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A COMPARISON OF SPONTANEOUS COMBUSTION SUSCEPTIBILITY OF COAL FROM THE BENUE TROUGH ACCORDING TO RANK

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ABSTRACT

This study investigated spontaneous combustion susceptibility of coal according to the rank. To estimate the spontaneous combustion susceptibility of coal, both crossing-point temperature (CPT) measurement and gas analysis by using gas chromatography (GC) were performed. For the experiment, Ihuoma coal, Orlu lignite, and Onyeama coal and Owukpa coal that are sub- bituminous coal, Obi/Lafia bituminous coal was used. The lignite such as Ihuoma coal contains more functional groups that easily react to oxygen more so than Onyeama and Owukpa coals. Also, Onyeama and Owukpa coal more functional groups than Obi/Lafia coal. For this reason, the lignite is more easily oxidized than sub-bituminous coal and sub-bituminous more easily oxidized than bituminous at low temperature, which results in high O_2 consumption, increase in CO and CO_2 generation, and low CPT. Although the CPT of Onyeama coal and Owukpa coal is identical to each other as they are the sub-bituminous, Owukpa coal has a lower initial oxidation temperature (IOT) and maximum oxidation temperature (MOT) than those of Onyeama coal. This means that although each coal has the same rank and CPT, spontaneous combustion susceptibility of coal may vary because the initial temperature of the coal at which oxidation begins may be different due to the substances that participate in oxidation.

Key Words: Coal rank, cross-point-temperature, spontaneous combustion, low temperature oxidation.

INTRODUCTION

The spontaneous combustion susceptibility of coal causes difficulties in transporting, sorting, and using coal, which is critical in terms of safety and economy (Goodarzi and Gentzis, 1991; Wei et al., 2013). Several methods are available to determine the spontaneous combustion susceptibility of coal, such as adiabatic oxidation (Banerjee, 1985); high temperature activation energy (Münzner and Peters, 1965; Banerjee, 1985; Postzednik et al., 1988); crossing-point temperature (Oresko, 1959; Banerjee, 1985; Handa et al., 1985; Kaymakçi and Didari, 2002); and gas analysis methods (Lang and Fu-bao, 2010; Xie et al., 2011). Crossing point temperature (CPT), average heating rate (AHR) and slope of the time-temperature curve on the crossing point (CPS) (Fig. 1) are taken as the spontaneous combustion parameters.

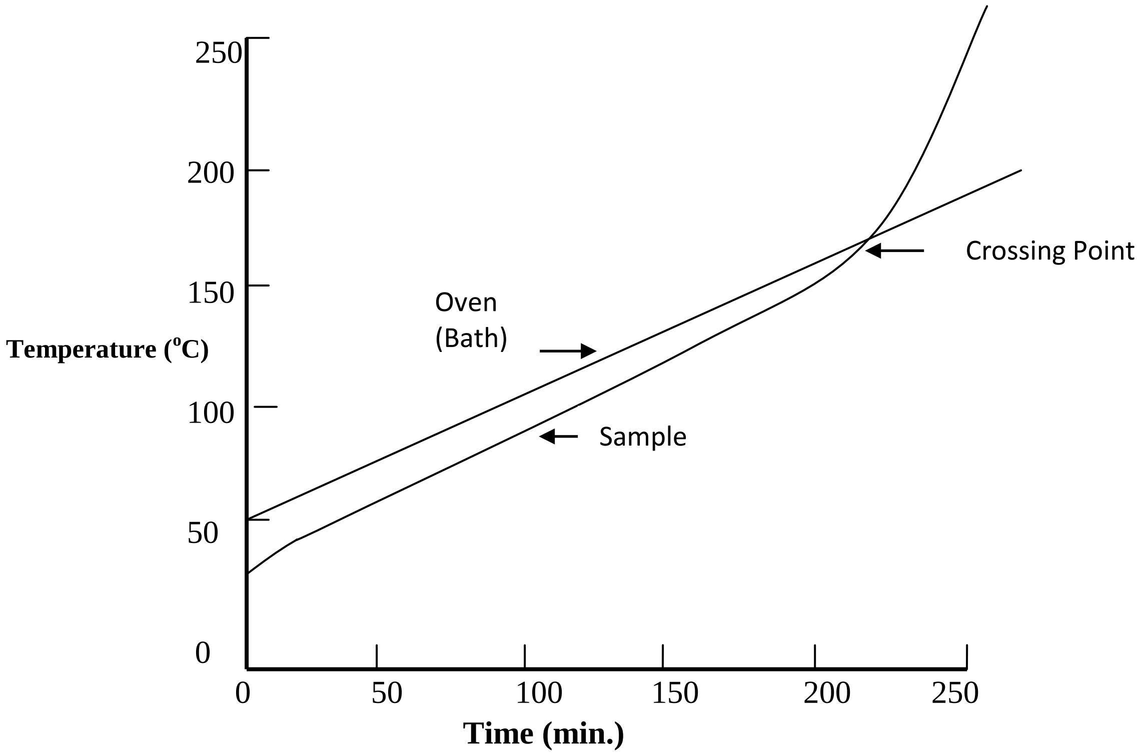

Fig. 1: Time – temperature curve (modified from Kaymakçi and Didari, 2002).

If air or oxygen is supplied in the oven that contains the coal by increasing the temperature inside the oven, the temperature of the coal will increase along with the temperature of the oven, and at a certain point, the coal temperature will exceed the oven temperature due to self-heating through the oxidation of coal. This point at which the coal temperature crosses that of the oven is called crossing-point temperature (CPT).

Research on CPT measurement was first done by Postzednik et al. (1988). CPT measurement has been used for evaluating spontaneous combustion susceptibility of coal. CPT measurement in previous studies was conducted according to coal properties such as rank, particle size, moisture content, and application conditions such as the flow rate of oxygen, heating rate, and the amount coal samples (Oresko, 1959; Banerjee, 1985; Handa et al., 1985; Kaymakçi and Didari, 2002). Since CPT measurement methods measures the temperature of part of all coal contained in the vessel, it may decrease the reliability of the results due to the property of coal that is not a single substance.

Gas analysis using gas chromatography (GC) is used to estimate spontaneous combustion susceptibility of coal through analysis the concentration of CO and CO_2 that is generated when coal reacts with oxygen and the oxygen consumption (Goodarzi and Gentzis, 1991; Lang and Fu-bao, 2010; Xie et al., 2011). In general, the higher the spontaneous combustion susceptibility of coal the more temperature (Kröger and Beier, 1962; Güney, 1968; Chamberlain and Hall, 1973a; Feng et al., 1973; Beier, 1973; Kim, 1977; Banerjee, 1982; Didari, 1988; Goodarzi and Gentzis,

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1991; Didari and Õkten, 1994; Lang and Fu-bao, 2010; Xie et al., 2011).

By combining the CPT measurement and GC analysis method, we can analyze the spontaneous combustion susceptibility of coal and compliment any short-coming that only we can measure the part of coal temperature during CPT measurement.
In this study, we measured CPT of two ranks of coal samples from three locations in Benue Trough (Fig. 2) made up of lignite and sub-bituminous in order to estimate the spontaneous combustion susceptibility of coal according to rank. In the process, we analyzed the CO and CO_2 concentration that generated in the process by using the GC.
During the experiments, we fixed the factors that affect the result of CPT measurement: gas flow rate, heating rate; moisture content, and size of the coal.
In this paper we assess coal samples from three regions in the Benue Trough (Fig. 2). The Benue Trough geographically subdivided into Southern, Central, and Northern Benue Trough is strictly a sedimentary basin extending from the Gulf of Guinea in the south to the Chad Basin in the north (Offodile, 1976; Dike, 1976a,b, 1993, 2002; Adeleye and Fayose,1978; Enu, 1987 and Petters, 1978, 1979b, 1980, 1982, 1991). The origin and tectonic history of the Benue Trough is associated with the break-up of the continents of Africa and South America. This break-up was followed by the drifting apart of these continents, the opening of the South Atlantic, and the growth of the mid-Atlantic ridge (Read and Watson, 1978; Freeth, 1990).

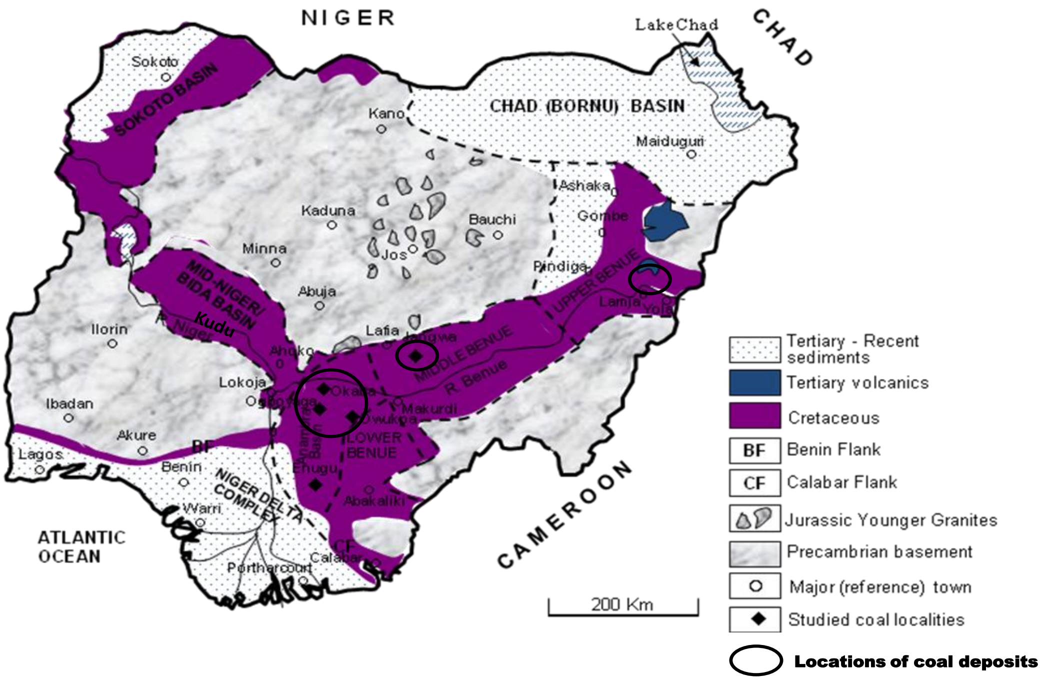

Fig. 2: Generalized geological map of Nigeria showing some coal deposit locations and locations of studied coal deposits

Synopsis of the Geology of the Benue Trough

The Benue Trough of Nigeria is a rift basin in central West Africa that extends NNE – SSW for about 800 km in length and 150 km in width (Fig. 1). The trough contains up to 6000m of Cretaceous – Tertiary sediments of which those pre-dating the mid-Santonian have been compressionally deformed, faulted and uplifted in several places. Compressional folding during the mid-Santonian tectonic episode affected the whole of the Benue Trough and was quite intense, producing over 100 anticlines and synclines (Benkhelil, 1989). Following mid-Santonian tectonism and magmatism, depositional axis in the Benue Trough was displaced westward resulting in subsidence of the Anambra basin. The Anambra basin, therefore, is a part of the Southern Benue Trough containing post-deformational sediments of Campano – Maastrichtian to Eocene ages. It is logical to include the Anambra basin in the Benue Trough, being a related structure that developed after the compressional stages (Akande and Erdtmann, 1998). The Benue trough is subdivided into a Southern, Central and an Nortern portion (Figs. 2, 3 and 4). A generalized stratigraphic succession in the Benue Trough and the relationship to the Chad basin and the Niger Delta is given on Figs. 3 and 4. Details on the geology and stratigraphy of the Benue trough have been comprehensively discussed, reviewed and presented by Carter et al. (1963), Whiteman, (1982), Akande et al. (1998), Obaje et al. (1999), Obaje and Hamza (2000), Pearson and Obaje (2000) and Obaje et al. (2004), amongst others.

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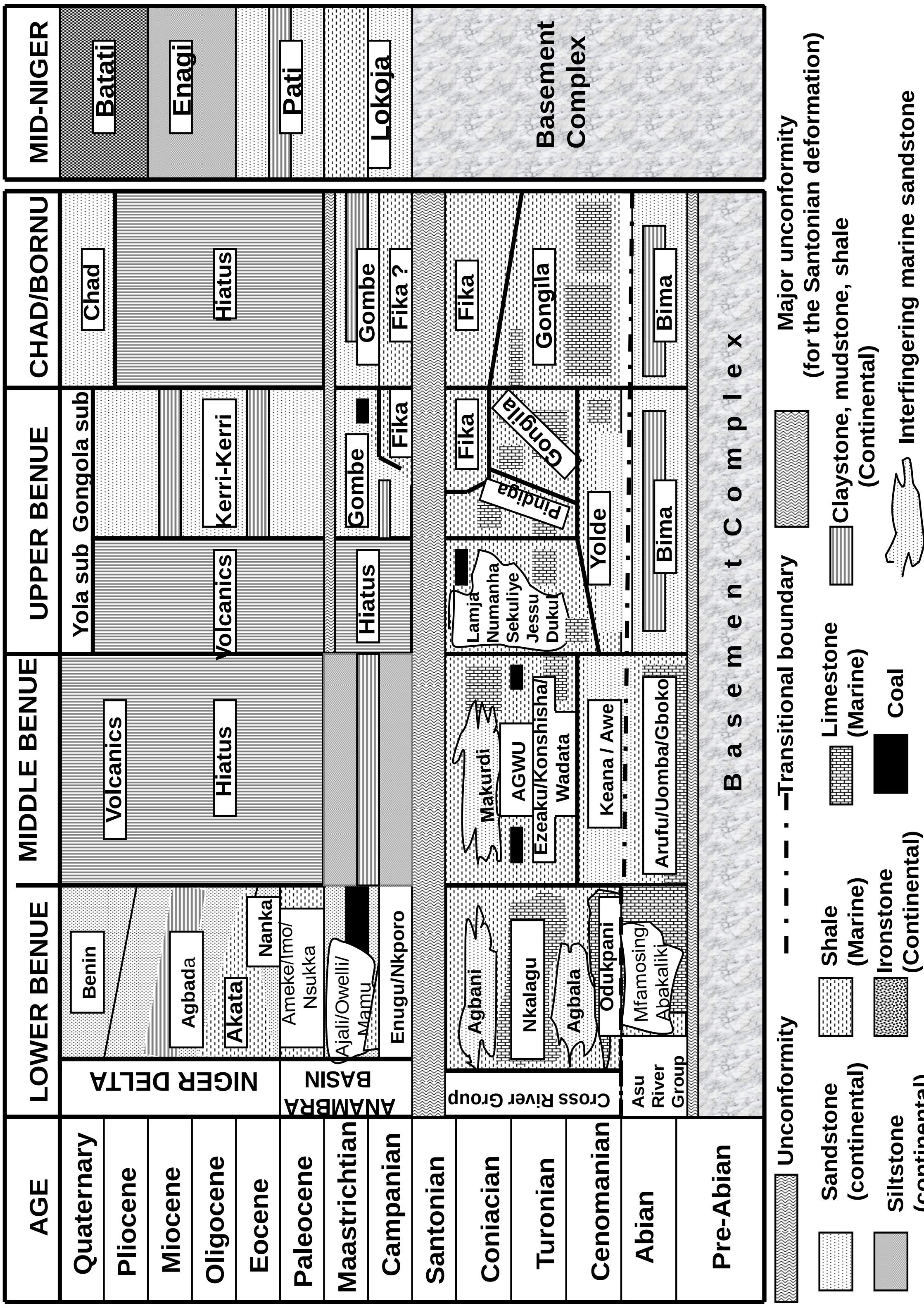

Fig. 3: Stratigraphic succession in the Benue trough, the Nigerian sector of the Chad Basin, the Mid-Niger Basin and relationship to the Niger delta (after Obaie et al.. 2006).

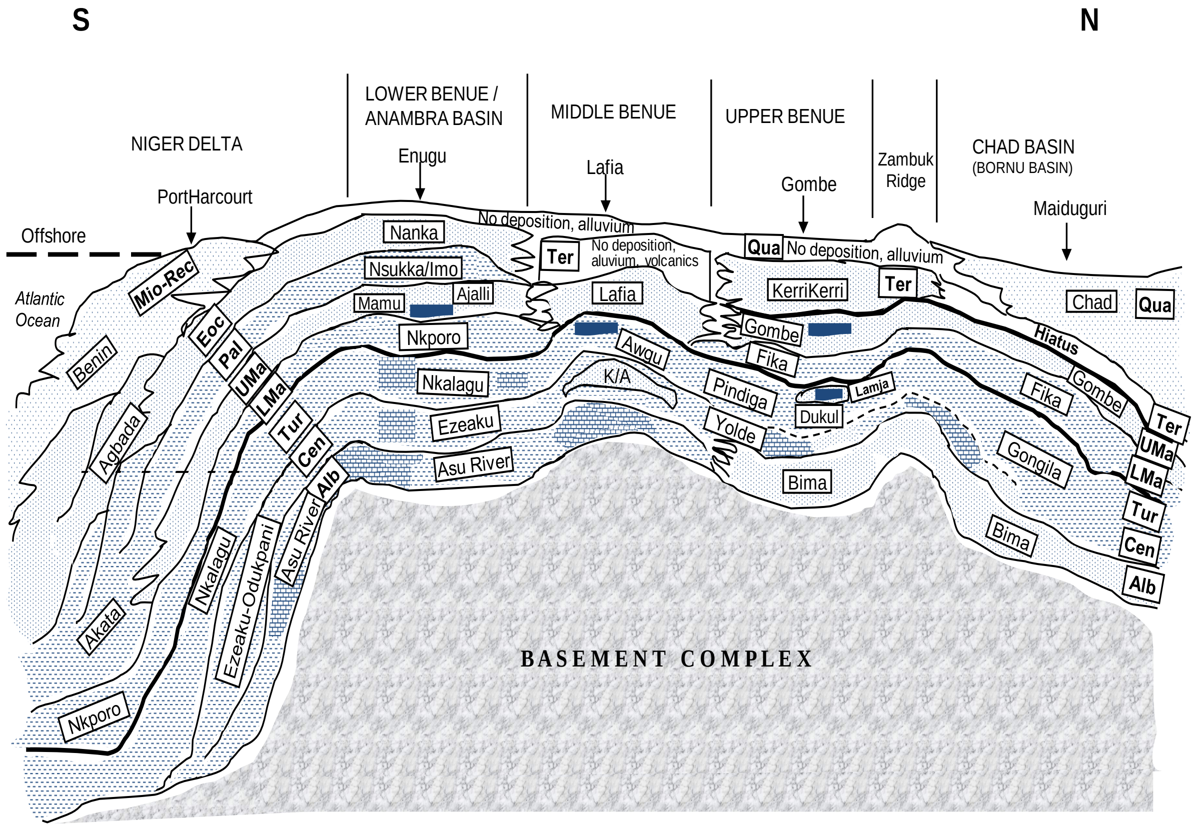

Fig. 4: Idealized N-S stratigraphic cross-section across the Benue Trough and the relationship to the Niger Delta and the Chad Basin (vertical scale exaggerated; erosion and uplift not considered, after Obaje et. al, 2004).

EXPERIMENTAL

Coal Sample

Ihuoma coal samples, which is lignite, Onyeama and Owukpa coal that are, sub-bituminous and Obi/Lafia coal of bituminous coal were used in this study. Among the coals selected lump coal that was more than 10cm in diameter was selected for the samples to minimize the effect of weathering. The lump coal was crushed and sieved in order to obtain particles that were 180-425 μm in size for use. To remove any effect caused by moisture during the CPT measurement, the coal was dried at 107^{o}C under a nitrogen atmosphere for 12 hours.

Experimental Apparatus and Procedure

A conceptual diagram of the CPT measurement apparatus used in the experiment is shown in Fig. 5. Two vessels that contain a coal sample are located inside an oven in which the temperature is programmable. To each vessel, nitrogen or air is selectively supplied pass through

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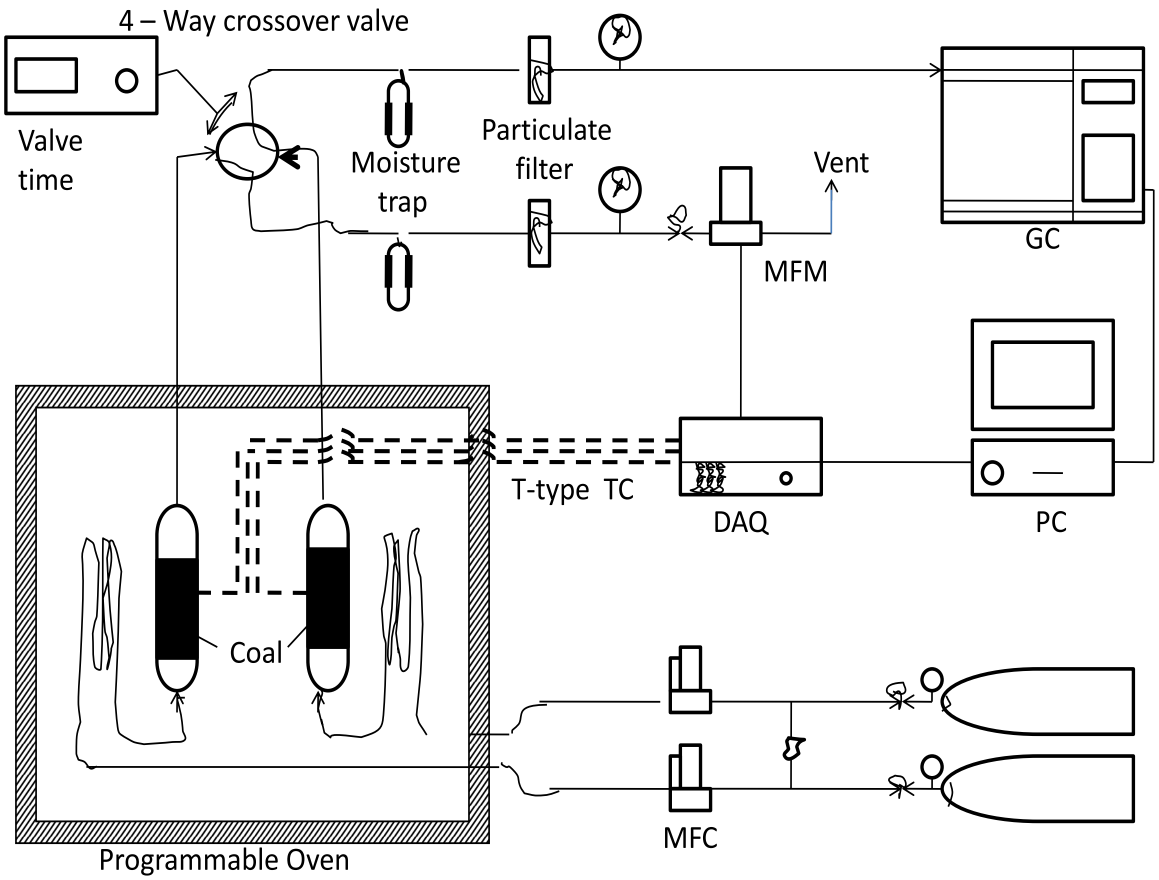

Fig. 5: Schematic diagram of experimental apparatus.

The vessels are sent to the GC 96890N, for measuring components, or the mass flow meter (MFM) for measuring the flow rate after reaction, according to the selected 4-way crossover valve location that changes periodically. The O_2, CO and CO_2 gases emitted from each vessel were measured in the GC to which the thermal conductivity detector and two columns (60/80 Molecular Sieve 13X, 80/100 Parapak N, SUPELCO) are installed.
35g of the coal sample dried in each vessel was loaded and installed them inside the oven that was set at 40^{o}C. Then, nitrogen gas of 75ml/min was supplied for two hours in order to cause the temperature to achieve equilibrium. When the temperature of the coal inside the vessel was 40^oC, nitrogen gas to one vessel of 75ml/min and the air to another vessel with the same flow rate and increased the oven temperature to 180^{o}C by 0.5^{o}C /min.

RESULT AND DISCUSSION

Since the generation of heat is the main purpose of industrial combustion, the coal's calorific value or specific energy is of particular interest. The property is governed by the proportion between non-combustible (ash, water) and combustible matter in the coal. The latter consists mainly of the elements carbon, hydrogen and sulphur. The exothermic values of the reaction of these elements with oxygen differ quite considerably, hydrogen having the highest value. Variations in the elemental composition of coals affect their heat values in response to changes in rank and type. One reason for an increase in calorific value during coalification is the decrease in the moisture content of lignite and sub-bituminous coals. The decline in the oxygen content during the transition from high-volatile to medium-volatile bituminous coals also contributes to an increase in calorific value. The latter peaks in the low-volatile bituminous coals but begins to drop again with the loss of much of its hydrogen as the coal approaches anthracite rank.

The infrared spectrum (Fig. 6) shows the principal functional groups related to organic matter include the aliphatics (2800 - 3200 cm^{-1}) and aromatics (800 - 1200 cm-1). The response at 600-3800 cm-1, attributed the hydroxyl group, is also influenced by clay minerals and cannot be considered as wholly from organic matter. The principal application of infra-red spectroscopy is to evaluate the relative importance in organic matter of C=O and C-C functional groups (Tissot and Welte, 1984).

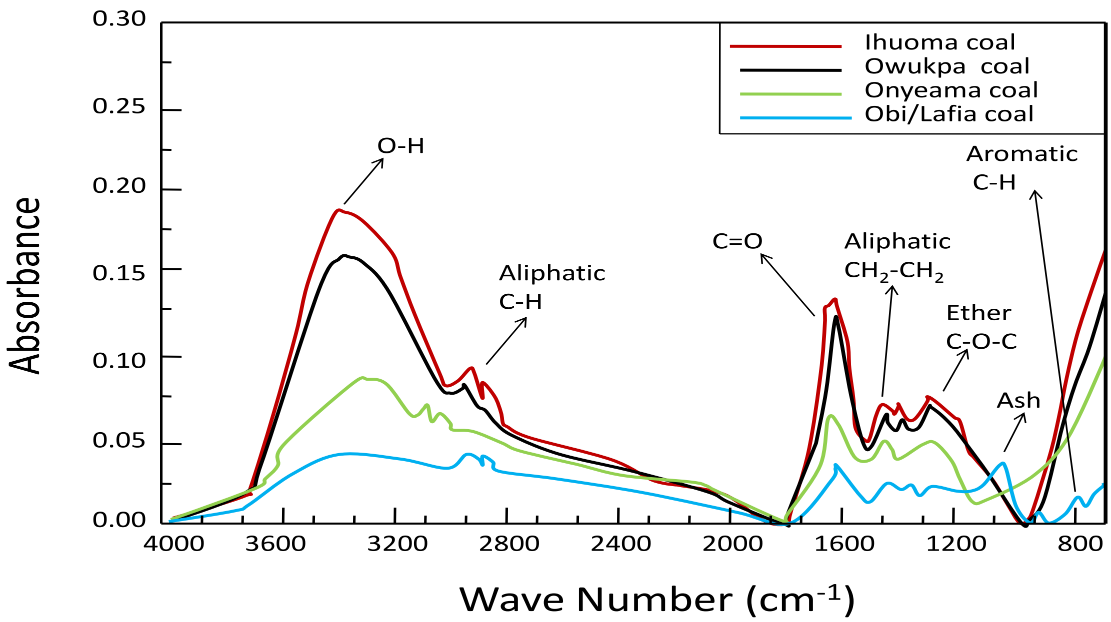

Fig. 6: FT-IR spectrum of coal from the Benue Trough.

Infrared spectrum shown in Fig. 6 is prominent functional groups related to organic matter include the hydroxyl groups at 3400 - 3500 cm^{-1}, the aliphatic at 2800 - 3200 cm^{-1} and 1400 - 1600 cm^{-1}, as well as the aromatics at 1600 - 1700 cm-1 and 700 - 850 cm^{-1}. The C=O groups attributed to the spectral region of 1700 - 1710 cm^{-1} [Mongenot *et al.*, 1999] are absent in Fig. 6, but some, attributed to 1040 - 1400 cm^{-1} [Tissot and Welte, 1984], may be masked by the broad band attributed silicates. Both the fact that the C=O groups have a high molecular absorption coefficient that is well resolved by FT-IR, and the low intensity of the 1700 - 1710 cm^{-1} band relative to other functional groups with lower molecular absorption coefficient, indicate the presence of few carboxyl and carbonyl groups.

Properties of Samples

Table 1 shows the results of proximate analysis and calorific value analysis of the coal samples. The moisture content of the

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lignite coal, Ihuoma coal was 36.55 wt%. The Onyeama coal and Owukpa coal, sub-bituminous coal, was 3.90 and 6.70 wt%, respectively. The Obi/Lafia coal, the bituminous coal, was 8.41 wt%. The content of volatile matter in Ihuoma coal (39.15 wt%) was higher than that of fixed carbon (20.34 wt%). However, the content of fixed carbon in Obi/Lafia coal (58.00 wt%) was higher than that of the Onyeama coal (41.20 wt%) and Owukpa coal (42.75 wt%) and also was higher than that of the volatile matter; the fixed carbon showed a higher amount than volatile matter (31.83 wt%). As for the ashes, Obi/Lafia coal contained percentage of ash content and it was higher than Onyeama, Owukpa and Ihuoma coal samples, which contained 9.90, 7.92 and 4.07 wt%, respectively. It has been known that the lower the rank, the more the coal will have high moisture content and the higher the volatile matter compared to the higher rank coals.

Table 1: Proximate and calorific values of minimum, maximum and average for aggregate of each coal type.

			Proximate Analysis (wt%)															
Sample Name	Type	Locality	%Moisture Content			%Ash Content (wf)			%Volatile Matter (waf)			% Fixed Carbon			Calorific Values (kcal/kg) wf		
			Min.	Max.	Av.	Min.	Max.	Av.	Min.	Max.	Av.	Min.	Max.	Av.	Min.	Max.	Av.
ORLU	Lignite	Ihuoma	35.40	37.50	36.55	3.98	4.20	4.07	37.76	40.33	39.15	19.59	21.05	20.34	5098.00	5342.00	5189.75
ONYE	Sub-bituminous	Onyeama	3.15	4.15	3.90	3.16	21.18	9.90	43.39	47.30	45.07	32.28	47.48	41.20	6067.00	7621.80	7025.00
OWUK	Sub-bituminous	Owukpa	6.36	7.23	6.70	6.96	8.60	7.92	41.98	47.90	45.22	42.33	43.23	42.75	6430.00	6475.60	6433.15
OLAF	Bituminous	Obi/Lafia	7.20	8.93	8.41	12.30	14.70	13.93	31.65	32.23	31.83	57.65	58.23	58.00	7524.00	7745.00	7633.25

The Mechanism of Spontaneous Combustion of Coal

Although many factors affect heat producing reactions, the oxidation of carbonaceous matter in coal of ambient temperatures is the major cause for the initiation of spontaneous combustion (Güney, 1968; Banerjee, 1985; Goodarzi and Gentzis, 1991; Wei et al., 2013).

The oxidation of coal, like all oxidation reactions, is exothermic in character. The exact mechanism of the reaction is still not well understood. However, scientists agree that the nature of the interaction between coal and oxygen at very low temperatures is fully physical (adsorption) and changes into a chemisorption form starting from an ambient temperature (Münzner and Peters, 1965; Banerjee, 1985; Postzednik et al., 1988). The rate of oxygen consumption is extremely high during the first few days (particularly the first few hours) following the exposure of a fresh coal surface to the atmosphere. It then decreases very slowly without causing problems unless generated heat is allowed to accumulate in the environment. Under certain conditions, the accumulation of heat cannot be prevented, and with sufficient oxygen supply, the process may reach higher stages.

The loose coal-oxygen-water complex formed during the initial stage (peroxy-complexes) decomposes above 70-85°C, yielding CO, CO_2 and H_2O molecules. The rate of chemical reactions and exothermicity change with the rise in temperature, and radical changes take place, starting at about 100°C, mainly due to loss of moisture (Oresko, 1959; Banerjee, 1985; Handa et al., 1985; Kaymakçi and Didari, 2002). This process continues with the rise in temperature, yielding more stable coal-oxygen complexes until the critical temperature is reached. From then on, it is fairly safe to assume that an actual fire incident will result.

Factors Affecting the Spontaneous Combustion of Coal

The main reason for the difficulties in understanding the mechanism of spontaneous combustion is the pressure of many internal and external factors affecting the initiation and development of the phenomenon (Table 2). These factors have been reviewed by various researchers (Kröger and Beier, 1962; Güney, 1968; Chamberlain and Hall, 1973a; Feng et al., 1973; Beier, 1973; Kim, 1977; Banerjee, 1982; Didari, 1988; Goodarzi and Gentzis, 1991; Didari and Õkten, 1994; Lang and Fu-bao, 2010; Xie et al., 2011). The main factors which have significant effect on the process according to Güney (1968) are summarized below:

i) Pyrite content may accelerate spontaneous combustion,

Table 2: Factors affecting the spontaneous combustion of coal (Güney, 1968)

Intrinsic Factors (Nature of Coal)	Extrinsic Factors (Atmospheric, Geological and Mining Conditions
i) Pyrites	i) Temperature
ii) Moisture	ii) Moisture
iii) Particle size and surface area	iii) Barometric pressure
iv) Rank and petrographic constituents	iv) Oxygen concentration
	v) Bacteria
v) Chemical constituents	vi) Coal seam and surrounding strata
vi) Mineral matter	vii) Method of working
	viii) Ventilation system and air flow rate
	ix) Timbering
	x) Roadways

ii) Changes in moisture content; i.e. the drying or wetting of coal, have apparent effects,

iii). As the particle size decreases and the exposed surface area increases, the tendency of coal towards spontaneous combustion increases,

iv). It is widely recognized that lower rank coals are more susceptible to spontaneous combustion than higher

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rank coals. The abnormalities in this relationship may be attributed to the petrographic constituents of coal. This is because lower rank coals contain more liptinite (more hydrogen) that enhance increase in hydrocarbon generation than the higher rank coals which might lose some hydrogen on high maturity stage. Coals of high hydrogen contents are more prone to spontaneous combustion,

v). Ash content generally decreases the liability of coal to spontaneous heating. Certain parts of the ash, such as lime, soda and iron compounds, may have an accelerating effect, while others, such as alumina and silica, produce a retarding effect. It is clear that some chemicals promote combustion while others inhibit its development. Also, it is known that oil shale bands adjoining coal seams play an important role in mine fires,

vi). The temperature of the underground atmosphere is a direct factor,

vii). The presence of faults and zones of weakness around faults may contribute to the danger by allowing air leakage into coal mass,

viii). Mining methods with partial extraction, in which part of the coal seam is left in the goaf and pillars (designed for several purposes), can contribute to the potential for spontaneous combustion,

ix). Air flow rate is a complex factor because an air supply provides oxygen while it carries away the heat produced. There is a critical air quantity which allows the coal to oxidize and also allows the generated heat to accumulate. Therefore, it favours the process,

x). High ventilation differentials and changes in the mine ventilation system also affect the development of the spontaneous combustion process.

According to Querol et al. (2008) oxidation of organic matter is mostly the main cause of coal self-ignition. When the ignition temperature of coal is reached, it starts to burn by spontaneous combustion (Gervet, 2007; Quintero et al., 2009). The temperature at which the coal oxidation reaction becomes self-sustaining and spontaneous combustion occurs generally depends on the type of coal and surrounding conditions of heat dissipation (Gervet, 2007; Wei et al., 2013). Although some authors found a relationship between maceral composition and the potential for spontaneous combustion of coal, a general rule is not clear (Querol et al., 2008; Wantaek et al., 2013). The coal oxidizes and self-heating is accelerated until the combustion occurs. With decreasing in coal rank, the tendency to the spontaneous combustion increases. For example, lignites and sub-bituminous coals from Orlu and Onyeama can begin their spontaneous combustion at 573 K, whereas bituminous coals as those of the Obi/Lafia coal mine at 873 K.

The results of this unique and comprehensive research are given in Table 1. Since the study is related to a specific coal region (Benue Trough), and any reported information on similar studies may not be available, a comparison equally may not be possible. Based on this reality, CPT can be taken as the major parameter to evaluate the relations. As seen in Fig. 1, there are high correlations between CPT and the volatile matter, ash, carbon of these coals from these areas (Orlu, Onyeama and Obia/Lafia) of the Benue Trough.

Kaymakçi and Didari (2002) demonstrated that through the linear regression analyses ash, volatile matter, carbon, hydrogen, exinite, inertinite and mineral matter are the major factors affecting spontaneous combustion. This report has confirmed the results that have been discussed for Table 1 and Fig. 1 above in which CPT has been correlated with all the proximate data obtained from this study. To further in confirming those realistic factor of CPT and proximate data obtained from coals of the Benue Trough through calculation of the multiple regression analyses Kaymakçi and Didari (2002), also found these major factors to be volatile matter, carbon, hydrogen, nitrogen, oxygen, sulphur and inertinite. With all these supportive and confirmative results and calculations a comparison of spontaneous combustion susceptibility of coals from Orlu, Onyeama and Obi/Lafia has been achieved through the recognition of these major factors.

There is no doubt that spontaneous combustion susceptibility has a negative impact in world coal deposits (such as the Benue trough coal mines) on mining operations, coal beneficiation and safety, health and environment of the mine and surrounding communities. CPT measurement in this study was conducted according to coal properties such as rank, moisture, ash, volatile matter contents, calorific value, and application conditions such as the flow rate of oxygen, heating rate, and the amount coal samples. From the results and discussions CPT has being taken as the major parameter to evaluate the relations of coal combustion susceptibility. It was found that there are high correlations between CPT and the volatile matter, ash, carbon of these coals from these areas (Orlu, Onyeama and Obia/Lafia) of the Benue Trough. With the recognition of these major factors a comparison of spontaneous combustion susceptibility of coals from Orlu, Onyeama and Obi/Lafia has been achieved.

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