A Detailed ABA Study of the Coal-Bearing Formations in the Waterberg Coalfield, Limpopo Province, South Africa

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ABSTRACT

Coal constitutes 77% of the primary energy needs in the country, with the Waterberg Coalfield estimated to host about 40% of the remaining South African coal resources. The Karoo coals were deposited in a reduced environment that have the potential to produce sulphides within the sediments they are hosted. The sulphur content within the coal can range from 0.1 wt.% to as high as 10 wt.%. Mining generates a disturbance in the natural groundwater levels and affects the surrounding water chemistry when sulphate is produced as a result of pyrite oxidation.

Acid base accounting (ABA) was used to determine the balance between the acid producing potential (AP) and acid neutralizing potential (NP). From the analysis the Net Neutralising Potential (NNP) classified samples as either acid or non-acid producing. ARD does not only result in the generation of acid but is accompanied by decreased pH and increased values of specific conductance, dissolved metals and sulphate.

The ABA results showed that interburden and coal samples have higher risks of producing acid upon oxidation than overburden samples. Higher concentrations of neutralising minerals are present in overburden samples. ABA indicated that the material 60m below ground surface had a higher acid producing potential than the material above. The analysis from kinetic tests showed the long-term behaviour of different samples, with the electrical conductivity (EC) and pH changing over time. Samples with lower pH continued to produce more sulphate, while calcium continued to increase until it was depleted from the samples. Inductively coupled plasma analysis determined the release of the heavy metals which can be detrimental to the environment, such as As, Co, Ni and Pb.

The water demand will increase as mining continues in the area, with inter-catchment transfers identified to overcome local water scarcity issues. ARD poses a big threat to both groundwater and surface water resources.

Keywords: New coal mine, ABA, lithology classified

INTRODUCTION

The Waterberg coalfield (Figure 1) has been noted as the continental powerhouse of coal fuelled electricity production (Waterberg Municipality, 2013). The shortage of water is a problem as new mining communities are emerging. The impacts of acid mine drainage has been under investigation for years, in both operating and closed mines around the world.

Different methods are available for the prevention, management, treatment and prediction of ARD but no method can guarantee the remediation of ARD impact. No impacts have been noticed so far in the Grootegeluk mine, but according to the ABA done in the area (Vermeulen *et al.*, 2011) some of the lithology shows a very high acid potential.
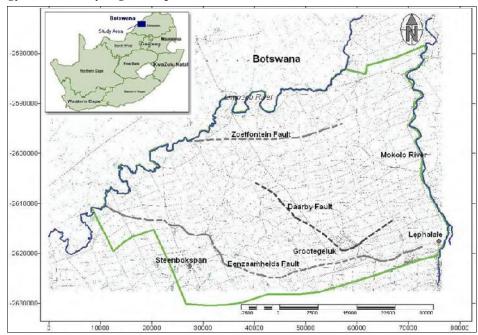

Figure 1 Location of study area. Insert: Map of South Africa indicating the position of the coalfield

Acid base accounting is used as a prediction tool to determine the acid potential of the lithological units. The 11 coal-bearing zones in the Waterberg coalfield are mined in benches and handled according to their intended use. There are two main coal-bearing formations in the Waterberg coalfield (Jeffrey, 2005); these coal zones extend from the Volksrust formation into the Vryheid formation. Vermeulen *et al.* (2011) described three main categories of coal thickness (identified by Bester, 2009), based on the weathered phase of the geology in the area. Figure 3 shows the three categories, green for full succession of coal, yellow for Middle Ecca and pink where area has been weathered in parts.

METHODOLOGY

Samples from different exploratian companies were collected from the study área in Figure 1. The investigation was conducted on various zones of borehole stratigraphy and on discards from the only active mine's beneficiation plant. The sampled material consisted of overburden, the mined interburden and discard after the coal material was liberated, as well as fly ash sampled form the Matimba Power Station.

Acid-Base Accounting was designed to evaluate the acid producing capability of coal mine wastes, and it is now used in a broader field to evaluate both the coal and the metal mine wastes (Lapakko *et al.*, 1999). The detection limits and outlines of static ABA criteria is shown in Table 1. The criteria is used during the analyses of the overburden and interburden. The balance between the acid producing potential (AP) and the neutralising potential (NP) is measured. The acid potential is calculated from the determination of the sulphur content and also represents the sulphur contained in the acid generating sulphide minerals within the sample. NP is determined by subjecting the mine waste samples to some form of acid digestion and represents the amount of acid-neutralising carbonate minerals present in the sample. Net Neutralising Potential (NNP) is the difference between NP and AP. NNP measurements classify rock or coal samples as potentially acid or non-acid producing. A greater NP than AP produces a positive NNP; on the contrary, if NP is less than AP, the resulting NNP will be negative.

Table 1 Detection limits and outlines of the Static ABA analyses

	Static ABA
Analyses / Used for	Used to determine the acid generating potential.
Data format	Expressed as calcium carbonate equivalents and compared to compute net acid-producing or neutralising potential.
Standards used	According to Usher et al. (2002), "material exhibiting a net acid production potential of 5 tons/ 1000 tons of overburden material or more as calcium carbonate equivalent, is classed as toxic or potentially toxic".
Sample preparation	Crushed by Retsch KG 5657 Haan BB100, milled by Giebtechnik Labor-Scheibenschwingmmuhle.

Oxidation products are analysed by using peroxide where only the reactive species (Usher, 2003) will oxidise. The reaction will produce acid and provides an indication of the heavy metal content. The final water quality is also provided as a result of the oxidation reaction.

Metal release

Metal and trace element content were analysed using the inductively coupled plasma optical emission spectroscopy (ICP-OES). This was used to analyse the release of the elements Ag, Al, As, Ba, Be, Ca, Cd, Cr, Co, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Na, Ni, Sb, Se, Sn, Sr, Pb, V, Zn and SO_4. As the oxidation process takes place, metals are mobilized and some become soluble. In the case of the iron species it converts from Fe^{3+} to Fe^{2+} at a pH of 3.1 (Hodgson *et al.*, 1999). If the sample has not yet oxidized in the field the initial pH of a sample will have a higher concentration of sodium carbonate. Calcium/magnesium carbonate exists at a pH of 6 and decreases as the pH decreases during acidification.

Kinetic test

Table 2 lists the kinetic cells used in the study and the ABA results of the same samples. A total of 13 samples used in the analysis were taken from Grootegeluk, Sasol and Sekoko cored boreholes. The criteria that were used to select the samples are the ones that gave inconclusive ABA results, and samples which had extremely high or low NNP from the ABA analysis.

Table 2 Kinetic cell samples as selected from the ABA results

Kinetic cell	Initial pH	Final pH	AP (Open)	AP (Closed)	NNP (Open)	NNP (Closed)
S8	7.86	1.98	32.35	64.71	7.25	14.51
S14	7.11	1.88	20.47	40.94	-11.28	-21.74
S15	6.77	1.72	11	22	-10.12	-21.12
S16	6.80	1.81	1.19	2.38	-0.07	-1.26
S34	6.8	1.8	2.09	4.18	2.64	4.73
SEK 1	6.94	2.08	0.82	1.63	-4.18	-4.99
SEK 3	6.17	2.97	0.58	1.16	-1.60	-2.18
S38	8.53	7.88	1.67	3.34	49.16	50.83
G10	8.14	6.73	0.04	0.08	3.34	3.38
G12	8.62	8.6	0.14	0.27	310.8	310.9
G18	7.71	4.82	0.50	1.00	-5.18	-5.68
SEK 2	7.18	4.07	0.20	0.41	3.16	3.37
SEK 4	6.95	6.54	21.01	42.01	18.29	39.29

Comparison of Leco (%S) with H_2O_2 (%S)

The total sulphur content of the samples was determined by the Leco method. Total S will overestimate the acid potential if a significant amount of sulphate S, organic S or other non-pyritic forms occur in the sample. The same samples were selected and analysed by both the Leco and H_2O_2 method. The relationship of the different methods of analyses for %S is illustrated in Figure 2.
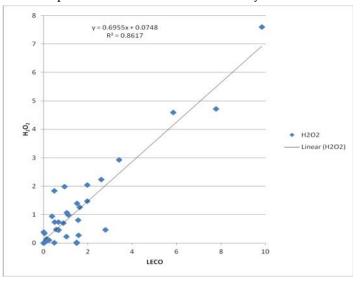

Figure 2 Comparison between the %S of the LECO method and the %S H_2O_2 method. The linear correlation between the two methods establishes that both methods %S can be used to determine the acid potentials of samples.

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The Leco method has higher sulphur values than those obtained for the H_2O_2 method; however, the values for the H_2O_2 method compared well to the values of the Leco analyser. Due to this similarity, the H_2O_2 methods sulphur values can be used to determine the total sulphur present in the samples.

RESULTS AND DISCUSSION

Acid Base Accounting

The ABA and kinetic tests highlighted the lithological units that are prone to become acidic or poses a neutralising potential. The geological units with a NP above the coal units included calcrete and sandstone. These units typically consist of quartz, kaolinite, muscovite, calcite, dolomite, rutile and feldspar. The minerals identified within the mudstone and shale units included quartz, kaolinite, muscovite, pyrite, hematite, marcasite and calcite. A summary of the geological logs, the minerals within the units as well as their AP and NP, are illustrated in Figure 3.

Mineralogy

The samples with a NP mainly contained the minerals calcite and ankerite/dolomite. This is due to the carbonate content within the mineral chemistry. Pyrite is the dominant mineral in the acidifying samples. The mineral chemistry of pyrite contains iron and sulphide elements, which under favourable conditions, contribute to the production of acidic waters.

There is a difference in the mineral distribution of the three successions over the various lithologies. The different minerals found in the mudstones are shown in Table 3.

Table 3 The mineral distribution found in mudstones in the different successions.

Minerals	Full succession	Partly weathered	Middle Ecca
Quartz	■	■	■
Kaolinite	■	■	■
Siderite	■		
Calcite	■	■	
Muscovite	■	■	■
Rutile	■	■	■
Pyrite	■	■	
Marcasite	■		
Hematite	■		
Apatite		■	
Pyrrothite		■	
Feldspar			■

The mudstones have both neutralising and acid generating potential, which are associated with their mineral chemistry. The mineral chemistry for the sandstones from the three successions appears to be relatively similar with minor exceptions for hematite, ilmenite and feldspar (Table 4).

Table 4 The mineral distribution found in sandstones in the different successions.

Minerals	Full succession	Partly weathered	Middle Ecca
Quartz	■	■	■
Kaolinite	■	■	■
Siderite	■	■	■
Muscovite	■	■	■
Rutile	■	■	■
Pyrite	■	■	■
Hematite		■	
Ilmenite			■
Feldspar			■

On average, the samples with an acidifying potential and those with a base potential, contain similar concentrations of SiO_2. The major difference is found between the CaO and MgO of the samples, with a base potential to be 50% less in the samples which acidify.
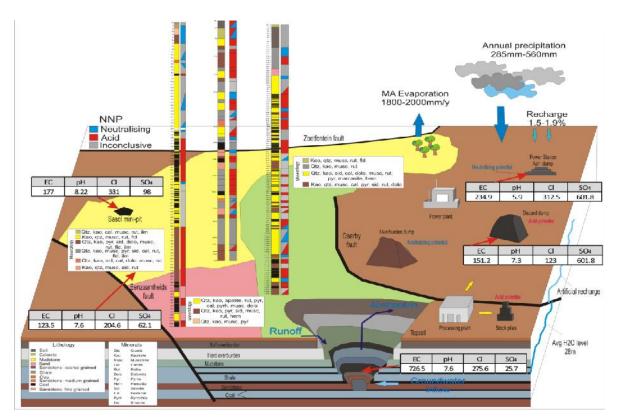

Figure 3 Summary of the acid and neutralising potential of the various contributing factors over the extent of the study area (Macdonald, 2014).
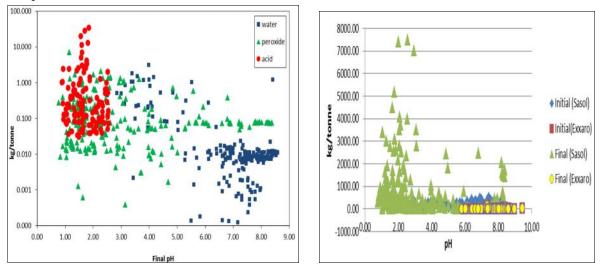
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Metal release

Elements such as sodium, potassium and nickel (Figure 4) are present in quantities below their solubility limits. This is denoted by a decrease in concentration with a drop in the pH (Hodgson *et al.*, 1999). Calcium/ magnesium carbonate exist as a form of a buffer which will cause the pH to increase, and will again decrease when the buffering species are depleted.

Iron and sulphate (Figure 5) exhibit an associated rise indicating that pyrite is the most common sulphide mineral present. Calcium and magnesium concentrations increase until they are depleted from the rock and coal through reaction with the acid. Sodium ranges from low to intermediate levels, and the higher sodium levels often coincide with high pH-levels, suggesting the presence of sodium carbonate species.

Figure 4 shows that the sulphate values will increase as the pH decreases in the case of Sasol samples; Exxaro samples show very low concentrations of sulphate even when the pH drops. This might be because of the dominance of the carbonaceous material that is found in the Exxaro samples.
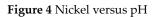

Figure 4 Nickel versus pH   Figure 5 Sulphate versus pH

Kinetic test results

The thirteen samples are from different areas. Eight of the samples provided a closed NNP (-20 to 20 kg/tonne $CaCO_3$) which indicates inconclusive results and the other five had a higher closed NNP which classifies the samples as having the potential to turn acidic or basic. The Sasol samples (S6 to S38) were collected from the interburden and overburden (shale/mudstone) and interburden (sandstone). The Grootegeluk samples (G10 to G18) were taken from the partly weathered overburden which consists of carbonaceous mudstone and the Sekoko samples (SEK1 to SEK4) were taken from the Middle Ecca interburden. Figure 6 shows the pH of the 13 samples that were analysed. Throughout the leaching test the pH values of the samples remained high; an exception being S14 and S15 which showed a drop in pH over 20 weeks and which remained acidic.

Figure 7 shows the release of sulphate from the interburden samples; the analysis shows that the production rate for all the samples was lower at the beginning of the analysis. The highest recorded sulphate concentration at the end of week 20 is in S14 and S15, with an approximate production rate between 250 mg/kg/wk and 50 mg/kg/wk. The majority of the samples showed an increase in

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the first three weeks, then decreased again thereafter due to the removal of already oxidised products. The increase in sulphate coincided with the decrease in pH. The samples which gave inconclusive closed NNP all produced rates that were below 150 mg/kg/wk in the first three weeks, and the production rate continued to decrease, which is indicative of the oxidised products that continued to be removed.
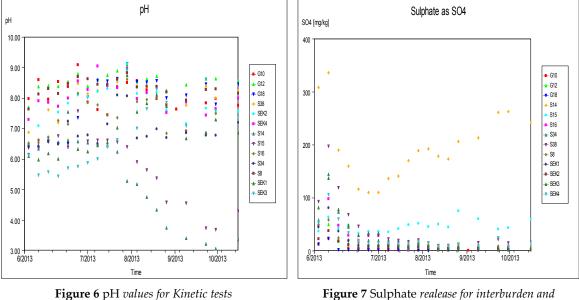

Figure 6 pH *values for Kinetic tests*

Figure 7 Sulphate *realease for interburden and overburden*

CONCLUSION

The ABA results showed that the coal seam has the highest AP, whilst overburden and shale have a NP. Leaching of heavy metals is evident when the pH decreases. This provides an idea of the post mining environment. Sample analyses was done on the interburden, overburden as well as the coal samples. The results indicated that over 35% to 50% of the samples in the different lithologies (interburden, overburden) have an excess AP. This classifies the samples as having a higher risk for acid generation. About 30% to 40% of the samples have a NP. The remainder of the samples have a medium risk for acid generation.

The ABA results determined that the interburden and discards used as backfill material has a potential to generate acid. This includes the composite samples with various densities from the processing plant. Overburden material has a NP. The ABA illustrated that the shale and mudstone samples have both acid and neutralising potentials and the majority of sandstone samples beneath 60 m have a distinct AP.

The composite material can be used as backfill material within the pit. It only becomes a risk upon exposure to oxygen and water. The Waterberg area has a relatively low annual rainfall, high evaporation and deep groundwater levels, thus the risk to produce acid from infiltrating water is limited. The risk of exposure to oxygen leads to the spontaneous combustion of discard material.

Kinetic test analysis were carried out on the overburden and interburden samples. The pH results showed variations throughout the analysis with all the samples having pH values above 5.5 for the first nine weeks of analysis. The majority of the samples exhibit a high pH throughout the course of

the analysis; a difference is only seen in two samples from the interburden which showed a drop below pH 4. According to ABA results the seven weathered interburden samples from the Middle Ecca have a higher risk of generating acid with the highest at pH of 2.97. An increase in sulphate production is directly related to an increase in EC. Six samples in the kinetic test results were classified as medium to lower risk of acid generation according to results with the lowest pH of 4.07. From the kinetic results, the samples also showed a final pH >7. After week 14 the sulphate production was below 50 mg/kg and the calcium production below 5 mg/kg, The EC values also remained low at constant values.

The pH variations of sandstone and mudstone within the overburden and interburden allowed the materials to be combined at an optimal ratio to ensure a neutral pH within the pit. The initial pH results before testing indicated no oxidation before ABA analysis were done. The interburden material with pH below 4 had correspondingly high sulphate loads (20000-350000 mg/kg). The overburden materials had a lower sulphate load between 5000-35000 mg/kg. The analyses indicated that the interburden material is more likely to generate acid.

The mining industry is always seeking new, cost-effective methods of improving operational efficiency within the mining infrastructure. There have been considerable improvements made to the various aspects of backfilling systems, including the availability and supply of fill materials, different types of backfill systems, backfill preparation procedures for transportation and placement, and the use of alternative cementing agents used in underground mining.

The preferred handling method for the waste generated at the coal mines is the *high and dry* method. Waste is placed layered in between non-reactive material (acidic potential and spontaneous combustion). At the bottom of the mined pit non-reactive interburden is placed in compartments and sealed on the sides and along the top of the backfilled heap. The individual heaps should not exceed 22° and not stacked higher than 17 m. The interburden must be the initial layer, stacked above the natural groundwater level. The stacking will not be affected as on-going mining operations will result in lowering the groundwater levels. The high and dry method places the acid prone material above the natural groundwater level, ensuring minimal to no contamination to groundwater bodies. The individual heaps are stacked to the specified heights and angles and then continue towards the mining advance. Overburden is used as sealing material, due to the shale and clay's low propensity of spontaneously combust or generate acid.

One suggestion to the handling would be to interlayer ash from Matimba Power Station close by. The ash is available, and the research showed it has a high neutralizing potential in ratios of 2:1 and 5:1, and it would help to reduce the volume of the final mine void. Ash reduces the iron and sulphate from acidic waters and continues to increase the pH by precipitating the problematic elements from the solution. It is however, a great challenge to transport ash in the study area due to the limited amount of water and the availability of ash material. The fine particles should be kept wet during transport. Cost of transport can be reduced when the same conveyor system that is already in place, is used to take ash back to the mine.

NOMENCLATURE

ABA   Acid base accounting

NP   Neutral potential

AP   Acid potential

Ovb   Overburden

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