

Acid-Base Accounting (ABA) of Coal Tailings from Middleburg Area, South Africa

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Abstract—Acid generated by coal mine tailing can have a detrimental effect on the environment and human health. Determining whether acid mine drainage (AMD) will occur is of the utmost importance to develop a remediation strategy. Acid base accounting (ABA) is one of the most popular test protocols used today to assess and classify the acid generating potential of mine waste rock (tailings)

The results of the ABA test revealed the potential of coal tailings of a wash plant situated in Middleburg to generate acid, which will lead to AMD if adequate steps are not taken to slow down the rate of acid generations and if suitable management plans are not implemented.

Keywords— Acid Base Accounting (ABA), Acid generation potential, Coal mine tailings, Middleburg.

I. INTRODUCTION

South Africa has mined coal commercially from 1857, and is one of the largest coal producers in the world. Coal is currently the primary source of energy for power generation in South Africa and will continue to be the primary source for the foreseeable future [1]. Acid mine drainage (AMD) is the second largest threat to sustainable life after global warming [2].

AMD is a result of oxidation of sulphide minerals in the presence of oxygen and water. The AMD effluents are usually very acidic and contain high concentration of sulphate (SO_4^{2-}) and dissolved metal ions. The AMD effluent is highly acidic due to the sulphuric acid (H_2SO_4) that is produced during the oxidation of sulphide minerals [3-18]

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ABA assesses the chemical and physical properties of waste rock tailings [19]. Acid base accounting (ABA) tests evaluate the balance between acid neutralizing processes and acid generating processes. ABA is used as preliminary method to classify samples as potential acid forming (PAF), non-acid forming (NAF) or uncertain (US) [20].

Mineral extraction and mining are common practice around the world. Mining for metal ore, salts or coal results in excavation and transport of large quantities of ore to the surface. The excavated ore is then subject to beneficiation process. If the ore deposits contain large quantities of the desired element, mining continues for prolonged periods of time. The beneficiation process removes the gangue material from the ore increasing the economic value of the ore. The beneficiation produces large quantities of by-products (waste rock) which must be transported and placed somewhere in the surrounding environment [21].

Mines are usually surrounded by piles containing waste rock that was produced by the beneficiation process. These waste rock piles are known as stockpiles, tailings or waste rock dumps. These tailings generally contain materials with a low ore grade, but still contain high concentration of sulphide minerals. These sulphide minerals undergo weathering and oxidation to produce acid and dissolved metals [22].

Most often these tailings are piled in large repositories, these mine tailings can reach hundreds of meters high and kilometers in length. Tailings have unique properties based on the original geological deposit that was mined and the type of beneficiation process that it has undergone. The properties of the tailings vary from mine to mine, but may differ across tailings deposited from the same mine [21].

AMD is the second biggest threat to sustainable life after global warming. The two aspects of AMD that affect the environment are the high concentration of sulphuric acid and dissolved metals released into underground water, streams and rivers [2-39].

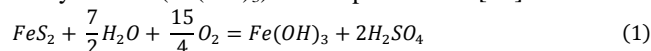
AMD destroys ecosystems and contaminates water in areas where freshwater is in short supply and is toxic to most forms of life [20].

During coal mining the coal mine tailings are exposed to oxygen and water rich environment. In the presence of oxygen saturated water, oxidation of sulphur bearing minerals will most likely occur and generate acid. Sulphur-bearing minerals that can contribute to acid generation include sulphide minerals, and sulphate minerals. Sulphide minerals contribute the most to acid generation. Pyrite (FeS_2) is considered to be the sulphide mineral that contributes the most to acid

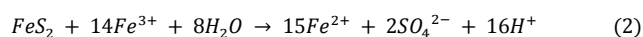
generation [3], [20], [22], [40].

Not all sulphide minerals generate acid under normal conditions. However, all sulphide minerals are capable of producing acid if oxidized by aqueous ferric iron.

Reaction equation (1) represents the general acid generation reaction associated with AMD. In (1) the sulphide mineral pyrite (FeS_2) is oxidized by oxygen and water to produce ferric iron hydroxide ($\text{Fe}(\text{OH})_3$) and sulphuric acid [41].



The rate of oxidation of sulphide minerals in the presence of ferric iron solution at low pH levels are up to two order magnitude higher than that of oxidation by oxygen. Ferric iron is seen as the more aggressive and effective oxidizer. However, oxygen still plays a vital role in sustaining the ferric iron concentration through oxidation of ferrous iron to ferric iron. The reaction mechanism of pyrite oxidation by ferric iron can be seen in (2) [42-44].



The acid generated during the oxidation of sulphide minerals will react with acid neutralizing minerals within the waste rock tailings. The acid neutralizing or pH-buffering reaction that contribute the most are the dissolution of carbonate minerals, aluminosilicate minerals and ferric iron hydroxides.

To predict whether AMD will occur, the acid generating potential of a waste rock tailing are evaluated. ABA test procedure are relatively dependable and inexpensive analytical procedure to assess chemical and physical properties of waste rock tailings [19].

ABA test evaluates the balance between acid neutralising processes (dissolution of alkaline carbonates, weathering of silicates and displacement of exchangeable bases) and acid generating processes (oxidation of sulphide minerals) [20]. The ABA tests procedure was first published in 1978, and had become the method of choice to assess the acid generating capabilities of waste rock [45].

In this study the potential of a tailing sample from the Middleburg area of South Africa to generate acid was evaluated using the acid-base accounting procedure.

II. METHODOLOGY

A. Sample collection and preparation.

The sample (SP1) was collected from the top of a 5 m high coal tailing pile from an undisclosed coal washing plant in the Middelburg. The sample was air dried and pulverized using a ball mill. The sample was then sieved to obtain a sample with a partial size smaller than 75 μm .

B. Paste pH

The paste pH was measured using a pH meter (Multiparameter pH meter, Hanna). Which was calibrated beforehand using standard buffer solutions with a pH of 4.0 and 7.0.

C. Maximum Potential Acidity (MPA)

The MPA was calculated using (3) [46].

$$\text{MPA} = (\text{Total } S\%) \times 30.6 \quad (3)$$

Total sulphur weight % ($S\%$) and sulphide weight % (S_i) were determined using an ELTRA CS 2000 Carbon Sulfur Determinator, PC controlled.

D. Acid Neutralization Capacity (ANC)

The following procedure was obtained from [46].

An accurately weighed 2.00 g (m) sample was placed into a 250 mL beaker. 20 mL (V_{HCl}) of 0.5 M (M_{HCl}) hydrochloric acid was pipetted into the flask and 20 mL of distilled water was added thereafter.

The beaker was covered with a watch glass and heated at 85°C for 2 hours. And then allowed to cool down to room temperature. Distilled water was added to the solution to obtain a solution volume of 125 mL.

The pH of the solution was measured to be 1, which should be between 0.8 and 1.5 to ensure that the correct volume and concentration of hydrochloric acid was used [47].

The solution was back titrated with 0.5 M sodium hydroxide to a pH of 5. Two drops of 30% hydrogen peroxide was added at a pH of 5 to encourage the oxidation of dissolved ferrous iron (Fe^{2+}) as well as the precipitation of iron (III) hydroxide ($\text{Fe}(\text{OH})_3$) [47]. The solution was further titrated to a pH of 7. The volume of sodium hydroxide used to titrate the solution to a pH of 7 (V_{NaOH}) was then recorded. The procedure was repeated on a blank sample [46].

Equation (4) was used to determine the ANC of the sample, where the constant C has a value 49 to obtain the ANC value in ($\text{kg H}_2\text{SO}_4/\text{t}$) [17].

$$\text{ANC} = \frac{\left(V_{\text{HCl}} - \left[V_{\text{NaOH}} \times \frac{V_{\text{HCl,Blank}}}{V_{\text{NaOH,Blank}}} \right] \right) \times M_{\text{HCl}}}{m} \times C \quad (4)$$

E. Mineralogy

The mineralogy of the sample was determined using X-ray powder diffraction (XRD).

The ANC based on mineralogy (ANC_{min}) is the sum of the ANC values calculated for each respective acid neutralizing mineral. The samples were subject to a XRD analysis using Panalytical X'Pert Pro multipurpose Diffractometer, 3 kW powered used at 40kV. Programmable divergence and anti-scatter slits; primarily Soller slits:0.04 rads; 2θ range:4-100. The interpretation was done using Rietveld method.

The sample was subject to X-ray fluorescence (XRF) to determine the porting of elements in the suspended solids. The XRF analysis was conducted using Axios^{mAX} spectrometer with SuperQ Version 6.

F. Mineral ANC calculations.

The ANC_{min} of carbonate minerals was calculated using (5) [48].

$$\text{ANC}_{\text{min}} = 10 \times [C_{\text{wt,frac}}] \times \left[\frac{MW_{\text{cal}}}{MW_c} \right] \times \text{wt}\% \times RC \times CF \quad (5)$$

Where $C_{\text{wt,frac}}$ is the weight fraction of carbon in the carbonate mineral.

CF is a conversion factor with a value of 0.98 to convert the ANC from kg CaCO₃/t to kg H₂SO₄/t.

wt% is the weight percentage of the mineral in the sample.

MW_{cal} and MW_c are the molar weights of calcite and carbon respectively.

R represents the relative reactivity.

The ANC_{min} of non-carbonate minerals were calculated using (6) [48].

$$ANC_{min} = 10 \times \left[\frac{MW_{cal}}{MW_m} \right] \times wt\% \times R \times CF \quad (5)$$

MW_m represents the molar weight of the neutralizing mineral.

III. RESULTS AND DISCUSSION.

The results of the ABA test are summarized in Table I.

TABLE I: RESULTS

| Sample | Past pH | S% | S _i % | MPA (kg H ₂ SO ₄ /t) | ANC (kg H ₂ SO ₄ /t) |
|--------|---------|-----|------------------|---|---|
| SP1 | 2.5 | 1.6 | 1.5 | 49 | 0 |

The paste pH is extremely low providing a strong indication that the sample will be PAF with a high quantity of stored acidity in the form of sulphates. Furthermore, the mobility of heavy metals will be high.

The MPA was determined using the total sulphur content instead of the total sulphide content due to following reasons. There is not a large difference between the sulphur and sulphide content thus using total sulphur that the MPA is not underestimated and may compensate for the stored acidity that is not incorporated in the MPA calculations. All sulphide-bearing minerals have the ability of generating acid when oxidized by Ferric iron (Fe³⁺) at low pH levels, thus at a pH of 2.5 it can be assumed that all the sulphide minerals shall produce acid [49-50].

The calculated ANC value had a negative value thus an ANC value of 0 was used as reference value. Based on the MPA and ANC results, a NAPP and ANC/MPA ratio of 49 kg H₂SO₄/t and 0 were obtained respectively.

The sample is classified as PAF, as the NAPP is well above 20 kg H₂SO₄/t and ANC/MPA is well below 1.

The mineralogy of the sample is presented in Table II.

| Mineral | Molecular formula | Weight percentage (wt%) |
|------------|--|-------------------------------|
| Calcite | CaCO ₃ | 0.3 |
| Dolomite | CaMg(CO ₃) ₂ | 0.2 |
| Muscovite | KAl ₂ (Si ₃ Al)O ₁₀ (OH,F) ₂ | 1.3 |
| Kaolinite | Al ₂ Si ₂ O ₅ (OH) ₄ | 29.2 |
| Pyrrhotite | Fe _{1-x} S | 0.4 |
| Gypsum | CaSO ₄ · 2H ₂ O | 0.8 |
| Quartz | SiO ₂ | 13.5 |
| Amorphous | | 52.9 |

Calcite, dolomite, muscovite and kaolinite are acid neutralizing minerals present in the sample. Muscovite and

kaolinite will not contribute to the ANC_{min} as their relative reactivities are extremely low. Calcite and dolomite will contribute to the ANC_{min}. Using (5) and the calcite and dolomite content of 0.3 wt% and 0.2 wt% respectively, an ANC_{min} of 5 kgH₂SO₄/t was obtained. ANC_{min} is extremely low due to the low content of neutralizing minerals, confirming the ANC obtained from the ABA.

As seen in Table II, the iron sulphide consists mainly of pyrrhotite. The sample has pyrrhotite content of 0.4 wt% which is relatively low compared to the sulphide content of 1.5 wt%. The MPA of the sample may be overestimated due to the low pyrrhotite content and thus additional tests such as the net acid generation (NAG) test should be conducted to evaluate the MPA predicted by the ABA. However, the sample can be classified as PAF with certainty due to the following reasons:

- Extremely low pH.
- Relatively high sulphur and sulphide content.
- Extremely low ANC.
- High acid generation due to oxidation via ferric iron.

IV. CONCLUSION

In brief, the ABA results show that the tailings are generating acid and that the rate of acid generation is at its peak or approaching its peak and thus immediate step should be taken to slow down the rate of acid generation and implement suitable management strategies.

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