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AN EVALUATION OF MULTISTAGE CAUSTIC DE-ASHING OF A SUB-BITUMINOUS COAL

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Abstract

Fractions of Okaba coal OK±250 ground to pass the 250 μm sieve in three stages were subjected to proximate/ash composition analyses, hot aqueous leaching de-ashing with sodium carbonate and water (at atmospheric pressure) in multiple stages and in a Na_2CO_3-H_2O- H_2O sequence. The results obtained indicate that the ash contents percent of Okaba fractions OK-250(1), OK-250(2) and OK+250(2) were further reduced below the upper limit of 10 by 1.9, 2.2 and 1.8, respectively. These reductions translate to an average overall ash decrease percent of 12.57, while the volatile reduced by 9.85 and fixed carbon increased by 18.65. The ash content of the concentrates blend at 8% is lower than 10% required for coal blends for standard coking practice. The light transmission microscopy of the coal fractions showed that the chemical leaching process produced finer coal particles and caused a decrease in the ash content of the coal fractions. The reduction in ash content obtained at the atmospheric pressure treatment of Okaba coal will further enhance its participation in blend formulation for cokemaking.

Keywords: coal, ash; de-ashing; leaching; concentrate.

1. Introduction

Coal is a combustible sedimentary organic rock which is composed mainly of carbon, hydrogen and oxygen. It is formed from vegetation which has been consolidated between other rock strata and altered by the combined effects of pressure, temperature and bacteria over millions of years to form the coal seams. The temperature effect mainly operates after bacteria action has ceased while pressure increases with increasing depth of burial. Coal is a fossil fuel with reserves far larger than oil and gas and that is estimated to last 119 more years. The proven reserves of coal worldwide have been estimated at about 847 billion tons, with coal being available in almost every country of the world. However, recoverable reserves are only found in about seventy countries with the biggest deposits in the United States, Russia, China and India [1-2].

Nigeria is endowed with notable coal deposits at Ogboyoga, Okaba, Orukpa, Ezimo, Enugu and Lafia-Obi with total reportable and non-reportable reserves of 411, 635, 299, 349, 209 and 33 million metric tons, respectively. Lafia-Obi, the only medium coking of the six contains very high inherent ash and sulphur contents of 26% to 57.8% and 2.4%, respectively. In view of its medium coking nature, there is a need to reduce the high ash and sulphur contents of Lafia-Obi coal and thus enhance its maximum participation in blends for metallurgical coke-making [3, 4].

In view of the scarcity of coking coal worldwide and its high cost in the international market, methods to upgrade low grade coals by reducing their ash and sulphur contents have been developed. These methods can be classed as physical, chemical and biological and their successful application has led to the production of coal concentrates with low ash, low sulphur and improved thermoplastic properties [4]. The use of low ash/sulphur coke in the blast furnace at Ajaokuta will reduce coke consumption, improve productivity, reduce the cost of ladle metallurgy desulphurization, makes local steel production economical and lower the unit cost of steel to be produced.

Adeleke *et al.* [5], has reported the atmospheric leaching de-ashing of Lafia-Obi coal on a hotplate. In this work, the response of Okaba coal to hot leaching with sodium carbonate, a cheap base not commonly used as a leachant for coal was studied.

2. Materials and Methods

2.1 Materials

The collection and preparation of the samples were carried out as described in the following sub-sections.

2.1.1 Sample collection

The coal samples used for this study was obtained from Okaba in Ankpa Local Government Area of Kogi state. About 25 kg of Okaba bulk samples were obtained from the open pit mine opened by the Nigerian Mining Corporation (NMC) from seams number 2 and 3 at depths varying between about 10 and 15 m.

2.1.2 Sample preparation

The clean bulk samples of Okaba coal obtained from seams 2 and 3 were mixed in 3:2 proportions. The coal mixtures were then air dried in the laboratory overnight. About 4 kg of the air dried clean mixtures was thereafter crushed with Pascall Engineering 18862 crusher to obtain the experimental samples.

2.2 Methods

The pulverized sample was subjected to the tests described below.

2.2.1 Particle size analysis

The sample obtained from primary crushing was passed through a 250 μm sieve to obtain OK-250(1) sample constituting 43.27% of the starting sample. The oversize was further subjected to secondary crushing and passed through the 250 μm sieve aperture to obtain the OK-250(2) sample; while the oversize residue was labelled OK+250(2) to form 32.41% and 11.29% of the starting sample; respectively.

2.2.2 Proximate Analysis

The determination of moisture, volatile matter, ash and fixed carbon contents of the coal was carried out with modified forms of methods described in ASTM D 3177-89 and Francis and Peters [2].

2.2.3 Determination of coal ash Mean Basicity Index (MBI)

The determination of MBI was carried out as described by Adeleke *et al.* [5]

2.2.4 Aqueous leaching of coals

It has been shown that the pH of sodium carbonate aqueous solutions do not increase linearly with increasing molar concentrations [5]. About 3 g of the OK-250(1) sample mixed with sodium carbonate (in a 1:40 mass ratio of reagent to coal) to form 75 ml derivative 0.0095M aqueous solution in a 250 ml beaker was allowed to soak for about 15 minutes (that is, preliminary water soaking, PWS) to produce a 40 g/litre solid-liquid ratio slurry. The procedure was again repeated for OK-250(2) and OK+250(2) samples. The OK-250(1) coal slurry prepared with lid on was then heated only on the Stuart SM3 magnetic stirrer hot plate at a setting such that a temperature of about 100^{o}C was attained in 17 minutes. The heated coal slurry was removed from the hot plate after 20 minutes and allowed to cool for the same period. The cooled leached slurry was then filtered with a Dr Watt filter paper into a 250 ml conical flask. The coal was dried and weighed on the electronic balance. The procedure described was again repeated for the slurries of OK-250(2) and OK+250(2) samples. The whole procedure was further repeated but with initial solution homogenization (ISH) by stirring of the slurry for about 5 minutes. The second and third stages of the whole procedure was then carried out but with water only as leached coal cleaning steps to form the leaching sequence Na_2CO_3-H_2O- H_2O (SHH). The three stages procedure was repeated with initial solution homogenization and in both Na_2CO_3-H_2O- H_2O (SHH) and H_2O-Na_2CO_3-H_2O (HSH) sequences.

2.2.5 Micro-structural examination of the coal particles

The glass slide placed on the hot plate was smeared with a mixture of Araldite resin and hardener when it was moderately hot. The pulverized coal was then sprayed on the mixture. The excess coal particles were shaken off the slide and the stuck particles was lightly tapped with a spatula to drive off air bubbles from the particles. The slide laden with the coal particles was washed with water and allowed to dry in air overnight to ensure it permanently stuck to the slide. The coal laden face of the slide was ground on a motor driven grinding wheel until its thickness was sufficiently reduced. The coal surface was then polished with silicon carbide until it became transparent to transmit light. The polished surface was thereafter viewed under LEICA 02-520.703 DMLM/P BZ.00 light transmission microscope and the thin section photomicrographs obtained are presented in Plates 1 and 2.

3. Results and Discussion

3.1 Results

The results obtained on proximate analysis, ash composition test and two-stage leaching of Okaba coal are presented in Tables 1, 2 and 3, respectively; while Figures 1 and 2 show the effects of pulp preparation methods on the leaching of Okaba coal in three stages. The micrographs obtained for the light transmission microscopy of Okaba coal fractions are presented in Plates 1 and 2.

Table 1 Proximate analysis of Okaba coal as-received and as-leached

Parameters (%)	OK±250	Parameters (%)	OK±250
Volatile matter (R)	53.38	Ash(R)	9.15
Volatile matter (daf)	62.44	Ash (C)	8.00
Volatile matter (C)	48.12	**Ash (Reduction)**	**12.57**
Volatile matter (daf)	54.94	Fixed Carbon (R)	32.10
VM (Reduction)	**9.85**	Fixed Carbon (C)	39.46
		FC (Increase)	**18.65**

R -as-received coal, C - coal concentrate, daf - dried ash free, OK±250 -Okaba sample consisting of fractions OK-250(1), OK-250(2) and OK+300(2)

Table 2 Main ash composition of Okaba coal

Parameters (%)	Okaba coal	Parameters (%)	Okaba coal
Alumina(Al_2O_3)	10.09	Iron oxide (Fe_2O_3)	24.20
Silica (SiO_2)	21.68	Copper oxide (CuO)	0.32
Phosporous oxide(P_2O_5)	0.50	Zinc oxide (ZnO)	0.30
Sodium oxide (Na_2O)	0.13	Vanadium oxide (V_2O_5)	0.24
Potassium oxide (K_2O)	0.46	Magnesium oxide (MgO)	0.17
Calcium oxide (CaO)	3.10	Lime Ratio	0.143
Titanium oxide (TiO_2)	4.26	Acidity Ratio	1.16
Chromium oxide(Cr_2O_3)	0.11	Mean Basicity Index (MBI)	66.08
Manganese oxide (MnO)	0.52		

ad = as determined (or as received); daf = dried ash free, nd = not determined

Table 3 Effects of two-stage oven and hot plate (hp) leaching on Okaba (OK-250 μm)

1st Leaching	2nd Leaching	1st Weight loss, (%)	2nd Weight loss, (%)	Weight loss (tot)	Observed filtrate colour
H_2O(hp):Okaba	1:40 Na_2CO_3	15.60	7.20	22.80	Light yellow
1:40 Na_2CO_3	H_2O:Okaba	15.17	9.20	24.37	Light yellow

OK-250 μm = a fraction of Okaba coal sieved to pass 250 μm aperture

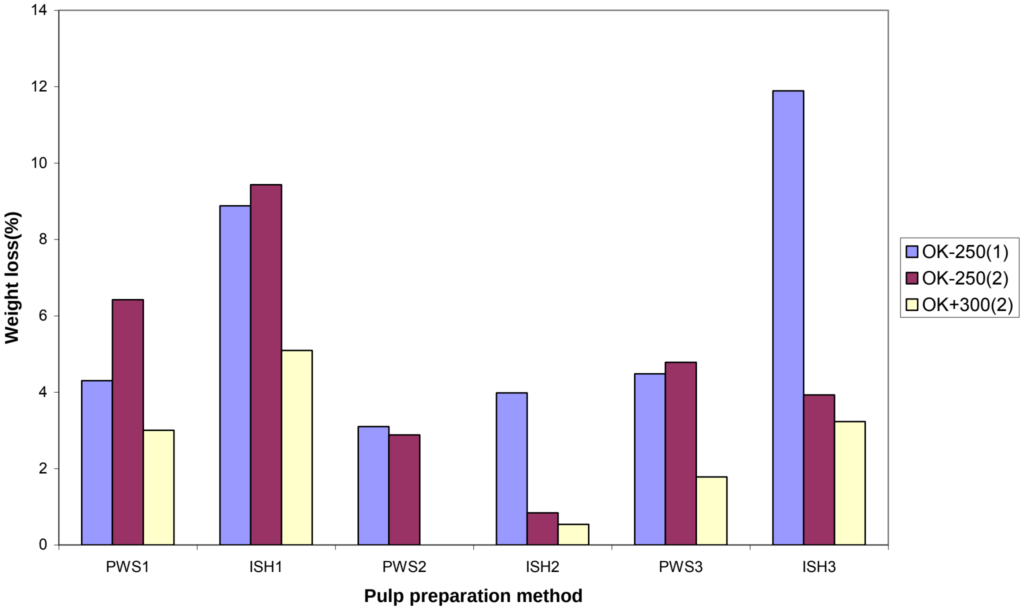

Fig. 1 Effects of pulp preparation methods on weight loss % in three-stage (SHH) hot plate leaching of Okaba coal fractions, DS, 40g/L

Legend: OK-250(1), OK-250(2), OK+250(2) - coal fractions of Okaba; PWS1, PWS2, PWS3 - preliminary water soaking for stages 1, 2 and 3, resp.; ISH1, ISH2, ISH3 -initial solution homogenization for stages 1, 2 and 3, respectively

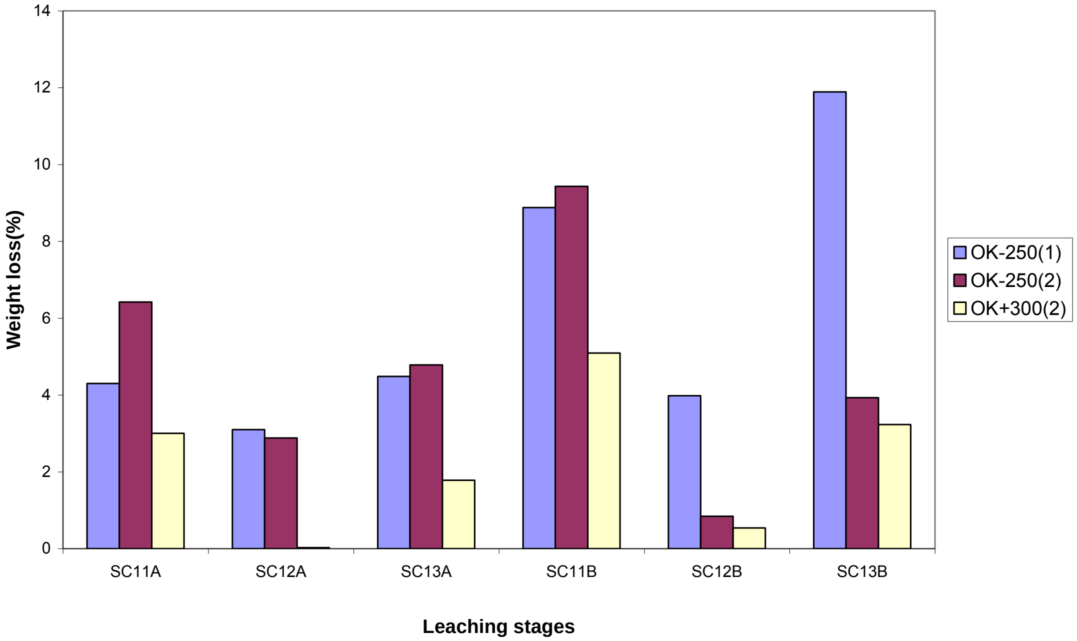

Fig. 2 Effects of three-stage (SHH) PWS(A) and ISH(B) leaching of Okaba coal fractions, DS, 40g/L with 1:40 Na_2CO_3:OK

Legend:

SC11 = 1:40 Na_2CO_3: Okaba 1st stage PWS leaching treatment
SC12 = 1:40 Na_2CO_3: Okaba 2nd PWS leaching treatment
SC13 = 1:40 Na_2CO_3: Okaba 3rd stage PWS leaching treatment
SC21 = 1:40 Na_2CO_3: Okaba 1st stage ISH leaching treatment
SC22 = 1:20 Na_2CO_3: Okaba 2nd ISH leaching treatment
SC23 = 1:20 Na_2CO_3: Okaba: L 3rd ISH stage leaching treatment
SHH = Na_2CO_3-H_2O- H_2O leaching sequence for Okaba coal
DS = oven dried sample; OK-250(1), OK-250(2) and OK+250(2) - fractions of Okaba coal that comprise; OK±250 sample.

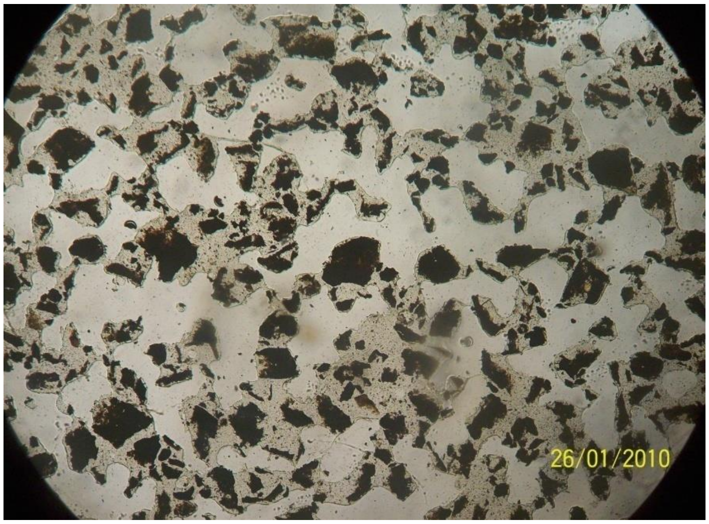

Plate 1a Okaba coal–250 (1), DS (X 100)

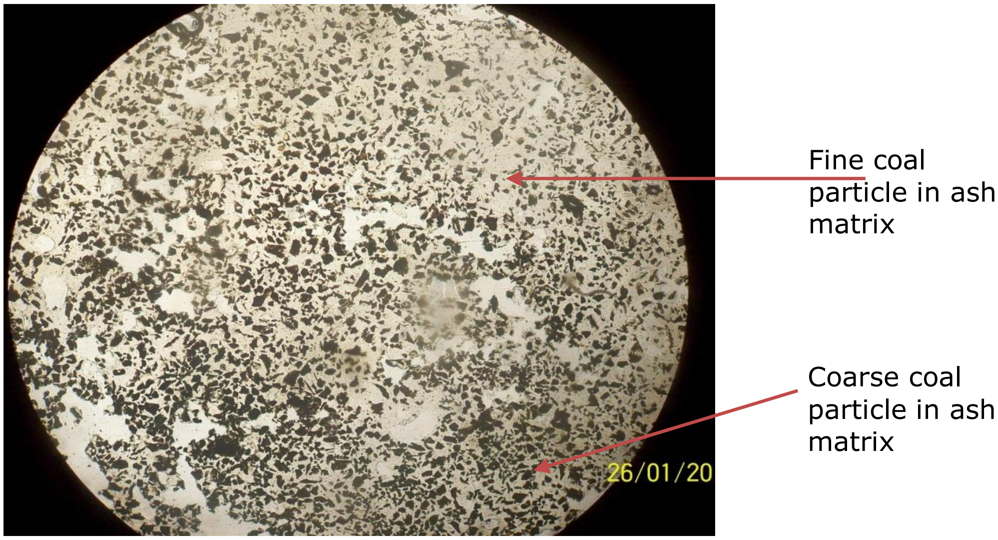

Plate 1(b) Okaba coal – 250 (1), Concentrates (X 50)

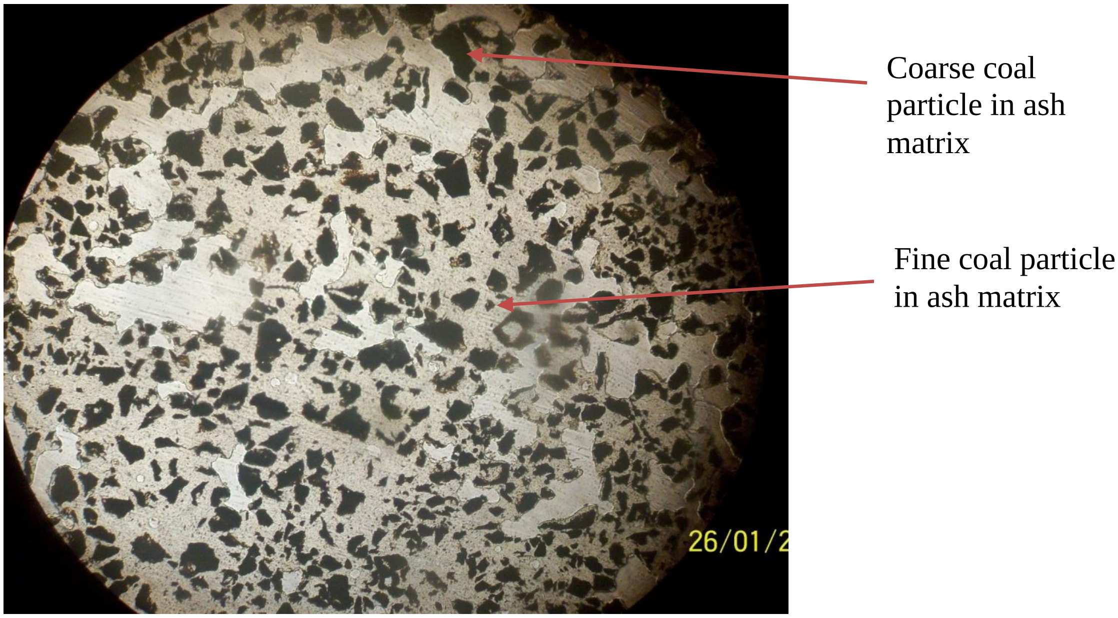

Plate 2(a) Okaba coal–250 (2), DS (X 50)

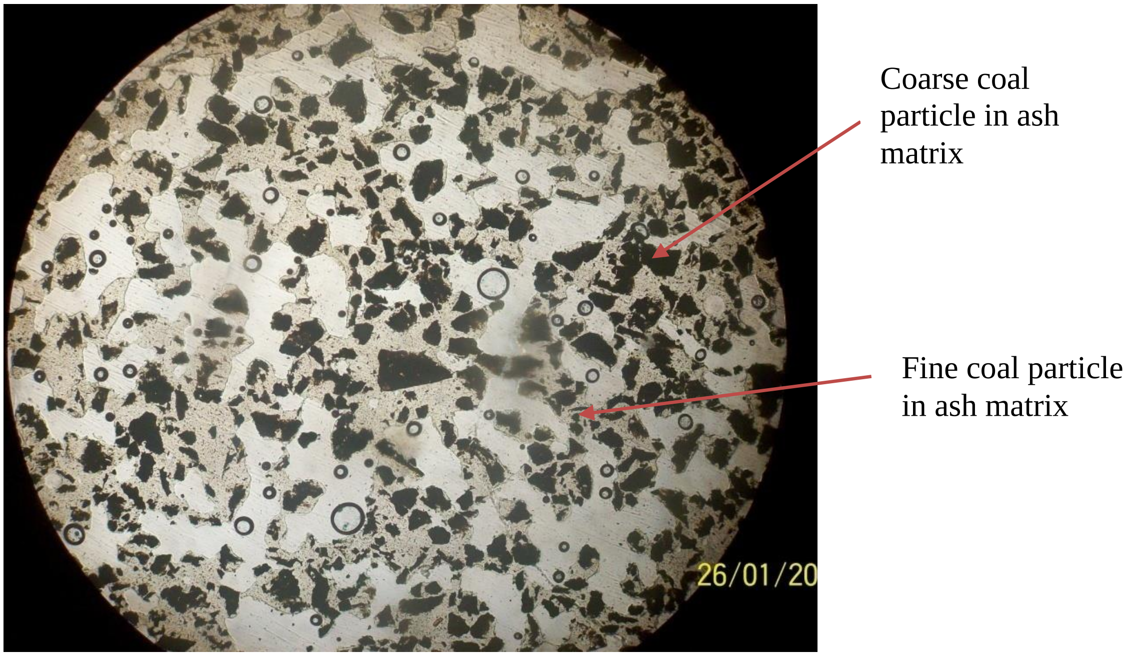

Plate 2(b) Okaba coal -250(2), concentrates (X50)

3.2 Discussion of results

From the results of the ash composition analysis, the acidity ratio calculated for Okaba coal is 1.16, while the total percentage of acidic oxides; that is, silica, alumina and titanium oxides was 36.03%. The high concentrations of silica and alumina cause the formation of highly viscous slag during ironmaking and the contents of both oxides are normally required to be low in both iron ore concentrates and by extension in other charge inputs such as coal/coke. For example, in direct reduction ironmaking, the contents of silica, alumina and titanium oxide in iron ore charge are required to be lower than 3.0% [6]. Considering the high contents of acidic oxides in Okaba coal, there is a need to reduce the contents of these deleterious oxides in order to minimize their adverse effects in ironmaking. Lower contents of the acidic oxides in the selected coals will also decrease their proportions in cokeable coal blends (including the coals) for metallurgical coke production.

The results for two stages leaching of Okaba coal fractions showed that initial leaching of Okaba coal with sodium carbonate followed with the second stage water cleaning produced the highest weight loss %. Also, the two stages leaching produced far greater total weight losses than single stage leaching without additional expense on reagents in the water leaching phase. The more intense yellowish filtrate obtained on the second water cleaning stage of Okaba showed that much of the leached sulphur still remained entrapped in the coal residue after the initial leaching coupled with hot water filtration. The second cleaning stage was thus necessary. Furthermore, the three-stages ISH hotplate leaching of OK-250(1) and OK-250(2) with 1:40: Na_2CO_3:Okaba as the first stage, followed by two stages of agitation water leaching gave very good leach weight losses of 24.75% and 14.20% (as against 8.88% and 5.09% for the first stages), respectively.

Figures 1 and 2 show the effects of coal pulp preparation methods on the weight loss percent for both Okaba coal fractions. The results obtained showed that for the three stages of Na_2CO_3-H_2O-H_2O (SHH) treatment sequence for Okaba fractions, the total weight loss percents for the coal fractions were higher for initial solution homogenization (ISH) pre-treatments than preliminary water soaking (PWS). The better leaching effect obtained with ISH treated samples may be due to the intimate contact between the leachant and the coal particles due to the homogenization treatment. The results obtained therefore indicate ISH treatment as the preliminary pulp preparation method for the efficient leaching of the two coals. From the results obtained, it can be deduced that multistage leaching in the correct sequence was required for the efficient demineralization of the selected coals and that leaching sequences of Na_2CO_3-H_2O-H_2O (S-H-H) gave the best leaching of Okaba coal.

The better leaching obtained for Okaba coal with the S-H-H leaching sequence suggests that the lower rank coals like Okaba with higher equilibrium moisture retaining capacity may be leached with reagent directly without prior water treatment. The leaching sequence also suggest that Okaba coal with two consecutive water cleaning stages required more intense water washing to strip it of the dissolved in-organics. The leaching of the Italian Sulcis coal was conducted in two stages of potassium hydroxide and hydrogen peroxide leaching, in that order [7].

Ash analysis for Okaba concentrates at 1:40: Na_2CO_3Okaba for OK-250(1) (DS, ISH), OK-250(2) (DS, ISH) and OK+300(2) (DS, ISH) gave ash % contents of 8.10, 7.80 and 8.20 (which translates to ash reduction % of 14.74, 4.55 and 6.82), respectively. The results indicate that the ash contents of Okaba fractions (OK-250(1), OK-250(2) and OK+250(2)) were further reduced below the upper limit of 10% ash specified for prime grade coal (by 1.9%, 2.2% and 1.8%, respectively [8- 9]. Higher concentrations of sodium concentrate produced only marginal increase in weight loss % and at much higher concentration an increase in ash content.

A reduction in the ash contents of coals for cokemaking is critical as high ash in the blast furnace requires higher energy consumption for slagging and lowers the productivity of the blast furnace. High contents of alkali oxides causes serious incidents like frozen hearth and burnt tuyere [8, 10-11]. Reduction in coke ash content decreases the mean basicity index (MBI) of coke and this may cause a decrease in coke strength after reaction (CSR); an important parameter for coke of metallurgical grade. Volatile matter analysis for Okaba concentrates OK-250(1), OK-250(2) and OK+250(2) yielded volatile contents average % (daf) of 48.12% which translates to an average volatile reduction of 9.85%. The results obtained showed that the leaching of Okaba coal with sodium carbonate caused a reduction in volatile matter. Leaching tests conducted on Okaba samples OK-250(1), OK-250(2) and OK250 (2) with 1:40 Na_2CO_3: Okaba showed that the fixed carbon increased from an average of 32.10% to 39.46% which translates to an overall increase of 18.65%.

From the micrographs of OK-250(1) DS, sparsely distributed coarse dark particles of coal in a large grayish matrix of ash/sulphur bearing inorganics can be seen, while the micrographs of the concentrate showed dense fine coal particles (with occasional large dark patches) in relatively small grayish background. These results suggest that the chemical leaching process produced finer coal particles and caused a decrease in the ash content of the OK-250(1) DS sample. Similarly, the micrographs of OK-250(2) DS, indicate sparsely distributed coarse dark particles of coal in a large grayish matrix of ash/sulphur bearing inorganic, while the concentrates show smaller particles with less sparse distribution. The results indicate that chemical leaching of OK-250(2) DS have caused less grain refinement and ash reduction (Plates 1 to 3).

4. Conclusions

The ash contents of the fractions of Okaba coal fractions OK±250 were successfully reduced by hot aqueous state leaching with sodium carbonate that is not commonly used as a leaching agent. The average ash reduction of 12.57% obtained is significant because of the low molar concentration of the reagent at 1:40 mass ratio of reagent to coal at atmospheric leaching of the low ash coal. The marginal reduction in the ash content of the low ash coal will further enhance its use as a blend component in cokemaking. The light transmission microscopy of the coal fractions showed that the chemical leaching process produced finer coal particles and caused a decrease in the ash content of the coal fractions.

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