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Air-Steam Gasification of Nigerian Lignite Coals for Hydrogen and Syngas Production

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Abstract - Coal is the fuel that drives most emerging economies. The gasification along with carbon capture and sequestration could ameliorate the harmful effects of coal utilisation. In this study, two recently discovered lignite coals; Obomkpa (BMK) and Ihioma (IHM) were examined for hydrogen and synthesis gas production through air-steam gasification using a non-stoichiometric model simulated in ASPEN Plus. The results showed that H₂ production from BMK and IHM was highly sensitive to temperature and air but moderately sensitive to steam during gasification. The optimal operating conditions for BMK and IHM are; temperature 1125 °C and 1350 °C; equivalence ratio (ER) 0.26 and 0.26; steam/carbon ratio (S/C) 2.25 and 2.19, respectively. The optimal gas composition for BMK was; H₂ (0.66), CO (0.18) and CO₂ (0.18) mole-fraction, whereas for IHM was; H₂ (0.65), CO (0.17) and CO₂ (0.17). The findings indicate that both lignite coals are suitable feedstock for H₂ and syngas production.

Keywords: Air-Steam, Gasification, Lignite Coal, Hydrogen, Syngas, Nigeria.

1. Introduction

Coal is the solid fossil fuel that birthed the industrial revolution and sustains human civilisation (OECD Working Paper 2012; Freese 2016). It is the most abundant and distributed solid fossil fuel in the world. Hence, coal accounts for over 60% of all economically recoverable energy resource compared to 19% and 17% for petroleum and natural gas (IEA 2018). Besides, coal accounts for over 40% or 8,200 terawatt-hours (TWh) of the electricity generated annually around the globe. Hence, coal is considered the cornerstone of global electricity and plays a crucial role in the global energy mix (OECD Working Paper 2012). However, over the years, there has been increased objection to the utilisation of coal for power generation due to its adverse effects on the environment (Fallows 2010). Intrinsically, many emerging and developed economies still depend on coal as the fuel that drives growth and sustains development.

One way of ameliorating the adverse effects of coal on the environment is the conversion into environmentally friendly fuels and chemicals such as syngas ($\text{H}_2 + \text{CO}$) with simultaneous carbon dioxide (CO_2) capture and sequestration. The most widely used process for converting coal into syngas is gasification. Gasification is a thermochemical process in which heat is utilised to convert carbonaceous feedstocks such as biomass, coal and petroleum coke into useful gaseous products and chemicals (Higman and van der Burgt 2008; Speight 2012). Over the years, numerous empirical studies have investigated coal gasification for syngas and synthetic natural gas (SNG) production. Recently, the mathematical modelling and simulation of coal gasification using the non-stoichiometry model and Gibbs free analysis can satisfactorily predict the product gas yield and composition.

Duan et al. (2015) simulated coal gasification with slag in the presence of steam in a blast furnace with a constraint on mass and energy for heat recovery. The study showed that clean syngas could be effectively recovered from the process. The flow processes for

pyrolysis, gasification, and combustion units from ASPEN Plus were integrated into the Texaco coal gasifier for the comprehensive conversion of coal into product gas. The simulation successfully predicted the product gas composition, carbon conversion, and gasification temperatures (Kong et al. 2013). Preciado et al. (2012) investigated the steam and oxy-fuel gasification of coal from Colombia through steady-state mathematical modelling and simulation. The findings showed that the ratio of oxygen to carbon is a crucial factor in the process. Similarly, the fixed bed catalytic coal gasification process of Exxon was successfully modelled and simulated, and the kinetic analysis result validated experimentally by Jang et al. (2013).

The ASPEN simulation of the catalytic co-gasification of coal and organic wastewater with an alkaline pH was investigated by Xiao et al. (2019) using a fluidised bed gasifier. The findings revealed that the optimal ratio of steam to coal, temperature, and equivalence ratio for the process. The co-firing of coal and dried sewage sludge (DSS) through gasification was examined in a dual-step gasification reactor comprising a fluidised bed and tar-cracking reactors (Jeong et al. 2019). The findings showed that the product gas composition improved with increasing mixing ratio of coal and DSS. Overall, the review of the literature indicates that modelling and simulation of various ranks of coal can be effectively utilised to predict product gas, yield, and composition.

Over the years, massive new deposits of coal have been discovered in Nigeria. As a result, there is renewed attention in converting the nation's coal resources into chemicals, industrial materials, and electricity (Nyakuma and Jauro 2016; Ryemshak and Jauro 2013; Nyakuma 2016). The nation's proven coal reserves are estimated at 640 million tonnes, whereas the projected resources are approximately 2.8 billion tonnes (Chukwu et al. 2016). The distribution of Nigeria's reserves of coal are comprised of lignite (12%), sub-bituminous (49%), and bituminous (39%), which are distributed across the six geopolitical zones (Obaje

2009). Despite the abundant resources in the country, industrial utilisation of coal is virtually non-existent (Nyakuma 2019; Ohimain 2014).

Amidst the growing calls for energy generation from renewable and sustainable sources, cleaner technologies are required for the utilisation of Nigeria's coal resources (Oboirien et al. 2018; Amoo 2015). The gasification of low-ranked coals such as lignite is a potential alternative to pulverised coal combustion (PCC) widely utilised for power generation. However, there is currently no study in the literature on the gasification of lignite coals from Nigeria. Furthermore, the product gas yield, composition, and optimal operating conditions for the gasification of Nigerian lignite coals are yet to be determined by researchers in the field.

Therefore, this study presents novel findings on the modelling and simulation of air-stream gasification of two newly discovered lignite coals from Obomkpa (BMK) and Ihioma (IHM) using ASPEN Plus. Likewise, the hydrogen and syngas production potential of BMK and IHM coals are investigated based on empirical data on ultimate, proximate, and sulphanal analyses. Lastly, the sensitivity and optimisation analyses of the lignite samples were performed in ASPEN Plus to determine the optimal operating gasification conditions for future application.

2. Theory and Methods

2.1 Theory

The process was modelled and simulated by reducing the Gibbs free energy of all the constituents involved in the primary gasification reactions. Typically, these reactions consist of water gas shift, steam reforming, and methanation. The Gibbs free energy for the N species ($i = 1 \dots N$) is based on the fundamental equation presented as (Basu 2010):

$$G_{total} = \sum_{i=1}^N n_i \Delta G_{f,i}^0 + \sum_{i=1}^N n_i RT \ln \left[\frac{n_i}{\sum n_i} \right] \quad (1)$$

Where the term $\Delta G_{f,i}^0$ denotes the Gibbs free energy for the formation of the i th species at standard pressure. The minimisation of the objective function was performed by solving n_i in Eq. 1. The carbon content deduced from ultimate analysis must equal the total carbon in the mixture of gases. Therefore, the j th component is expressed by the relation;

$$\sum_{i=1}^N a_{i,j} n_i = A_j \quad (2)$$

Where $a_{i,j}$ represents the atoms of the j th element of the i th species, whereas A_j is the total amount of atoms of the j element in going to the reactor. Based on the Lagrange multiplier, the term λ methods is given by the relation:

$$L = G_{total} - \sum_{j=1}^K \lambda_j \left(\sum_{i=1}^N a_{i,j} n_i - A_j \right) \quad (3)$$

Therefore, the final values can be deduced by substituting Eq. 3 into Eq. 1;

$$\frac{\partial L}{\partial n_i} = \frac{\Delta G_{f,i}^0}{RT} + \sum_{i=1}^N \ln \left(\frac{n_i}{n_{total}} \right) + \frac{1}{RT} \sum_{j=1}^K \lambda_j \left(\sum_{i=1}^N a_{i,j} n_i \right) \quad (4)$$

In this study, the critical products considered were H₂, CO, CO₂, CH₄, C (graphite) and excess steam during the reforming reaction. Besides, the prospective ancillary constituents identified were; ethane, ethylene, acetylene, and ethanol not previously reported in the literature (Vagia and Lemonidou 2007). In practice, the secondary products are considered precursors of coke despite their presence in minor concentration. Conversely, the high content of carbon in the constituent reactions was taken into account in this study. The

steam reforming of coal is governed primarily by the following reactions; water gas shift, steam reforming, and methanation, which are presented as (Liu et al. 2013):

Combining Eq. 5 and Eq. 6 gives the overall reaction for coal steam reforming in this study;

Lastly, the product gas and yield of the reaction are comprised of H_2 , CO , CO_2 and CH_4 , as computed from to Eq. (8).

2.2 Process Description

The simulation of the coal gasification process was developed in ASPEN software. The process flow sheet depicted in Figure 1 consists of a three-model unit: the RYield (DECOMP), RGibbs (BURN), Separator (SEPARATE) and Calculator unit.

Figure 1. Flowsheet of Lignite Coal Gasification in ASPEN

The RYield was used to convert the proximate, ultimate, and sulphanal analyses of the lignite coals (presented in Table 1) into potential chemical compounds. On the other hand, the Calculator module was employed to normalise the output of the RYield comprising; water (H_2O), ash, carbon (C), hydrogen (H_2), nitrogen (N_2), chlorine (Cl_2), sulphur (S) and oxygen (O_2). The outlined components serve as the inlet components for the RGibbs or gasification unit. The Gibb free equilibrium analysis was used to compute the moles of each component present in the reactor at the specified operating conditions. Apart from the inlet components from the RYield, the products in the RGibbs consist of all potential gasification gas products, namely; hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4) and

ethylene (C_2H_4). The product of the RGibbs unit was then separated in the stream separator into gases and solid products of the gasification process.

Table 1. Proximate, Ultimate and Sulphanal analysis of BMK and IHM (Nyakuma et al. 2017)

3. Results and Discussion

3.1 Obomkpa (BMK) coal analysis

3.1.1 Effect of temperature on gas composition

The effect of temperature on the product gas yield and composition of H_2 , CO_2 , CO , and CH_4 during the gasification of BMK is presented in Figure 2. For the simulation, the air (oxygen) and steam values were fixed at 10 kmol and 1500 kg/h, respectively. The findings indicate that the composition of H_2 rapidly increases with temperature from zero at 200 °C to the maximum mole-fraction of 0.71 at 650 °C. Similarly, the composition of CO increased from 0.0 – 0.07 in the temperature range from 200 °C to 600 °C, whereas the CO_2 and CH_4 decreased from 0.61 – 0.22 and 0.35 – 0.0, respectively (Oladokun et al. 2016). The findings suggest that the temperature range from 200 °C to 600 °C favours all three major reactions reforming reactions. In addition, it accounts for why the composition of CH_4 is higher than CO (Oladokun et al. 2017). However, a further increase in temperature from 600 °C to 1600 °C resulted in a decline in H_2 and CO_2 compositions to 0.63 and 0.14 mole-fraction or 3% and 2.5%, respectively.

Figure 2. BMK Product Gas Composition variation with Temperature

However, the CO increased steadily from 0.07 to 0.22 mole-fraction. This can be explained by the increase in temperature, which favours the steam reforming reaction and the reversible methanation reaction resulting in more CO production. Lastly, the optimal operating conditions for the BMK gasification were determined. The optimal temperature is defined as the lowest temperature when CO is greater and/or equal to CO_2 . For the

gasification of BMK, the optimal temperature deduced at ER (0.26), S/C (2.3), Pressure (1 bar) is 1125 °C based on Figure 2. Hence, the optimal gas composition at the optimum temperature was; H₂ (0.66), CO (0.17), CO₂ (0.17) and CH₄ (0.0).

3.1.2 Effect of ER and S/C on H₂ composition

Figure 3 presents the effects of ER and S/C on the H₂ composition (Mole-fraction) at the simulated temperature and pressure conditions of 1100 °C and 1 bar, respectively.

Figure 3. BMK H₂ Mole-fraction variation with ER and S/C

The findings indicate that the composition of H₂ decreased at higher equivalence ratio (ER) for all steam to carbon (S/C) ratios. In addition, the profile revealed that changes to the S/C at ER 1.05 had an insignificant effect on the H₂ composition. The ER at 1.05 is the maximum range for BMK gasification and the beginning of combustion, as observed by the absence of H₂. Furthermore, the composition of H₂ increased with the increase in steam due to the steam reforming reaction. However, the percentage H₂ decreased with an increase in S/C for all the ER considered. The optimal S/C determined at 2.25 was the point at which the sensitivity of the H₂ composition was insignificant. Therefore, the H₂ composition at the optimum S/C of 2.25 was 0.74 at the ER of 0.0 for the maximum value, whereas the minimum H₂ was 0.0 at the ER of 1.05. The ER for gasification occurs at values above 0.2 but below 1.0. Therefore, the range of H₂ composition is 0.66 at ER 0.26 and 0.54 at the ER of 0.52.

3.1.3 Effect of S/C on H₂ Composition

The profile of H₂ composition with S/C and ER at a constant temperature of 1100 °C and pressure of 1 bar is presented in Figure 4. The increase in steam caused a corresponding increase in H₂ production for all the ER except for ER higher than 0.92. As explained earlier,

H₂ will not be produced when the value of ER is at 0.92 despite an increase in steam. The sensitivity of H₂ composition to S/C was appreciable from 0 till 2.25. However, the percentage increase was subsequently constant with further increase in S/C for all ER values.

Figure 4. BMK H₂ Mole-fraction variation with SC and ER

Based on the comparison of the profiles for all ER at S/C of 2.25, the composition of H₂ decreased with increasing ER. The optimum ER was determined at the least significant percentage decrease, which was between 0.26 and 0.39. The composition of H₂ at the optimum range of ER (0.26 – 0.39) was 0.66 – 0.61.

3.2 Ihioma (IHM) coal analysis

3.2.1 Effect of temperature on gas composition

Figure 5 shows the profile of the gas product composition of IHM gasification with temperature, Air at 10 kmol and Steam at 2000 kg/h. The temperature profile is divided into two regions beginning with the rapid increase in H₂ production (200 °C to 650 °C) and the region of significant CO production (650 °C to 1600 °C).

Figure 5. IHM Product Gas Composition variation with Temperature

The first region was characterised by the three gasification reactions: steam reforming, water gas shift, and methanation reactions. Similarly, the second region experienced similar effects; however, the water shift and methanation reactions favoured the backward reaction of the reversible reaction. The gas compositions generated from the first region showed that the H₂ mole-fraction increased from 0.03 to 0.71, whereas the mole fraction of CO was 0.0 in this region until 500 °C but increased to 0.05 at 650 °C.

The composition of CO₂ and CH₄ in the region from 200 °C to 650 °C showed a rapid decrease in composition compared to the rapid increase in H₂ composition. Similarly, the

CO₂ decreased from 0.62 mole-fraction to 0.24, whereas the CH₄ decreased from 0.33 to 0.0. The second region from 650 °C to 1600 °C showed a steady rise in the CO composition from 0.05 to 0.20, whereas the compositions of H₂, CO₂, and CH₄ decreased to 0.63, 0.16, and 0.0, respectively. Hence, the optimum temperature for IHM gasification was 1350 °C, which was determined when the minimum temperature of CO was above and/or equal to CO₂. The optimal gas composition at these conditions was H₂ (0.65), CO (0.17), CO₂ (0.17), and CH₄ (0.0).

3.2.2 Effect of ER and S/C on H₂ Composition

Figure 6 presents the profile of H₂ composition with the variation in ER for IHM at the simulated temperature of 1200 °C and pressure of 1 bar. As observed, the composition of H₂ decreased with increasing ER but increased with S/C.

Figure 6. H₂ Mole-fraction variation with ER and S/C

Furthermore, the results showed that the composition of H₂ became zero at the ER gasification limit of 1.06 for all the S/C values considered in the study. The optimum S/C from the profile was 2.19 and determined from the first least significant percentage change. The composition of H₂ was in the range from 0.64 to 0.05 for the ER range between 0.26 and 0.93 during gasification and the profile of H₂ composition at an S/C of 2.19.

3.2.3 Effect of S/C and ER on H₂ Composition

The composition of H₂ at the simulated temperature of 1200 °C and pressure of 1 bar is presented in Figure 7. As observed, the H₂ composition increased with increasing S/C but decreased with increasing ER.

Figure 7. H₂ Mole-fraction variation with S/C and ER

The limiting S/C was 2.19 for all ER values examined in the study. The S/C limit was determined at the point where further supply in steam did not result in any significant increase in the production of H_2 via the steam reforming reaction. Based on the optimum S/C of 2.19 determined for IHM earlier, the H_2 composition decreased from 0.64 to 0.05 at the ER values of 0.26 and 0.93, respectively. However, this does not include the values of ER greater than or equal to one, which represents combustion. The ER limit is between 0.26 and 0.53, and the optimum ER is determined as 0.26.

4 Conclusion

The air-steam gasification of two newly discovered lignite coals from Obomkpa (BMK) and Ihioma (IHM) in Nigeria was successfully modelled in ASPEN. For the gasification process, a non-stoichiometric model was developed based on a combination of model units and the Gibbs free energy reactor (RGibbs). The proposed ASPEN model was used to investigate the hydrogen and syngas potential of BMK and IHM. The results showed that BMK and IHM are highly sensitive to temperature and equivalence ratio (ER) but only moderately sensitive to steam/carbon (S/C) ratio. The optimal gasification temperature for maximum hydrogen and syngas production at minimum CO_2 production was 1125 °C for BMK and 1350 °C for IHM. The gas composition at the optimal operating conditions showed higher H_2 values (or 26% increase) compared to CO and CO_2 for the samples. Overall, the result showed that BMK and IHM are suitable feedstock for the production of hydrogen and syngas through gasification.

Declarations section

Availability of data and materials

The authors hereby declare that all the data used in this manuscript is ours and provided within the manuscript except otherwise referenced thereof.

Competing interests

The authors hereby declare that there are ZERO competing.

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Authors' contributions

Olagoke Oladokun – Original Manuscript Writing, Mathematical Modelling, Data Analysis;

Bemgba B. Nyakuma – Original Manuscript Writing, Editing, Fact Checking, Formatting and Submission.

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Figures

Figure 1

Flowsheet of Lignite Coal Gasification in ASPEN

Figure 2

BMK Product Gas Composition variation with Temperature

Figure 3

BMK H2 Mole-fraction variation with ER and S/C

Figure 4

BMK H2 Mole-fraction variation with SC and ER

Figure 5

IHM Product Gas Composition variation with Temperature

Figure 6

IHM H₂ Mole-fraction variation with ER and S/C

Figure 7

IHM H2 Mole-fraction variation with S/C and ER