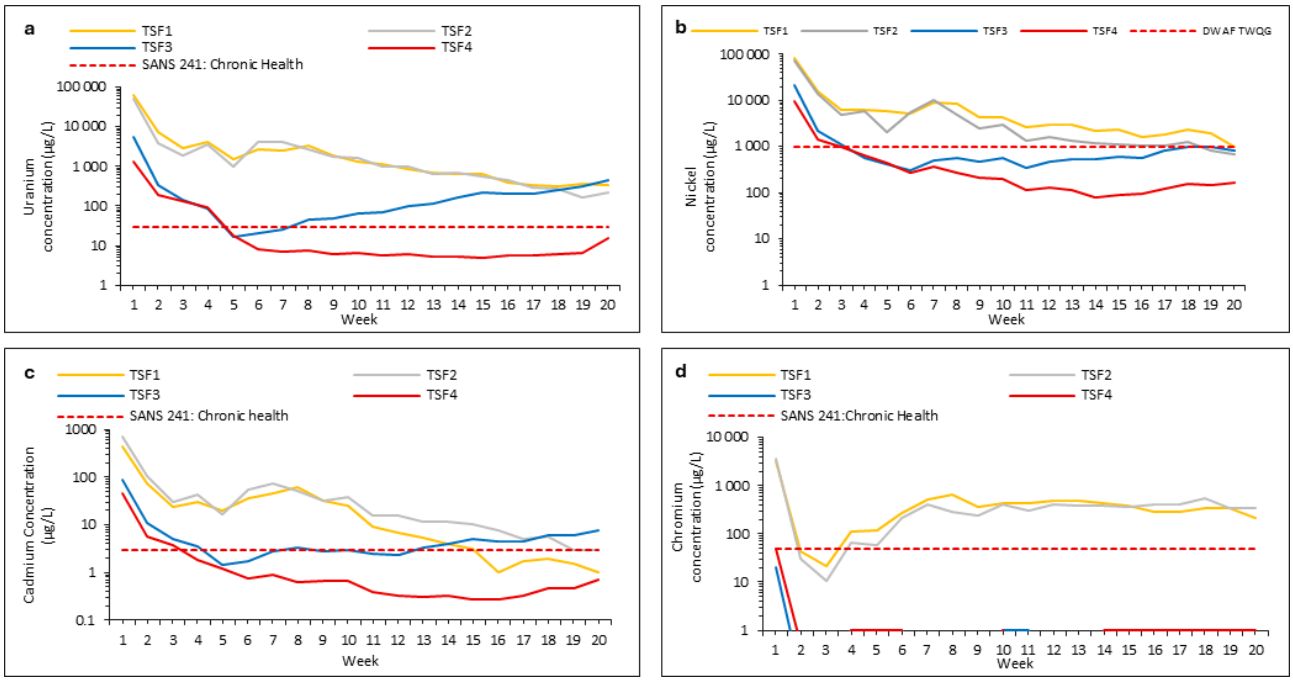

Figure 7 Old tailings storage facility (TSF) complex composites' metal leaching profiles. (a) Uranium; (b) Nickel; (c) Cadmium; (d) Chromium

3.3.6 Scaling the kinetic test result

There has been a concerted effort over several years to understand how the results of various laboratory-based geochemical testing can be used to predict drainage water generated from mine waste byproduct as part of extension or closure. However, no established methodology exists to scale up microscale laboratory results to predict drainage quality for larger-scale field structures. Furthermore, due to a lack of biological activity in laboratory-based tests, processes such as microbial catalysis of Fe^{2+} oxidation, sulphur cycling and sulphide weathering are not adequately captured under controlled laboratory conditions.

Notwithstanding this, ongoing research is examining the correlation between laboratory testing results and measured drainage quality in the field, with 2 lines of thought on developing appropriate scaling factors.

These are typified by the approach adopted by Stromberg & Banwart (1999), which applies scaling factors for individual mechanistic phenomena to account for differences such as rock composition, reaction environment and solute transport by reviewing many studies in this area. The second approach estimates a scaling factor by comparing the results of field studies with those from laboratory-based procedures.

When selecting the appropriate scaling factor, the literature has been reviewed to determine whether similar correlation efforts have been conducted for sites such as the SDGM Old TSF complex. The research undertaken suggests that a scale factor of 0.001–0.04 is appropriate for adjusting HC test results, based on the empirical approach by Kirchner & Mattson (2015). This is suggested because the SDGM Old TSF complex composites reported total S% between 0.22 and 2.9%.

However, if a scaling factor is calculated using the mean concentrations of common ions and metals detected in downgradient monitoring boreholes around the Old TSF complex, and given SLR's experience with gold mining operations in the Witwatersrand Basin, it is suggested that a scaling factor of 0.016 be applied to the HC results.

This was determined using chloride concentrations in downgradient monitoring boreholes as a conservative tracer. If this factor is applied to the HC testing results, except for Fe in TSF1 and 2, all determinants are either non-detected or are below the national and international water quality and effluent guideline limits referenced for this study.

3.3.7 Closure planning implications

Due to the challenges of upscaling laboratory tests to field scale, closure planning should focus on restricting geochemical evolution of the waste materials by isolating residual pyrite waste materials. Different engineered or vegetated soil covers should be assessed for their efficacy to limit the infiltration of contact water (precipitation) and the ingress of oxygen into tailings complexes to reduce the risk of AMD and metal leaching after closure. This study shows that acidic conditions persist in TSF1 and 2 tailings that are actively being reclaimed, with the dormant TSF3 and 4 tailings showing potential for acid generation. This potential is likely to materialise once their reprocessing begins, which will lead to acidic conditions dominating across the entire complex, with a risk of metal leaching that could impact local water resources. Consequently, this assessment should be considered when advising on closure options and applied in a dedicated feasibility and multicriteria analysis study facilitated by several specialists.

4 Conclusion

The pre- and post- humidity cell (HC) X-ray diffraction (XRD) mineralogy analyses and acid-base accounting (ABA) tests confirmed that TSF1 and 2 tailings are AG. The HC pH and alkalinity profiles for TSF1 and 2 tailings displayed consistently acidic pH trends, indicating that these materials are likely to have a long-term acid-generating (AG) potential. The long-term carbonate molar ratio (CMR) for TSF1 and 2 are both < 1, suggesting that neutralisation capacity will not keep up with the rate of pyrite oxidation over the long term, supporting the AG character of these tailings. The sulphate production rates for TSF1 and 2 suggest that there is a readily available source of residual sulphate in the materials that will emanate from the oxidation of unreacted pyrite. This corresponds to the long-term acidic pH and negligible alkalinity profiles of TSF1 and 2. The low FMR (< 1) for TSF1 and 2 infers that the materials are unlikely to present sufficient neutralisation potential (NP) in the long term.

The XRD results for TSF3 and 4 reported lower pyrite contents, indicating the materials as being potential acid-generating (PAG), reinforced by the ABA tests. TSF3 tailings displayed a pH profile that fluctuated between moderately to weakly acidic to circumneutral, supporting its classification as PAG over the long term. However, the TSF4 tailings displayed a long-term pH profile within a circumneutral range, with moderate alkalinity availability. This indicates that this material poses a low long-term risk of acid rock drainage and is unlikely to generate net acidity in its current setting. The CMR values for TSF3 and 4 are marginally greater than 1, indicating that NP can keep up with pyrite oxidation over the long term. High FMR values for TSF3 and 4 indicate a significant presence of aluminosilicate minerals relative to other minerals and support the long-term potential to neutralise acid, albeit at slower rates compared to when carbonate minerals dominate.

The molar ratios of the major ions indicate that chlorite and talc are the main minerals controlling NP in the Old TSF. The long-term metal leaching risks of the Old TSF complex tailings reported several metal and metalloid exceedances of water quality and effluent standards for local water resources, namely Al, As, Cd, Cr, Mn, Ni, Pb and U. By referencing literature and calibrating the kinetic test results with water-monitoring data at the site, we suggest a laboratory-to-field scaling factor of 0.016. When this scaling factor is applied to the stable leaching kinetic results, except for Fe for TSF1 and 2, all determinants can be reported as either non-detected or below national and international effluent/drinking water guidelines.

Although predicting drainage quality for the Old TSF complex from laboratory tests is useful in identifying and predicting constituents of concern, it does have its challenges when extrapolating the results to field scales. Therefore, closure planning should focus on restricting the geochemical evolution of the waste

materials. Possible options are engineered or vegetated soil covers that limit water infiltration and ingress of oxygen into the Old TSF complex that will retard sulphide oxidisation reactions and thereby assist in mitigating acid mine drainage and metal leaching risks.

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