

Static Test-Based Acid Rock Drainage Formation Potential of Metalliferous Tailings from O’Kiep, South Africa

I.G. Erdogan^{a, b, c}, E. Fosso-Kankeu^{a*}, S.K.O. Ntwampe^{b, c}, F.B. Waanders^a, N. Hoth^d and A. Rand^b

Abstract — One of the environmental challenges in O’Kiep is the increased levels of potentially toxic elements (PTEs) in metalliferous tailings (MTs), which may result in acid rock drainage (ARD) through oxidation. In this study, laboratory-based static tests were conducted on MTs to predict the risk of acid rock drainage potential (ARDP). Mineralogical analysis of the MTs showed that the MTs consisted mainly of quartz, microcline, muscovite, albite, anorthite, magnetite, magnesioferrite, gypsum and halloysite-10A. The major oxides, i.e. $\text{SiO}_2 > \text{Al}_2\text{O}_3 > \text{Fe}_2\text{O}_3 > \text{CaO} > \text{MgO} > \text{K}_2\text{O} > \text{Na}_2\text{O} > \text{TiO}_2 > \text{P}_2\text{O}_5 > \text{SO}_3 > \text{Cr}_2\text{O}_3 > \text{MnO} > \text{NiO}$, in the MTs were higher; while the concentrations of PTEs were in the following sequence: $\text{F} > \text{Cu} > \text{Cr} > \text{Mn} > \text{S} > \text{Zr} > \text{Ba} > \text{Sr} > \text{V} > \text{Cl} > \text{Rb} > \text{Ni} > \text{Zn} > \text{Y} > \text{Co}$. The mineralogical and geochemical characteristics of the MTs indicated oxidative weathering of the PTEs. The MTs paste averaged a pH of 3.1, with the net neutralization potential of - 210 kg CaCO_3/t , neutralizing potential ratio of 0.1 and a total sulfur of 6.1%, an indication that the MTs would be prone to ARDP. Furthermore, the leaching of the MTs increases the risk of contamination of surrounding groundwater bodies in O’Kiep. Additionally, there are increasing concerns over significant potential environmental health effects of the MTs, including the mobility of the PTEs, which necessitates the development of suitable strategies for reclamation and the rehabilitation of the mining site in the O’Kiep area.

Keywords— Acid rock drainage potential, metalliferous tailings, O’Kiep, potentially toxic elements, static tests.

I. INTRODUCTION

Metalliferous mining operations generate large volumes of finely crushed rock, commonly referred to as metalliferous tailings (MTs). It is characterized by fine particle size ($>100 \mu\text{m}$) and is stored in the tailings storage facilities (TSFs) [1]. MTs, especially those found in arid or semiarid

environments, pose a long-term health hazard for nearby communities through exposure to potentially toxic elements (PTEs) containing dust originating from the MTs with observed minimal plant growth due to soil toxicity [2]. The reactivity of MTs can result in acidic, highly charged waters [3]. Disposal of MTs is a major concern for many countries as it contaminates the environment including the atmosphere, water resources and soil [4, 5]. The MTs always contain PTEs concentrated during ore processing, which may disperse into the nearby environment and threaten public health [6]. Elements such as Al, Cd, Cu, Fe, Mn, Pb and Zn, Mn are essential for cell growth and metabolism; however, at high levels, they are toxic for living organisms [5]; hence, referred to as PTEs. Under watery acidic conditions, these PTEs will be released from MTs [7], as they will undergo chemical alteration under such conditions. The chemical alterations are most often a function of exposure to atmospheric oxidation[8].

TSF containing copper sulfide MTs in O’Kiep has been exposed to the weathering reactions for years. Although the environmental impact of MTs on the nearby environments, including windblown is well known [9], only a few comprehensive studies have been carried out in O’Kiep. In general, the primary minerals in the MTs are quartz and pyrite, with secondary minerals such as goethite. Acid rock drainage (ARD) from such MTs is generated by the moisture-based weathering and oxidation of sulfide minerals such as pyrite [10]. ARD management approach is neutralization by an alkaline reagent such as limestone to raise the pH and remove most of the PTEs through precipitation reactions [10, 11]. O’Kiep MTs are potentially primary contributors to ARD due to their high sulfide content [12]. Generally, studies on acid rock drainage formation potential (ARDP) of the MTs in O’Kiep are limited. ARD prediction is a significant subject in order to predict and prevent long-term deleterious environmental contamination. However, the extent of the acid generation, as well as the sources and dynamics of PTEs from O’Kieps MTs, have not been determined thus far. Therefore, the geochemical and mineralogical characterization of these MTs was carried out in order to evaluate the capacity for PTEs release over time. In addition, static tests such as acid-base accounting (ABA) were conducted in order to determine the O’Kieps MTs acid generating potential.

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^aWater Pollution Monitoring and Remediation Initiatives Research Group in the CoE C-based fuels School of Chemical and Minerals Engineering, Faculty of Engineering, North-West University, Potchefstroom – South Africa.

^bBioresource Engineering Research Group (BioERG), Department of Biotechnology, Cape Peninsula University of Technology, Cape Town, South Africa.

^cDepartment of Chemical Engineering, Cape Peninsula University of Technology, Cape Town South Africa.

^dInstitute of Mining and Special Civil Engineering, Technische Universität Bergakademie Freiberg, Saxony, Germany.

II. MATERIALS AND METHODS

A. Study area and sample collection

The MTs used for this study were collected from a tailings storage facility (TSF) in the arid town of O'Kiep [29°35'45.0"S 17°52'51.0"E]. The main climatic and geological characteristics of the study area have been described by Erdogan [13]. Approximately 5 kg of MTs representative of grab samples (n=10) using generic field sampling guidelines described by USEPA Operating Protocol for Soil sampling USEPA [14] and Moyle and Causey [15]. Each sample was collected at a depth of 0.6 m from the surface, using a Draper 24414 steel auger [16], during August 2017 (wet season). The sampling points were marked (MT1 to MT10) using a Global Positioning System (GPS), as shown in Fig.1.

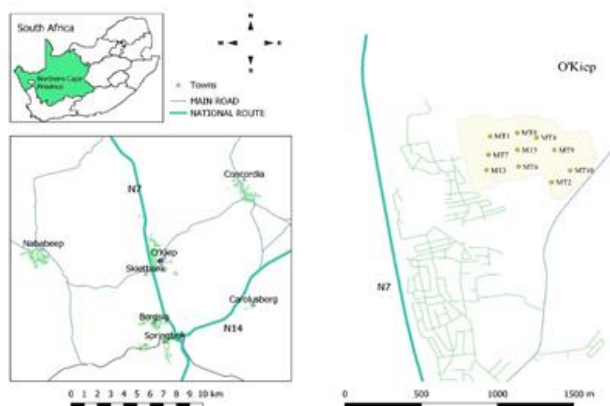


Fig. 1: Sampling points generated using Quantum GIS software (v. 2.18.11) and data from National Geo-Spatial Information (NGI).

The samples were air-dried and were homogenized and sieved to 2 mm in the laboratory. All samples (n=10) were heterogeneously mixed to form one composite sample of approximately 5 kg. The composite sample was stored in double-sealed plastic zipper bags and further analyses were conducted. A portion of the MTs sample was used for mineralogical and geochemical characterization and was analyzed at the University of Cape Town, Department of Geological Sciences, South Africa. The mineral phases present in the MTs samples were identified and quantified using the Philips PW1130/90 X-ray diffractometer (XRD), and the geochemical composition oxides and potentially toxic elements (PTEs) of the MTs sample were measured by Panalytical Axios wavelength-dispersive X-ray fluorescence (XRF). The details of sample preparation, instrumentation condition for each method are described by Beckhoff [17] and Compton [18].

B. Static tests

A modified field and laboratory test method for ABA Sobek [19] and test modified by Lawrence and Wang [20] was used in static tests. Additionally, a prediction test was performed using a paste for pH testing to assess individual samples' acid-forming characteristics. Static tests are empirical techniques usually conducted to predict ARDP. The most common static tests ABA [21], which is based on the determination of three main factors: 1] acid consumption 2] acid formation and 3] determination of

the net acid consumption or production [22]. Subsequently, the determination of acid potential (AP) and neutralization potential (NP) allowed for the computation of the net neutralization potential (NNP), including the net potential neutralizing ratio (NPR) - a criteria used for the determination of ARDP of the MTs sample.

III. RESULTS

A. Mineralogy and geochemical characterization

The primary minerals in the MTs were quartz, microcline, albite, magnesioferrite, magnetite, gypsum, clinochlore-1MIIb, anorthite, Muscovite-1M and halloysite-10A as presented in Table I. These were interlinked to the geochemical conditions at the surface of the TSF, which revealed a heterogenic environment that can be categorized divided into an acidic and oxidizing zone. The major oxides and PTEs are presented in Table II.

TABLE I: MINERALOGY OF THE METALLIFEROUS TAILINGS FROM O'KIEP

Minerals	Formula	%
Quartz	SiO ₂	26.3
Microcline	KAlSi ₃ O ₈	10.4
Albite	NaAlSi ₃ O ₈	8.34
Magnesioferrite	MgFe ₂ O	4.17
Magnetite	Fe ₂ O ₄	4.17
Gypsum	CaSO ₄ ·2H ₂ O	2.6
Clinochlore-1MIIb	(Mg ₅ Al)(Si,Al) ₄ O ₁₀ (OH) ₈	1.13
Anorthite	(Ca,Na)(Al,Si) ₂ Si ₂ O ₈	0.65
Muscovite-1M	KAl ₂ Si ₃ AlO ₁₀ (OH) ₂	0.33
Halloysite-10A	Al ₂ Si ₂ O ₅ (OH) ₄ ·2H ₂	0.26

TABLE II: OXIDES AND POTENTIALLY TOXIC ELEMENTS FROM METALLIFEROUS TAILINGS

Oxides	wt. %	PTEs)	mg/kg
SiO ₂	57.4	F	1177
Al ₂ O ₃	13.5	Cu	921
Fe ₂ O ₃	12.5	Cr	877
CaO	3.41	Mn	830
MgO	2.57	S	786
K ₂ O	2.56	Zr	753
Na ₂ O	2.49	Ba	686
TiO ₂	1.56	Sr	249
P ₂ O ₅	0.48	V	247
SO ₃	0.27	Cl	168
Cr ₂ O ₃	0.12	Rb	158
MnO	0.11	Ni	114
NiO	0.02	Zn	98.0
LOL	1.92	Y	86.9
Total	99.4	Co	37.5

A. Static test predictions

Static tests were carried out in the MTs to measure the acidic and neutralization potentials of the sampled MTs [19]. The paste pH test was also used to indicate the nature of the studied MTs in terms of pH [23]. The ABA static test was conducted to predict the acid-generating potential of the MTs. ABA measurements indicate AP and NP of a given sample. The ABA results according to the modified static test proposed by Sobek

[19] and Lawrence, and Wang [20] are shown in Table III.

TABLE III: ACID-BASE ACCOUNTING ANALYSES OF METALLIFEROUS TAILINGS FROM O'KIEP

Paste pH	3.1
Total Sulphur (%)	6.09
Acid Potential (AP) (kg/t)	190
Neutralization Potential (NP)	-19
Nett Neutralization Potential (NNP)	-210
Neutralising Potential Ratio (NPR)	0.102

IV. DISCUSSION

A. Metalliferous Tailings Characterisation

MTs from the former closed metalliferous mine of O'Kiep in Namaqualand showed coarse-fine textures with a specific gravity of 3.104 m²/g [12]. Similar results were obtained from an underground copper mine tailings in southern China [24]. Aluminosilicates such as microcline, albite clinocllore-1MIIB, anorthite, muscovite-1M, and halloysite-10A were abundant in the sample, which is in agreement with the gneiss nature of the granitic host rock in O'Kiep [25]. Aluminosilicates such as albite, clinocllore-1MIIB, anorthite, muscovite-1M and halloysite-10A may raise the pH, subsequently the dissolution of chloride [26]. Secondary Fe minerals can be formed from the reduction and dissolution of Fe(III) in crystalline Fe minerals, i.e., magnesioferrite and magnetite commonly present in the MTs via transformation processes catalyzed by Fe(III)-reducing bacteria [27]. The MTs sample also contained magnesioferrite and magnetite, which are primary sources of ARDP. Magnesioferrite and magnetite are the major acid producing minerals in copper MTs [4].

Quartz is the most dominant primary mineral and this shows that it is less reactive in oxidizing conditions and have minimal potential to neutralize acid, while muscovite belongs to a very slow weathering group due to their physical characteristics [28 - 30]. Furthermore, the presence of Halloysite10A was indicative of constituents that normally occur in arid climates [31]. However, the presence of CaO can potentially provide neutralization potential to the acid being formed [28]. Silicate minerals were found to be the most important contributors to the neutralization potential. Of the silicate minerals, Anorthite was observed to be the most important contributor to the total NP, followed by the other silicate minerals of the fast weathering groups [30]. Currently, there is a community that lives within the surroundings of the TSF in O'Kiep wind-borne, certainly being inhaled by the populace. Oxides observed in the MTs were SiO₂ > Al₂O₃ > Fe₂O₃ > CaO > MgO > K₂O > Na₂O > TiO₂ > P₂O₅ > SO₃ > Cr₂O₃ > MnO > NiO. The MTs were free of primary sulfide constituents. However, exhibited significantly higher average concentrations of F > Cu > Cr > Mn > S > Zr > Ba > Sr > V > Cl > Rb > Ni > Zn > Y > Co.

B. Static tests predictions

Static tests (Table II) were conducted in order to define the potential acidity generated by the MTs disposed-off in TSF in O'Kiep. The MTs samples used in this study were collected in acidic zones of the TSF which was confirmed by the paste pH (3.1) of the MTs, revealing the elevated acidity of the MTs. Low paste pH can be related to iron sulfide oxidation and also to soluble mineral dissolution [32]. The modified ABA test classified the MTs sample as having low sulphide-S content (6.1%) and highly negative values of NNP (-210 kg CaCO₃/t) indication that the MTs are likely to generate ARD when exposed to atmospheric conditions [33].

Additionally, based on the NPR ratio of 0.1 and a total sulfur content (S) of > 3%, there can be a higher chance of ARDP [34]. The value of the NP obtained was -19 in MTs studied which is in accordance with the low values of acid-neutralizing minerals obtained, such as CaO (3.41 mg/kg) and gypsum (2.6 wt%); albeit, these conditions could enhance PTEs leachability and cause an increased environmental risk for the nearby groundwater sources. ARD seepage effects can manifest in low pH waters (1.4 to 4), with subsequent precipitates of hydrous ferric oxides including efflorescent sulfate salts, and blooms of green filamentous biofilms on the TSF [35] which is the case in O'Kiep. Consequently, the effects of ARD and aggradation by mineralized MTs would affect many communities further downstream [36], therefore reducing the amount of usable water in a country where physical water scarcity is already a concern [37-55]. Additionally, the chemical and physical dispersion of PTEs might continue from the TSF for the predictable future unless a mitigation strategy is implemented.

V. CONCLUSION

The MTs of O'Kiep contains PTEs such as F > Cu > Cr > Mn > S > Zr > Ba > Sr > V > Cl > Rb > Ni > Zn > Y > Co. While the, major oxides were found to be SiO₂ > Al₂O₃ > Fe₂O₃ > CaO > MgO > K₂O > Na₂O > TiO₂ > P₂O₅ > SO₃ > Cr₂O₃ > MnO > NiO. The NNP values of the tailings were -210 kg CaCO₃/t, and sulfur content was high (6.1%), hence, the MTs were classified as acid-producing by the ABA test. Although the tailings were disposed years ago, the MTs are likely to produce ARD containing PTEs for a long period unless remedial measures are taken into consideration. It is recommended that a kinetic test be designed to mimic weathering at the laboratory scale to determine the rate of acid-generation, the variation over time in leachate water quality and thus allow to develop mitigating strategies.

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Ms. Innocentia Erdogan is currently a Lecturer at Cape Peninsula University of Technology in the Department of Chemical Engineering. Innocentia is also a PhD candidate at the School of Chemical and Minerals Engineering, North West University.