

ESTIMATION AND ASSESMENT OF FREE SWELLING INDEX AND SOME PETROGRAPHIC PROPERTIES FROM CHEMICAL ANALYSIS OF COALS ACROSS RIVER NIGER

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Abstract

Free-swelling index (FSI) and dilation are useful indicators to predict the strength of coke that may be derived from a particular coal. FSI is a measure of a coal ability to swell and cake during heating. Both coal from Ovbiowun and Oloku are considered to have poor coking and swelling tendencies as inferred from the derived FSI values (2.04 and 1.48 respectively) as well as > 36% in volatiles. GCV/HHV is determined from both proximate and ultimate analyses values as input parameters using a generally proven formula. GCV of Ovbiowun coal is 28.7MJ/Kg which is within range of 26.7 – 30.2 MJ/Kg for "high volatile C bituminous". Oloku coal has GCV of 25.3 MJ/Kg which is within the range of 24.4 – 26.7 MJ/Kg for "Sub bituminous A grade". For Ovbiowun coals, the Ibs of CO₂/MMBtu (gross) = 167.44 Ibs/Btu, 4596 Ib/tonne and for Oloku coals, Ibs of CO₂/MMBtu (gross) = 164.21 Ibs/Btu, 4376 Ib/tonne. This strongly agrees with Oloku coals as sub-bituminous and Ovbiowun coals as bituminous, having higher carbon content and thus more CO₂ production. The fuel ratio for both Ovbiowun and Oloku is 0.98 and 0.91 respectively which is a referral to bituminous grade. The vitrinite reflectance as determined for the studied samples was 0.48 for Ovbiowun coals and 0.43 for Oloku coals implying bituminous and sub-bituminous coals respectively. No coal bedded methane is produced from both coals. The Roga index as well as their dilation is inferred to be around 0 -20. Ovbiowun coals have 65 HGI and Oloku coal is 87 HGI due to higher moisture content. The moisture content of Oloku coal (15.0) is higher than Ovbiowun (10.8) while the volatile matter (as received) for Oloku (40.7) is lower than Ovbiowun (41.7). From the > 10% moisture and ash content, coal in both study area is unsuitable for coke production. Also the derived Hydrogen % for Ovbiowun (63.3%) is greater than Oloku (57.43) implying more maturity in Ovbiowun coals.

Keywords: Coal; Gross Calorific Value; FSI; HGI; Vitrinite reflectance; CO₂; Proximate and Ultimate analysis; Fuel ratio.

1. Introduction

1.1. Proximate analysis

The proximate analysis of coal is a simple means of determining the distribution of products obtained when the coal sample is heated under specified conditions. As defined by ASTM proximate analysis separates the products into four groups: moisture, volatile matter consisting of gases and vapours driven off during pyrolysis, fixed carbon, (the non-volatile fraction of coal) and ash, (inorganic residue after combustion). Proximate analyses (moisture, volatile matter, fixed carbon by difference and ash) are conducted at atmospheric pressure.

1.1.1. Moisture

The most elusive constituent of coal to be measured in the laboratory is moisture. The moisture in coal ranges from 2–15% in bituminous coal and may be up to 50% in lignite. Varying amounts of water are still present at different stages of the coalification process. The overall result of coalification was to eliminate much of the water particularly in the later stages,

as is evident from a comparison of the moisture contents of different ranks of coal from lignite to anthracite. The moisture in coal may be divided into four types: inherent moisture, surface moisture, decomposition moisture, and water of hydration of mineral matter. Inherent moisture is equally termed bed moisture or equilibrium moisture. Surface moisture has a vapour pressure equal to that of free water at the same temperature. Decomposition moisture is generated from the thermal decomposition of organic constituents of coal. The water of hydration of mineral matter is incorporated into the crystal lattices of the inorganic and claylike materials found in coal. Pore sized has been found to decrease as rank decreases (due to compression and increased heat gradient) hence low-rank coals are more susceptible to more moisture content. Excessive moisture can be of disadvantage in pulverizing and handling operations. A wet coal is very difficult, and in some instances almost impossible to pulverize. Total moisture is important in assessing and controlling the commercial processing of coals. Coals with high moisture content require more heat for vaporization of the moisture.

1.1.2. Ash

Coal ash is the residue remaining after the combustion of coal under specified conditions. It is a by-product of chemical changes that take place in the mineral matter during the ashing/burning process. The quantity of ash can be more than, equal to, or less than the quantity of mineral matter in coal, depending on the nature of the mineral matter and the chemical changes that take place in ashing. There are two types of ash forming materials in coal, extraneous mineral matter and inherent mineral matter. The extraneous mineral matter consists of materials such as calcium, magnesium, and ferrous carbonates, pyrite, marcasite, clays, shale, sand, and gypsum. Inherent mineral matter represents the inorganic elements combined with organic components of coal. The origin of such materials is probably the plant materials from which the coal was formed. The ash value is to evaluating sampling procedures and its value commonly specified in coal contracts. In combustion, high ash content is an indicator of reduced Gross calorific value (GCV) obtainable from a given quantity of coal. The Ash Content is determined in the suite of proximate analyses as representing the residue after combustion, and used to infer the total mineral composition of the coal. Ash represents impurities that cannot burn and usually present in coal in the range is 5 to 40%. In the case of some flow gasifier, higher ash content coals will result in a decrease in gasifier efficiency. This is due to an increase in oxygen demand and loss of heat in the slag that cannot be fully recovered, as well as a possible reduction in throughput due to the additional slag that needs to be removed via the slag tap.

1.1.3. Volatile Matter

This is the loss of mass (provided it is corrected for moisture) which results when coal is heated in specified equipment under prescribed conditions. The matter lost is composed of materials that form upon the thermal decomposition of the various components of coal. Some of the constituents of coal volatile matter are hydrogen, carbon monoxide, methane and other hydrocarbons, tar vapours, ammonia, some organic sulphur- and oxygen-containing compounds, and some incombustible gases, such as carbon dioxide and water vapour. All of which come from the decomposition of organic materials in coal. Inorganic materials in coal contribute the water of hydration of mineral matter, carbon dioxide from carbonates, and hydrogen chloride from inorganic chlorides to the volatile matter. Volatile matter does not include the residual moisture. Pressure, however causes volatiles to stay in a particle for a longer time, which has implications when extrapolating proximate analysis data for gasification performance. Messenbock *et al.* [1] demonstrated that certain coal types are more reactive to pressure increases than other coals.

Chemical analysis of coal provides data gathered from "whole-coal" materials, embracing moisture and mineral matter as well as the organic constituents. The data from ultimate analysis (C, H, O, N, and S percentages) may be corrected to a moist, mineral matter-free (mmmf); dry, mineral matter-free (dmmf); or dry, ash-free (daf) basis to assess composition

of the organic matter alone, but even so the composition of the organic matter determined in this way inherently represents an aggregation of the composition of the different maceral components. Volatile matter can be used to infer the quantity of smoke to be emitted per tonnage from furnaces or other types of coal burning equipment. With a very high volatile matter, the more will be the flame length, and the faster the ignition of coal. According to Peng [2] volatile matter is a major factor in determining the suitability of coals for various applications. It can be used to prescribe the optimum limit and conditions as to furnace height and volume. It may also influence secondary air requirements. Excessive volatile matter content of a coal may cause the coal to be banned for use in gasification processes in order to control smoke emissions.

1.1.4. Fixed Carbon

The fixed carbon value is obtained by subtracting the sum of the percentages of moisture, ash, and volatile matter from 100. This value is considered to be the amount of carbon residue that remains after the volatile matter test. The residue is the product of the thermal decomposition of the coal. The fixed carbon value is one of the values used in determining the efficiency of coal burning equipment. It is a measure of the solid combustible material that remains after the volatile matter in coal has been removed. Fixed carbon values, corrected to a dry, mineral-matter-free basis are used as parameters in the ASTM coal classification system. Fixed carbon gives a rough estimate of heating value of coal.

1.2. Ultimate analysis

Carbon-to-hydrogen ratios are determined from the ultimate analysis. The total sulphur, Nitrogen, as well as oxygen by difference are included in this suite of analyses. During gasification, the total convertible sulphur exits with the raw gas, mostly as hydrogen sulphide, and is subsequently removed during gas cleaning. Sulphur form analysis (determination of pyritic, inorganic, and organic sulphur) is useful when beneficiation or abrasiveness is a concern.

Ultimate analysis of coal is defined as the determination of the carbon and hydrogen in the material, as found in the gaseous products of its complete combustion, the determination of sulphur, nitrogen, in the material as a whole, and the estimation of oxygen by difference. The carbon determination includes that present in the organic coal substance and any present as mineral carbonate. The hydrogen determination includes that in the organic materials in coal and in all water associated with the coal. All nitrogen determined is assumed to be part of the organic materials in coal. For practical reasons, sulphur is assumed to occur in three forms in coal: as organic sulphur compounds, as inorganic sulphides, which are mostly the iron sulphides pyrite and marcasite, and as inorganic sulphates. The total sulphur value is used for ultimate analysis.

The aim of this study is to determine and estimate some major petrographic properties and chemical analysis of the Ovbiowun and Oloku coals and their suitability for coking in furnaces as well as quality assessment in terms of calorific value, environmental pollution, ease of grinding and suitability for use in boilers

2. Materials, methodology and procedures

2.1. Material

The study area are Ovbiowun (near Afuze) Edo state and Oloku (near Ogboyoga) Kogi state East and West of R. Niger as shown in Figure 1

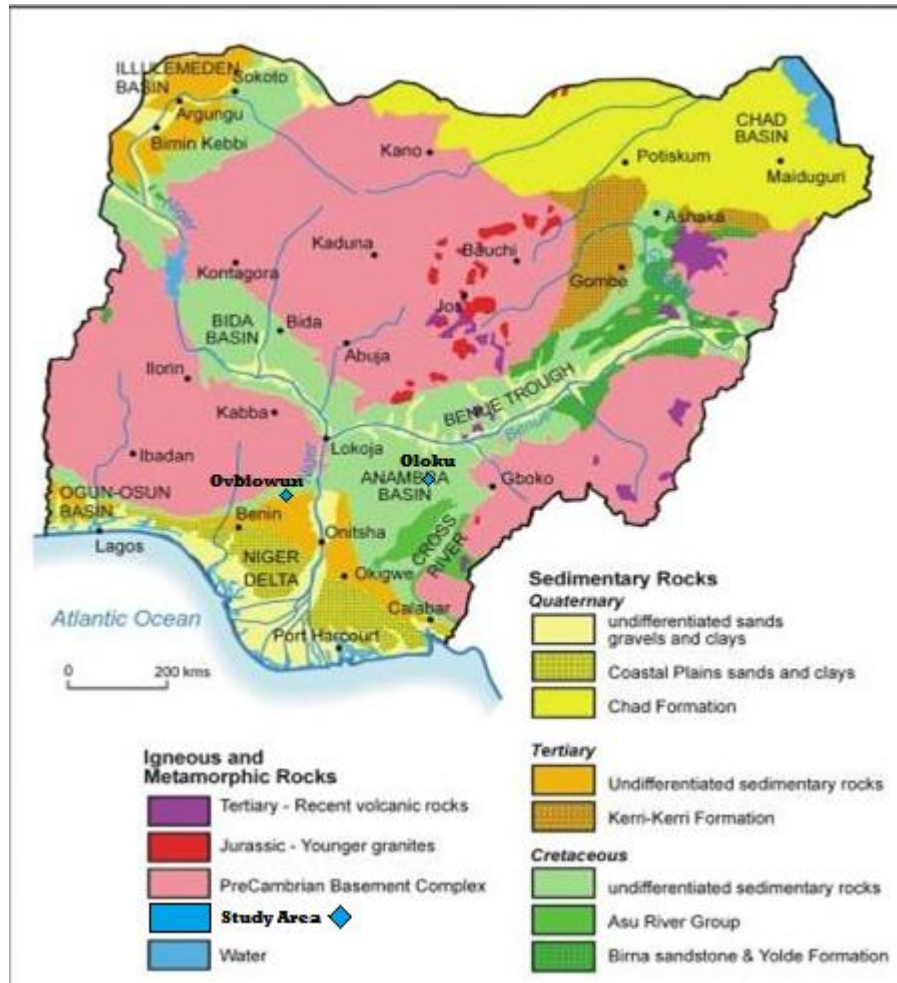


Figure 1: Map of Nigeria showing the study area in sedimentary terrain East and West of R. Niger.
 Source: <http://www.intechopen.com/source/html/48084/media/image1.jpeg>

The study area in Oviowun town in Owan East LGA of Edo State may be accessed through road from Afuze from Sabongida-Ora or Auchu. The co-ordinates of the study area will not be detailed due to the propriety nature of the data. Geomorphological features are undulating red sand hills of ferruginized Ajali sandstone, the false bedded sandstone and low lying plains of sometimes exposed Mamu formation. The drainage pattern is dendritic with individual streams, some of which are seasonal, merging to form rivers. The main river in the study area is River Owan, with several tributaries notably R. Okwuaeken along which coal exposures occurs. Thick fissile shale with thickness (4-15m) but usually more than 10m makes the coal almost un-prospective. However in most areas the coal is exposed to the surface. The annual rainfall ranges from 1500 to 2000 millimetres with prominent peak period in September. The temperature ranges from 27°C to 32°C and it has a relative humidity of about 75-95% annually. The vegetation is typically that of rain forest characterized by palms and shrubs, however, the original vegetation have been tampered with by human activities which include farming, palm wine tapping, palm oil making and trading. The people are typically Owan language speaking ethnic group of Edo State, particularly of the Emai Clan which extends from Obviowun to Afuze. They are engaged mainly in farming lumbering and trading, with typical crops grown including cashew, cocoa, plantain, oil palm, cassava etc.

The study area in Oloku town is the most northerly of the coal deposits on the northeast flank of the Anambra Coal Basin, an extension of the Ogboyaga coal district with thickness estimate of from 1.9 meters to 3.0 meters and averaged 1.68 meters thick. The Oloku coal is

near Okobo-Enejema which is just 4 km North of Ankpa located in eastern Kogi state. Oloku can be accessed from both Anyigba and Ankpa both intersecting at Inye-Ofugo junction, the major towns around the area. It can also be accessed from Abejukolo, the headquarters of Omala LGA passing through Ikaligwo. Because of the propriety nature of the data for now, the precise coordinates will not be specified. The people are Igala speaking language. They are engaged mainly in farming (typical crops grown include extensive cashew plantation, oil palm, cassava) lumbering, fishing and artisanal/crude coal digging. They are friendly and welcoming people. The geomorphologic features are flat undulating plains of Mamu formation, exposing the weathered light pink false bedded sandstones, an extension from of cretaceous sediments from Ovbiowun. There are occasional deep steeping valleys incised on the almost gentle plains by some immature rivers especially where the coal is outcropping. The annual rainfall ranges from 1000 to 1500 millimetres. The rainfall assumes peak of 1500mm mostly from late August to September. The temperature ranges from 25°C to 31°C and it has a relative humidity of about 70-85%. Shale thickness (usually less than 2m) is minimal and hardly exposed to the surface. Some of the coal is capped by fine sub-rounded to rounded grains of iron rich lateritic cover. The soil cover/decayed humus is equally thin in most area that the false bedded sandstone of the Mamu Formation is exposed on the surface and is farmed intensively. The soil cover/decayed humus is equally thin in most area that the false bedded sandstone over the Mamu Formation is exposed on the surface and is farmed intensively. The study area falls extensively between the thickest and most extensive part of the Ajali/Mamu cretaceous sediments of middle Benue trough of Anambra basin. It is considered to be very prolific with economical and viable coal resources.

2.2. Methodology

It is much easier to determine proximate analysis with a much more simple apparatus while the ultimate analysis is determined in a properly equipped laboratory by a highly skilled and precision conscious chemist. The calorific value and ultimate analysis of South African coals has been predicted from their proximate analyses with reasonable accuracy by Kock [3]. Same determination was done by Gill [4], who proposed that the proximate analysis may be used to deduce an acceptable approximation and correct values for ultimate analysis using the 'Gebhardt' Formula of several other formula. Several authors confirmed this with differently proved and accurate derivatives. According to Gill [4], given the proximate analysis in percentage as FC - % of Fixed Carbon; A - % of Ash; VM - % of Volatile matter; M - % of Moisture; S - % of Sulphur from the analysis.

Hydrogen H = $(7.35/(v + 10)) - 0.013$; where V = volatile matter

Nitrogen N = 0.07V for anthracite and semi-anthracite, N = $2.10 - 0.012V$ for bituminous coal and lignite; where V = volatile matter.

Total Carbon C = Fixed carbon (FC) + volatile carbon (V). The volatile carbon is calculated from volatile matter percentage as observed from the proximate analysis. The volatile carbon differs for different grades of coal and is as illustrated bellow

Total Carbon C = Fixed carbon + volatile carbon

= FC + 0.02V² for anthracite

= FC + 0.9(V - 10) for semi-anthracite

= FC + 0.9(V - 14) for bituminous coal.

= FC + 0.9(V - 18) for lignites

Sulphur increases the value of volatile carbon; hence the formula may not be accurate with very high sulphur content coals. Oxygen is determined by difference by subtracting the sum of values of H, C, N (derived for the ultimate analysis), ash, sulphur and moisture (from proximate analysis) from 100.

Oxygen O = 100 - (H + C + N + S + A + M). However for the sake of this study, the 'Gebhardt' formula for determination of elements (Hydrogen, Total Carbon, and Nitrogen) will not be used (best suited for bituminous coals and higher grades). A derivative which is more suitable for sub-bituminous and bituminous coal is used. The table 1 illustrates the interrelationships

between the proximate and ultimate analysis according to a proven formula used on Indian, Indonesian and South African Coal and as used by Elizabeth *et al.* [5] 2015 on Nigerian coals.

Table 1. Relationship between Ultimate Analysis and Proximate Analysis

%Carbon	$0.97\mathbf{FC} + 0.7(\mathbf{VM} - 0.1\mathbf{A}) - \mathbf{M}(0.6-0.01\mathbf{M})$
%Hydrogen	$0.036\mathbf{FC} + 0.086(\mathbf{VM} - 0.1\mathbf{A}) - 0.0035\mathbf{M}^2(1-0.02\mathbf{M})$
%Nitrogen	$2.10 - 0.020\mathbf{VM}$
% Oxygen	$100 - \%C - \%H - \%N - \%S - A\% - M\%$

c-% of Fixed Carbon; *A* - % of ash; *VM* - % of volatile matter; *M*- % of moisture; *S*- % of sulphur from the analysis

The elemental derivative of ultimate analysis equation in Table 1 is valid for lignite and sub bituminous and high volatile grade C and B coals with higher moisture contents as observed in the analysed samples. It will not apply to "low volatile higher grade A" bituminous and anthracite containing less than 10% Moisture content.

Table 2. Ovbiowun coal analysis report

Parameter	As received	Dray basis	Dry ash free
Total moisture, %, M	10.8		
Ash content, %, A	6.3	7.1	
Volatile matter, %, VM	41.7	46.8	50.4
Fixed carbon, %, FC	41.1	46.1	49.6
Total sulphur, %, S	1.66	1.66	2.00
GCV, kcal/kg	6864	7695	8283
GCV, MJ/Kg	28.739	32.219	34.641

Table 3. Equivalent elemental derivatives for Ovbiowun coal analysis generated from proximate parameters according to the Table 1

Element	Derivative	Value
% Carbon	$0.97\mathbf{FC} + 0.7(\mathbf{VM} - 0.1\mathbf{A}) - \mathbf{M}(0.6-0.01\mathbf{M})$	63.30
% Hydrogen	$0.036\mathbf{FC} + 0.086(\mathbf{VM} - 0.1\mathbf{A}) - 0.0035\mathbf{M}^2(1-0.02\mathbf{M})$	4.70
% Nitrogen	$2.10 - 0.020\mathbf{VM}$	1.27
% Oxygen	$100 - \%C - \%H - \%N - \%S - A\% - M\%$	11.97

Table 4. Oloku coal analysis report

Parameter	As received	Dray basis	Dry ash free
Total moisture, %, M	15.0		
Ash content, %, A	7.0	8.2	
Volatile matter, %, VM	40.7	47.9	52.2
Fixed carbon, %, FC	37.3	43.9	47.8
Total sulphur, %, S	0.36	0.42	0.46
GCV, kcal/kg	6051	7119	7755
GCV, MJ/Kg	25.333	29.804	32.466

Table 5. Equivalent elemental derivatives for Oloku coal analysis generated from proximate parameters according to the Table 1

Element	Derivative	Value
% Carbon	$0.97\mathbf{FC} + 0.7(\mathbf{VM} - 0.1\mathbf{A}) - \mathbf{M}(0.6-0.01\mathbf{M})$	57.43
% Hydrogen	$0.036\mathbf{C} + 0.086(\mathbf{VM} - 0.1\mathbf{A}) - 0.0035\mathbf{M}^2(1-0.02\mathbf{M})$	4.23
% Nitrogen	$2.10 - 0.020\mathbf{VM}$	1.29
% Oxygen	$100 - \%C - \%H - \%N - \%S - A\% - M\%$	14.69

2.3. Procedure

2.3.1. Gross caloric value (GCV)/Net caloric value (NCV)

The quantity termed Gross Calorific Value (GCV) is otherwise called the higher heating value (HHV), Upper Heating Value (UHV) or Higher Calorific Value (HCV). The net calorific

value (NCV) is same as lower heating value (LHV) or lower calorific value (LCV), determined by deducting the heat of vaporization of the water vapour from the GCV or HHV. The gross calorific value which is laboratory-determined calorific value is the heat produced by combustion of a unit quantity of coal at constant volume in an oxygen bomb calorimeter under specified conditions such that the end products of the combustion are in the form of ash, gaseous carbon dioxide, sulphur dioxide, nitrogen, and *liquid water*. Burning coal as a fuel does not produce as much heat per unit quantity. In coal or any fuel containing hydrogen the calorific value as found by the calorimeter is higher than that obtainable under most working conditions in boiler practice by an amount equal to the latent heat of the vaporization of water. This is because in the closed system of the laboratory calorimeter, this heat would reappear when the vapour was condensed, though in ordinary practice the vapour passes away uncondensed. Corrections are made to the gross calorific values for this difference between the laboratory and coal-burning facility. The corrected value is referred to as the net calorific value, NCV. This is defined as the heat produced by combustion of a unit quantity of coal at constant atmospheric pressure under conditions such that all water in the products remains in the form of vapour (in coal burning facilities exits as vapour, not condensed like laboratory). The net calorific value is lower than the gross calorific value. This fact gives rise to a distinction in heat values into the so-called "higher" and "lower" calorific values. The higher value, i.e., the one determined by the calorimeter is theoretical calorific value of coal while the lower value is the practical heating value a certain mass of coal will produce when burnt in standard facilities.

Both are estimated by attempt to reverse all combustion products back to its pre-combustion temperature state. The LHV/NCV assumes the water as a vapour is lost to atmosphere hence no heat energy is re-supplied to the system by vapour cooling to liquid (hence the deduction of the heat of vaporization energy of water from GCV). The water component of a combustion process will assumes liquid state after the combustion process hence GCV will include heat of vaporization energy of water. The water vapour produced holds the heat of vaporization of the water. The GCV of a fuel includes the heat released if all of the water vapours in the combustion products were condensed, releasing the heat of vaporization of the water in the combustion products. This is typically the situation that exists when a bomb calorimeter is used to measure the heat of combustion.

GCV/HHV Calculations

Several authors had attempted the calculation of HHV/GCV from numerous derived equations. According to Parikh *et al.* [6], the entire spectrum of solid carbonaceous materials like coals, lignite, all types of biomass material, and char to residue-derived fuels have been considered in derivation of present correlation which is given as:

$$\text{GCV/HHV} = 0.3536\text{FC} + 0.1559\text{VM} - 0.0078\text{A}$$

where FC is fixed carbon between 1.0–91.5%, VM is volatile matter between 0.92–90.6% and ash is 0.12–77.7% ash content.

Several other derived theoretical physical constants developed in relation to determine the gross calorific value are as follows:

$$\text{GCV} = 0.3491\text{C} + 1.1783\text{H} + 0.1005\text{S} - 0.1034\text{O} - 0.0151\text{N} - 0.0211\text{A} \text{ (MJ/kg)}$$

with the ranges as, represents carbon, hydrogen, oxygen, nitrogen, sulphur and ash contents of material respectively, expressed in mass percentages on dry basis [7]

$$\text{GCV} = -0.03\text{A} - 0.11\text{M} + 0.33\text{VM} + 0.35\text{FC} \text{ (Majumder et al. [8]) based on proximate analysis.}$$

$$\text{GCV} = 3.55\text{C}^2 - 232\text{C} - 2230\text{H} + 51.2\text{C} \times \text{H} + 131\text{N} + 20,600 \text{ (Friedl et al. [9]) based on ultimate analysis; unit in KJ/Kg.}$$

Another formula to calculate the GCV is Dulong's Formula. Dulong [10] derived GCV on dry basis as follows:

$$\text{GCV(db)} = 333 \times \text{C(db)} + 1442(\text{H(db)} - \text{O(db)} / 8) + 93 \times \text{S(db)} \text{ GCV is in (KJ/Kg dry basis) - C, H, O, S are percentage dry basis.}$$

$GCV = -1.3675 + 0.3137C + 0.7009H + 0.0318O$ according to Sheng and Azevedo [11] from his ultimate analysis.

$GCV = -0.763 + 0.301C + 0.525H + 0.064O$ According to Jenkins and Ebeling [12] from his ultimate analysis.

$HHV = 0.561M^{-6.137} \times VM^{0.381} \times FC^{0.666}$ (Akkaya [13]) using proximate analysis

All the HHV/GCV is in (MJ/kg) except for Fried *et al.* [9] and Dulong's Formula. All the parameters are on weight percentage (%wt). Any derivatives that do not include moisture (M) imply that the parameters are to be used as on-dry-basis.

The best correlated multivariable equation for determining GCV (for this particular grade of coal) is developed by Given *et al.* [14] by using Artificial Neural Network (ANN) to remodel and remove some errors and supply non-provided conditions as well as combining the precision in the work of several existing authors as mentioned. He derived a GCV equation with R^2 value of 0.9952 implying overall good fit for all data with up to 95% confidence. This is one of the best-correlated multivariable equations, between the various ultimate and proximate parameters and is given as

$GCV/HHV \text{ (MJ/kg)} = 91.4621 - 0.0556M + 0.02800VM - 0.9039A - 0.5687C - 0.6972N - 1.1252O - 0.8775S.$

The amounts of fixed carbon and volatile combustible matter directly contribute to the heating value of coal. Fixed carbon acts as a main heat generator during burning. High volatile matter content indicates easy ignition of fuel. Oxygen content of coal have been found to have relatively maximum influence and nitrogen content have been found to be minimum influence over GCV (MJ/kg). The net calorific value NCV is given as

$NCV = \text{Gross CV} - 50.6H - 5.85M - 0.191O$ in kcal/kg unit.

$NCV = \text{Gross CV} - 0.212H - 0.0245M - 0.0008O$ in MJ/kg unit

$NCV = \text{Gross CV} - 91.2H - 10.5M - 0.34O$ in Btu/lb unit.

where M is % moisture; H is % hydrogen; O is % oxygen (from ultimate analysis, as received or calculated).

GCV of 25.3 MJ/Kg of Oloku coal is within range of 24.4 – 26.7 MJ/Kg for the highest grade of sub-bituminous coal termed the "Sub bituminous A grade" according to the ASTM classification table. Ovbiowun coal with GCV of 28.7 MJ/Kg is within the range of 26.7 – 30.2 MJ/Kg as the least grade of bituminous coal termed the "high volatile C bituminous" by the same ASTM classification table. The volatile content of coal is an indicator of its level of maturity and it is observed that as volatiles in bituminous (and by extension, all coals) reduces from high-to-low, fixed carbon increases, the rank advances (from sub-bituminous, bituminous, semi anthracite to anthracite) and calorific value increases.

CO₂ emission factor

There is a general growing concern about carbon dioxide (CO₂) emissions resulting from the combustion of fossil fuels and the potential effects of the emissions on future global climate. There have been several proposal and strategies for reducing the CO₂ emissions from fossil fuel combustion which include reducing end-use energy demand (conservation), improving the efficiency of engines and generators and shifting the mix of fossil fuels consumed. Natural gas produces the lowest CO₂ emissions per unit of energy output of all available fossil fuels while coal produces relatively the highest. A small percentage of the carbon in the coal remains un-burnt due to factors, such as reactivity of the coal particles, milling, air to fuel ratio, flame turbulence, fuel residence time etc. A small portion of the un-burnt carbon goes with the fly ash (FA) and the remaining un-burnt carbon goes in the bottom ash (BA). Carbon dioxide (CO₂) emissions may be expressed in form of either of the four equations:

$t \text{ CO}_2/TJ = 3.667C \times [NCV/10000]$ in kJ/kg

$t \text{ CO}_2/TJ = 3.667C \times [NCV/2388.46]$ in kcal/kg.

Ibs of CO₂/ MMBtu (gross) = % Carbon/GCV X 36,640 in Ibs/MMBtu

CO₂ emissions factor (in lb/ton coal) = % Carbon x 72.6 in Ib/tonne

Carbon dioxide (CO₂) emissions for the sake of this study are calculated in form of gross calorific value as pounds of CO₂ emission per million British thermal units (lbs CO₂/MMBtu) and lb/tonne in the last two equations. The % carbon is the total carbon from the ultimate analysis and gross calorific value is expressed in Btu/lb, both on a dry basis. The constant, 36,640, is simply the ratio of the molecular weight of CO₂ to the atomic weight of carbon, multiplied by the ratio of 10⁶ (a million) to 100 percent. In this calculation assumption is made that all the carbon in the coal converts to CO₂ during combustion. The gross calorific value used to calculate CO₂ emissions in this study is determined at constant volume and with water in the condensed state.

For Ovbiowun coals, the Ibs of CO₂/ MMBtu (gross) = 167.44 Ibs/Btu, 4596 Ib/tonne

For Oloku coals, Ibs of CO₂/ MMBtu (gross) = 164.21 Ibs/Btu, 4376 Ib/tonne

Fuel ratio

Fuel ratio is given by FR = Fixed Carbon / Volatile Matter. The fuel ratio may be calculated based on as received or dry ash free and for this project, both gave same results.

Ovbiowun coal (as received) = 41.1/41.7 = 0.98; (dry ash free) = 49.6/50.4 = 0.98

Oloku coal (as received) = 37.3/40.7 = 0.91; (dry ash free) = 47.6/52.2 = 0.91

From Frazer's [16] fuel ratio, coals of lower rank than sub-bituminous coal were not considered in detail and his classification was based on the two proximate analysis derivatives (fixed carbon and volatile matter) as shown in table 6.

Table 6. Frazer's fuel ratio coal classifications [16]

S/No	Coal Type	Fuel Ratio
1	Anthracite	12-100
2	Semi Anthracite	9-12
3	Low volatile bituminous	5-8
4	Medium to High volatile bituminous & All Sub-bituminous	0-5

Vitrinite reflectance

The Vitrinite reflectance of the studied coal was determined using the formula given by Rice [17], whose applicability is limited to a defined range of VM, i.e., 10% < VM < 40% [17]. The VM% of the coal sampled is within this range. Where V_o is vitrinite reflectance, VM_{das} is the volatile matter calculated on dry ash free basis, the formula is given by:

$$V_o\% = -2.712 \times \log (VM_{daf}) + 5.092$$

For Ovbiowun coals VM_{daf} is 50.4, hence V_o% = -2.712 × log (50.4) + 5.092 = 0.48

For Oloku coals VM_{daf} is 52.2, hence V_o% = -2.712 × log (52.2) + 5.092 = 0.43

From correlation chart for vitrinite maturity indices for coal (Hayes 1991), 0.25 – 0.45 is sub-bituminous while 0.5 – 0.6 is congruous with high volatile, lowest grade C bituminous.

Methane content

The generation of methane gas depends on the temperature, pressure and composition of coal seams. Generally, with increasing depth, pressure and temperature conditions, enhancement in the rank and maturity of the coal is observed along with increase in carbon percentage. Several workers gave empirical formulae for the estimation of Coal bedded methane (CBM) in the coal seams depending on various parameters. Meisner [18] observed that the amount of methane gas is related to volatile matter, calculated on dry ash free basis, by the following equation:

$$V_{CH_4} = -325.6 \times \log (VM_{daf}/37.8) \text{ (m}^3\text{/tonne)}$$

The estimated volume of methane generated is in (m³/tonne) according to Meisner [18]. During coalification, coal becomes progressively enriched in carbon and continues to expel volatile matter. It leads to progressively enriched methane content due to the thermal maturation which confirms the potentiality of CBM as depth of overburden increases [19]. Gas

content of coal was found increasing with increase of fixed carbon (daf). Hence we can conclude that higher percentage of fixed carbon (daf basis), higher vitrinite reflectance and lower percentage of volatile matter (daf basis) reflects more methane in coal. However for this study, there will be no coal-bedded methane since the volatile matter on dry-ash-free basis is greater than 37.8 i.e. methane production in coals starts when the volatile matter approximates less than 37.8%

Free swelling index

The free-swelling index (FSI) is a measure of the increase in volume of coal when heated under specified conditions (ASTM D-720; ISO 335) [20-21]. The free-swelling index number is an indicator of the capability of coal to form coke. An index number of 6 or higher indicates good coke-forming ability with the higher numbers suggesting greater coke-forming ability. FSI is classified into different range of caking/swelling capacities and assigned several standard values from 1 to 9. According to Speight [22], he classified coals FSI into (0-2), (2-4), and (4-9) caking ranges representing weak, moderate and strong caking tendencies as shown in table 7.

Table 7. Plastic properties of coal after Baughman [23]

Coal Type	Swelling Index	Dilation (%)	Roga Index
Non-caking	0	0	0-5
Weakly caking	1-2	0	5-20
Medium caking	2-4	0-40	20-50
Strongly caking	>4	>50	>50
Metallurgical coals	>7		

The impurities present in coke reduce available carbon for reduction properties necessary to produce good coke. The impurities majorly are ash, volatile matter, sulphur, Iron oxides and alkali oxides. Some mineral matter, most especially calcium-containing substances reduce coking properties/tendencies by deteriorating thermoplastic properties hence decrease their swelling index [24]. Equally, Sulphur has the same reducing effect because most of the sulphur content of coal remains in coke and as it increases coke productivity in the blast furnace decreases [25]. On the contrary, the alkali content (Fe_2O_3 and CaO) in coal may favour coke reactivity. When these oxides are much available in optimal/maximum proportions the catalytic effect of the ash on coke reactivity is enhanced. Higher moisture contents result in lower FSIs, also the higher the volatile matter the lesser the FSI and the higher the alkali oxides (Fe_2O_3 and CaO) the higher the FSI. Chelgani [26] assessed the properties of a wide range of American coals with reference to its determined FSI and its variations with respect to proximate, ultimate and ash analysis of coals using regression and artificial neural networks. Based on the FSI feed-forward artificial neural network (FANN) equations (of improved and accurate FSI prediction model), the free swelling index is re-modified for the calculation in this study. According to Chelgani *et al.* [26]:

$$\text{FSI} = 33.379 - 0.779\text{O} - 0.23\text{C} - 1.787\text{N} + 0.437\text{H} - 0.318\text{A} - 0.362\text{Sp} - 0.361\text{So} \quad R^2 = 0.62$$

Chelgani. [26] explains according to inter correlations using a stepwise procedure between the ultimate parameters and FSI, it can be shown that hydrogen exclusive of moisture have positive effect while total hydrogen has negative effect on FSIs. However for the sake of this project, the "total hydrogen" and not "hydrogen exclusive of moisture" is used hence the "+0.437" is changed to "-0.437". The hydrogen used for the above equation is total hydrogen (H_{total}) which is Hydrogen dry basis from ultimate analysis (H) + Hydrogen in moisture Hydrogen in Moisture ($H_{\text{moist}} = 0.1119 \times \text{Moisture} = 0.1119M$). Total hydrogen (H) = Hydrogen on dry basis from ultimate analysis (H_{ult}) + Hydrogen in Moisture (H_{moist})

$$H = H_{ult} + H_{moist} = H_{ult} + 0.1119M$$

The equation of Chelgani [26] for this study is modified as

$$FSI = 33.379 - 0.779O - 0.23C - 1.787N - 0.437(H_{ult} + 0.1119M) - 0.318A - 0.362Sp - 0.361So \quad R^2 = 0.62$$

where A = Ash content; O = Oxygen; C = Carbon; N = Nitrogen; H_{ult} = Hydrogen on dry basis from ultimate analysis; Sp = Pyritic Sulphur and So = Organic Sulphur.

Using a stepwise procedure while integrating with ultimate parameters, the equation is further re-derived. The total sulphur as analysed from the result of the analysis is Sp + So and since their coefficients are approximately equal 0.362/0.361, then the equation may be factorized by the coefficients as

$$FSI = 33.379 - 0.779O - 0.23C - 1.787N - 0.437(H_{ult} + 0.1119M) - 0.318A - 0.362 (Sp + So)$$

Substituting S, total sulphur = Sp (pyritic sulphur) + So (organic sulphur) into the equation, we have

$$FSI = 33.379 - 0.779O - 0.23C - 1.787N - 0.437(H_{ult} + 0.1119M) - 0.318A - 0.362S$$

For Ovbiowun coals (Table 2 and 3) FSI = 2.04; for Oloku coals (Table 4 and 5) FSI = 1.48

Correlations between ultimate and proximate analysis as studied by Chelgani *et al.* [26] proves that ash, moisture, volatile matter, oxygen and sulphur in coal negatively correlated with FSI, the higher these parameters the lower the FSI. They all have a negative effect on the FSI value. Carbon content and hydrogen (not including that in moisture) as well as K_2O in coal have positive effects on FSI. Lower-volatile coals with volatile matter (31 to 33%) are strongly expanding, creates strong pressure during coking and forms or makes a strong coke, due to high fixed carbon and other coking characteristics.

Hardgroove grindability index

The grindability index or Hardgroove Grindability Index HGI determines how pulverisable/millable a coal is. Hardgroove Grindability Index (HGI) is of great consideration as a predictive tool to determine the performance capacity of industrial pulverizers in power station boilers because it gives an expression of the relative ease of grinding coals or the power required for grinding coals in those pulverizers. A high value of HGI indicates a soft and easily grindable coal i. e. the higher HGI values, the greater the ease of milling the coal and the lower the costs of the milling operation. HGI of coal may reach a maximum of about 105 for bright coals and then falls sharply to about 35 for anthracites. The Hardgroove index of coal is largely determined by the coal's moisture content and it controls mill performance. This index is a factor in deciding the size of the crushers and grinders that would be required in coal preparation plants. Although the HGI testing device is not too costly, the measuring procedure to get a HGI value is time consuming and consequently some researchers have investigated the prediction of HGI based on proximate analysis, petrography, vitrinite reflectance and artificial neural networks (ANNs). In particular is the feed-forward artificial neural network (FANNs) which has been extensively studied to present process models whose use in coal industry has been rapidly growing [31]. The following equation resulted from FANNs as derived between HGI proximate and ultimate analysis by Khoshjavan *et al.* [27] as:

$$HGI = 67 - 3.16 O - 3.02 N - 0.23 H - 2.37 C - 0.00585 GCV(Btu/Ib) - 1.53 S + 3 FC - 1A + 3 VM + 1.13M.$$

All the values are calculated on dry basis including the GCV (in Btu/Ib) except the moisture content. Virtually all coal mills are designed for HGI greater than or equal to 50. Anything lesser will cause mill wear. Bituminous coal ranges generally from 45 – 95 HGI and Anthracite 30 to 50 HGI. Some lesser rank lignites might be at 90 HGI or higher. From table 2 and 3 Ovbiowun coals have 65 HGI, and from table 4 and 5 Oloku coal is 87 HGI using the equation of Khoshjavan *et al.* [27].

Volatile matter

The classification of degree of suitability of volatile matter in coking coals for blending was proposed by Gray *et al.* [28]. He prescribed volatile matter content between 31.0 to 33.0% is described as good, 33.0 to 36.0% as medium, and greater than 36.0% is termed as poor. Sequel to his proposal, Blackmore [29] observed that coals with over 4 FSI are generally considered to have appreciable coking abilities. For the coal to be classified as metallurgical coals, its FSI must be greater than or equals 7. The highest FSI according to Blackmore is 9, at the top of the scale.

3. Results and discussion

For both Ovbiowun near Afuze (Edo state, western R.Niger) and Oloku near Ogboyoga (Kogi state, eastern R.Niger) analysed in this project, the results is as shown in the table 8.

Table 8. Results for the various parameters analysed with their derivatives.

Derivatives	Ovbiowun Coal (Edo State)	Oloku Coal (Kogi State)
GCV/HHV (MJ/kg) = 91.4621 - 0.0556M + 0.02800VM - 0.9039A - 0.5687C - 0.6972N - 1.1252O - 0.8775S	28.739 MJ/Kg	25.333 MJ/Kg
NCV = GCV - 50.6H - 5.85M - 0.191O in kcal/kg unit.	27.342 MJ/Kg	23.939 MJ/Kg
CO ₂ EMISSION FACTOR	167.44	164.21
Ibs of CO ₂ / MMBtu (gross) = % Carbon/GCV X 36,640 in Ibs/MMBtu	Ibs/Btu,	Ibs/Btu,
CO ₂ emissions factor (in lb/ton coal) = % Carbon x 72.6 in Ib/tonne	4596 Ib/tonne	4376 Ib/tonne
FUEL RATIO		
FR = Fixed Carbon / Volatile Matter	0.98	0.91
VITRINITE REFLECTANCE		
Vo% = -2.712 × log (VM _{das}) + 5.092	0.48	0.43
METHANE CONTENT		
V _{CH₄} = -325.6 × log(VM/37.8) (m ³ /tonne)	0.00	0.00
FREE SWELLING INDEX		
FSI = 33.379 - 0.779O - 0.23C - 1.787N + 0.437H - 0.318A - 0.362S	2.57	2.23
HARDGROVE GRINDABILITY INDEX		
HGI = 67- 3.16 O- 3.02 N- 0.23 H -2.37 C - 0.00585 GCV(Btu/Ib) - 1.53 S + 3 FC - 1A + 3 VM + 1.13M.	65	87
VOLATILE MATTER (dry basis)	46.8	47.9

3.1. GCV

According to table 6, GCV of 25.3 MJ/Kg of Oloku coal is within range of 24.4 – 26.7 MJ/Kg for the highest grade of sub-bituminous coal termed the “Sub bituminous A grade” according to the ASTM classification. Ovbiowun coal with GCV of 28.7 MJ/Kg is congruous to the range 26.7 – 30.2 MJ/Kg for the least grade of bituminous coal termed the “high volatile C bituminous” equally by the ASTM. For Ovbiowun coals, the GCV calculated from the formula (GCV/HHV (MJ/kg) = 91.4621 - 0.0556M + 0.02800VM - 0.9039A - 0.5687C - 0.6972N - 1.1252O - 0.8775S.) approximates 28.19 (MJ/Kg) which is very close to the analysis report of 28.739 and Oloku coals approximates 25.67 (MJ/Kg) very close to 25.333 (MJ/Kg) as observed from the analysis.

3.2. CO₂ Emission factor

Generally, bituminous coals have heating values of 10,500 Btu/lb (24.423MJ/Kg) to 14,000 Btu/lb (32.564 MJ/Kg) on a wet, mineral-matter-free basis. The heating values of sub-bituminous coals range from 8,300 Btu/lb (19.306 MJ/Kg) to 11,500 Btu/lb (26.749 MJ/Kg)

on a wet, mineral-matter-free basis, and from 9,420 Btu/lb (21.911 MJ/Kg) to 10,130 Btu/lb (23.562 MJ/Kg) on an as-mined basis (Windsor, 1981). This strongly agrees with Oloku coals as sub-bituminous and Ovbiowun coals as bituminous. The CO₂ emission factor of both coals (167.44 and 164.21 Ibs/Btu) is within the acceptable range of accepted value by the EPA (Environmental protection Agency).

According to External combustion sources, the result tallies with expected CO₂ emissions for sub-bituminous and high volatile bituminous coals. With rank advance in the coal from low-volatile bituminous to semi-anthracite to anthracite, calorific value tends to decrease though carbon content continues to increase. Hence anthracites would be expected to have more CO₂ emissions even greater than other grade of coals [32].

3.3. Fuel ratio

According to Frazer's classification (Table 7) using Pennsylvania and Indian coals, he ranged bituminous coal between 0-5 which contained the calculated values for the study. The fuel ratio for both Ovbiowun and Oloku is 0.98 and 0.91 respectively which is a referral to bituminous grade.

3.4. Vitrinite reflectance

According to the ASTM classification, sub-bituminous coal has a vitrinite reflectance within the range of 0.34 – 0.47 while high volatile C bituminous coal in range of 0.47 – 0.6. The vitrinite reflectance as determined for the studied samples as determined according to Rice [17] was 0.43 for Ovbiowun coals and 0.48 for Oloku coals implying bituminous and sub-bituminous coals respectively.

3.5. Methane content

The volume of methane produced by various coals according to Meisner [18] stipulates generation of gas starts at 37.8 VM_{daf} corresponding to 0.73 vitrinite reflectance and continues with decrease in volatile matter/increase in vitrinite reflectance. Since the VM_{daf} is greater than 37.8 and less than 0.73 R_o, no coal-bedded methane will be generated. From correlation chart for Vitrinite maturity indices for coal after Hayes [32], only "low volatile bituminous A grade" will production of methane gas starts. No coal-bedded methane is produced from both coals.

3.6. Free swelling index

From results in table 7, the FSI of Ovbiowun (2.04) and Oloku (1.48) implies very weak swelling and caking characteristics of the coal. The Roga index is inferred to be around "20 – 40" and their dilation approximates "zero". Free swelling index is a rank-dependent parameter and when correlated with volatile matter, moisture and ash content makes determinant in prescribing the effectiveness of coal coking ability. Both coal from Ovbiowun and Oloku are considered to have poor coking and swelling tendencies as inferred from the derived FSI values. They cannot be used as coke since the coal in this study exceeds 36% in volatiles which makes them in range of poor coking abilities as explained by Blackmore [29]. Volatile has been known to provide rank estimation and burning characteristics as well as price parameter for selling and buying coals. More so, the > 10% moisture and >10% ash content coal of both coal makes them unsuitable for coke production.

3.7. HARDGROVE grindability index

Ovbiowun coals have 65 HGI and Oloku coal is 87 HGI. This is within bituminous range and as expected, Ovbiowun has less volatile, more vitrinite reflectance value as well as of higher bituminous grade than Oloku. Higher moisture content in coal can result in higher HGI and lower volatile matter content in coal results in higher HGI [30]. The moisture content of Oloku coal (15.0) is higher than Ovbiowun (10.8) while the volatile matter (as received) for Oloku (40.7) is lower than Ovbiowun (41.7). The increase of hydrogen content in coal can result in lower HGI and higher total sulphur (S_{total}) result in higher HGI (Chelgani *et al.* [30]). Also the

derived Hydrogen % for Ovbiowun (63.3%) is greater than Oloku (57.43) confirmed by the lesser HGI values.

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Appendices conversions – units

From kcal/kg to MJ/kg multiply by 0.004187 From kcal/kg to Btu/lb multiply by 1.800
 kcal/kg = Btu/lb/1.800 From MJ/kg to kcal/kg multiply MJ/kg by 238.846
 MJ/kg = kcal/kg/238.846 From MJ/kg to Btu/lb multiply MJ/kg by 429.923
 MJ/kg = Btu/lb/429.923 From Btu/lb to kcal/kg multiply Btu/lb by 0.5556
 From Btu/lb to MJ/kg multiply Btu/lb by 0.002326
 Tons- A "short" or net ton is equal to 2,000 pounds. A "long" or British ton is 2,240 pounds; a "metric" ton is approximately 2,205 pounds.

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