Petroleum and Coal

Article **Open Access**

Gasification of Nigerian Lignite Coals under Air-Steam Conditions using ASPEN Plus for the Production of Hydrogen and Syngas

Olagoke Oladokun [1], Bemgba B. Nyakuma [2], Tertsegha J-P. Ivase [3]

[1] *Department of Chemical Engineering, College of Engineering,Covenant University, Canaan Land, Ota, Ogun State, Nigeria*
[2] *Research Initiative for Sustainable Energy Technologies (RISET), North-Bank, Makurdi, Benue State, Nigeria*
[3] *Bio-Resource Development Centre, National Biotechnology Development Agency, Makurdi, Benue State, Nigeria*

Received November 18, 2020; Accepted January 26, 2021

Abstract

Coal is the fuel that drives most emerging economies. 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 the production of hydrogen (H_2) and synthesis gas (syngas) under air-steam gasification conditions through a non-stoichiometric model simulated in ASPEN Plus. Results revealed that the production of H_2 from BMK and IHM was highly sensitive to temperature and air but moderately sensitive to steam during the gasification process. The optimal conditions for gasification of BMK and IHM are; temperatures of 1125°C and 1350°C, equivalence ratio (ER) of 0.26, steam/carbon ratio (S/C) 2.25 and 2.19, respectively. The optimal composition of gases for BMK are; H_2 (0.66), CO (0.18) and CO_2 (0.18) mole-fraction, whereas it was; H_2 (0.65), CO (0.17) and CO_2 (0.17) for IHM. Overall, the results of the study showed that the lignite coals are suitable feedstock for the production of H_2 and syngas.

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

1. Introduction

Coal is the fossil-based fuel that birthed the industrial revolution and sustains human civilisation [1-2]. It is the most abundant and distributed solid fossil fuel on the planet. As a result, coal accounts for above 60% of all economically recoverable energy resources compared to 19% and 17% for petroleum and natural gas [3]. Furthermore, coal accounts for over 38% or 8200 terawatt-hours (TWh) of the electric power generated yearly worldwide. It is thus considered the keystone of global electricity and plays a crucial role in the world-wide energy mix [1]. Conversely, there has been increased objection to the utilisation of coal for power generation over the years, which is largely due to its adverse effects on the environment [4]. Intrinsically, many emerging and developed economies still depend on coal as the fuel that drives economic 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 synthesis gas or syngas (H_2+CO) with simultaneous carbon dioxide (CO_2) capture and sequestration. The most widely used route for the conversion of 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 [5-6]. Numerous empirical studies have investigated the gasification of coal 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, built into software packages such as ASPEN Plus, can satisfactorily predict the product gas yield and composition.

The ASPEN simulation of the catalytic co-gasification of coal and organic wastewater with an alkaline pH was investigated by Xiao *et al.,* [7] 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 fluidised bed and tar-cracking reactors [8]. The findings showed that the product gas composition improved with increasing mixing ratio of coal and DSS. Duan *et al.,* [9] simulated coal gasification with slag in the presence of steam in a blast furnace with a constraint on mass and energy for the recovery of heat. 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 coal conversion into product gas. The simulation successfully predicted the product gas composition, carbon conversion, and gasification temperatures [10]. Preciado *et al.,* [11] 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 modelled and simulated successfully with the kinetic analysis certified experimentally [12]. 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.

The discovery of massive new coal deposits has reinvigorated the energy industry in Nigeria. Hence, there is a renewed focus on converting the country's coal resources into chemicals, industrial materials, and electricity [13-15]. The nation's proven coal reserves are estimated at 640 million tonnes, whereas the projected resources are approximately 2.8 billion tonnes [16]. The distribution of Nigeria's reserves of coal are comprised of lignite (12%), sub-bituminous (49%), and bituminous (39%), which are spread throughout the nation's geopolitical zones [17]. Despite Nigeria's abundant resources, the industrial-scale utilisation of coal is almost absent [18-19]. Amidst the growing calls for energy generation from renewable and sustainable sources, cleaner technologies are likewise necessary for the exploitation of the coal resources in Nigeria [20-21]. The gasification of low-ranked coals (LRC) such as lignite is a potential alternative to pulverised coal combustion (PCC) widely utilised for power generation worldwide.

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. Finally, the sensitivity analyses and optimisation of the samples were performed to determine the optimal operating conditions of gasification for future applications.

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…N) is based on the fundamental equation presented as [22]:

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

where the term $\Delta G_{f.i}^{0}$ denotes the Gibbs free energy for the formation of the ith 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;

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

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:

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

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

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

In this study, the critical products considered were H_2, CO, CO_2, CH_4, 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 [23]. 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 [24]:

$C_xH_yO_z + (x - z)H_2O \boxed{\leftrightarrow} xCO + (x + y/2 - z)H_2$ (5)
$CO + H_2O \boxed{\leftrightarrow} CO_2 + H_2$ (6)
$CO + 3H_2 \boxed{\leftrightarrow} CH_4 + H_2O$ (7)

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

$C_xH_yO_z + (2x - z)H_2O \boxed{\leftrightarrow} xCO_2 + (2x + y/2 - z)H_2$ (8)

Lastly, the product gas and yield of the reaction 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.

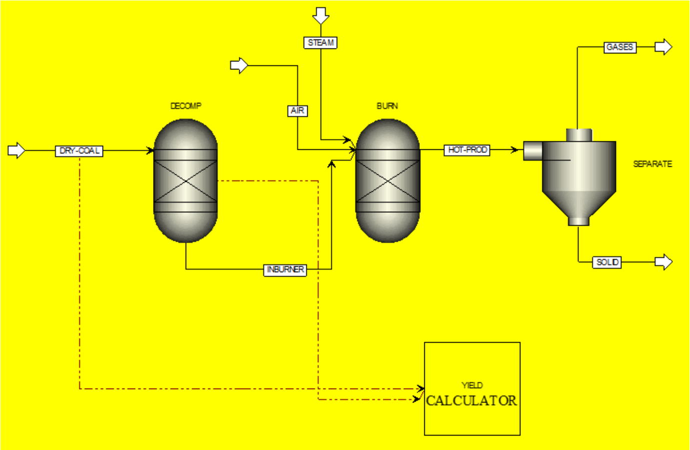

Figure 1. Flowsheet of lignite coal gasification in ASPEN

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 [25]

Analyses	Element	Symbol	Obomkpa (BMK, wt.%)	Ihioma (IHM, wt.%)
Ultimate	Carbon	C	44.40	45.60
	Hydrogen	H	5.00	5.30
	Nitrogen	N	0.50	0.60
	Sulphur	S	0.90	1.50
	Oxygen	O	37.40	44.50
	Chlorine	Cl	0.10	0.10
Proximate	Moisture	M	3.70	4.80
	Volatile Matter	VM	58.00	69.50
	Ash	A	11.70	2.40
	Fixed Carbon	FC	26.60	23.30
Sulphanal	Pyritic	S1	0.41	0.68
	Sulfate	S2	0.09	0.15
	Organic	S3	0.40	0.67

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.

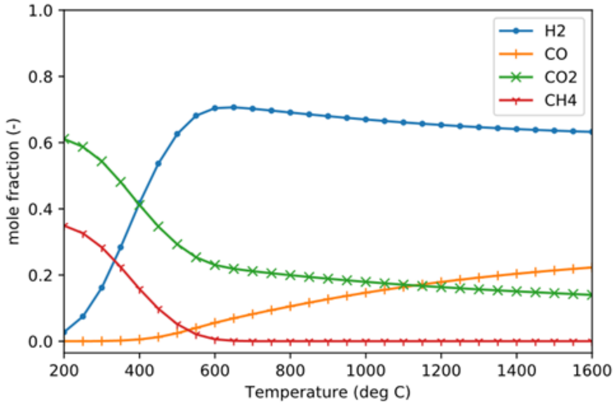

Figure 2. BMK product gas composition variation with temperature

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 [26]. The findings suggest that the temperature range from 200°C to 600°C favours all three major reforming reactions. In addition, this accounts for why the composition of CH_4 is higher than CO [27]. 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.

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; H_2 (0.66), CO (0.17), CO_2 (0.17) and CH_4 (0.0).

3.1.2. Effect of ER and S/C on H_2 composition

Figure 3 presents the effects of ER and S/C on the H_2 composition (Mole-fraction) at the simulated temperature and pressure conditions of 1100°C and 1 bar, respectively.

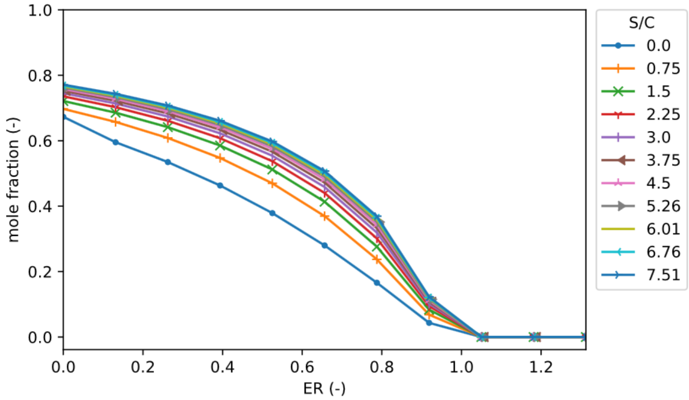

Figure 3. BMK H_2 Mole-fraction variation with ER and S/C

The findings indicate that the composition of H_2 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 H_2 composition. The ER at 1.05 is the maximum range for BMK gasification and the beginning of combustion, as observed by the absence of H_2. Furthermore, the composition of H_2 increased with the increase in steam due to the steam reforming reaction.

However, the percentage H_2 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 H_2 composition was insignificant. Therefore, the H_2 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 H_2 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 H_2 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 H_2 composition

The profile of H_2 composition with S/C and ER at a constant temperature of 1100°C and pressure of 1 bar is presented in Figure 4.

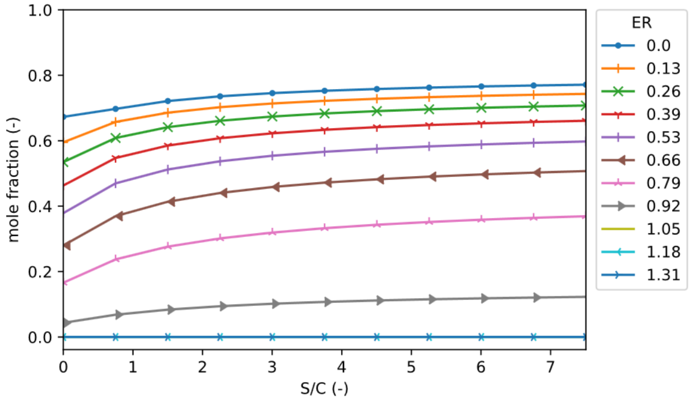

Figure 4. BMK H_2 Mole-fraction variation with SC and ER

The increase in steam caused a corresponding increase in H_2 production for all the ER except for ER higher than 0.92. As explained earlier, H_2 will not be produced when the value of ER is at 0.92 despite an increase in steam. The sensitivity of H_2 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.

Based on the comparison of the profiles for all ER at S/C of 2.25, the composition of H_2 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 H_2 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 H_2 production (200°C to 650°C) and the region of significant CO production (650°C to 1600°C).

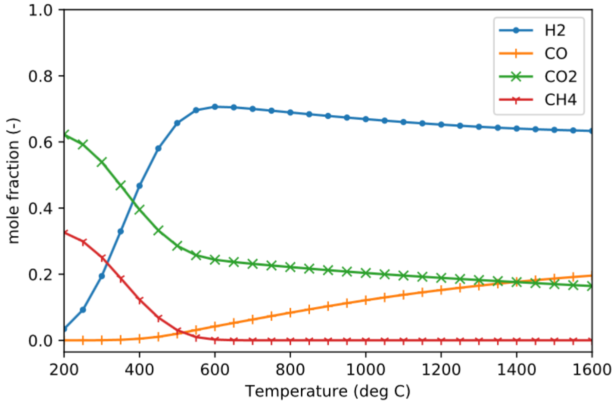

Figure 5. IHM product gas composition variation with temperature

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 H_2 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 CO_2 and CH_4 in the region from 200°C to 650°C showed a rapid decrease in composition compared to the rapid increase in H_2 composition. Similarly, the CO_2 decreased from 0.62 mole-fraction to 0.24, whereas the CH_4 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 H_2, CO_2, and CH_4 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 CO_2. The optimal gas composition at these conditions was H_2 (0.65), CO (0.17), CO_2 (0.17), and CH_4 (0.0).

3.2.2. Effect of ER and S/C on H_2 composition

Figure 6 presents the profile of H_2 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 H_2 decreased with increasing ER but increased with S/C.

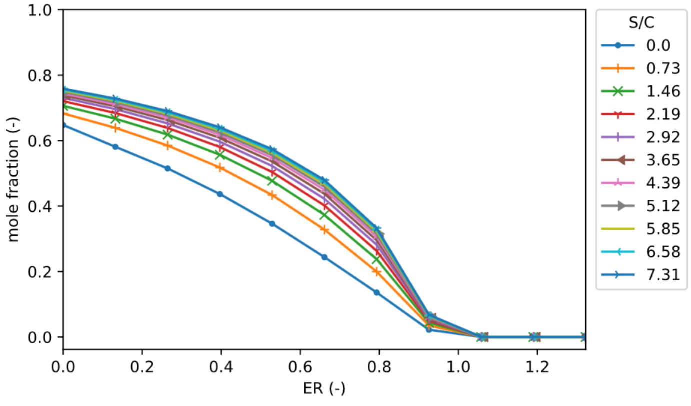

Figure 6. H_2 mole-fraction variation with ER and S/C

Furthermore, the results showed that the composition of H_2 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 H_2 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 H_2 composition at an S/C of 2.19.

3.2.3. Effect of S/C and ER on H_2 composition

The composition of H_2 at the simulated temperature of 1200°C and pressure of 1 bar is presented in Figure 7. As observed, the H_2 composition increased with increasing S/C but decreased with increasing ER.

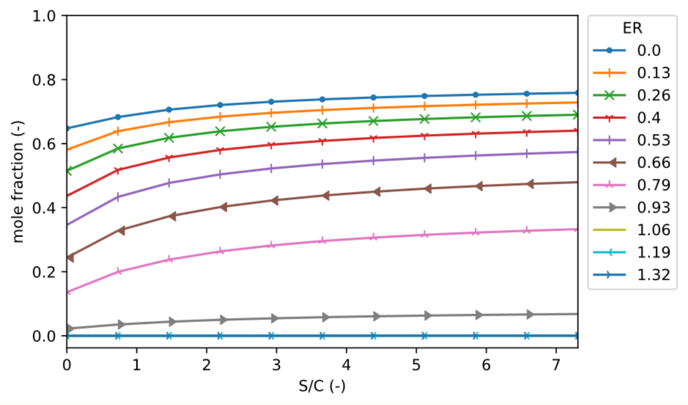

Figure 7. H_2 mole-fraction variation with S/C and ER

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 between0.26 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 results showed that BMK and IHM are suitable feedstock for the production of hydrogen and syngas through gasification. Future studies can be performed to examine the product yield, distribution and waste profiles of other ranks of Nigeria coals through mathematical simulation and experimental studies.

References

[1] OECD Working Paper, The Global Value of Coal 2012.
[2] Freese, B. Coal: A Human History. 2016.
[3] IEA, Market Report Series: Coal 2018. 2018; International Energy Agency: Paris, France.
[4] Fallows J. Dirty coal, clean future. The Atlantic, 2010; 306(4): 64-78.
[5] Higman C, and van der Burgt M. Gasifiation 2008.
[6] Speight JG. The Chemistry and Technology of Coal. Chemical Industries, 2012: 8.
[7] Xiao X, Wang X, Zheng Z, Qin W, and Zhou Y. Catalytic Coal Gasification Process Simulation with Alkaline Organic Wastewater in a Fluidized Bed Reactor Using Aspen Plus. Energies, 2019; 12(7).
[8] Jeong YS, Choi YK, Park KB, and Kim JS. Air co-gasification of coal and dried sewage sludge in a two-stage gasifier: Effect of blending ratio on the producer gas composition and tar removal. Energy, 2019; 185: 708-716.
[9] Duan W, Yu Q, Wang K, Qin Q, Hou L, Yao X, and Wu T. ASPEN Plus simulation of coal integrated gasification combined blast furnace slag waste heat recovery system. Energy Conversion and Management, 2015; 100: 30-36.
[10] Kong X, Zhong W, Du W, and Qian F. Three Stage Equilibrium Model for Coal Gasification in Entrained Flow Gasifiers Based on Aspen Plus. Chinese Journal of Chemical Engineering, 2013; 21(1): 79-84.
[11] Preciado JE, Ortiz-Martinez JJ, Gonzalez-Rivera JC, Sierra-Ramirez R, and Gordillo G. Simulation of Synthesis Gas Production from Steam Oxygen Gasification of Colombian Coal Using Aspen Plus (R). Energies, 2012; 5(12): 4924-4940.

[12] Jang D-H, Kim H-T, Lee C, and Kim S-H. Kinetic analysis of catalytic coal gasification process in fixed bed condition using Aspen Plus. International Journal of Hydrogen Energy, 2013; 38(14): 6021-6026.

[13] Nyakuma BB and Jauro A. Physicochemical Characterization and Thermal Decomposition of Garin Maiganga Coal. GeoScience Engineering, 2016; 62(3): 6-11.

[14] Ryemshak SA and Jauro A. Proximate analysis, rheological properties and technological applications of some Nigerian coals. International Journal of Industrial Chemistry, 2013; 4(1): 7.

[15] Nyakuma B. Physicochemical Characterization and Thermal Analysis of newly discovered Nigerian coals. Bulgarian Chemical Communications, 2016; 48(4): 746 – 752.

[16] Chukwu M, Folayan C, Pam G, and Obada D. Characterization of some Nigerian coals for power generation. Journal of Combustion, 2016; 2016.

[17] Obaje NG. Geology and mineral resources of Nigeria. Lecture Notes in Earth Sciences, 2009; 120: 221.

[18] Nyakuma BB. Physicochemical, Geomineralogical, and Evolved Gas Analyses of Newly Discovered Nigerian Lignite Coals. Coke & Chemistry, 2019; 62(9): 394-401.

[19] Ohimain EI. Can Nigeria generate 30% of her electricity from coal? International Journal of Energy and Power Engineering, 2014; 3(1): 28-37.

[20] Oboirien B, North B, Obayopo S, Odusote J, and Sadiku E. Analysis of clean coal technology in Nigeria for energy generation. Energy Strategy Reviews, 2018; 20: 64-70.

[21] Amoo LM. Computational fluid dynamics simulation of Lafia–Obi bituminous coal in a fluidized-bed chamber for air-and oxy-fuel combustion technologies. Fuel, 2015; 140: 178-191.

[22] Basu P. Biomass Gasification and Pyrolysis: Practical Design and Theory. 2010.

[23] Vagia E and Lemonidou A. Thermodynamic Analysis of Hydrogen Production via Steam Reforming of Selected Components of Aqueous Bio-oil Fraction. International Journal of Hydrogen Energy, 2007; 32(2): 212-223.

[24] Liu S, Chen M, Hu Q, Wang J, and Kong L. The kinetics model and pyrolysis behavior of the aqueous fraction of bio-oil. Bioresource Technology, 2013; 129: 381-386.

[25] Nyakuma BB, Oladokun O, Jauro A, and Nyakuma DD. Fuel Characterization of Newly Discovered Nigerian Coals. IOP Conference Series: Materials Science and Engineering, 2017; 217: 012012.

[26] Oladokun O, Ahmad A, Tuan Abdullah TA, Nyakuma BB, Kamaroddin MFA, Ahmed M, and Alkali H. Sensitivity Analysis of Biohydrogen Production from Imperata Cylindrica Using Stoichiometric Equilibrium Model. Jurnal Teknologi, 2016; 78(8-3): 137-142.

[27] Oladokun O, Ahmad A, Abdullah TAT, Nyakuma BB, Kamaroddin MFA, and Nor SHM. Biohydrogen production from Imperata cylindrica bio-oil using non-stoichiometric and thermodynamic model. International Journal of Hydrogen Energy, 2017; 42(14): 9011-9023.

To whom correspondence should be addressed: Dr. Bemgba B. Nyakuma, Research Initiative for Sustainable Energy Technologies (RISET), Court 5 North-Bank, Makurdi, Benue State, Nigeria, E-mail: bbnyax1@gmail.com