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**Research Article** 

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# Effect of Process Variables on the Controlled Combustion of Nigerian Natural Gas

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Abstract The effect of process variables on the controlled combustion of Nigerian natural gas has been studied. Atmospheric air was the source of oxidant used for the combustion process. Flue gas analysis was performed using instrumental method of analysis on wet basis within a range of air temperature and percentage excess/deficient air. Formation of pollutants and their concentrations at various combustion conditions were investigated. The concentration levels of carbon dioxide and water vapour were discovered to be maximum at stoichiometric condition, and decreased with percentage excess/deficient air. The concentration of CO<sub>2</sub> also decreased slightly from 7.01 % to 6.60 % as air temperature increased from 28 °C to 120°C. Generation of major air pollutants (oxides of nitrogen) was found to be higher as air temperature increased from 28 °C to 120 °C. It was also discovered that carbon monoxide was not formed during combustion with excess air but its concentration became significant (42.55 mg/m<sup>3</sup>) during combustion with inadequate air. Methane ( $CH_4$ ) and other unburned hydrocarbons were substantially higher in their concentrations at inadequate air condition. Their concentrations at stoichiometric and gas-lean conditions were insignificant. Formation of CO was maximum (28.75mg/m<sup>3</sup>) as air temperature increased from 28 °C to 120 °C following decomposition of carbon dioxide. The concentration values of combustion products from this study are well within the emission limits from stationary sources as prescribed by Federal Environmental Protection Agency (FEPA). The study concludes that combustion of Nigerian natural gas in industrial burner/boiler system is environmental friendly when performed under ambient air temperature and 2 to 5 % excess air.

Keywords Process variables, Combustion, Natural gas, Flue gas, Concentration

# Introduction

The sources and byproducts of energy use in any nation is a matter that requires attention of all. In the past, and even now, fossil fuels have been the major energy source for satisfaction of man's energy demands. Chemical energy in fossil fuels (coals, petroleum, natural gas etc.) can only be converted to thermal (heat) energy through combustion process. During the process of converting chemical energy of natural gas into heat energy, harmful flue products are emitted into our environment [1]. These combustion products are mainly carbon dioxide (CO<sub>2</sub>), (CO) and others including greenhouse gases [2]. To comprehend fully the formation of these pollutants, we must discern the nature of fuels being burned and their general stoichiometry [3]. Among the various fossil fuels currently in use, the simplest in nature and composition is natural gas, which has methane as major constituent and insignificant amount of other components such as ethane, propane, nitrogen and carbon dioxide [4].

Activities of man since the beginning of the industrial revolution have produced a 42% increase in atmospheric concentration of carbon dioxide, from 281ppm in 1751 to 400ppm in 2002 [3]. The increase occurred despite the absorption of large portion of the emission by many natural sinks involved in carbon cycle. Scientists have predicted that if greenhouse gas emission continue at this rate, the earth's surface temperature could go higher than recorded value by 2050. The earth by then shall be unconducive for man.

However, man needs energy to sustain his life and numerous activities. Adequate supply of energy is the base of several activities of man on earth. The first source of available energy to man was the sun. Energy from the sun reaches the earth surface at a reasonable intensity but the intensity and rate at which it falls on a square meter of earth surface is negligible or very small [1]. So the difficulty is how to concentrate the energy gathered over a wide area so that its use in the production of work or power becomes practical. The sun could not alone serve or meet up with man's demand for energy because it is not steady at all times. Other sources of energy such as wind mills and potential energy of tides were also exploited but power production from these sources was not reasonable compared to demand. The advantages derived from the analysis of exhaust gases emitted into atmosphere from natural gas fired boilers are enormous. Exhaust gas analysis is an important step to adequately run and manipulate any combustion system in order to attain reasonable efficiency with the least emissions of greenhouse gases and other toxic products. In other to ensure complete safety of any combustion process, it is most important to monitor the concentration levels of carbon monoxide and hydrocarbon via flue gas analysis. Emissions from natural gas-fired boilers may include carbon dioxide, volatile organic matter, methane, soot and particulate matter. Methane generation is high during low combustion temperature particularly at plant start-up and shut-down periods for combustors [5]. Incomplete combustion in gas fired boilers and moving trucks are major emitters of CO [6].

Combustion of fossil fuel such as natural gas in industrial boiler to generate steam and electricity constitutes serious problems to people and their environment. These combustion systems increase the concentration levels of greenhouse gases and air pollutants present in atmosphere. Some of the problems are environmental pollution, climate change, global warming, formation of acidic rain, poor visibility and low quality of ambient air. The consequences of these problems on people are poor health conditions and short life span. To mitigate these problems mentioned above, there is urgent need to drastically minimize the generation and emission of  $CO_2$ , CO,  $CH_4$ , NO,  $NO_2$ ,  $SO_2$ , and particulate matter during operation of boilers in industries. This can be done through adequate flue gas analysis, hence, the aim of this paper.

#### **Materials and Methods**

In this paper, industrial steam boiler fitted with a dual purpose, two stage burner was used to burn the natural gas to generate combustion products (flue gases) without any modification. Combustion of the gas was done on installation (Carbon dioxide Production Plant) under practical conditions of operation as found in industries.

The materials used in this paper are natural gas, gas regulator, 2mm sample pipes and calibration gas. Equipment used are industrial steam boiler, fitted with a dual purpose, two stage burner, Testo 330 gas analyzer, Servomex gas detector and Teledyne hydrocarbon analyzer.

## **Boiler Operation Procedure**

The boiler used during this experiment was a fire tube boiler with a safe working pressure of one bar and a total volume of 9790 litres. The boiler consists of two major parts- burner and combustion chamber. The burner is a dual purpose type because it was designed to combust natural gas as well as diesel oil. The boiler has facilities to control or regulate process variables such as air volume and temperature.

The First step taken before the combustion process was to put the combustion air regulating unit on manual mode so that it could allow changes or alterations to vary the combustion air flow rates. The flow rate of natural gas was constant, at each burner load, and maximum at burner full load of 100%. The process plant was then started and loading of the burner continued gradually till fully loaded (100%). After an hour of stable operation, gas flow rate was read and recorded as  $240m^3$ /hr from the gas metering unit. The air heater was not "ON" during the first stage of the experiment and as such, the combustion air temperature was measured to be 26 °C with a dry bulb thermometer in the production hall. Flow rate of air was indicated by the flowmeter and displayed on the Human Machine Interface (HMI). The air blower has a variable frequency drive (VFD) and inlet guide vane (IGV) that operate in both manual and automatic modes and controlled by the plant Programmable logic Controller (PLC) when in automatic mode to regulate flow rate or volume of air into the burner.

#### **Gas Analyzer Description**

Gas analyzer used were three, each with special feature to detect and measure specific gas in the boiler flue gases.

Testo 330 - 1LL gas analyzer measures the level of a target gas at an electrode through current output measurement. When a gas comes in a contact with the working electrode, a chemical reaction occurs at the electrode. The electric current generated is proportional to target gas concentration.

Servomex gas detector was used to detect and measure oxygen and carbon dioxide concentrations in flue gas. Its sensor and transducer module for measuring oxygen concentration works in the same principle as paramagnetic analyzers [7]. It measures carbon dioxide concentration using non- dispersive infra red (NDIR) absorption principle.

Teledyne hydrocarbon gas detector was used to identify and measures unburned hydrocarbons such as  $CH_4$ ,  $C_2H_4$ ,  $C_2H_6$ ,  $C_3H_8$ ,  $C_2H_2$  in flue gas .It is a microprocessor based, oven heated hydrocarbon analyzer which uses flame ionization detector and thermal conductivity meter to measure the amount of target hydrocarbon gas

## **Flue Gas Analysis**

Flue gas analysis can be performed efficiently by either chemical method or instrumental method. In this paper, instrumental method was employed to identify composition and concentration of each gas in flue gas sample. Such instruments used were Servomex process analyzer, Teledyne analytical instrument and Testo 330 - 1LL. These instruments were chosen for this analysis because of their suitability and sensitivity to detect trace components of exhaust gases.

## Measurement of Flue Gas Composition and Concentration

The process variables considered were percentage excess air and air inlet temperature. During the first stage of experiment, the ambient air flow rate was increased/ decreased through a regulating knob. This adjustment translated to varying percentage excess air since the inlet gas flow rate is constant at 240  $m^3$ /hr at burner full load. After one hour operation, the maximum, stable, flame temperature in the combustion chamber was taken.

Thereafter, the sample probe of Testo 330 analyzer was inserted into flue gas sampling point to identify and measure the concentration of CO,  $H_2O$ ,  $N_2$ , NO,  $NO_2$ , and  $SO_2$ . This analyzer also measured flue gas temperature. Again, the flue gas sampling point was connected to the servomex analyzer permanent sampling line to heat it up by purging. After about five minutes purging, the flue gas sample flow was reduced and made to flow into servomex analyzer for ten minutes. This analyzer identified and measured the concentration of  $O_2$  and  $CO_2$ .

In the same way, Teledyne analytical instrument was used to identify and measure the concentration of any unburned hydrocarbons in the flue gas after about  $1/\frac{1}{2}$  hours of flue gas flow. Identified and measured hydrocarbons were CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>2</sub>.For the second combustion process experiment, the combustion air temperature was varied to 40, 60, 80, 100, 110, 115, and 120 °C during stoichiometric and fuel – lean combustion conditions. Flue gas component and concentrations were also identified and measured using the analyzers mentioned above during each air temperature setting with air pre- heater.



Figure 1: Schematic Diagram of Experimental

Journal of Scientific and Engineering Research

#### **Measurement of Suspended Particulate Matter**

A Riken Keiki particulate monitor with model No NP -237H was used to measure or detect suspended particulate matter (PM<sub>10</sub>and PM<sub>5.0</sub>) in the flue gases at stoichiometric, excess air and deficient air combustion conditions

## **Results and Discussion**

Table 1: Concentrations of CO <sub>2</sub> , O <sub>2</sub> , CO, N <sub>2</sub> , H <sub>2</sub> O, NO AND NO <sub>2</sub> in Flue Gases with Percentage excess air												
Excess Air	15.04	12.00	9.47	8.48	7.06	5.44	2.40	0.10	-5	-10	-15	FEPA
(%)												
Constituents												
CO <sub>2</sub> (%)	7.00	7.50	8.10	8.36	8.76	9.30	10.60	11.80	7.40	3.60	0.10	10%
$O_2(\%)$	2.73	2.24	1.81	1.63	1.38	1.08	0.49	0.02	ND	ND	ND	-
$N_{2}(\%)$	73.8	73.6	73.4	73.2	73.0	72.8	72.6	72.5	72.1	72.0	71.7	-
NO $(mg/m^3)$	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1000mg/m <sup>3</sup>
$NO_{2}$ , (mg/m <sup>3</sup> )	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	1000mg/m <sup>3</sup>
H <sub>2</sub> O Conc.(%)	5.50	5.80	6.10	6.20	6.45	6.65	7.10	7.50	6.50	6.00	5.10	-
Flue gas temp. ( <sup>0</sup> c)	202	205	208	208	214	216	217	218	212	206	200	-
$CO (mg/m^3)$	ND	ND	ND	ND	ND	ND	ND	0.012	21.85	35.65	42.55	5mg/m <sup>3</sup>
Particulate matter	-	-	-	-	-	-	-	-	-	-	0.02	$250 \text{mg/m}^3$
ND N. I.												

N.D = Not detected

Table 2: Concentrations of CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, SO<sub>2</sub>, C<sub>2</sub>H<sub>2</sub> and C<sub>2</sub>H<sub>4</sub> in Flue gas with Percentage Excess Air

Excess Air	15.04	12.00	9.47	8.48	7.06	5.44	2.40	0.10	-5.00	-10.00	-15.00	FEPA
(%)												
Constituents												
$CH_4 (mg/m^3)$	ND	ND	ND	ND	ND	ND	ND	0.00	39.3	105.06	183.40	50mg/m <sup>3</sup>
$C_2H_6 (mg/m^3)$	ND	ND	ND	ND	ND	ND	ND	ND	12.27	73.99	177.91	$50 \text{mg/m}^3$
$C_{3}H_{8} (mg/m^{3})$	ND	ND	ND	ND	ND	ND	ND	ND	1.87	10.98	27.36	50mg/m <sup>3</sup>
$SO_2(mg/m^3)$	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	$3000 \text{mg/m}^3$
$C_2H_2 (mg/m^3)$	ND	ND	ND	ND	ND	ND	ND	ND	0.44	1.19	2.50	$50 \text{mg/m}^3$
$C_2H_4 (mg/m^3)$	ND	ND	ND	ND	ND	ND	ND	ND	0.06	0.29	0.80	$50 \text{mg/m}^3$
Flue Gas Temp.	202	205	208	208	214	216	217	218	212	206	201	-
(°C)												

N.D = Not detected

Table 3: Concentrations of O2, CO2 and CO in Flue gases with Combustion Air Temperature at Fuel Lean

-
Condition
CONTINUOL

Gas Flow (m <sup>3</sup> /hr)	Air Flow (m <sup>3</sup> /hr)	Air Temp. (°C)	Flame Temp. (°C)	O <sub>2</sub> Conc. (%)	CO <sub>2</sub> Conc. (%)	CO Conc. mg/m <sup>3</sup>
240	3595	28	331	2.73	7.01	-
240	3595	40	365	2.70	6.97	-
240	3595	60	379	2.68	6.91	-
240	3595	80	405	2.66	6.90	-
240	3595	100	442	2.64	6.88	-
240	3595	110	481	2.60	6.83	0.012
240	3595	115	493	2.52	6.80	0.120
240	3595	120	501	2.45	6.60	0.184

Table 4: Concentrations of CO<sub>2</sub>, CO and H<sub>2</sub>O in Flue Gas with Combustion Air Temperature at Stoichiometric

Region

Gas	Air Flow	Air Tomp	Flame Temp.	$CO_2$	CO Cono	H <sub>2</sub> O Conc.
(m <sup>3</sup> /hr)	(m <sup>3</sup> /hr)	(°C)	(°C)	(%)	mg/m <sup>3</sup>	(%)
240	3200	28	390	10.60	-	7.2
240	3200	40	415	10.30	1.84	6.0
240	3200	60	440	10.00	4.60	5.5
240	3200	80	466	9.80	9.20	5.1
240	3200	100	502	9.50	17.25	4.9
240	3200	110	516	9.00	23.00	4.7
240	3200	115	525	8.70	25.88	4.5
240	3200	120	532	8.30	28.75	4.2



		27	2		1			0
Expt.	Gas Flow	Air Flow	Air	Flame	$N_2$	NO	NO <sub>2</sub>	Flue Gas
No.	Rate	Rate	Temp.	Temp.	Conc.	Conc.	Conc.	Temp.
	(m <sup>°</sup> /hr)	(m <sup>2</sup> /hr)	(°C)	(°C)	(%)	mg/m <sup>o</sup>	mg/m <sup>°</sup>	(°C)
A1	240	3200	28	378	72.6	0.012	0.000	216
B1	240	3200	40	400	71.4	0.935	0.752	225
C1	240	3200	60	421	70.0	1.378	1.128	232
D1	240	3200	80	450	69.1	1.870	1.504	240
E1	240	3200	100	469	68.0	2.460	2.256	246
F1	240	3200	110	480	67.5	3.075	2.632	252
G1	240	3200	115	507	67.0	3.690	2.820	257
H1	240	3200	120	520	66.6	4.317	3.384	261
	Table 6	: Particulate m	atter sizes id	lentified at di	fferent comb	oustion cond	ditions	
	-				3			

Table !	5: (	Concentrations	of N <sub>2</sub> ,	NO	and NO	in F	lue Ga	s with	Air	Tem	perature	at S	Stoicl	niome	etric ]	Regi	on
			21								1					0	

Combustion condition	PM <sub>10</sub> (mg/m <sup>3</sup> )	$PM_{5.0} (mg/m^3)$
Fuel lean condition	0.00	0.00
Stoichiometric	0.00	0.00
Fuel- rich condition	0.00	0.02

# **Flue Gas Composition and Concentration**

The flue gas constituents identified during this study are shown in Tables 1 and 2. It was observed that during combustion with excess air, flue gas constituents were mostly nitrogen, oxygen and carbon dioxide. Nitrogen concentration in the flue gas was fair higher than that of oxygen and  $CO_2$  probably because of its volume in air and its inert nature.

But under combustion with inadequate volume of air, flue gas composition became more of CO and lower paraffins such as CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>. C<sub>3</sub>H<sub>8</sub> and other unsaturated hydrocarbons. Among the paraffins detected, methane and ethane concentrations in the flue gases were more than other higher paraffins in the flue gases. This may be attributed to the fact that methyl radical recombination is a direct one step path to stable paraffin formation. Higher concentration of methane than ethane from this study may also be attributed to the fact that the stability of paraffins decreases with increasing molecular weight.

# Effect of the Volume Flowrate of Air on the Concentrations of O<sub>2</sub>, CO<sub>2</sub> and Co in the Flue Gas

Table 1 above shows the concentration profile of oxygen  $(O_2)$ , at various percentage excess air. Air temperature was constant at 28 °C while the natural gas flow was constant at 240m<sup>3</sup>/hr. Experimental data show that oxygen level or amount in percentage decreases with decreasing excess air. The Oxygen concentration at stoichiometric region was almost zero where the air flow rate was 3128 m<sup>3</sup>/hr and percentage excess air of 0.10. The result further shows that as combustion process goes into fuel-rich region (region with inadequate air), no oxygen was found in flue gases. This means that the volume of air/oxygen supplied was grossly inadequate to oxidize all the natural gas molecules. The increasing concentration of oxygen in flue gases as percentage excess air increases is owing to unused oxygen from excess air.

However, carbon monoxide concentration increases as percentage excess air decreases from stoichiometric region to fuel-rich region. No carbon monoxide was found in flue gases as excess air increases from stoichiometric region to fuel-lean condition. The reason is that there was enough molecules of oxygen to completely oxidize all the molecules of natural gas into Carbon dioxide and water vapor. The concentration of carbon monoxide in flue gases in part per million volume at various percentage excess air is also shown in Table 1 above.

For Carbon dioxide (CO<sub>2</sub>), experimental results showed that its concentration in flue gases is maximum at the stoichiometric region (where exact theoretical volume of air was supplied). On the fuel-lean region, its concentration decreases as the excess air increases. This is owing to the dilution effect of so much nitrogen and oxygen as the percentage excess air increases. Again, at the fuel-rich region, CO<sub>2</sub>concentration in flue gases decreases as the excess air decreases. The reason is practically because of inadequate air to oxidize intermediate product (carbon monoxide) into carbon dioxide. This observation on the concentration of CO<sub>2</sub> at the deficient and excess air conditions is depicted in Table 1.



#### Effect of Air Temperature on O<sub>2</sub>, Co and CO<sub>2</sub> Concentrations in the Exhaust Gas

The experimental results on Table 4 show that combustion chamber temperature increases as the air temperature increases. Oxygen and carbon dioxide concentrations decrease slightly as the air temperature increases in region of excess air (Table 3). But the concentration profile of carbon monoxide on Table 4 shows a sharp increase at air temperature of 40 °C and above at stoichiometric region. This increase occurred despite presence of little excess air. It therefore proves that the concentration of Carbon monoxide is also temperature dependent. This increase in CO concentration and slight decrease in  $CO_2$  concentration is attributed to decomposition of carbon dioxide at elevated temperature.

# Effect of Volume and Temperature of Air on the Concentrations of N<sub>2</sub>, NO, NO<sub>2</sub>, H<sub>2</sub>O and SO<sub>2</sub> in the Flue Gas

From experimental data on Table 1, the concentration profile for Nitrogen  $(N_2)$  in the flue gas increases as the percentage excess air supplied for combustion increases. This marginal, steady increase in  $N_2$  concentration is attributed to the fact that nitrogen is inert and does not partake in combustion process.

Another reason for the observed increase in  $N_2$  concentration is that the flame temperature is not enough to cause thermal decomposition of nitrogen and formation of nitrogen oxides. Thus, oxides of nitrogen (NO and NO<sub>2</sub>) were not found in the flue gas even as excess air increases or decreases at ambient temperature of 28 °C. Again, oxides of sulphur are often known components of exhaust gases. From the experimental data on Table 2, oxide of sulphur was not detected in flue gases. The reason for this observation may be linked to the absence of sulphur or any of its compounds in the natural gas under study

However, the effect of air temperature on the concentrations of nitrogen ( $N_2$ ), nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>) was very noticeable. From the experimental data on Table 6, nitrogen N<sub>2</sub>concentration in the exhaust gases decreases slightly as the combustion air temperature increases from 28 °C to 120 °C around stoichiometric condition. Table 5 shows the variation of N<sub>2</sub> concentration in flue gas at different air temperatures.

The concentration profiles for nitric oxide and nitrogen dioxide increase sharply as air and flame temperature increases. Concentration of these air pollutants are therefore temperature dependent. In fact, the decreasing concentration of nitrogen and increasing concentration of its oxides can perfectly be linked to thermal decomposition of nitrogen at high temperature and subsequent reaction with oxygen atoms to form nitrogen oxides. This experimental data showing sharp increase in concentrations of nitrogen oxides (NO and NO<sub>2</sub>) as air temperature increases is in line with thermodynamic view that higher flame temperature in the combustion chamber favour the formation of oxides of nitrogen. Table 5 shows the effect of increasing combustion air temperature on the concentration of trace constituents (NO and NO<sub>2</sub>) in the flue gas.

#### Effect of Natural Gas Combustion on Environmental Performance

The concentrations of  $CH_4$ , CO, NO,  $NO_2$ ,  $SO_2$  and NMHC, from this paper are below their emission limit level of  $50 \text{mg/m}^3$ ,  $5 \text{ mg/m}^3$ ,  $1000 \text{ mg/m}^3$ ,  $300 \text{ mg/m}^3$  and  $50 \text{mg/m}^3$  respectively [8]. The only exception is  $CO_2$  concentration, which is 18% above the FEPA limit of 10% by volume. Carbon dioxide is a pollutant, as well as a heat trapping gas that constitute global warming. From the above comparison, it can be seem that the combustion of Nigerian natural gas in industrial burners is environmental friendly except for the volume of  $CO_2$ released.  $CO_2$  released during combustion can be absorbed with mono ethanolamine solution and further purified into food grade  $CO_2$ . The purified  $CO_2$  is extensively used in breweries, bottling and beverage companie5 and in dry ice production.

However, the results on Table 6 show that suspended particulate matter from natural gas combustion is very insignificant but tends to increase at fuel – rich condition. The range of the detection of the instrument is  $0-2.5 \text{ mg/m}^3$ .

#### Conclusion

The combustion of Nigerian natural gas in an industrial boiler under various temperatures and aeration conditions have been studied. The combustion chamber temperatures were observed to generally increase with

increasing air temperature at a given air-natural gas ratio. The rising combustion chamber temperature was instrumental to the formation of some critical air pollutants such as nitrogen oxides. Carbon monoxide (CO) formation increases as the air-gas mixture becomes gas-rich and decreases with increasing excess air. Its concentration in flue gases shows a strong dependence on the volume/flow rate of combustion air.

Oxides of nitrogen (NO and  $NO_2$ ) formation and level in flue gases demonstrate their strong dependence oncombustion chamber temperature. Their peak concentration values were observed at conditions generating highest temperature. The concentration of these pollutants were very small and within the emission limits.

Carbon dioxide  $(CO_2)$  is highest around stoichiometric region at ambient condition. Its concentration in the flue gases decreases with increasing percentage excess/deficient combustion air and flame temperature. The maximum carbon dioxide concentration of 11.8% by volume is higher than the FEPA emission limit of 10.0% by volume.

Particulate matter ( $PM_5$  and  $PM_{10}$ ) concentration levels in flue gases are very negligible and far below 250mg/m<sup>3</sup>limit set by FEPA. Sulphur oxides were not detected in the flue gases. This is owing to the fact that Nigerian natural gas does not contain sulphur or any of its compounds.

This paper reveals that the contribution to air pollution and global warming arising from the combustion of Nigeria natural gas in boilers is insignificant under stoichiometric or excess air condition.

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