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CHARACTERIZATION AND ASH CHEMISTRY OF SELECTED NIGERIAN COALS FOR SOLID FUEL COMBUSTION

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Abstract

In this paper, characterization and ash chemistry of selected Nigeria coal samples were investigated for it potential as a solid fuel combustion. The three coals considered originated from Northern Benue trough, Central Benue trough and Anambra Basin of Nigeria where proven reserve deposits are found. The coal samples were analysed using various analytical methods such as thermogravimetric analysis (TGA) and X-ray Fluorescence spectroscopy (XRF) analyses. The TGA profile suggests that Enugu coal showed high thermal stability than that of Okaba and Maiganga coals but had an ash content higher by a factor of 2.0 when compared with Okaba and Maiganga coals. Maiganga coal however has the highest heating value of 27.40 MJ/kg when compared to Okaba coal (25.74 MJ/kg) and Enugu coal (22.92 MJ/kg). The ash chemistry indices used in predicting the performance of the coal samples includes basic to acidic oxide (B/A), silica ratio, iron index and sulphur slagging index (Rs). While silica ratio indicated that Enugu coal has high slag volume, the B/A suggest that all the coals were found to be less than 0.6 indicating low slagging potential. The ash chemistry suggests that Maiganga coal has lowest slagging potential while Enugu coal has severe slagging potential.

Keywords: Coal; fuel; thermogravimetric; combustion; ash chemistry; slagging.

1. Introduction

Coal is supposed to be the dominant source of power generation all over the world and should remain relevant even in the near future. Nigerian current power generation of about 5000 MW of electricity is grossly inadequate. This situation has adversely affected the electricity supply to residential and commercial consumers. Electricity is a solid based on which social and economic life strives. It cannot be stored and as such it must be consumed as soon as it is produced. With the population of approximate 170 million people, only about 40% of Nigerians are connected to the national grid, despite the abundant reserve of over 2.6 billion tonnes predominantly sub-bituminous coal type [1].

Nigeria is endowed with a large coal deposits most of which are reported to be within the Anambra Basin and Benue trough [2-3]. Anambra Basin is the largest coal producing basin where intensive exploration and exploitation activities have been reported while new discovered coal deposit exist. Coals are of divers in nature, and the individual characterizations are very important in deciding their suitable applications [4-5].The contemporary global concern for a clean source of energy has rekindle interest in coal. Large coal deposits which have been abandoned due to the availability of petroleum are now being investigated for their clean coal technology [6].

There is an obvious undeniable need to utilize and improve on the available coal resources in Nigeria. Coal power plants could give a high energy capability and pose little or minimal threat to life, property and environment if properly used. Most of the existing research work on coal focus on the combustion [7], liquefaction [8] and coke making [9], and they have found varying physical, chemical and thermal characteristics for coal mined at different geographical locations in Nigeria. Most Nigerian coal have been reported to be non-coking except for Obi-Lafia coal deposit that has been described as coking coal [10]. Obaje *et al.* [11] have worked on the suitability of Nigerian coals for liquefaction. The combustion properties of Nigeria coals have been limited to the determination of proximate, ultimate and calorific values [12]. The combustibility, calorific values and ash chemistry are genetically linked to one another and they all depend on the coal rank and maceral composition.

Over the years, ash chemistry has been developed to predict the performance of coals prior to actual solid fuel combustion. The majority of ash chemistries relate the chemical composition of the ash with the slagging tendency, for example, the ratio of the basic metal oxides to the acidic metal oxides (B/A). This index is important for the melting behaviour of coal ash systems. Bad coals are said to have a low silicon oxide level of equal and less than 87% or a high level of iron oxide, above 6% [13-14]. B/A is also useful in distinguishing between good and bad coals prior to actual combustion. Lawrence *et al*. [14] reported that a decrease in the B/A of coal raise its fusibility and hence decreases it is slagging potential and reported that good coals have B/A ≤ 0.11. An empirical slagging index which referred to as the product of B/A and the dry sulphur content in coal was developed and tested on United State coals because of the presence of iron sulphide (FeS_2) as pyrite in the coal samples. But in low sulphur coals, where there is strong correlation between iron and carbonate ($FeCO_3$) as in the case of Australian coals, slagging index is better defined as the product of B/A and Fe_2O_3. According to McLennan *et al.* [13], if the sulphur slagging index (Rs) of a coal, assessed as the product of B/A and dry S is greater than 0.6, the coal is said to have high slagging potential. Other slagging indices include the silica ratio and the iron oxide. However, these indices have been established based on particular coal properties, and therefore, can be used to assess Nigerian coals.

There are only a few experimental investigations on the combustion profile of Nigerian coals, for example. Fatoye *et al*. [12] and their slagging potentials are even rarer. An in-depth understanding of the characterization and ash chemistry of the Nigerian coals is necessary in order to assess high combustion efficiency. In this paper, characterization and ash chemistry of selected Nigerian coals were carried out. The selection of coal sample was done in view of disparity of coal from different geographical locations as coal formation largely depends on certain factors such as; constituent's formation, compression temperature and pressure which plays a major role in coal formation. Hence three different locations were selected to study and research on their inherent disparity and look into how their thermal and chemical properties differ. The three coals considered originated from Northern Benue trough, Central Benue trough and Anambra Basin of Nigeria where there are proven reserves of coal deposits.

2. Materials and method

Choice of Coal Samples and preparation: The coal samples used sourced from Maiganga (Northern Benue trough), Okaba (Central Benue trough) and Enugu (Anambra Basin) resp. All samples were initially crushed and pulverized. The coal samples were sieved to a particle size of 150 µm in preparation for further analysis. These experimental methods were carried out explicitly to investigate and determine certain thermal, chemical and ash characteristic of the coal samples.

2.1. Proximate and ultimate analysis

The moisture, volatile matter and ash contents were determined based on American Society for Testing and Materials D3172-13. The fixed carbon (dry basis) in the coal samples was obtained by subtracting the percentages of moisture, volatile matter and ash from 100. The ultimate analysis (sulphur, nitrogen, carbon and hydrogen) were also based on D3176-15. The results obtained from the ultimate analysis were used explicitly in determining the calorific values of each coal sample. High heating value (HHV) of coal was calculated using the formula below by [15].

HHV = 0.3491C + 1.1783H + 0.1005S – 0.1034O – 0.0151N – 0.0211A (MJ/Kg) (1)

where; C = carbon, H = hydrogen, S = sulphur, O = oxygen N = nitrogen and A = ash

2.2. Thermal and chemical characterization

Thermal Characterization: The samples were analyzed in purely nitrogen environment having a flow rate of 20 ml/min, with a pressure of about 2.5 bars and a constant heating rate controlled at 10°C/min for a wide temperature range from 23°C to 950°C. The sample was loaded into the equipment with the use of Pyris manager software and the analysis was initiated and performed after a constant weight of sample was observed using the created heating profile.

Chemical Characterization: The ash compositions of the samples were carried out using X-ray fluorescence (XRF). The sample was placed in a sample holder contained in the XRF equipment and oriented to an angle of 45°. The X-ray machine was closed and the shutter was opened to open the window of the X-ray tube. The filament voltage was then set progressively to 40 kV and the current to 20 mA. A silicon drift detector was used to detect the secondary X-rays (X-ray detector) and to acquire and record the spectrum (acquisition and processing system). The elements were displayed in their oxide forms for each sample.

2.3. Ash chemistry prediction

Following ASTM D3174-12, the samples were heated at 800°C for 5 hours and cooled. The oxide composition of the ash samples were analyzed by XRF and were then used as input into silica ratio, basic to acidic oxide [13-14], iron percentage of iron index [14] and sulphur index in Equations (2), (3), (4) and (5) respectively.

$$S_R = 100\left(\frac{SiO_2}{SiO_2 + Fe_2O_3 + CaO + MgO}\right) \quad (2)$$

$$B/A = \frac{Fe_2O_3 + CaO + MgO + NaO + K_2O}{SiO + Al_2O_3 + TiO_2} \quad (3)$$

$$Iron\,Index = \%Fe_2O_3 \quad (4)$$

$$R_S = \left(\frac{B}{A}\right) \times S \quad (5)$$

3. Results and discussion

3.1. Proximate and ultimate analyses

*Moisture content**:*** Table 1 displays the result obtained from the proximate analysis carried out on the three (3) coal samples (Okaba, Maiganga and Enugu samples). In studying the effect of moisture content of coal, the result showed that the moisture contents range from 5.17% to 6.200%. Enugu coal has the highest moisture content of 6.2% followed by Okaba and then Maiganga coal samples with 5.44% and 5.17% respectively. These values were considered to be within the range for good coking coals [9]. Low moisture content is an indication that the coal is of a high rank and good quality, possibly the rank of bituminous grade [15]. It could also be inferred that a low moisture content represents a significant improve-ment in coal's quality because moisture affects the calorific value, the concentration of other constituents, decreases system capacity and increases operational cost. The moisture content of coal depends on the degree of maturity [15]. Therefore Maiganga coal sample with the lowest moisture content (5.171%) may be the most matured, followed by Okaba and then Enugu coal samples.

Ash Content: Enugu coal contains a relatively low ash content of 20.02%, while ash contents of Maiganga and Okaba coals were 13.981% and 10.719% respectively. High ash content is undesirable and harmful because it influences slag volume and composition [16]. These results agree with Fatoye *et al*. [12] who found that typical ash content in Nigerian coals are within the range of 5-40%. High ash content in coals presents an undesirable character due to lower fusion temperature which can give rise to a high slag volume and low blast furnace efficiency. The high ash content is also an indication of low degree of coalification and hence immaturity of the coal [16].

Volatile Matter: The analysis shows that the volatile matter of Maiganga, Okaba and Enugu coals are 19.19%, 21.54% and 28.98 % respectively. Enugu coal from this result shows that it has the highest amount of volatile matter of 28.98% and Maiganga coal bears the lowest with 19.19 %. The coals with the higher volatile matter ignite easily because it has low ignition temperature and burns with long smoky yellow flame [17]. Typically the percentage of volatiles from high-rank coals at 1300 K is less than 30% [15]. The higher the volatile matter, the lower the coal rank. Beside the use of volatility in coal ranking, it is one of the key para-meters used in determining their suitable applications [18]. Although high volatile matter could generates high pressure during combustion which is detrimental to the boiler walls.

Fixed Carbon Content (FC): The Fixed carbon contents of Maiganga, Okaba and Enugu coals are 61.66%, 54.86% and 52.65% respectively. The fixed carbon content of a bituminous material is the solid residue other than ash obtained by destructive distillation. It is the carbon found in organic materials after volatile materials have been driven off [18-19]. The Fixed carbon content of coal also determines the rank and quality of a coal sample. High carbon content is essential for coke making coal because it is the mass that forms the actual coke on carbonization [18]. Furthermore, it is used to estimate the amount of coke that will be obtained on carbonization [18-19]. All these suggests that Maiganga coal sample with the highest value of 61.66% FC has more potential for coke formation, followed by Okaba and then Enugu coal samples.

Ultimate Analysis: From the result of this analysis, it showed as in Table 1 that Enugu coal has the highest sulphur content of 1.82% while Okaba and Maiganga were 0.89% and 0.58% respectively. Low sulphur content signifies that it is a good fuel which can be used to generate power with less environmental challenges. The analysis result table shown in Table 1 showed that Maiganga coal has the highest carbon content of 59.23%. Maiganga and Enugu coals carbon content were 57.94% and 53.27% respectively. The carbon content of coal affects the heating value of the coal. The oxygen content of the selected coal samples is displayed in the result table shown in Table 1. Okaba coal had the highest oxygen content of 13.76% while Maiganga and Enugu coal had 12.07% and 12.43% respectively. From the analysis result displayed in Table 1. The result showed that Maiganga coal had a hydrogen content of 7.10% which is the highest, followed by Enugu of 4.97% and Okaba of 6.02%.

Table 1. Data obtained from the proximate and ultimate analyses of some Nigerian coal samples.

Coal sample	Maiganga coal (%)	Okaba coal (%)	Volatile Matter (%)
	Proximate analysis		
Moisture Content (%)	5.17	5.442	6.200
Ash Content (%)	13.98	10.72	20.02
Volatile Matter (%)	19.19	21.54	28.98
Fixed Carbon (%)	61.66	54.86	52.65
	Ultimate analysis		
Nitrogen	1.37	2.20	2.67
Carbon	59.23	57.94	53.27
Hydrogen	7.10	6.02	4.97
Sulphur	0.58	0.98	1.82
Oxygen	13.53	13.76	12.43
HHV(MJ/kg)	27.40	25.74	22.92

From the result of the calorific value calculated from formula (equation 1) on heating value of coal determination shown in Table 1, it was observed that Enugu coal had the lowest heating value of 22.92 MJ/kg. Maiganga coal showed the highest heating value of 27.40 MJ/kg then followed by Okaba coal with 25.74MJ/kg. Good coals are said to have a HHV, 26 MJ/kg and above [15]. Furthermore, it is inferred that the Maiganga coal with HHV is a better the coal for solid fuel combustion. This result show that only Maiganga coal has caloric value > 26 hence can be considered as good coal.

3.2. Thermal analysis

The pyrolysis for coal shows a typical three region profile. The mass loss is processed slowly at low temperatures, where dehydration of water occurs. This is follow by decomposition reaction which shows maximum weight loss rate, after which it approaches a constant residual weight.

The thermogram (TG) curves of Enugu, Okaba and Maiganga coals are shown in Figure 1. For Maiganga coal, about 53.89% of the sample was left undecomposed at 823.55°C which indicated that only about 46.109% of the coal sample was decomposed. This further implied that 53.89% of the sample that was left undecomposed consisted of certain materials such as clay, silt, and sand stone. For Okaba and Maiganga coals, about 46.923% and 84.647% of the sample were left undecomposed at 825.31 and 845.06°C indicating that only about 53.077% and 15.353% of the coal were decomposable respectively. The TGA used in this study has a tolerance limit of < 1000°C while pulverized coal combustors which is the dominant method for burning coal for power generation worldwide has combustion tempe-rature in the of range 1300–1700ºC [7].

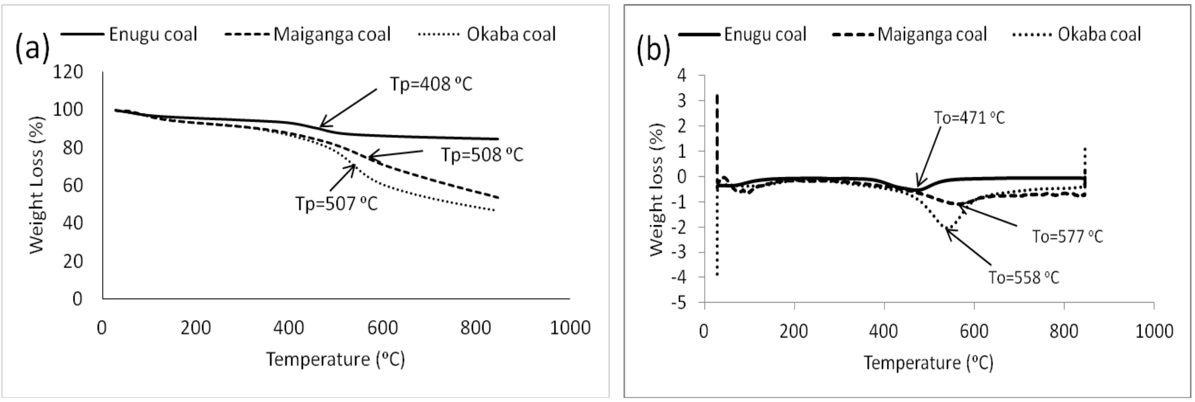

Figure 1. Thermal profile of the coal samples showing (a) TG and (b) DTG curves.

In this first stage, Maiganga and Okaba TG curves showed visible steep fall approximately in the range of 28.26 -187.29ºC (extracted from TGA and shown in Table 2) which can be described as a region where the drying of surface water occurred as result of the heat treatment to the coal sample which is described as an endothermic process in which heat is absorbed. Enugu coal showed a visible steep fall between 28.99ºC and 117.77ºC during drying of surface water.

Table 2. Thermal decomposition characteristics extracted from TGA

Coal sample	Degradation tempe-rature range (°C)	To (°C)	Tp (°C)
Maiganga	432.53-643.33	508.22	577.28
Okaba	417.65-641.74	506.87	558.27
Enugu	239.75-686.75	407.57	470.83

During dehydration, Maganga coal shows visible peak temperature of 96.96ºC due to the loss of surface water. The second stage is called the fairly constant region observed between

178.88ºC and 417.65ºC as shown in the TG in Figure 1(b). This occurrence is probably because nothing was released, hence, no decomposition peak was observed in this region.

The degradation temperature range (417.65ºC and 641.741ºC) describes the third stage where volatiles were released. From the DTG in Figure 1(b), decomposition peak describing mass loss was observed at a peak temperature of 558.27ºC with a visible downward major peak, indicating devolatilisation process in which compounds containing carbon, hydrogen and oxygen were released and according to Crelling *et al.* [20], it is identified as a primary decomposition process. This is followed by a downward fall to 825.31ºC. The onset temperature is defined as that temperature at which the degradation started which was at 506.87ºC. Maiganga coal had the highest onset temperature of 508.22ºC in contrast to Okaba and Enugu coal at 506.87ºC and 407.570ºC respectively. From Figure 1(a), Enugu coal was the earliest to decompose leaving high percentage of undecomposed matter, followed by Okaba coal and then Maiganga coal. Because the volatile content in the Enugu coal is very high, the volatile can be easily decomposed and released at a low temperature of about 407.570ºC, compared with about 508.22ºC and 407.57ºC for Maiganga and Okaba coals. Consequently, the onset temperature and decomposition of the Enugu coal should be much more stable. It was used in determining the onset temperature. It can also be seen in Figure 1 that Enugu coal showed more thermal stability than either of Okaba and Maiganga coal and the ash content is enlarged by factor 2.0 when compared with the other two coal samples. More so, it can be inferred that Enugu coal at 845.06°C had most amount of undecomposed matter (84.647%) whereas, that of Maiganga was 53.891% and then Okaba was 46.923% which was the least.

Figure 1(b) shows the derivative of thermogravimetric (DTG) of coal samples (Maiganga, Enugu and Okaba). Coals of higher rank generally have a higher peak maximum temperature as this trend occurs because coals of higher rank contain less mineral matter effectively raising the calorific values. Figure 1(b) shows that the peak temperature follows a pattern as a function of carbon content [17]. Amongst the coal samples, Maiganga coal had the highest peak temperature of 577.28°C indicating that it's the highest in rank between the three samples followed by Okaba and then Enugu coal with peak temperature of 558.27°C and 470.830°C respectively. The peak temperature is defined as the temperature at which nearly all the volatiles were removed.

3.3. Chemical analysis

The results of the elemental analysis of the coal ash obtained with the aid of XRF (X-ray fluorescence) are given in Table 3. It is evident from the results that there was variation in the distribution of various inorganic elements in the coal samples (Maiganga, Enugu and Okaba) analyzed. XRF analysis indicated that sulfur was present as compound SO_3. Contrary to what was expected, the analysis did not detect any presence of pyrite/marcasite (FeS_2). Those compounds are generally present in coals in variable amounts. Sulphur being the element of interest was seen to be occurring in large amount of 5.91% in Enugu coal then followed by 2.78% in Okaba coal and very little amount of 2.31% in Maiganga coal. Maiganga showed very acceptable level of sulphur content which is occurring in a minimal amount.

Table 3. Ash chemical composition of Maiganga, Okaba and Enugu coal samples.

Oxides	Maiganga Coal	Okaba Coal	Enugu coal	Oxides	Maiganga Coal	Okaba Coal	Enugu coal
TiO_2	4.33	4.11	3.85	SO_3	2.31	2.78	5.91
Al_2O_3	23.6	26.02	28.12	MnO	1.72	2.58	2.08
SiO_2	49.4	44.8	39.0	P_2O_5	0.57	0.44	0.83
Fe_2O_3	5.52	5.68	7.2	CuO	0.019	0.07	0.02
CaO	8.06	10.34	12.01	ZnO	0.210	0.95	0.10
MgO	2.4	2.7	3.4	Ag_2O	0.550	0.39	0.45
Na_2O	0.73	0.85	1.03	BaO	0.210	0.19	0.38
K_2O	1.35	1.70	2.93				

From the result in Table 3, silicon oxide occurred as the major element predominantly in all the coal samples. Maiganga coal had the highest amount of silicon oxide. More so, Fe, Ca, K and P in their oxide forms were found occurring as minor elements. These elements do not evaporate easily on combustion and share similar characteristics. Furthermore, Cu, Zn, Ag and Ba, occurred in trace elements in all the coal samples.

3.4. Ash chemistry of coal samples

The ash chemistry of the coal samples in Table 2 showed high levels of silicon and aluminum. The oxides of these elements constitute more than seventy percent except for Enugu coal. Iron oxide and calcium oxide are a dominate basic oxide in the ash of the coal samples. When titanium dioxide is added silicon and aluminium it is reported as acidic oxides. The basic oxides equal the sum of the percentages of iron, calcium, sodium, magnesium and potassium components. High content of silica results in a higher degree of covalence, and this behaviour yields a high temperature melting phase and hence a low deposition potential [21-22]. From Table 4 coal Maiganga coal has the silica ratio, followed by Okaba coal, the least is Enugu coal. Guided by the silica ratio, Maiganga and Okaba coals are expected to show low slagging propensities, while Enugu coal show high slagging propensities. Enugu coal has the highest percentage of alumina. It is believed that silicon combine with alumina alumina-silicate is formed which is known to increase slag volume [22]. A significantly higher potassium content in Enugu coal than the other of the samples ashes (Table 4) could be the reason for severe slagging [23]. During combustion potassium is released to the gas phase KOH and KCl. Transformation of KOH and K_2SO_4 can be carried out through the following global reactions [24].

Table 4. Summary of existing coal slagging indices employed and this study.

Expression	Slagging potential				Coal samples in this study		
	Low	Medium	High	Severe	Maiganga	Okaba	Enugu
B/A	<0.5	0.5< B/A <1	1	≥1.75	0.24	0.31	0.42
S_R	72–80	65–72	50–65	–	76	69	59
Iron Index	< 6 %	6–7 %	> 7 %	–	5.52	5.68	7.2
$Rs=(B/A)\times S$	< 0.6	0.6–2.0	2.0–2.6	> 2.6	0.13	0.30	0.76

The content of iron is another important parameter in determining the ash slagging potential. High iron content usually lowers the melting point of slag [22], which results in a high slagging potential. Iron content lower than 6% in coal ash translate to a low slagging tendency [13]. The percentage of iron oxides for the three coal samples has been computed and the results obtained are also presented in Table 4. The results indicate that coal Maiganga coal has the lowest Fe_2O_3 content, followed by Okaba coal, then Enugu coal coal, and therefore, we expect their deposition potential to be in the same ascending order. Iron oxide is a low melting phase compound and act as a strong fluxing agent [13,22]. Where the iron index is high slagging of heating surface is controlled by pyrite behaviour which may react with clay and quartz to form alumino-silicate slags. The dissolution of iron into alumino-silicate reduces the melting point of fly ash in the combustion system [25].

The basic to acidic oxides ratio (B/A) of the coals also aids in describing good and bad coals prior to combustion. An decrease in the B/A of the coal will raise its fusion temperature. The calculated values for the B/A for three coals been investigated, are also shown Table 4. The results indicate that the slagging potential of Enugu coal is expected to be the worst, then Okaba coal, and Maiganga coal, in decreasing order. If a portion of the sulfur in coal exists with more pyrite than siderite B/A. B/A is multiplied by the percentage of dry sulphur in the coal in order to obtain Rs shown in Table 4. Coal with Rs > 0.6 is said to have high slagging propensities [22,26-27]. The slagging indices for coal samples are also presented in Table 4. The calculated results suggest that Maganga and Okaba have lower slagging potential than Enugu coal.

4. Conclusion

The different geographical locations as well as mode of occurrence coal constituents play a major role in coal formation. Hence three coals were selected from Northern Benue trough, Central Benue trough and Anambra Basin. The coal samples were first characterized and then combusted to ash samples. The thermal profiles suggest that Enugu has higher thermal stability and lower calorific value than Maiganga and Okaba coals. The ash chemistry indices used to predict the performance of the coal samples prior to combustion shows that silica ratio indicated that Enugu coal high slagging tendency, B/A suggest that all the coals were found to be less than 0.6 indicating low slagging potential. The ash chemistry suggests that Maiganga coal has low slagging potential while Enugu coal has severe slagging potential.

Nomenclatures

HHV	*high heating value*
TGA	*thermogravimetric analysis*
XRF	*X-ray Fluorescence spectroscopy*
B/A	*basic to acidic oxides ratio*
Rs	*sulphur slagging index*
S_R	*silica index*

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