Physicochemical characterization and thermal analysis of newly discovered Nigerian coals

B. B. Nyakuma

Centre of Hydrogen Energy, Institute of Future Energy,
Universiti Teknologi Malaysia, 81310 Skudai, Johor Bahru, Malaysia.

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The physicochemical properties and thermal degradation behaviour of three newly discovered sources of coals; Afuze, Garin Maiganga and Shankodi-Jangwa from Nigeria were examined. Characterization was performed to determine the rank, classification and quality of the coals using ultimate analysis, proximate analysis and higher heating value (HHV). Ultimate analysis revealed high Carbon (*C*), Hydrogen (*H*) and Oxygen (*O*), but low Nitrogen (*N*) and Sulphur (*S*) content. Proximate analysis demonstrated that the Garin Maiganga (GMG) coal sample exhibited the highest volatile matter (51.16 %) and Moisture (5.28 %) content. The highest ash content 30.99 % was observed in the Afuze coal; whereas the highest FC content was observed in the Shankodi-Jangwa coal sample. The heating values (HHV) of the different samples were; Garin Maiganga (23.74 MJ/kg); Shankodi-Jangwa (27.34 MJ/kg) and Afuze (30.52 MJ/kg) which confirmed Afuze and Shankodi-Jangwa as high rank coals and Garin Maiganga as low rank. Consequently, Afuze coal is classified as Bituminous and NOT Sub-Bituminous as previously reported. The results also demonstrate the reactivity and maturity of the coal samples by increasing order; Garin Maiganga>Shankodi-Jangwa>Afuze. Overall, the physicochemical and low rank properties of GMG coal make it suitable for coal power generation. The high fixed carbon content and thermochemical reactivity of SKJ indicate a good coking potential for steel and cement production. Last, the low moisture content and high carbon and heating values indicate AFZ coal can be utilized for power generation.

Keywords: Physicochemical, Thermogravimetric, Characterization, Low Rank Coal, Nigeria.

INTRODUCTION

Coal is the world's cheapest, most abundant and widely distributed source of energy with a global consumption of 5,544.3 Mtce [1, 2].Consequently, coal is the largest primary source of fossil fuels utilized for electric power generation and the production of chemicals, fuels and steel [3]. Despite the global importance of coal resources, the low cost, sustainable supply and self-sufficient electricity generation in sub-Saharan Africa remains a colossal conundrum.

Nigeria is the largest economy and exporter of crude oil in Africa with estimated crude oil reserves of 35 billion barrels, 187 trillion cubic feet of natural gas, and over 4 billion metric tonnes of coal [4]. In spite of Nigeria's vast energy potential, the country remains perennially plagued by energy crises resulting from low electricity generation, poor distribution and transmission losses [5]. This unfortunate scenario has greatly undermined Nigeria's potential for sustained socioeconomic growth, infrastructural development and energy security. Therefore, there is a critical need for cheap, sustained and consistent electric power supply generated from inexpensive sources of energy such as coal. Nigerian coals are typically low rank sub-bituminous in nature, with deposits located predominantly in the Lower, Middle and Upper Benue Trough of the Nigerian sedimentary basin [6].

Conversely, the search for higher ranked coals has resulted in the discovery of new deposits in Afuze, Garin Maiganga and Shankodi-Jangwa [7]. The Afuze coal is categorised as sub-bituminous with significant deposits located South West of the Benin Flank in the Anambra Basin in Afuze, Edo State [8]. The Shankodi-Jangwa coal is bituminous in nature with deposits situated in the Middle Benue Trough of Obi, Nasarawa state [9]. Last, Garin Maiganga coal is classified as sub-bituminous with deposits in the Akko, Gombe state [10].

However, lack of comprehensive scientific data on Nigerian coal has greatly hampered its utilization for electric power generation and industrial applications [11]. In addition, coal accounts for 37% of global carbon dioxide (CO_2) emissions prompting concerns about the long term socioeconomic and environmental impacts of coal power generation particularly in developing countries like Nigeria. Consequently, clean coal technologies such as underground coal gasification (UCG) [12], carbon capture storage (CCS) [13, 14] and the Integrated Gasification Combined Cycle (IGCC) [15] are currently under investigation for future coal power generation [16-18]. The development and diffusion

* To whom all correspondence should be sent:
E-mail: bbnyax1@gmail.com, bnbevan2@live.utm.my

of these clean coal technologies into the current energy mix of nations around the globe will greatly increase the prospects of cheap, efficient and sustainable energy.

However, the effective application of low-carbon emission techniques for coal power generation in Nigeria will require comprehensive knowledge of the physicochemical and thermal properties of indigenous coal. This is vital for examining the rank classification, environmental impacts and feasibility of the coal feedstocks for future applications [19]. Furthermore, the coal property data will be essential for the engineering design, process optimization and project costing of future energy conversion systems.

Consequently, this study is aimed at describing the physicochemical and thermal properties of the newly discovered Afuze, Garin Maiganga and Shankodi-Jangwa coals from Nigeria. The study presents novel results of the thermal degradation behaviour of the coals using thermogravimetric analysis (TGA) under inert conditions. Finally, the study attempts to propose potential future applications of the newly discovered coals.

EXPERIMENTAL

The coal samples examined in this study were obtained from the Afuze coal field in Afuze, Edo state; the Shankodi-Jangwa coal field in Obi, Nasarawa state and Garin Maiganga coal field in the Akko, Gombe state of Nigeria. The coals were subsequently labelled; AFZ - Afuze coal; SKJ – Shankodi-Jangwa and GMG – Garin Maiganga. The coal mines are all located in various sedimentary basins in Nigeria as illustrated in Figure 1 [21].

The coal samples were subsequently crushed into small particles using a dry miller. Next, the pulverized coal particles were sifted using an analytical sieve (Retsch™250 μm mesh) to obtain homogeneous particles for physicochemical characterization and thermal analysis.

Next, elemental coal analysis was carried out using an EL Vario MICRO Cube Elementar™ CHNS analyser in accordance with the specifications of the American Society for Testing and Materials (ASTM) D5291 standard. Proximate analysis was done using ASTM D3173, D3174 and D3175 standard techniques for moisture (M), volatile matter (VM) and ash (AC) content, respectively. The fixed carbon (FC) and Oxygen (O) content were determined by difference. The mineral matter was calculated using the *Parr formula* ($Mm = 1.08A + 0.55S$); where A and S represent the ash and sulphur content, respectively [21].

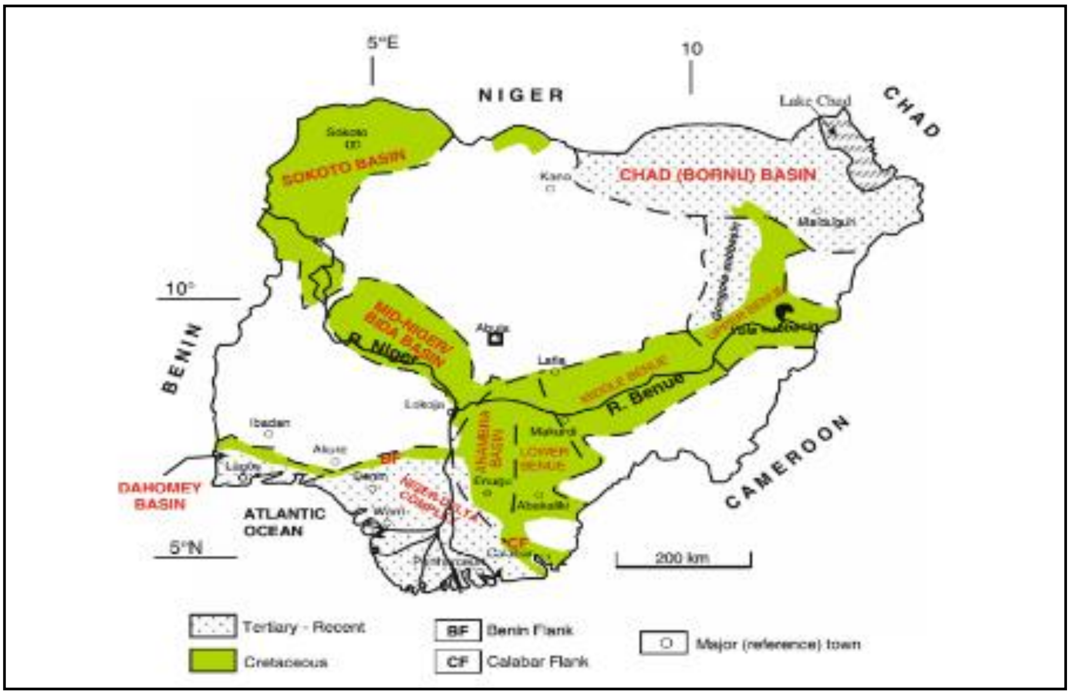

Fig.1. Sedimentary basins of Nigeria [20].

Table 1. Physicochemical Properties of Nigerian Coals.

Symbol	C	H	N	S	O	M	VM	A	FC	HHV	Mm
AFZ	72.46	6.07	1.63	1.41	18.43	1.97	45.80	30.99	21.24	30.52	34.24
GMG	61.96	4.42	1.07	0.39	32.16	5.28	51.16	21.05	22.52	23.74	22.95
SKJ	71.46	6.40	1.37	2.03	18.76	5.14	40.73	14.94	39.18	27.34	17.25

The higher heating value (HHV) of the coal samples was determined using bomb calorimetry (IKA C2000 Bomb Calorimeter). All the tests were repeated at least three times to ensure the reliability of the results. It is important to state that the coal samples were not subjected to any thermal or chemical pre-treatment prior to physicochemical and thermal characterization, consequently all the results are presented in *as received* (ar) basis. The rank and agglomeration classification of the coals were examined using the heating value (HHV) as described in the ASTM D388 standard for coal classification [22].

The thermal degradation behaviour of the coal samples was investigated using a Perkin Elmer STA 8000 Simultaneous Thermogravimetric (TG) analyser. About 12-16 mg of each sample was placed in an alumina pan and heated at 25°C/min from 50 to 1000°C to examine the thermal degradation in an inert atmosphere.

The evolved gases from the coals were purged using ultra-pure nitrogen gas at a flow rate 100 mL per min. The resulting thermograms were analysed using the Pyris Thermal Analysis Software (Version 11) to determine the temperature profile characteristics; peak decomposition temperature, T_p, residual mass R_m and total mass of the sample, T_m decomposed during thermal analysis.

RESULTS AND DISCUSSION

Physicochemical properties

The elemental composition, proximate analysis, mineral matter and heating value of the coal samples; Afuze (AFZ), Garin Maiganga (GMG) and Shankodi-Jangwa (SKJ) are presented in Table 1 in *as received* basis (ar) as also reported in the literature [3, 10].

The elemental composition presented in Table 1 indicates that the coal samples possess a high *C* and *H* content but relatively low *O* content. The highest carbon *C* content was observed in AFZ coal, followed by SKJ and GMG. The *H* content of the coal samples was in the range 4.42 to 6.40 % with the highest value observed in SKJ while the lowest value was observed in the GMG coal sample. The *O* content of the coal samples was in the range 18.43 to 32.16 % with GMG exhibiting the highest value. In general, the *C*, *H* and *O* content significantly influences the heating value, maturity and chemical reactivity of the coal [10, 21].

The *N* and *S* content, which serves as a measure of the environmental friendliness of coals, was observed to be low in all samples. The lowest *N* content was observed in GMG coal whereas the values for SKJ and AFZ were 1.37 wt.% and 1.63 wt.%, respectively. Conversely, the lowest *S* content was observed in GMG, whereas the values for SKJ and AFZ were 2.03 wt. % and 1.41 wt. %, respectively. The typical acceptable range for *N* and *S* in coals should not exceed 2.0 % [23] due to the risk of NO_x and SO_x pollutant emissions. Consequently, the high *S* content may hamper the potential application of the coals in steel manufacturing where the limits are 0.8 – 1.0 % [23]. Comparatively, GMG coal showed the lowest *N* and *S* content thereby posing the lowest risk for environmental pollutant emissions. Consequently, GMG may be suitable for cement and steel manufacture, power generation as well as industrial and domestic heating due to its favourable properties [10].

The highest HHV of 30.52 MJ/kg was observed for AFZ coal; whereas SKJ was 27.34 MJ/kg and GMG 23.74 MJ/kg. The observed results are in good agreement with the values for GMG and SKJ reported in the literature [10]. The high heating values of AFZ and SKJ can be attributed to the high *C*, *H* and low *O* content in the elemental composition of the coals. In contrast, the low HHV in GMG is primarily due to its high *O* and low *C* and *H* contents. In addition, with the HHV lower than 24 MJ/kg, the GMG coal can be categorized as low rank (LRC) whereas SKJ and AFZ coals are high rank coals (HRC) [21].

Proximate Analysis

The proximate analysis revealed AFZ coal has the lowest moisture, *M* content 1.97 wt.%, while SKJ was 5.14 wt.% and GMG 5.28 wt.%. Moisture content in coals is undesirable due to its chemical inactivity and tendency to absorb heat during combustion. This typically results in a low heating value and low thermal efficiency during conversion along with handling and transportation problems [23]. Furthermore, *M* and *VM* content is an index for evaluating the maturity, quality and potential application of coals. Hence, it is fundamentally established that coal rank or maturity increases with decrease in *M* and *VM* content [21, 23]. The highest *VM* content of 51.16 wt.% was observed for GMG, SKJ coal was 40.73 % and AFZ coal recorded as 45.80 %. Consequently, based on *M* and *VM*, the maturity of the coals increases in the order; GMG > SKJ > AFZ on the basis of *M* content; while the order is GMG > AFZ > SKJ based on the *VM* content.

The ash content typically determines the fouling tendency and slagging potential of coals during thermal conversion [21]. Furthermore, ash content can be used to determine the composition, volume and performance of blast furnace coke [10, 24]. The ash analysis results for the coals indicate that the highest ash content of 30.99 % was observed in AFZ coal; compared to 21.05 % for GMG and 14.94 % for SKJ coal. The mineral matter, *Mm*, of the coals also showed a similar trend as observed for ash content. Hence, it can be inferred that the fouling potential of the coals increases in the order; SKJ>GMG>AFZ.

The highest FC content was observed in SKJ coal followed by GMG and AFZ. Since, FC is the solid residue leftover after devolatilization and can be used to estimate the amount of coke obtained from coal carbonization [10, 21]. Therefore, SKJ has the highest coke potential among the coals investigated demonstrating it could be a good source of coking coal required for the manufacture of steel [10].

Coal Rank Classification

The agglomerating or non-agglomerating property of coal samples can be determined from *VM* and HHV, according to the ASTM D388-15 standard [22]. Based on this criteria, the HHV of non-agglomerating coals typically range from 14.7 to 26.7 MJ/kg, however agglomerating coals range from 26.7 to 32.4 MJ/kg [20]. Hence, SKJ with HHV (27.34 MJ/kg) and AFZ (30.52 MJ/kg) can be classified as agglomerating coals while GMG (23.74 MJ/kg) is non-agglomerating.

Furthermore, the coals can be classified into sub groups or ranks based on HHV. The GMG coal can thus be classified as *Sub-Bituminous B coal* with HHV typically from 22.1 to 24.4 MJ/kg [22, 23]. The SKJ coal can be classified as *High Volatile C Bituminous Coal* with an HHV typically from 26.7 to 30.2 MJ/kg [22, 23], which agrees with the Bituminous classification described in the literature by *Ryemshak* & *Jauro* [10]. Last, the AFZ coal (HHV = 30.52 MJ/kg) can be classified as *High Volatile B Bituminous Coal* with a typical HHV from 30.2 to 32.6 MJ/kg [22, 23] which distinctly contrasts with the classification widely reported in the literature [4, 8]. Hence, the results indicate that AFZ coal has been hitherto, incorrectly classified or ranked as a *Sub-Bituminous Coal* by the academics and researchers in Nigeria. Consequently, these findings provide not only reference material vital for future studies but also valuable data for the future utilization of AFZ coal in the Nigerian energy industry.

Thermogravimetric (TG) Analysis of Nigerian Coals

The thermogravimetric (TG) curves for the Nigerian coals investigated are presented in Fig. 2. The curves exhibit the slanted *Z*-type downward sloping curves typically observed for thermally degrading materials which demonstrate the effect of increasing temperature on the weight loss characteristics of the coals.

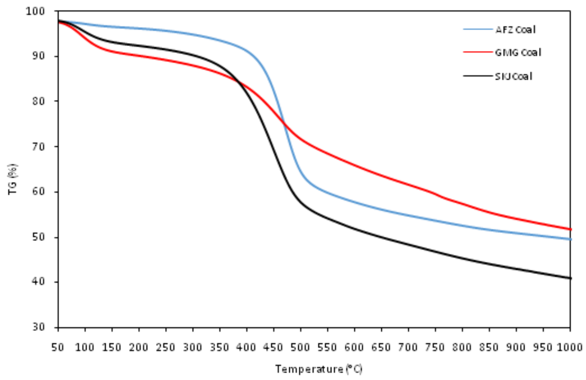

Fig.2. TG curves of Nigerian Coals.

From Fig. 2, it can be observed that the TG curves for AFZ and SKJ are similar in shape and orientation whereas the GMG coal is steeply sloped. This suggests that the reaction mechanisms for the conversion of AFZ and SKJ coal are similar as expected for coals of similar rank, but different from GMG. However, the analysis of the temperature profile characteristics revealed that the ignition temperature, T_i of SKJ coal, 375.74°C is considerably lower than for AFZ, 426.14°C and; GMG coal, 381.36°C. The results indicate that the SKJ is the most reactive while AFZ is the least reactive coal. The observed difference (50.40°C) in the reactivity of SKJ coal and AFZ coal may be due to the ash and mineral matter content of the coal samples. At 30.99 wt.% the AFZ coal has twice the ash content of the SKJ coal (14.94 wt.%). High ash content in coal is known to lower the calorific value, thermal efficiency and results in operational problems such as slagging and agglomeration [21, 23]. Consequently, the lower reactivity of the AFZ coal relative to SKJ may be due to the difference in ash content which affects the thermal degradation of the samples as observed during thermal analysis.

Derivative Thermogravimetric (DTG) Analysis of Nigerian Coals

The Derivative Thermogravimetric (DTG) curves of the Nigerian coals investigated are presented in Fig. 3. The curves are typically used to examine the devolatilization profiles of the potential solid fuels required for assessing their suitability and potential applications [25, 26]. In addition, the gasification and coking potential of the coals can be examined using DTG curves from the thermal analysis under inert atmosphere [27].

The DTG curves for AFZ and SKJ coals each revealed two peaks within the temperature range 50-200°C and 250-700°C. In contrast, the thermal degradation of the GMG coal revealed three peaks at 50-200°C, 250-700°C and 700-850°C.

The degradation of coals from 50-200°C can be attributed to drying as typically characterised by the loss of chemically bonded water molecules and mineral hydrates [3]. The DTG peaks observed during this stage (50-200°C) are significantly smaller and broader than the DTG peaks between 250-700°C. In particular, the results showed a good correlation between the moisture content and size of the DTG peaks.

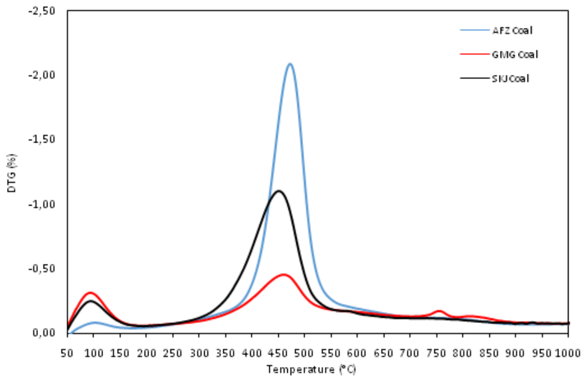

Fig.3. DTG curves of Nigerian Coals.

The weight loss observed from 250-700°C is primarily ascribed to the devolatilization of weakly bonded species into CO_2, CO, CH_4 and other non-condensable gases [27, 28]. The devolatilization of coal typically involves a significant loss of weight, therefore it is termed the active stage of pyrolytic decomposition. For the AFZ coal sample the weight loss observed was 41.58%, while GMG was 28.52% and SKJ 44.09 %. The results for the weight loss during devolatilization the coals confirms the higher reactivity of the SKJ coal compared to the AFZ and GMG coals. Table 2 presents the temperature profile characteristics of the coal samples in terms of the peak decomposition temperatures, T_p, Residual mass, R_m, and total mass D_m, of the sample decomposed during thermal analysis.

The peak decomposition temperature, T_p, is the most significant temperature profile characteristic deduced from DTG. It is used to examine the reactivity, thermal stability and temperature at which the maximum weight loss of the thermally decomposing material occurs. The T_p values for the coals increased in the order SKJ → GMG → AFZ. The low T_p value of SKJ indicates can be easily degraded compared to GMG and AFZ as presented by the results for the total mass decomposed D_m, during TGA. This trend is in good agreement with the results for other Nigerian coals reported in the literature [27].

In addition, the residual mass of the coal samples increased in the order SKJ→ AFZ → GMG. The increase in residual mass may be due to the reactivity of the coals based on the high ash and fixed carbon content [29]. However, the effect of ash can be remedied by pre-treatment with additives or co-firing with biomass [30] prior to future application. In general, based on the properties; GMG could be used in domestic heating and power generation while SKJ and AFZ coals for power generation, steel and cement production.

Table 2. Temperature profile characteristics of Nigerian Coals.

Coal Sample	Peak Decomposition (°C) T_p	Residual Mass (%) R_m	Mass Decomposed (%) D_m
AFZ	470.71	49.46	50.54
GMG	459.51	51.74	48.26
SKJ	450.29	40.94	59.06

CONCLUSION

The physicochemical properties and the thermal degradation behaviour of three newly discovered Nigerian coals were examined. The results indicate the coals contain high proportions of C, H, O and low N and S contents. The heating values confirmed the categorization of AFZ and SKJ as high rank coals (HRC) whereas GMG coal is low rank (LRC). Furthermore, the results revealed that AFZ coal is Bituminous NOT Sub-Bituminous as previously presented in the literature. The reactivity and maturity of the coals increased in the order: GMG > SKJ > AFZ. However, based on the devolatilization profile characteristics from thermal analysis, the reactivity of the coals increases in the order SKJ → GMG → AFZ. Additionally, the physicochemical and low rank properties; high moisture, volatile matter yet low Nitrogen and Sulphur content of GMG coal suggest it is suitable for clean coal applications in pyrolysis, gasification and power generation. The high fixed carbon content and thermochemical reactivity of SKJ indicate a good coking potential for application in steel and cement production. Last, the AFZ coal with its low moisture content and high carbon and heating value can be efficiently utilized in thermal conversion technologies for electric power generation.

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ФИЗИКОХИМИЧНО ОХАРАКТЕРИЗИРАНЕ И ТЕРМИЧЕН АНАЛИЗ НА НОВОТКРИТИ НИГЕРИЙСКИ ВЪГЛИЩА

Б. Б. Някума

Център за водородна енергетика, Институт по енергии на бъдещето, Технологичен университет в Малайзия, 81310 Скудай, Джохор Бахру, Малайзия

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(Резюме)

Изследвани са физико-химичните свойства и термичната деградация на каменни въглища от три ново-открити източника (Афузе, Гарин Маинганга и Шанкоди-Джангуа) от Нигерия. Охарактеризирането е проведено с елементен анализ, анализ на химическия състав и на горната калорична стойност, за да се установят степента, класификацията и качеството на въглищата. Анализът на химическия състав показва високо съдържание на летливи вещества (51.16 %) и влага (5.28 %). Най-високо съдържание на пепел се наблюдава в въглищата от Афузе, докато най-високо съдържание на свързан въглерод се забелязва в пробите от Шанкоди-Джангуа. Калоричната стойност (HHV) на различните въглища е: Гарин Маиганга (23.74 MJ/kg); Шанкоди-Джангуа (27.34 MJ/kg) и Афузе (30.52 MJ/kg), което потвърждава въглищата от Афузе и Шанкоди-Джангуа като висококачествени, а Гарин Маиганга – от по-ниско качество. Освен това, въглищата от Афузе са определени като битуминозни, но не суб-битуминозни, акто е било съобщено реди. Резултатите показват реактиво-способност и зрялост в следния възходящ ред: Гарин Майганга >Шанкоди Джангуа >Афуза. Общо взето, физикохимичните свойства и по-ниското качество на въглищата от Гарин Майганга ги правят подходящи за термоцентрали. Високото съдържание на свързан въглерод и термо-реактивоспособността на въглищата от Шанкоди Джангуа ги правят подходящи за коксуване при произвдството на стомана и в циментената промишленост. Накрая, ниското съдържание на влага, високо въглеродно съдържание и калоричност прави въглищата от Афузе подходящи за производство на енергия.