

PRELIMINARY INVESTIGATION ON ACID GENERATING POTENTIAL OF COALS FROM BENUE TROUGH, NIGERIA

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Abstract

Coals and other lithological units in the coal bearing successions of some selected areas of Benue Trough namely: the lower (LBC), middle (MBC) and upper (UBC) had been investigated. Major elements (lithium-metaborate/tetraborate fusion), inductively coupled plasma optical emission mass spectrometry (ICP-OES), combustion infra-red (LECO) and modified acid-base accounting were employed to investigate the potential of the coal and coal-bearing units to produce acid mine drainage conditions.

The results indicate that SiO₂ concentrations in the coal vary between 51.13 and 65.29 wt.%, Al₂O₃ between 16.94 and 31.31 wt.%, Fe₂O₃ between 1.75 and 11.15 wt.%, and S between 0.34 and 1.39 wt.%. Minor concentrations of CaO (0.09 to 3.17 wt %) and MgO (0.20 to 3.52 wt. %) are also present. P₂O₅ occurs in concentrations of 0.04 to 0.34 wt. % and K₂O is in the order of 0.45 to 1.85 wt. %. Na₂O varies between 0.05 and 1.74 wt. %. The average Net Neutralising Potential (NNP), low proportion of base neutralizing cations (CNK) and the high CIA values ranging between 64.9 to 97.5%, as well as high reactive sulphur content of greater than 3.4kg/tonne suggest that LBC and MBC coals are capable of generating acid.

The information obtained from this study could be used in predicting the types of situations that might arise concerning groundwater quality, and implement proper prevention or remediation techniques.

Keywords: Acid mine drainage; coal ash; weathering; sulphur; ; Benue trough

1. Introduction

The coal reserves of Nigeria are estimated at 2.75 billion tonnes, with occurrences spreading over 13 states of the country [1]. Reported occurrences of coal include the lower (LBC), middle (MBC) and upper (UBC) Benue Trough [2-6]. Another potential area for coal occurrence is in the Chad basin, which is stratigraphically and structurally related to the Benue and Anambra basins [7]. Smaller occurrences include Garin Maigangu (Bauchi), Afikpo area (Abia), Koton – Karfi (Kogi), Ute (Ondo) and many more [8]. Currently, exploration and exploitation of coal particularly at Okpara and Onyeama underground mines (in LBC), Okaba surface mine and Orukpa underground mine (in MBC) have been intensified with a bid to diversify the economy which is almost solely dependent on oil and gas [9-10]. However, coal mining and exploration may result in environmental degradation. To estimate some of this degradation, we measured the potential for acid mine drainage (AMD) which can be formed when sulphide minerals, notably pyrite, are oxidized on exposure to air and water in mines or spoil or piles. AMD does not occur if the sulphide minerals are non reactive or if the rock contains sufficient alkaline material to neutralize the resulting acidity. The environmental implications of AMD include an increase in heavy metals (for example arsenic) and sulphate in water, which in turn affects the quality of the aquatic life and other sectors of the ecosystem. Control of AMD can be achieved by preventing or inhibiting acid generation, by control of acid migration, or by treatment of AMD; as the case may be. There are many requests for information on the proximate and ultimate analyses, sulphur form data (AMD), trace element concentrations, modes of occurrence, mineralogy, washability, petrography, and so on from domestic and foreign coal companies,

manufacturers of coal related products and scientific researchers [6]. One of the easiest and less expensive ways to analyze for acid-generating potential of rocks is described by several authors [11-15].

The Nigerian economy is dependent almost solely on oil and gas, with about 90% of her foreign exchange earning coming from this sector. In order to diversify the economy, the Federal Government of Nigeria has established a Solid Mineral Development Ministry to look into other mineral resources. On the priority list of the solid minerals that are tagged for exploration and exploitation for export and domestic consumption, are the regionally extensive coal measures. It is therefore important to investigate, the environmental implications that may arise from the exploitation of coal. The present study is aimed at providing preliminary investigation on some coals and the associated strata in some coalfields of Benue trough and to predict their potential for producing acidic or alkaline mine waters during mining operations.

2. Geological setting

The main coal deposits of Nigeria occur in the Benue trough which is defined as an intracontinental Cretaceous basin about 1,000km in length, stretching in a NE-SW direction and resting unconformably on the Pre-Cambrian basement. It is subdivided into three main domains namely: lower (LBC), middle (MBC) and upper (UBC) Benue trough, corresponding to geological and geomorphologic partition [6,16-24]. The Benue trough has been described as a rift bounded tension structure produced as the south America and Africa drifted apart (triple rift system known as an aulacogen) and have been documented by some workers [18,22,23].

The Benue Trough was subjected to four main depositional cycles, each of which was associated with transgression and regression of the sea [25]. The first sedimentary cycle lasted from the middle Albian to upper Albian and is thought to have been initiated by the opening of the South Atlantic Ocean. It is associated with the deposition of the Asu River Group in the LBC, which is a lateral equivalent of the Bima Sandstones in the UBC, and Awe/Arufu/Uomba Formations in the MBC. The second sedimentary phase occurred between the upper Cenomanian and middle Turonian and it was associated with the deposition of Eze-Aku Shales in LBC. Its lateral equivalents are the Makurdi Sandstones in the MBC as well as the Gongila, Jessu and Dukul Formations in the UBC. The third sedimentary cycle ranged from the upper Turonian to the lower Santonian. It is associated with the deposition of Awgu Shale and Agbani Sandstones in the LBC and MBC, which are lateral equivalents of Fika/Sekunle Shale in the UBC. The Turonian transgression, which marked the start of this cycle, is believed to have commenced from the Gulf of Guinea through the Anambra basin to the Benue Trough [24]. Most of the deposits of this cycle have been eroded as a result of the upper Cretaceous tectonic activity [23-24]. The fourth sedimentary cycle was marked by deposition of the Nkporo Shales, Owelli Sandstones, Afikpo Sandstones and Enugu Shales during the Campanian-Maastrichtian transgressive phase in the LBC. This cycle also marked the deposition of the coal measures including: the Mamu Formation, Ajali Sandstones and Nsukka Formation in the LBC. Its lateral equivalents are the Lafia Formation in the MBC, and Numanha Shale, Gombe and Lamja Sandstones in the UBC.

3. Sampling and analytical techniques

Two coal samples were collected each from the UBC, MBC and LBC (Fig. 1). The UBC coal samples as well as associated underlying and overlying strata were collected from an outcrop along the Gombe-Yola road and Chikila town belonging to Lamja Sandstones which consists dominantly of sandstone with shale and coal interbeds (Fig 2). The MBC coal samples were collected from boreholes: BH 90 and BH 94 at depth of 160 and 350m respectively belonging to the Awgu Shale. It consists of dominantly dark grey shale with abundant plant remains usually alternating with limestone, sandstone and coal seam interbeds (Fig.3). The LBC coal samples were retrieved from Akwuka (LBC 1) and Okpara (LBC 2) from the coal seams of the Mamu Formation being exploited in coal mines near Enugu. It consists of calcareous dark grey shale, sandstone and coal seam interbeds (Fig. 4). The lithologic profiles of the study samples reflect different rock units which may have impact on the acid generating potential.

A total of six (6) pulverized coal rock samples, each weighing 50 grams, were stored in sealed plastic bags. The samples were then analyzed at the Activation Laboratories, Canada for ashing at 150°C for 50 hours, major elements (lithium-metaborate/tetraborate fusion), inductively coupled plasma optical emission mass spectrometry (ICP-OES), combustion infra-red (LECO) and modified acid-base accounting.

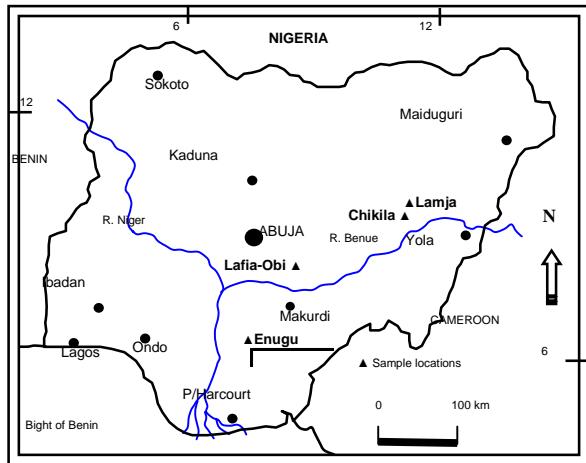


Fig. 1 Map of Nigeria showing the LBC, MBC and UBC sample locations

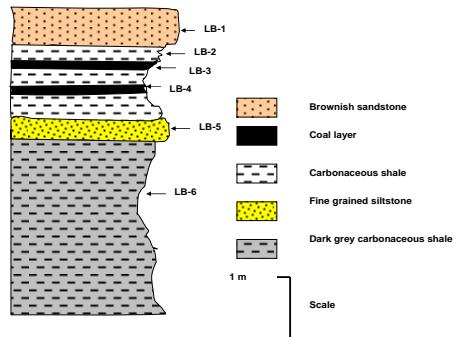


Fig. 2: Lithologic section of LBC showing sampling intervals of the Mamu Formation

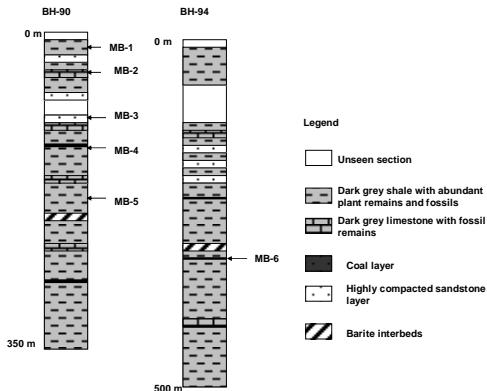


Fig. 3: Lithologic section of MBC showing sampling intervals of the Awgu Shale

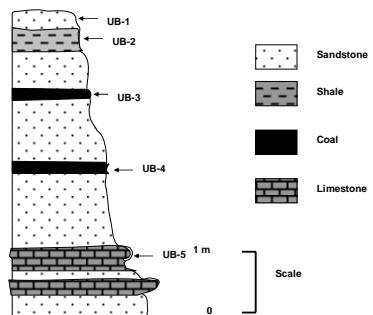


Fig. 4: Lithologic section of UBC showing sampling intervals of the Lamja Sandstone

4. Results and interpretation

4.1 Major and trace element concentrations

The result of the major elements in coal and non coal strata is presented in table 1. The SiO_2 values range from 9.91 to 76.12% while Al_2O_3 vary between 0.68 and 18.94%. The CaO and Fe_2O_3 values are between 0.24 to 51.96% and 1.36 to 11.15% respectively (table 1). SiO_2 , Al_2O_3 , CaO and Fe_2O_3 account for between 65-85% of the samples. The ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$ is low (between 2.69 and 11.23) in all the samples indicating a lower silt content and therefore a tendency towards marine condition, while the high content of CaO (>4%) in the underlying and overlying strata samples indicate a calcareous shale [26-27]. The higher Fe_2O_3 (7.57% in average) in the coal samples analyzed confirm the presence of iron bearing minerals such as pyrite. This also suggests that the shale units were deposited under a reducing environment. The higher Fe_2O_3 and MgO indicate more chlorite content [28]. The low percentage of K_2O (0.31 and 4.73%) indicates that montmorillonite and illite are present in relatively small proportion [29]. Also, it was observed that the oxides of Ca, Mg and Na are low in some of the strata in LBC and MBC and higher in some strata in the UBC samples (table 1). The oxides of Fe and Al are found to occur in higher concentrations in the LBC and MBC compared with that of the UBC. This observation corroborates with Krauskopf's theory [30] that as weathered material loses Ca, Na, Mg and K, there will be an apparent gain in Fe and Al. Also, the higher concentrations of acidic oxides of Si, Al and Fe as well as the comparatively

lower concentrations of basic oxides of Mg, Ca, Na and K imply a lower level of base neutralizing cations, for all the samples.

Table 1: Major element concentrations in the coals and non-coal strata from Benue trough, Nigeria

Benue Trough	Sample No	Lithology (*)	SiO ₂ %	Al ₂ O ₃ %	Fe ₂ O ₃ %	MnO %	MgO %	CaO %	Na ₂ O %	K ₂ O %	TiO ₂ %	P ₂ O ₅ %	LOI %	TOTAL %
LBC	LB-1	SS	76.12	6.78	1.93	0.08	0.03	1.94	0.47	2.67	0.32	0.06	9.60	100.00
	LB-2	CS	62.74	5.82	3.12	2.85	1.86	11.92	2.10	4.73	0.74	0.44	3.98	100.00
	LB-3	Coal	42.03	31.31	1.75	0.04	0.20	0.09	0.05	0.45	1.34	0.08	22.58	100.00
	LB-4	Coal	44.96	20.24	6.39	0.03	1.01	1.39	1.14	1.85	1.96	0.34	20.35	100.00
	LB-5	FGS	65.29	8.16	2.95	0.04	0.64	0.85	0.06	1.39	1.63	0.57	18.42	100.00
	LB-6	DGC	48.43	9.57	6.09	1.36	1.31	15.42	0.87	2.25	0.49	0.43	13.76	100.00
MBC	MB-1	DGS	63.13	8.39	3.53	2.30	1.35	4.53	2.14	4.58	0.13	0.15	9.62	100.00
	MB-2	DGL	25.67	2.89	1.78	0.78	1.16	42.18	0.19	0.31	0.24	0.19	24.42	100.00
	MB-3	HCS	64.35	10.30	6.01	0.20	0.50	0.53	1.35	0.45	1.48	0.03	9.80	100.00
	MB-4	Coal	41.13	18.94	11.15	0.05	1.67	0.95	0.64	1.22	0.78	0.04	23.39	100.00
	MB-5	DGS	57.18	7.29	2.87	0.63	1.25	6.35	1.42	3.85	0.57	0.02	18.55	100.00
	MB-6	Coal	56.60	16.94	9.48	0.03	1.34	0.35	1.40	1.09	0.90	0.13	21.24	100.00
UBC	UB-1	SS	65.61	12.37	3.19	0.02	0.53	0.24	0.04	2.18	0.94	0.07	14.81	100.00
	UB-2	Shale	57.31	7.58	6.28	1.32	2.35	7.83	0.54	1.10	0.31	0.41	14.97	100.00
	UB-3	Coal	48.17	18.17	10.34	0.03	1.75	0.53	0.31	1.37	0.96	0.04	18.45	100.00
	UB-4	Coal	47.18	17.68	4.60	0.07	3.52	3.17	1.74	1.02	0.97	0.04	20.04	100.00
	UB-5	LS	9.91	0.68	1.36	0.25	0.52	51.96	0.05	0.15	0.04	0.03	34.88	100.00

(*) Lithologic description: SS=Sandstone, CS=Carbonaceous shale, FGS=fine grained siltstone, DGC=dark grey carbonaceous shale, DGS=dark grey shale, DGL=dark grey limestone, HCS=highly compacted sandstone, and LS=limestone

4.2 A –CNK-FM Ternary Plot

In order to obtain a multivariate and visual analysis of the relationship between mineralogy, major element geochemistry and acid producing potential of coal ash, an A- CNK- FM MINPET ternary plot has been produced (Fig 5). The ternary plot indicates the normalized molar proportions of Al₂O₃, CaO + Na₂O + K₂O and Fe₂O₃ + MgO. The normalized molar proportions are obtained following the method of Nesbitt et al. [31]. Ternary plots have been used by various researchers [32-33] to show the correlation between coal ash compositions grouped into basic, fluxing acidic and non-fluxing acidic oxides, with ash fusion temperatures.

Examining the A-CNK –FM ternary plot of normalized molar proportions (Fig. 5) of the six composite coal ash samples and their non-coal strata, all the samples have low proportion of base neutralizing cations (CNK) and this is most pronounced in the LBC samples. The CNK proportions range between 2 and 19% and seems to be much lower than 30 to 56% reported on coals from New Zealand mines [34]. The reactive sulphur values also range between 3.4 and 12.9kg/tonne (Table 2) which is also very high than 0.5kg/tonne estimated for the New Zealand coals.

4.3 Chemical Index of Alteration (CIA)

CIA values indicate the degree of weathering of the source rock calculated (using molar proportions) for each sample using the following equation [31].

$$\text{CIA} = (\text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3 + \text{CaO} + \text{Na}_2\text{O} + \text{K}_2\text{O} + \text{MgO})) \times 100 .$$

Assuming that all the sulphur calculated for the rocks was reactive; a plot of total reactive sulphur versus CIA can be made (Fig. 6). Acid producing coal ash will have CIA value greater than 20% (hence low base proportion in ash) and high reactive sulphur content [34]. With this hypothesis, the LBC, MBC and UBC samples infer apparent acid potentials; with CIA values between 65% and 98% that are much greater than CIA lower limit value of New Zealand coals.

As earlier mentioned, CIA values indicate the degree of weathering of source rocks. Krauskopf [30] stated that as a weathered material loses Ca, Na, Mg and K, it will appear to gain Al and Fe. Hence, the CIA which is a ratio of these will increase, proportionally with the degree of weathering. From the CIA plots, all the samples have undergone various degrees of weathering (Fig. 6). Chemical weathering includes oxidation, one of the processes which may aid the production of acid from sulphide minerals. The

sample from the UBC (UB-4) appears to have undergone the lowest level of weathering compared with the other samples.

4.4 Acid-Base Accounting

Acid-base accounting allows determination of proportions of acid generating and neutralizing minerals present in the samples. Table 2 shows the results of the modified acid-base accounting tests. Parameters determined include paste pH, reactive sulphur (both in % and kg/tonne), neutralization potential (NP or NPT), maximum acid potential (AP or MPA), net neutralization potential (NNP) and net acid producing potential (NAPP). If the pH is less than 5, then acid generation would probably occurred in the sample (Campbell et al., 2001). From Table 2, paste pH of LB-3, MB-4, and UB-3 are respectively 3.18, 3.83 and 6.44. This shows that the coal samples from the LBC and MBC are capable of generating acid, while the coal sample from the UBC indicates lower acid generating capability. Table 2 indicates a total sulphur content range of between 0.07% (UBC2) and 2.0% (MBC2) for the coal samples collected from the Benue Trough. This reveals a smaller range of values compared to the 0.17% - 4.28% total sulphur content range established for the coals collected from New Zealand [34].

Table 2: Modified acid base accounting results for the selected coal samples from Benue Trough Nigeria

Sample No	Fizz pH rating	Reactive sulphur, % S	Reactive sulphur Kg/t	NPT	MPA Kg CaCO ₃ /t	NNPA	NCI NaOH Vol NaOH	0.11 N 0.50N Vol HCl	NAPP=(MPA-NPT)
NBM-1	Slight			43.15	<0.31	43.15	2.95	30.00	
LB-3	Slight 3.18	0.40	4.0	-1.04	<0.31	-1.04	1.15	5.00	1.35
MB-4	Slight 3.83	1.29	12.9	-4.00	<0.31	-4.00	1.60	6.00	4.31
UB-3	Slight 6.44	0.34	3.4	2.09	<0.31	2.09	0.90	5.00	-1.78

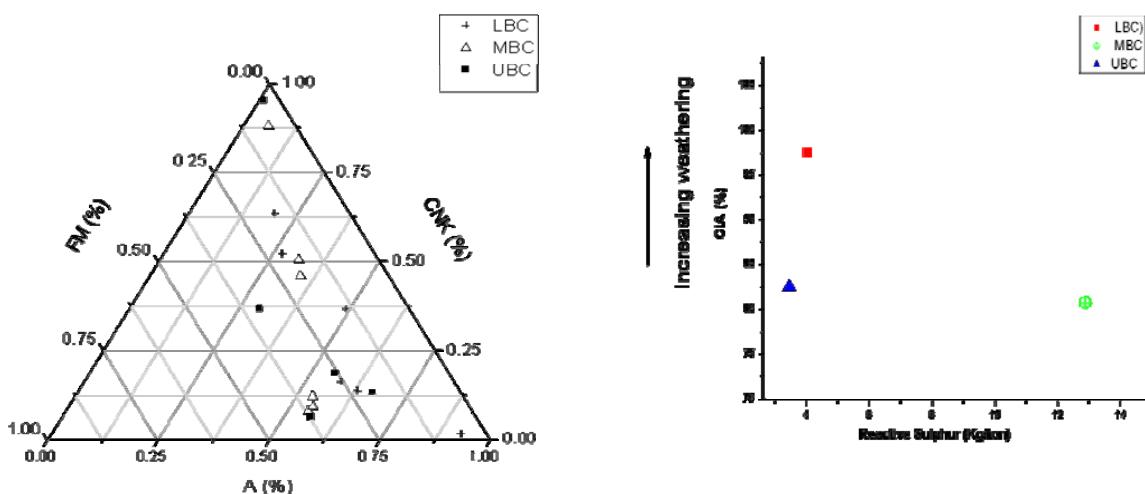


Fig. 5: A-CNK-FM Plot for the coal and the non-coal samples

Fig. 6: Plot of CIA against reactive sulphur

Recent research indicates that since mineral groups containing base cations have different relative reactivities at pH 5 (i.e carbonates have a relative reactivity of 1 whereas pyroxenes, amphiboles and sorosilicates have a relative reactivity of 0.02 and feldspars a relative reactivity of 0.01), then the control that the mineralogy has on "freeing" cations for neutralization is very significant [35]. The reduced neutralizing potential (NPT) is better predicted by the conservative 1:2 line [36] and this line was adopted as the neutral line for this study.

Figure 7 shows a widely distributed sample population, which indicates that the samples from the LBC and MBC (LB-3 and MB-4) are potentially acid producers, while the upper Benue Trough sample

(UB-3) is either non-acid producing or neutral. None of the samples lie within the zone of uncertainty. Other variables such as pyrite grain size, permeability and access of oxygen and moisture will control the rate of acid generation [14,15,34].

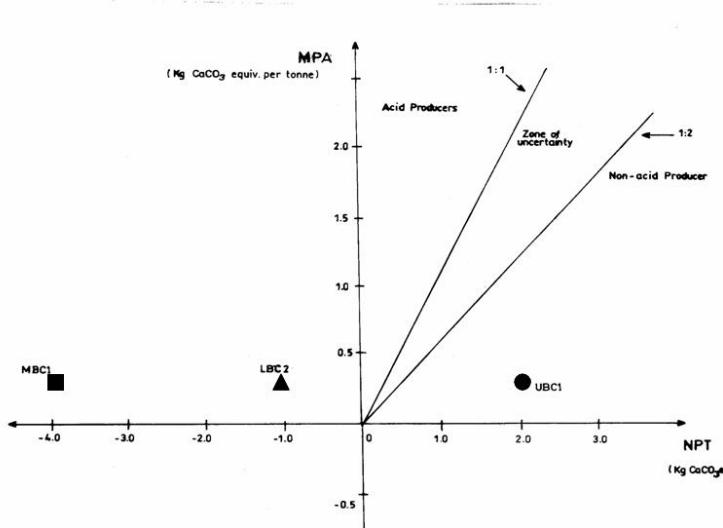


Fig. 7: Plot of MPA against NPT

The samples tend to decrease in their net acid potential as the net neutralization potential of the samples increase. The negative relationship indicates that two samples LB-3 and MB-4 are net acid producers while sample UB-3 from the UBC can be interpreted as a net alkaline producer.

5. Conclusion and recommendation

Currently, exploration and exploitation of coal particularly in the Benue Trough of Nigeria have been intensified to diversify the economy, which has been almost solely dependent on oil and gas. Attention should also be focused on the environmental aspect of coal exploitation too in order to prevent the harmful of acid mine drainage. The coal and non-coal samples from LBC, MBC and UBC have low values of neutralizing cations (CNK) and the reactive sulphur content of the coal layers indicates capability to generate acid. In fact, some of the non-coal strata could act as alkaline rocks. However, other variables such as pyrite grain size, permeability and access of oxygen and moisture which control the rate of acid generation cannot be ruled out in the study area. It is therefore recommended that, further studies and more detailed analyses be carried out on acid generation and control within the study area most especially on the underground water. The ground water levels within the working mines should be closely monitored to estimate the potential for acid generation.

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