Effects of Caustic Leaching Demineralization on the Crucible Swelling Properties of Lafia-Obi Coal, Nigeria

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ABSTRACT

Fractions of the high ash Nigerian Lafia-Obi coal L±250 and L±300 ground to pass the 250 and 300 µm sieves, respectively, in three stages were subjected to hot aqueous leaching de-mineralization with water and sodium carbonate in multiple stages and in a H_2O-Na_2CO_3-H_2O sequence. The samples, as-received and as-leached, were also analyzed for volatile matter and crucible swelling properties. The results obtained showed that ash, sulphur, and volatile contents were reduced by 38.86, 60.16, 15.73 and 24.52, 27.34, 8.56 percents in L±250 and L±300 samples, respectively. The crucible swelling indices and percent lump of semi-coke buttons of 1.5, 86.70 and 1.67, 89.17 were also obtained for Lafia-Obi ±250 and ±300 concentrates, respectively. These results translate to a high coke stability increase of about 13.04% in Lafia-Obi ±250 and an average caking index and semi-coke lump stability increases of 11.33% and 3.16% for Lafia-Obi±300, respectively.

(Key words: coal, high ash, demineralization, swelling, coke stability)

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 (World Coal Institute, 2010; Francis and Peters, 1980).

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 cokemaking (Federal Ministry of Mines and Steel Development, 2005; Afonja, 1976; 1977).

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 (Afonja, 1976; 1977). 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.

The chemical leaching methods include molten caustic leaching and agitation caustic leaching and have been successfully employed to reduce the ash and sulphur contents of coals (Yang *et al*, 1985; Hairprasad, 2009).

Chriswell *et al.* (1991) reported the use of the molten caustic leaching (MCL) method for the removal of over 90% of the sulphur and ash in coal. The molten caustic leaching method is an advanced chemical technique that involves the water washing of caustic leached coals to remove un-reacted caustic and impurities produced by reactions of oxides with the molten caustic. A counter current procedure designed for efficient washing with minimal water loss was used. The main problem encountered was the formation of massive precipitates of carbonates and this led to far lower than expected recoveries of coal concentrates.

The aim of this study is to examine the improvements in the crucible swelling numbers of Lafia-Obi coal fractions following hot caustic leaching de-mineralization.

MATERIALS AND METHODS

Materials

The Lafia-Obi sample was obtained from a virgin site of Lafia-Obi coal-field on the bank of Kogi River, in Shankodi Jangwa district of Awe Local Government Area of Nasarawa State. The Jangwa district is about 70 km East of Lafia, which is on Latitude 8°30' North and Longitude 8° 34' East. The coal site is about 81 km on the left side off Lafia-Obi highway. About 10 kg of the bulk samples of Lafia-Obi were removed from the top seams intersected at a depth of about 1 m from the ground surface in open pit mines A and B.

Similarly, the clean bulk samples of Lafia-Obi samples A and B were mixed in 1:1 proportions. The coal mixtures were then air dried in the laboratory overnight. About 4 kg of the air dried clean mixture of Lafia-Obi was thereafter crushed with Pascall Engineering 18862 primary crusher to obtain the experimental samples.

Methods

Determination of Free Moisture Content

About 1 g of the prepared coal sample was weighed on Mettler Toledo PB 153 electronic balance and placed on a clean paper surface in the laboratory for about 48 hours. The weight of the air dried sample was taken and the moisture content determined using Equation 1 (Francis and Peters, 1980).

$$\% Moisture = \frac{W_i - W_f}{W_i} 100 \qquad \textbf{(1)}$$

where,

W_i = Initial weight of sample

W_f = Final weight of sample after the moisture test

Determination of Equilibrium Moisture Content

About 1 g of the prepared coal sample was also taken, placed in a silica crucible and heated to 110^{o}C in Gallekamp 7B 16590 oven. The crucible and its content were then held at this temperature for 1 hour. The weight of the heated sample was taken and the moisture content determined using Equation 1.

Aqueous Leaching of Coals

The oven dried coal sample was crushed and ground to three size fractions labeled L-250(0), L-250(0)' and L+250(0)'' that are products of primary grinding, secondary grinding (passing 250 μm sieve) and residue after secondary grinding and constituting 43.06, 49.32, and 7.62%, respectively. Similarly, another batch of oven dried coal sample was crushed and ground to three size fractions labeled L-300(1), L-300(3), and L+300(3) that are products of primary grinding, secondary grinding (passing 300 μm sieve) and residue after secondary grinding and constituting 42.41, 26.14 and 31.45, respectively.

Mixture of L-250(0) size fraction of Lafia-Obi coal and sodium carbonate (in a 1:20 mass ratio of reagent to coal making a derivative 0.019M aqueous solution) was homogenized for about 5 minutes in a 75 ml aqueous solution (in a 250 ml beaker) to form the reaction aqueous mixture. The latter procedure was also repeated for the L±300 samples but in a 0.094M aqueous solution.

The initially homogenized coal slurry bearing solution with a lid was then heated only on the

Stuart SM3 magnetic stirrer hot plate such that a temperature of 90°C was attained in 25 minutes. The 40 g/litre L-250(0) coal slurry hot leached was then allowed to cool off the hot plate for the same 25 minutes. The cooled leached slurry was afterward filtered with a Dr. Watt filter paper into a 250 ml conical flask. The coal concentrate was dried and weighed on the electronic balance. The procedure described was again repeated for the slurries of L-250(0)', L+250(0), L-300(1), L-300(3)', and L+300(3) samples.

Multiple Stages Leaching

The procedure was further carried out in three stages for L-250(0) sample but with first stage water leaching, second stage leaching with sodium carbonate and third stage water leaching (that is, H_2O-Na_2CO_3-H_2O (HSH) leaching sequence). The three stage leaching was again repeated first stage leaching with sodium carbonate, second and third stages water leaching (that is, Na_2CO_3-H_2O-H_2O (SHH) leaching sequence). The procedure described was again repeated for the slurries of L-250(0)', L+250(0), L-300(1), L-300(3)', and L+300(3) samples (Adeleke *et al.*, 2011).

Crucible Swelling Index

The Vecstar LF3 muffle furnace was heated from ambient temperature to 900°C. From the prepared coal sample, about 1 g sample was similarly measured and put in a silica crucible. The sample in the crucible was soaked with several drops of benzene. The crucible and its content with the lid on were thereafter placed in the heated muffle furnace for 7 minutes. Equation 2 was then used to calculate the volatile matter of the coal sample.

$$\%\,Volatile = \frac{W_i - W_f}{W_i} x100 \qquad \textbf{(2)}$$

where,

W_i = Initial weight of sample

W_f = Final weight of sample after the volatile test

The coke buttons obtained from the volatile matter tests were compared with standard profiles on the free swelling index chart and the appropriate swelling number determined between 0 and 9 in accordance with International Standard Organization (ISO) standard profiles (Francis and Peters, 1990). The weight of the cake produced and its chips were also determined.

RESULTS

The average results obtained for moisture, volatile, ash and sulphur contents for Lafia-Obi±250 Lafia-Obi±300 fractions are presented in Table 1, while Table 2 shows the crucible swelling numbers obtained on the Lafia-Obi fractions as-received and as-leached.

Table 1: Proximate Analysis, Reduction in Ash, Volatile and Sulphur.

Parameters (%)	L±250	L±300
Moisture (R)	3.49	3.49
Moisture (C)	3.15	3.15
Moisture (Reduction)	**9.74**	**9.74**
Volatile matter (R)	26.99	26.99
Volatile matter (daf)	42.20	42.20
Volatile matter (C)	27.36	20.73
Volatile matter (daf)	35.56	37.85
VM (Reduction)	**15.73**	**8.56**
Ash(R)	32.55	32.55
Ash (C)	19.90	24.57
Ash (Reduction)	**38.86**	**24.52**
Sulphur (R)	1.28	1.28
Sulphur (C)	0.51	0.93
Sulphur (Reduction)	**60.16**	**27.34**

LEGEND: R= coal as-received, C= coal as-leached

Table 2: Free Swelling Indices of As-Received (DS) Coal/Concentrate(ISH)

Lafia-Obi coal	FSI No.	Weight of cake lump (%)	Weight of cake chips (%)	Remarks
L-250(0) DS	1.5	76.67	23.34	Caking
L-250(0)'	2	88.45	11.55	Caking
L+250(0)	1	94.65	5.35	Caking
Average Values	1.5	83.85	16.15	
L-250(0) C, ISH	2.5	92.84	7.16	Good Caking
L-250(0)'	0	0	100	Not Caking
L+250(0)	2	86.70	13.30	Caking
Average Values	1.5	46.58	53.42	
L-300(1) DS	1.5	86.44	13.56	Caking
L-300(3)	1	90.67	9.33	Caking
L+300(3)	1	88.19	11.81	Caking
Average Values	1.17	88.10	11.90	
L-300(1) ISH	2.5	93.05	6.95	Good Caking
L-300(3)	1.5	82.06	17.94	Caking
L+300(3)	1	89.84	10.16	Caking
Average Values	1.67	89.17	10.83	

DS: oven dried sample, ISH=initial solution homogenization

DISCUSSION

The results obtained showed that the moisture contents for the coal concentrates are lower than for the as-received Lafia-Obi coals. It has been reported that a decrease in moisture content is an indication of an increase in coal rank (CBM, 2010).

The results obtained for volatile matter shows that Lafia-Obi L±250 sample fractions gave higher reduction in volatile of 15.73%. The good volatile reduction obtained for the L±250 sample may be due to its finer size consists in comparison to the coarser L±300 sample. Volatile matter is an important parameter in the classification of coals and the volatile matter of the three coal concentrates exceed the upper limit of 30.30% for coals for cokemaking at Ajaokuta (Francis and Peters, 1990; Raw Materials Specification, 1994). The results obtained thus suggest that blend formulation of the coal concentrates with lower volatile prime coking coals will be required in using them for cokemaking.

The results obtained for the ash contents showed that Lafia-Obi L±250 sample gave far lower ash content than the L±300 sample and also a very high ash reduction % of 38.86. The high ash reduction obtained for Lafia-Obi L±250 sample might have been due to its smaller size consists that must have enhanced the leaching reactions (Umland and Bellama, 1999). The results obtained also showed that sulphur content was reduced and fixed carbon contents of the coals increased. The highest reduction in sulphur of 60.16% was obtained for Lafia-Obi L±250 sample, while the largest increase in fixed carbon of 25.44% was determined for L±250 sample. The trends in the results obtained is similar to those reported by Fois *et al.* (2010) and Kumar and Gupta (1997).

The results obtained showed that the crucible swelling tests on L-250(0), L-250(0)', and L+250(0) for as-received and leached fractions was 1.5. The highest swelling number of 2.5 was obtained for the most stable cake from L-250(0) concentrates with 92.84% cake lump. These results suggest that the L-250(0) fraction that constitutes about 43.06% of the L±250 coal is the most caking of all the coal fractions tested. However, for bulk samples, the L±300 sample concentrate showed the highest average FSI of 1.67 which exceed that of L±250 by 0.17 units.

The crucible swelling number of 1.5 for the as-received L±300 coal is 1.33 units below the lower limit for a typical bituminous grade coal, while its concentrate crucible swelling number of 1.67 is only 0.83 units lower (Panaitescu, 1994).

The average weight of lumps and crucible swelling numbers from the cakes formed from the as-received and leached L±300 samples were determined as 88.10%, 89.17%, and 1.17, 1.67, respectively. These results showed that the average FSI number and stability of the cake lump from the L±300 fractions increased by

1.07% and 42.74%, respectively. The results suggest that the leaching conducted increased the stability of the semi-coke obtained from the leached L±300 fractions appreciably. Rubiera *et al.* (2002) reported the increase in the free swelling index of a British coal subjected to chemical treatment in mixture of hydrofluoric and fluorisilicic acids.

The average caking index of L±300 coal increased from 1.5 to 1.67 on the international standard profile of crucible swelling index for coals. The increase in cakeability obtained must be due to reduction in ash contents that might have improved the potency of the coal inherent vitrinite. Decrease in ash contents has been reported to cause an increase in caking indices of Indian high ash coals. The reduction of the ash content of the Indian Bhojudih coal from 21% to 15% caused an increase in FSI from 2.5 to 3.33 (Parthasarathy *et al.*, 1992). The free swelling index of 1.67 determined for L±300 is however lower than 6.5 determined for the Australian prime coking Bulli coal and typical French coking coals. It is also lower than the lower limit of 4 for good coking coals in ISO chart (Podolak, 1987; Panaitescu, 1990; BHP, 1998). The results obtained showed that blending of the local concentrates with a highly caking coal is necessary to obtain cokeable blends.

CONCLUSIONS

Fractions of the high ash Nigerian Lafia-Obi coal L±250 and L±300 were appreciably de-mineralized with water and sodium carbonate in multiple stages and in a H_2O-Na_2CO_3-H_2O sequence and also analyzed for volatile matter and crucible swelling properties. The results obtained showed that ash, sulphur, volatile contents were reduced by about 38.86, 60.16, 15.73, and 24.52, 27.34, 8.56 percents for L±250 and L±300 samples, respectively; while coke stability increase of about 13.04% and 11.33% were obtained in Lafia-Obi ±250 and Lafia-Obi±300, respectively.

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