Critical evaluation of a Nigerian sub-bituminous coal potential for energy derivation

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Abstract. A good understanding of the chemical composition and structural characteristics of a carbonaceous material is essential in conversion processes. Understanding how the composition and structural changes influence the burning behaviour of coal is important when assessing a coal's potential for utilization. To explore the potentials of a typical Nigerian coal, both conventional and advanced analytical techniques such as proximate analysis, ultimate analysis, calorific value, surface area analyser, SEM, FTIR, XRD and SAXS were employed. The results obtained from these characterizations agree favourable well with a typical South African coal that is of enormous contribution to the gross domestic product (GDP) of the nation economy.

Keywords: coal; char; physical properties; chemical properties; petrographics; GDP

1. Introduction

Coal and coal products will continue to play an increasingly important role in fulfilling the energy needs and economies of nations. This is because of the abundant reserves of coal and its low cost (Crompton and Wu 2005, Ma *et al.* 2013, Stracher and Taylor 2004). Coal accounts for roughly 25% of the world energy supply and 40% of carbon emissions but even with the high percentage of emissions, it is very unlikely that any of these countries that are into coal exploration and production will turn their back on coal very soon (Jinke *et al.* 2008, Patzek and Croft 2010, Yilman and Ulsu 2007). Economic growth requires energy growth. With the recent concern for the environment and renewed interest in research on alternative energy from renewable sources, hydrogen from coal through the integrated gasification combined cycle has been considered for the proposed hydrogen economy (Attwood *et al.* 2003, Bartels *et al.* 2010, Veziroglu and Sahin 2008). Gasification has been tipped as the 21st century clean coal conversion technology than the other coal utilization processes such as liquefaction and combustion because it is high energy efficient, non-polluting and economical (Attwood *et al.* 2003, Bartels *et al.* 2010). It have also the merit of going beyond the use of coal for the generation of power, metal processing and the production of chemicals, as coal could be converted to useful gases and liquids (Levine *et al.* 1982, Schobert and

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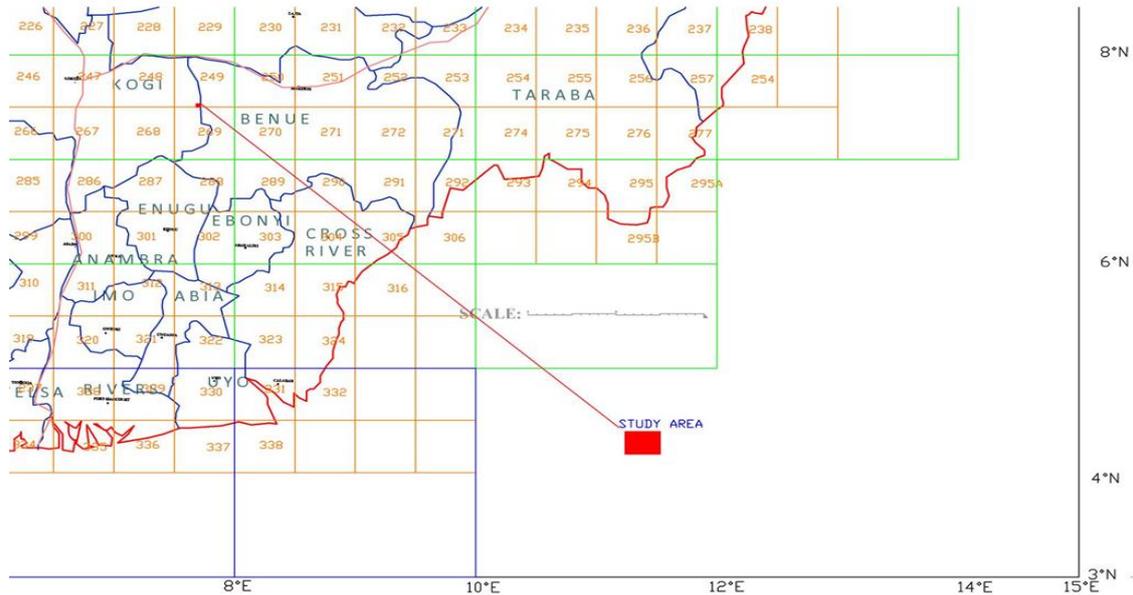∗Corresponding author, Ph.D., E-mail: odehandy@yahoo.com

Fig. 1 Field location for the sub-bituminous coal sample used

Song 2002). Coal is a complex carbonaceous material consisting of organic and inorganic matter. During gasification, the organic and inorganic matter undergoes various chemical and physical transformations. In order to maximise the gasification efficiency, there is the need to understand the mechanism for the chemical and physical transformation, as this will assist in the reduction of carbon emissions in the process especially when gasifying low rank coal (Collot 2006, Cousins *et al*. 2006). Several options are used to control the feed rate of coal during gasification: fixed bed fluidised bed and entrained flow gasifiers. Fluidised bed gasifiers have the potential advantage that low-grade coals rich in ash and inertinites, such as South African coals can be processed more efficiently than in conventional in pulverized coal boilers (Everson *et al*. 2008, van Niekerk *et al*. 2008).

Therefore, the design of coal utilization processes will require a deeper understanding of coal's intrinsic properties and the ways in which it is chemically transformed under process conditions. Hence a critical comparision of a Nigerian sub-bituminous coal with a typical South African bituminous coal in order to comprehend the coal to char transformation including their behaviour is the subject of this investigation. It is anticipated that the information obtained from this study would assist in optimising gasifier design and operation, most especially when it relates to a Nigerian sub-bituminous coal. Nigeria has been reported to have one of the largest coal deposits in Africa second to South Africa with a total estimated reserves of two and half billion tonnes and a proven reserves of about 200 million tonnes but unfortunately, the coal deposits have not been structurally investigated to explore the potentials (Akande *et al*. 1992, Sharma and Sharma 1991).

2. Experimental

2.1 Sample preparation

A series of coal covering all rank suite: Lignite to anthracite were subjected to heat treatment to investigate the both the physical and chemical transformation that takes place from coal to char and how this transformation impacts on coal reactivity. But in this paper, the discussion will be focused on the Nigerian coal, a sub-bituminous coded as NGR and a South African coal coded as BCH for comparison. Fig. 1 shows the geographical location of the Nigerian coal sample used in this study. The South African sample was collected from the highly exploited Witbank coal fields. The coal samples were subjected to coal preparation and pulverized to coal particle size of -75 μm and -1 mm as the analysis warrant. All the samples were stored under argon prior to analysis.

The prepared coal samples were demineralised to reduce the amount of mineral matter present in them as well as to minimise their influence during quantitative analysis. The procedure used for the chemical cleaning of the coal followed the sequential leaching with hydrofluoric acid (HF) and hydrochloric acid (HCl); a more detailed procedure is reported in Strydom *et al*. (2011).

Routine preparation of petrography coal blocks was conducted using ISO standard 7404/2. Parent coal was crushed to -1mm, and block for point counting and image analysis was produced by mounting the coal in resin. The blocks were polished using a Struers Tegraforce-1 polishing machine. Thereafter, detailed petrographic analysis (maceral analysis, total reflectance analysis and carbon form analysis) was carried out using ISO standard 7404/3. The standard entire sequence of steps for petrographic analysis was followed as described by Malumbazo *et al*. (2012).

2.2 Apparatus and procedure

The char production sequence from the parent coal samples are as follows: The measured coal samples (40 g) were placed in a boat and put in a horizontal tube furnace at 60°C and for 10 minutes so that the condensed moisture is driven off. The sample temperature was equilibrated to ambient temperature and pressure in a flow of nitrogen at a flow rate of 1 liter/min. The furnace was then heated non-isothermally at 20°C.min^{-1} to the target temperature, and held isothermally at the target temperature for 60 minutes. The target temperature varies from 450 C to 700°C, a temperature regime considered for char formation.

The conventional chemical analysis (both proximate and ultimate analyses) of the untreated coal, acid treated and heat treated samples were done according to the international ASTM 3172 and ASTM 3176 method respectively. The coal morphology and that of the resultant char obtained in the transition of coal to char was observed using scanning electron microscope (SEM) model FEI Quanta 250 with FEG emitter, equipped with an EDS detector that does the elemental composition analysis. The surface areas of the various samples were determined using the carbon dioxide adsorption BET method on a Micromeritics ASAP2020 Surface Area Analyzer.

The XRD was used to study the carbon crystallite properties of both the coal and char samples. The XRD scans were conducted on a PANalytical XRD X'Pert Pro powder diffractometer using both Co and Cu Kα radiation, a detailed procedure is reported by Wang *et al*. (2010). The spectra used in obtaining the structural properties of both the coal and char were obtained from the fourier-transform infrared spectrometer equipped with an attenuated total reflectance (FTIR-ATR), model Perkin-Elmer Spectrum 400. More information on the procedure and advantages of using FTIR-ATR technique has been reported by Li (2007).

The results of the proximate analysis, ultimate analysis and calorific values of the parent and demineralised coal, along with the gross calorific values, atomic H/C and atomic O/C ratio of the samples are presented in Table 1. Table 2 gives the calculated values of the atomic H/C ratio, atomic O/C ratio and the aromaticity (theoretically calculated from conventional analysis (Orrego-

Table 1 Proximate analysis, ultimate analysis, calorific values and computed H/C and aromaticity values for parent coal and demineralised coal

Coal	NGR	BCH	[a]NGR	[a]BCH
wt% Inherent moisture(air dried)	9.6	2.1	1.9	2.7
wt% Ash (air-dried)	9.0	16.2	2.0	1.2
wt% Volatile matter (air-dried)	37.6	26.7	43.2	27.2
wt% Fixed carbon (air-dried)	43.8	55.0	53.0	68.9
wt% Carbon (daf)	75.6	81.6	75.1	83.4
wt% Hydrogen (daf)	5.2	4.6	5.2	4.6
wt% Nitrogen (daf)	1.7	2.0	1.8	2.0
wt% Oxygen (daf)	16.9	10.7	17.4	9.1
wt% Sulphur (daf)	0.7	1.2	0.1	1.0
Gross calorific value (MJ/kg)	24.6	26.8	29.3	32.0
H/C	0.83	0.67	0.83	0.66
O/C	0.17	0.10	0.17	0.08
f_a	0.65	0.73	0.65	0.74

a: demineralised

Table 2 Computed H/C, O/C and aromaticity values for heat-treated coal from conventional analysis, FTIR and XRD

Coal	NGR					
	450	500	550	600	650	700
H/C	0.45	0.38	0.32	0.26	0.16	0.13
O/C	0.08	0.06	0.04	0.03	0.03	0.03
Gross calorific value (MJ/kg)	31.0	31.4	31.8	32.3	31.9	31.4
$f_{a\,(CA)}$	0.86	0.90	0.93	0.96	1.00	1.03
$f_{a\,(FTIR)}$	0.75	0.78	0.81	0.84	0.87	0.90
$f_{a\,(XRD)}$	0.67	0.69	0.70	0.74	0.78	0.80
	BCH					
	450	500	550	600	650	700
H/C	0.45	0.43	0.34	0.28	0.21	0.14
O/C	0.05	0.05	0.03	0.03	0.02	0.01
Gross calorific value (MJ/kg)	33.2	33.5	33.8	33.9	33.7	33.4
$f_{a\,(CA)}$	0.86	0.89	0.92	0.95	0.98	1.03
$f_{a\,(FTIR)}$	0.83	0.86	0.89	0.92	0.95	1.00
$f_{a\,(XRD)}$	0.93	0.94	0.97	0.98	0.99	0.99

Ruiz *et al.* 2011)) and experimental values from FTIR and XRD techniques) of the heat treated coal, simulating char formation.

3. Results and discussion

The coal samples are referred to as "NGR450, NGR700" to clearly indicate the sample identity and the temperature to which it was heat-treated. It is evident from our study that the Nigerian coal minerals have a lower degree of minerals when compared with the South African bituminous coal. The Nigerian coal has an ash content of 9 weight percent on air-dried basis while the South African ash content of 16.2 weight percent. And again, considering the inorganic elemental composition, the Nigerian coal has a lower percentage; 1.7 wt % N_2 and 0.7 wt% suphur for NGR while BCH has the values of 2 wt% N_2 and 1.2 wt% sulphur respectively (Table 1). Earlier work by Olajire *et al*. (2007), on the occurrence and distribution of metals and porphyrins in Nigerian coal minerals confirmed this finding and concluded in their report that the Nigerian coal is more environmentally friendly based on their low level of toxic metals compared to the coals from other parts of the world. Ogala *et al*. (2012) agreed to this finding by concluding that the Nigeria coal is of high grade, the low-rank classification not withstanding due to the low mineral content. This factor makes the Nigerian coal a choice coal in this era of getting clean energy from coal due to the fact that the level of emissions from the use of this coal in coal-fired utilities would be minimal. The impact of demineralization was more prominent in NGR (86% reduction in sulpur) compared to BCH (17% reduction in sulpur) which implies that the use of NGR in coal utilization systems would lead to reduced operational and environmental effects (corrosion, slagging, fouling, emission of particulate matter). The two coal samples tend to exhibit the same trend in their heating values. The gross calorific values of both coal samples increases maximally with increasing coalification temperature until the temperature of $600^{o}C$ (Table 2). At the maximum heating value of both coal samples at $600^{o}C$, as can be seen in Table 2, the atomic O/C ratio of both samples and virtually the atomic H/C ratio of both samples are the same, indicating indeed that the portion of the aromatic carbon in both samples is obtained at that temperature; char forming temperature for both samples is $600^{o}C$. The aromaticity determined at this temperature for both samples confirm the closeness in the properties of both coal samples being in agreement with the decreasing atomic O/C and H/C ratios. The aromaticity derived for NGR is 0.96, while BCH is 0.95. Basically, the major difference between the two coal samples which could traced to the vegetal deposition and to the coal classification system as seen in the volatile matter and the moisture contents. The volatile matter and the moisture contents (on-dried basis) for NGR is 37.6 wt% and 9.6 wt%, and 26.7 wt% and 2.1 wt% for BCH respectively. The van Krevelen diagram (Fig. 2) points to the same result as the high atomic H/C ratio obtained for both samples indicates high liptinite content and the coal samples can therefore be considered to be humic-like in nature.

A critical look at the distribution of the macerals in the two coals shows a closeness in the values of the total reactive macerals, as well as in the total number of non-reactive macerals; an indication which points to the fact that both coals will tend to exhibit similar reactivity in a reactor (Table 3, 4 and Fig. 3). The properties of macerals as described in Stach *et al*. (1982) elucidated the role of each lithotype and microlithotype; the liptinites are the most reactive macerals and rarely occur by themselves and are usually associated with either one or two other macerals. These associations referred to as microlithotypes could be bi-macerals between two macerals or tri-macerals between three macerals. The vitrinites are considered as reactive macerals while the inertinites are generally considered as unreactive. Though, of the inertinites, research has revealed that some part of the semi-fusinites and inertodetrinites has properties which range between those of vitrinite and fusinite and mineral impregnations are much rare, and, as such are classified as reactive macerals.
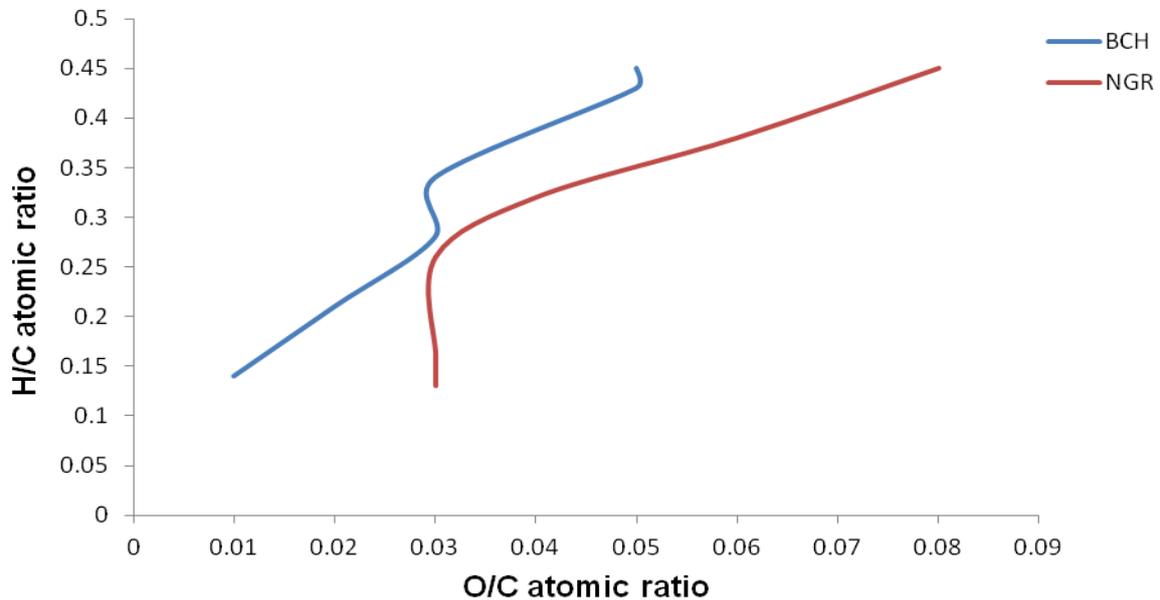
Fig. 2 The van Krevelen diagram for both coal samples during the coal to char transition
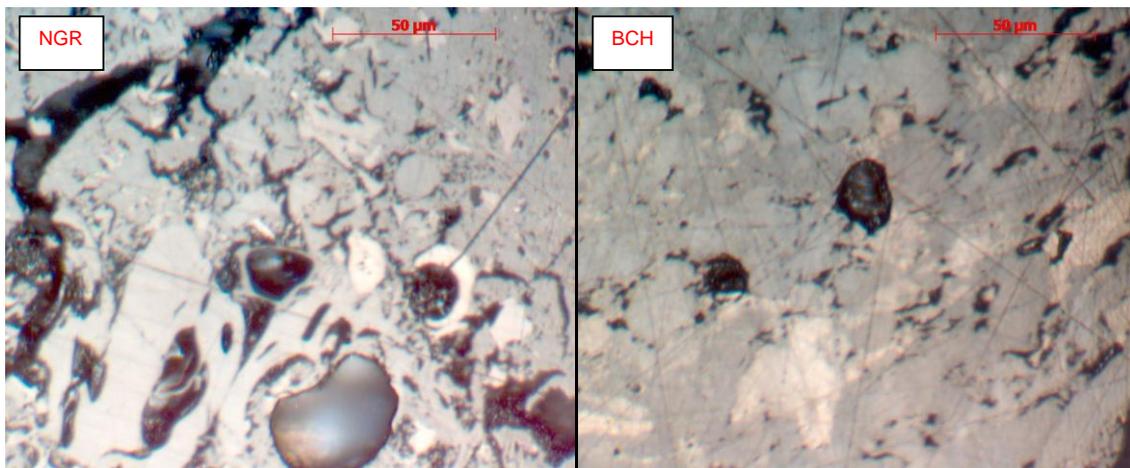

Fig. 3 Petrographic picture of the sub-bituminous and bituminous coal at 450°C

The reactivity to gasification can be affected broadly by the surface area and the pore size distribution of the coal material. The porosity and surface area, including the density has been identified as parameters that aids carbon burnout in coals (Fermoso *et al.* (2010)). Fig.4 shows the morphology of the Nigerian sub-bituminous coal and the South African bituminous coal, giving the surface view of the how the surface area and the pore size distribution lies within each coal material. However, Table 5 shows the values of the pore volumes, density, porosity and surface areas of both coal, and this undoubtedly gave close values for all parameters and similar trend. The trend displayed by both coals gave an indication that they will exhibit similar behaviour when subjected to coal conversion processes.

Table 3 Maceral group analysis and rank value for both coals

Macerals (Vol. %)	NGR	BCH
Vitrinite	56.5	32.0
Liptinite	10.5	6.0
Reactive semifusinite	3.0	9.0
Inert semifusinite	5.0	20.0
Fusite	13.0	8.0
Reactive inertodetrinite	1.8	8.0
Inert inertodetrinite	5.3	14.0
Micrinite	4.1	1.0
Minerals	0	2.0
Rank (mean random vitirinite reflectance %)	0.72	0.73
Rank (mean random total reflectance %)	0.77±0.52	1.23±0.61

Table 4 Total reactive and inert macerals for both coal samples vol. %

Macerals	NGR	BCH
Total reactive macerals	71.8	55.0
Total inert macerals	10.3	34.0
Total inertinites	15.1	51.0

Table 5 Determined properties obtained from SAXS, Pycnometer and Surface Area Analyser (ASAP 2020) for the heat treated samples

	NGR					
	450	500	550	600	650	700
Pore Volume (cm^3/g)	0.089637	0.103797	0.101829	0.128430	0.126650	0.128000
Density (kg/m^3)	1.239	1.258	1.349	1.489	1.544	1.483
Porosity (%)	11.1	13.1	13.7	19.1	19.6	19.0
CO_2 BET surface area (m^2/g)	155.78	182.61	183.19	234.10	238.14	239.74
	BCH					
	450	500	550	600	650	700
Pore Volume (cm^3/g)	0.080338	0.093108	0.104365	0.112853	0.112860	0.115407
Density (kg/m^3)	1.533	1.589	1.606	1.638	2.014	1.857
Porosity (%)	12.3	14.8	16.8	18.5	22.7	21.4
CO_2 BET surface area (m^2/g)	130.17	158.68	183.89	206.40	215.40	224.95

4. Conclusions

Nigeria is endowed with a large coal deposit, most of which are reported to be within the Benue trough. Despite the reported occurrence of these deposits, little attention has been paid to the exploration and exploitation of this resource since the discovery of crude oil and gas such that it
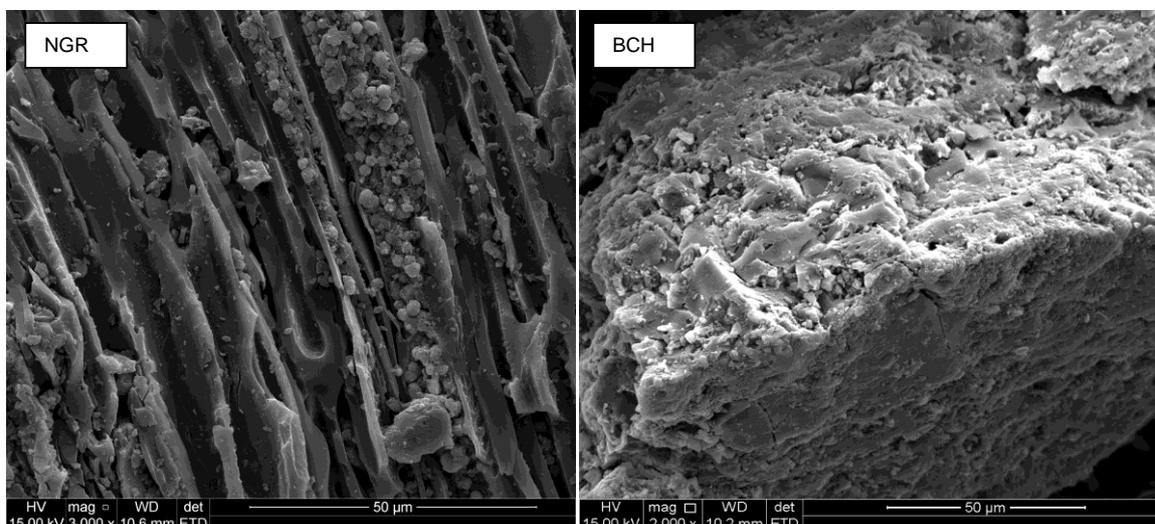
Fig. 4 SEM images for both coal samples

can be considered as a neglected prime resource. Based on the fact that coal is a resource that man will continue to look unto for energy generation for a very long time to come and coupled with depletion of this resource in some other countries, an exploration campaign in order to fully evaluate and characterize the Nigerian coal reserves is highly recommended. Despite the low rank of the Nigerian sub-bituminous coal, when compared with the highly exploited South African bituminous coal, It clearly demonstrated and constitute a high potential of good quality coal that could be used for coal gasification and other coal utilization processes.

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