Valorization of Sugarcane Bagasse for Hydrogen-Rich Gas Production using Thermodynamic Modeling Approach

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ABSTRACT: Hydrothermal gasification also known as supercritical water gasification (SWG) has been considered a promising approach for converting wet biomass such as sugarcane bagasse into high-quality syngas. This study presents the thermodynamic modeling of the hydrothermal gasification of sugarcane bagasse using Aspen Plus. The effects of process parameters on the composition and yield of product gases were also investigated. It was found that the effect of temperature and biomass concentration were significant in the production of hydrogen-rich gas, while less impact was observed with pressure. The hydrogen gas (H_2) produced with the highest mole fraction (56.70 mol%) and yield (103.26 kmol/kg) was obtained at 750°C and low biomass concentration of 10 wt%, while the lowest yield (1.52 kmol/kg) and mole fraction (2.45 mol%) of H_2 were obtained at 450°C and high biomass concentration of 50 wt%. Findings from this study also showed that the highest net calorific value (17.55MJ/kg) was reached at 450˚C and 50 wt% of biomass concentration. This study would help to consolidate research on hydrothermal gasification of sugarcane bagasse and optimization of experimental processes and also serve as an important benchmark in the utilization of biomass as a clean energy source for future projects.

KEYWORDS: Hydrothermal gasification, Thermodynamic modelling, Aspen Plus, Hydrogen gas, Sugarcane bagasse

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I. INTRODUCTION

The concentration of greenhouse gases in the earth's atmosphere has risen dramatically as a result of human activity such as the burning of fossil fuels as energy sources, fuel and chemicals for power generation, heating and transportation. This has led to the amplification of the natural greenhouse effect and further warming of the earth's surface and atmosphere (Caney, 2015; Tavares et al, 2020). As a result, the search for the utilization of pollution-free "green" energy sources such as hydropower, wind, solar, geothermal, and biomass to help solve environmental problems posed by the burning of fossil fuels is receiving much attention in the scientific community (Tavasoli *et al*, 2016). Biomass is one of the most abundant resources on earth with a vast potential to produce value-added chemicals and sustainable biofuels (Sattar *et al*, 2014). Evidence has shown that the use of biomass can contribute to about 10-14% of the global energy supply (Okolie *et al*, 2019). In recent years, the use of biomass as an alternative source of energy has emerged to complement fossil-based resources. Owing to its steady feedstock supply and cleaner nature than most traditional sources as it contains an infinitesimal amount of sulphur and nitrogen, less tar formation, or ash, resulting in lower sulphur dioxide, nitrogen oxides, and soot emissions than conventional fossil fuels (Im-orb *et al*, 2018; Yaghoubi *et al*, 2018).

Sugarcane bagasse is biomass made up of a mixture of hard fibres, soft and smooth parenchymal tissue (pith) with a high hygroscopic characteristic left after crushing sugar cane with a moisture level of 45-50% (Kumar *et al*, 2021). Rashidi and Tavasoli (2015), estimated that annually, 1.6 billion tons of sugarcane are processed, which generates approximately 279 million metric tons of biomass residues (leaves and bagasse). These biomass residues, which are always dumped on open land, affect every area of our lives, from making water unsafe to drink through run-off or being burnt by farmers as a way to clear land or fertilize the soil. Converting this abundant but underutilized biomass into useful products is worth investigating. Over the years, several conversion technologies such as fermentation (Chen *et al*, 2015), anaerobic digestion (Ahmad *et al*, 2016), combustion (Sikarwar *et al*, 2017), pyrolysis (Fremaux *et al*, 2015), and gasification (Watson *et al*, 2018) have been employed for converting sugarcane bagasse into value-added products including bioethanol, methane, bio-oil, and hydrogen (Cao *et al*, 2018). However, gasification has been considered the most promising technology due to its auto-thermal ability, high carbon conversion, flexibility of raw materials, and higher calorific value of syngas (Gökkaya *et al*, 2019).

Supercritical water gasification (SWG), also known as hydrothermal gasification (HTG), is an iteration of the traditional gasification process that uses water as the gasifying medium at supercritical conditions (Pc > 22.1 MPa, Tc > 374 ºC) to convert biomass into hydrogen-rich gases (Okolie *et al*, 2020). These conditions, which are above the critical pressure and temperature of water, change its thermo-physical

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properties (i.e dielectric constant, density, ionic product, and viscosity), allowing water to behave as a catalyst, green solvent, and reaction medium, that improves mass transfer and reaction rates (Okolie *et al*, 2021). Hydrothermal gasification, which occurs at a temperature range of 400°C - 750°C and pressure range of 24 MPa - 36 MPa, has been deemed a potential approach for the production of high-quality synthesis gas from biomass containing appreciable quantities of moisture, paving the way for the majority of its advantages over the traditional gasification process, as wet biomass can be used directly without the need for drying (Mustapha *et al*, 2021).

Hydrogen gas is considered a cleaner energy carrier with the highest energy density compared to other gases and energy efficiency of 122 KJ/kg, which is 2.75 times that of a typical hydrocarbon fuel. Thus, the present research focussed on determining the feasibility of meeting the global energy needs (Lamb and Pollet, 2020). This energy carrier is ideal because it produces water as the byproduct of combustion, so it emits no emissions despite its high energy density. Hydrogen can be utilized directly or as an intermediate storage fuel for manufacturing gasoline, methanol, ethanol, and other useful compounds. Hydrogen can be used as a gas or a liquid, depending on the application, making it a versatile fuel (Parthasarathy and Narayanan, 2014).

Numerous experimental investigations have been conducted on the SWG of sugarcane bagasse with the use of different catalysts to increase the rate of reaction as well as enhance gas yield and selectivity. For example, Rashidi and Tavasoli (2015) performed the SWG of sugarcane bagasse in the presence of unpromoted and copper-promoted carbon nanotubes supported nickel. The results showed that the promotion of Ni/CNTs catalysts with copper increased the total gas and hydrogen yield by 14.4% and 14.7%, respectively while the yield of methane decreased by 25.9%. Safari et al (2016) developed a novel process for the gasification of sugarcane bagasse under supercritical water conditions for the co-production of hydrogen and power. The result revealed that hydrogen production of 8.55 kg/h and electrical power generation of 56 kW were obtained for the 20 wt% mixture of bagasse with a mass flow rate of 1000 kg/h, reactor pressure of 300 bars and temperature of 700ºC. Sheikhdavoodi et al (2015) studied the SWG of sugarcane bagasse in a batch reactor and evaluated the effects of catalyst and process parameters on hydrogen production. They observed that an increase in reaction temperature to 800ºC favored hydrogen yield with the presence of KOH as catalyst. Zhang et al (2019) reviewed the SWG of sugarcane bagasse for hydrogen production from the exergy aspect. The results showed that exergy efficiencies of hydrogen production were mainly in the range of 0.04% - 42.05%. So far, only a few studies have been reported on the thermodynamic modelling of the SWG process. Okolie *et al* (2020) employed an experimental and thermodynamic modelling approach to study the hydrothermal gasification of soybean straw and flax straw for hydrogen rich-gas production. Recently, Mustapha et al (2021) reported the hydrothermal gasification of Scenedesmus obliquus microalgae using Aspen plus. However, there is a scarcity of modeling work on hydrothermal gasification of sugarcane bagasse using Aspen Plus or any other simulation software. Experimental processes involving biomass gasification are generally expensive and laborious, especially on larger scales; hence, modelling and simulation are necessary to save time and money while also assisting in the planning and optimization of experiments to be conducted in real systems. Aspen plus thermodynamic modelling and simulation of SWG of sugarcane bagasse can be effectively used to examine the technical difficulties of overcoming the high cost of hydrogen production to increase the commercial market for advanced gasification technology.

Hence, this research aimed to evaluate the production of hydrogen-rich gas from the gasification of sugarcane bagasse under SWG conditions. The process of SWG of sugarcane bagasse was modelled using Aspen plus. The effects of process variables such as biomass concentration, temperature, and pressure on hydrogen gas production were studied, and validation of the Aspen plus model results with experimental work was also carried out. This study would serve as an important benchmark in the utilization of biomass as a clean energy source for future projects.

II. MATERIALS AND METHODS

A. *Feedstock Characterization*

The proximate and ultimate values of sugarcane bagasse used in this study were obtained from Cao *et al* (2018) as presented in Table 1.

Table 1: Feedstock composition of sugarcane bagasse

Proximate Analysis	**(%)**
Moisture	4.46
Volatile matter	83.32
Fixed carbon	14.03
Ash (dry basis)	2.65
Ultimate Analysis	**(%)**
C	47.09
H	6.16
N	0.52
O	42.50
S	1.08
Cl	0.00

B. *Modelling Hydrothermal Gasification under Aspen Plus V.10*

1) *Lists of components*

The components utilized in the hydrothermal gasification model are shown in Table 2. The lists of the components are grouped into nonconventional, conventional and solids.

2) *Physical property method*

This simulation used a combination of the Peng–Robinson and Boston–Mathias function (PR-BM) property approach, which estimates every physical characteristic of the typical components in the gasification process. This property package's alpha parameter is a temperature-dependent variable. When the temperature is very high, this parameter increases the pure component vapour pressure (Mustapha *et al*, 2021). The major reason this property package was selected for the gasification process is that the temperature used was fairly high. The HCOALGEN and DCOALIGT were the enthalpy and density models used for both biomass and ash, which are non-conventional components.

Table 2: Components utilized in the hydrothermal gasification.

Nonconventional Element	Conventional Element	Solid Element
Biomass(Sugarcane bagasse)	Water	Carbon-Graphite
Ash	Methane	-
-	Carbon monoxide	-
-	Sulfur	-
-	Carbon dioxide	-
-	Oxygen	-
-	Nitrogen	-
-	Hydrogen	-

3) *Thermodynamic modeling assumptions*

The following assumptions were developed during the development of the model:

i. Hydrogen, Methane, Carbon dioxide, Carbon monoxide, Water, Hydrogen Sulfide and Ammonia were the gaseous products from the gasifier;
ii. The simulation model operated at a steady state;
iii. Char consisted of solid carbon and ash only;
iv. The gasifier temperature was uniform throughout the process;
v. Oxides of nitrogen or sulfur were not formed;
vi. Ash was considered inert.

4) *Gasification reactions*

The chemical reaction steps that make up the hydrothermal gasification process are listed using Eqns. 1–10 (Parthasarathy and Narayanan, 2014):

Biomass Devolatilization: $Biomass\ char + tar + H_2O + light\ gas\ (CO + CO_2 + H_2 + N_2 \ldots)$ (1)

Oxidation of Carbon: $C + \frac{1}{2}O_2 \rightarrow CO$ -111 MJ/ kmol. (2)

$C + O_2 \rightarrow CO_2$ -394 MJ/ kmol (3)

Char gasification: $C + CO_2 \rightarrow 2CO$ +172 MJ/ kmol. (4)

$C + H_2O \rightarrow CO + H_2$ +131 MJ/ kmol. (5)

$C + 2H_2 \rightarrow CH_4$ -75 MJ/ kmol. (6)

Volatile oxidation: $CO + \frac{1}{2}O_2 \rightarrow CO_2$ -283 MJ/ kmol. (7)

$H_2 + \frac{1}{2}O_2 \rightarrow H_2O$ -242 MJ/ kmol. (8)

$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$ +206MJ/ kmol. (9)

$CO + H_2O \rightarrow CO_2 + H_2$ -41 MJ/ kmol. (10)

5) *Description of biomass decomposition process*

The biomass was designated as an unconventional component, and the ultimate and proximate analyses as well as the mass flow were entered. The biomass feedstock used in all the simulations ran at $10kghr^{-1}$. By setting the product distribution based on the ultimate analysis, the RYIELD reactor was utilized to simulate the decomposition of biomass. In this step, biomass was converted into its constituent components, such as carbon (C), hydrogen (H_2), oxygen (O_2), nitrogen (N_2), sulfur (S), chlorine (Cl), and ash. For the product distribution, the FORTRAN statement was used, which is as follows:

FACT = (100-WATER) / 100
H_2O = WATER/100
ASH = ULT (1) / 100 * FACT
C = ULT (2) / 100 * FACT
H_2 = ULT (3) / 100 * FACT
N_2 = ULT (4) / 100 * FACT
O_2 = ULT (5) / 100 * FACT
S = ULT (6) / 100 * FACT

The GASIFIER (i.e the principal reaction unit block) was linked by various streams to the other unit blocks. CYCLONE, COOLER, and F-SEP in the simulation with each unit performing distinct functions as described in Table 3. The process flow diagram for the hydrothermal gasification process is shown in Figure 1.

C. *Validation of Model*

The model validation was carried out to verify that the proposed Aspen Plus model in this study was reliable, and the experimental data from Cao *et al* (2018) on hydrothermal gasification of sugarcane bagasse was utilized. The experiments were conducted with a sugarcane bagasse concentration of 6 wt% and a gasifier maintained at (600℃ - 750℃, 24 - 30 MPa). Using the vast nature of the Aspen Plus, a model was simulated to produce synthesis gas from hydrothermal gasification of sugarcane bagasse under similar empirical conditions as reported by Cao *et al* (2018). The Aspen Plus simulation results are in good accord with the empirical outcome of Cao *et al* (2018) as the simulation model produced a similar forecast of the produced gas composition (in mol.%) that is comparable to that of the experimental result as shown in Figure 2.

III. RESULTS AND DISCUSSION

A. *Influence of Temperature*

The effect of temperature on produced gas composition and yield as well as the net calorific was studied by varying the temperature in the range of 450 to 750℃. (CH_4), hydrogen (H_2), and carbon dioxide (CO_2), hydrogen (H_2), are the primary resulting gases recognized from the hydrothermal gasification with minute amounts of carbon monoxide (CO), hydrogen sulphide (H2S) and ammonia (NH_3). The current research will be focused on the major gaseous products of H_2, CH_4 and CO_2 obtained from the SWG process. The main reactions during the hydrothermal gasification process are steam reforming reaction (Equation 5), methanation reaction (Equation 6) and water-gas shift reaction (Equation 10). Each reaction has a significant impact on the outcome of the gasification process. The H_2 is produced by water-gas shift and steam-reforming reactions, while the CH_4 is produced through methanation reactions (Rashidi and Tavasoli, 2015).

Temperature plays a very significant impact in SWG as the gasifier temperature affects the entire end product composition. This occurs because of some chemical reactions in the gasifier, such as steam-reforming reactions which are endothermic. As a result, according to Le Chatelier's principle (Adar *et al*, 2020), a higher temperature favors the endothermic reaction product and suppresses exothermic reactions (Tavares *et al*, 2020). Figure 3 illustrates the produced gases H_2,

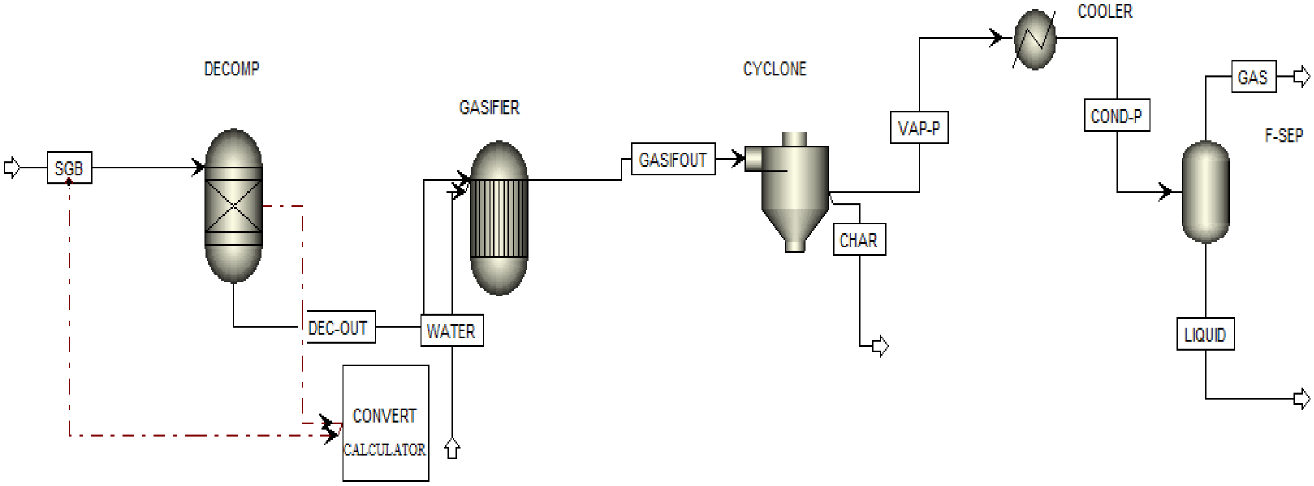

Figure 1: The flow diagram of Aspen Plus model for hydrothermal gasification of sugarcane bagasse.

Table 3: Description of Aspen plus unit operation blocks used in the simulation model.

Aspen plus ID	Block ID	DESCRIPTION
RYield	DECOMP	Based on their proximate and ultimate assessment, it converts nonconventional biomass into conventional components.
RGibbs	GASIFIER	Simulation of solid-gas reactions based on phase and chemical equilibrium calculations and the minimization of the system's Gibbs free energy.
SSplit	CYCLONE	Separation of solid product from the vapour product.
Heat Exchanger	COOLER	Reduction of the vapour stream temperature to induce condensation of liquid products.
Flash2	F-SEP	Separation of the non-condensable gaseous product from liquid product
Calculator	CONVERT	Calculation of mass yields obtained from DECOMP using FORTRAN statement with execution of the calculator block before DECOMP block.

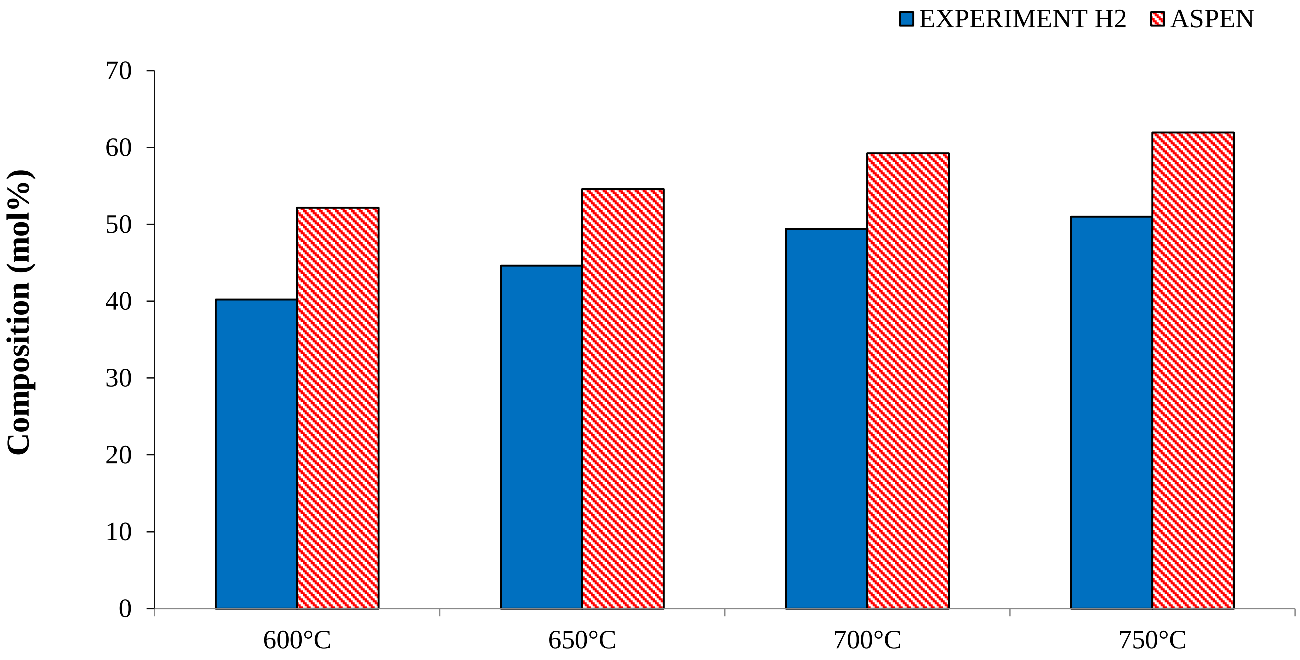

Figure 2: Composition of H_2 from the hydrothermal gasification of sugarcane bagasse (Temperature: 600 – 750 °C, Pressure: 25MPa, and 6 wt% biomass feed concentration).

CH_4 and CO_2 molar fraction as a function of the gasification temperature.

The mole fraction of H_2 obtained in Figure 3 increased from 6.22 to 42.86 mol% at 26 MPa, whereas the composition of CH_4 declined from 44.97 to 15.88 mol% and that of CO_2 from 43.93 to 32.79 mol%. Depicting an opposite trend in their production as the temperature elevated from 450°C to 750°C. The decrease in CH_4 with temperature is due to exothermic behaviour exhibited by the methane reaction formation (Eqn. 6). In contrast, the increase in H_2 was due to the steam-reforming reactions and water gas shift reactions (Eqns. 5 and 10), favoring the production of more H_2. These findings are consistent with the existing literature (Mustapha *et al*, 2021; Tavares *et al*, 2020), which also concluded that lower temperatures favored CH_4 production while H_2 production was favored at higher temperatures.

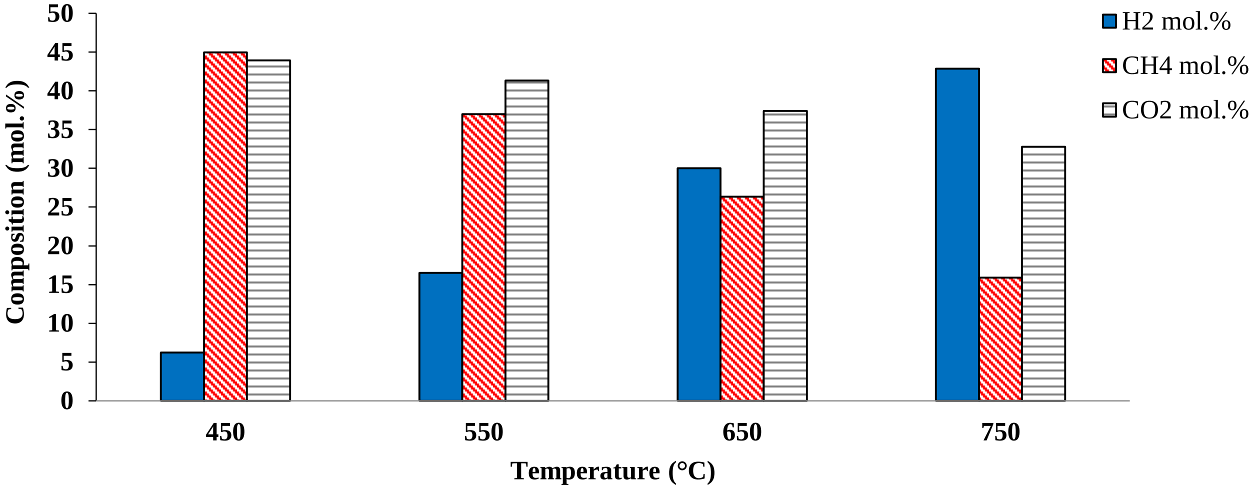

Figure 3: Effect of varying temperature on gas composition obtained from Hydrothermal Gasification of 20wt% biomass concentration at 26 MPa pressure.

B. *Influence of Pressure*

Figure 4 depicts the effect of changing the pressure from 24 to 30 MPa at 650°C on the gaseous product mole fraction. Utilization of a pressure range of 24 to 30 MPa resulted in a reduced hydrogen mole fraction from 56.70 to 54.71 mol%. Thermodynamically, it is expected that an increase in temperature would favor more H_2 production since the steam reforming reaction is endothermic. However, despite operating at a high temperature of 650ºC, a decrease in H_2 composition was observed when pressure was increased from 24 to 30 MPa (see Figure 4). This is a suggestion that the effects for the steam reforming reaction as a result of increasing temperature was offset by the significant retardation effect due to increasing reaction pressure. On the other hand, a methanation reaction is a volume-reducing reaction favored by high pressure. As shown in Figure 4, a slight increase was observed with an increase in pressure for CH_4 and CO_2 mole fraction respectively. This is an indication that the effect for the methanation reaction as a result of increasing temperature was offset by the significant promotion effect due to increasing reaction pressure. Because of their combinatorial effect, the pressure has the slightest effect on the composition of the gaseous product relative to the temperature. This slight effect on the composition of gaseous products is evidenced by the minor increase observed with CH_4 mole fraction and a slight decline in H_2 mole fraction. The work of Mustapha *et al* (2021) on hydrothermal gasification of microalgae also revealed H_2 generation declined while the CH_4 production was promoted with an increase in reaction pressure.

C. *Influence of Biomass Concentration*

Sugarcane bagasse concentration was varied within the range of 10 – 50 wt % attemperatures of 450°C, 550°C, 650°C, and 750°C, respectively. The mole fractions of the gases (i.e. H_2, CO_2, CH_4) and their yield at a range of temperature and biomass concentration are depicted in Figure 5. As shown in Figure 5, sugarcane bagasse of 10 wt% at 750°C generated the highest H_2 mole fraction of 56.70 mol% with a corresponding yield of 103.26 kmol/kg. Moreover, as the biomass concentration increased from 10 to 50 wt%, the H_2 mole fraction decreased from 56.70 to 21.00 mol% with the corresponding hydrogen yield also decreasing drastically from 103.26 kmol/kg to 20.14 kmol/kg due to the reduction in the moisture content as biomass concentration increases in the hydrothermal reaction. Water served as a reactant in the two major reactions (steam reforming and water-gas shift reactions) occurring in hydrothermal gasification. This reactant positively influences the yield of a lesser biomass concentration because more moisture is accessible for the supercritical gasification process while reduction of water content in the gasifier implies the feedstock, sugarcane bagasse, is at a higher concentration. Thus, the steam-reforming reactions and water-gas shift were hastened at a lesser biomass concentration due to sufficient availability of more moisture invariably favoring H_2 over CH_4 generation via the methanation reaction. Cao *et al* (2018) reported that with a lower biomass concentration, hydrogen gasification efficiency is higher and this could be due to the release of atoms of hydrogen from excess water to the gaseous product. The maximum CH_4 mole fraction of 48.23 mol% in the process was attained at 450˚C, 50 wt.% biomass concentration with a

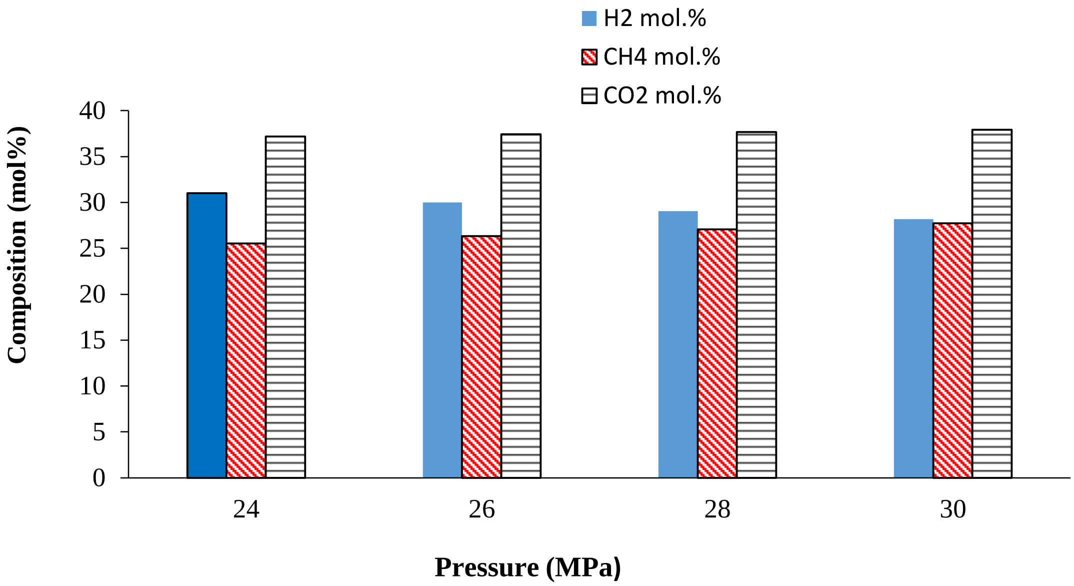

Figure 4: Effect of varying pressure on gas composition obtained from hydrothermal gasification of sugarcane bagasse with 40wt% biomass concentration.

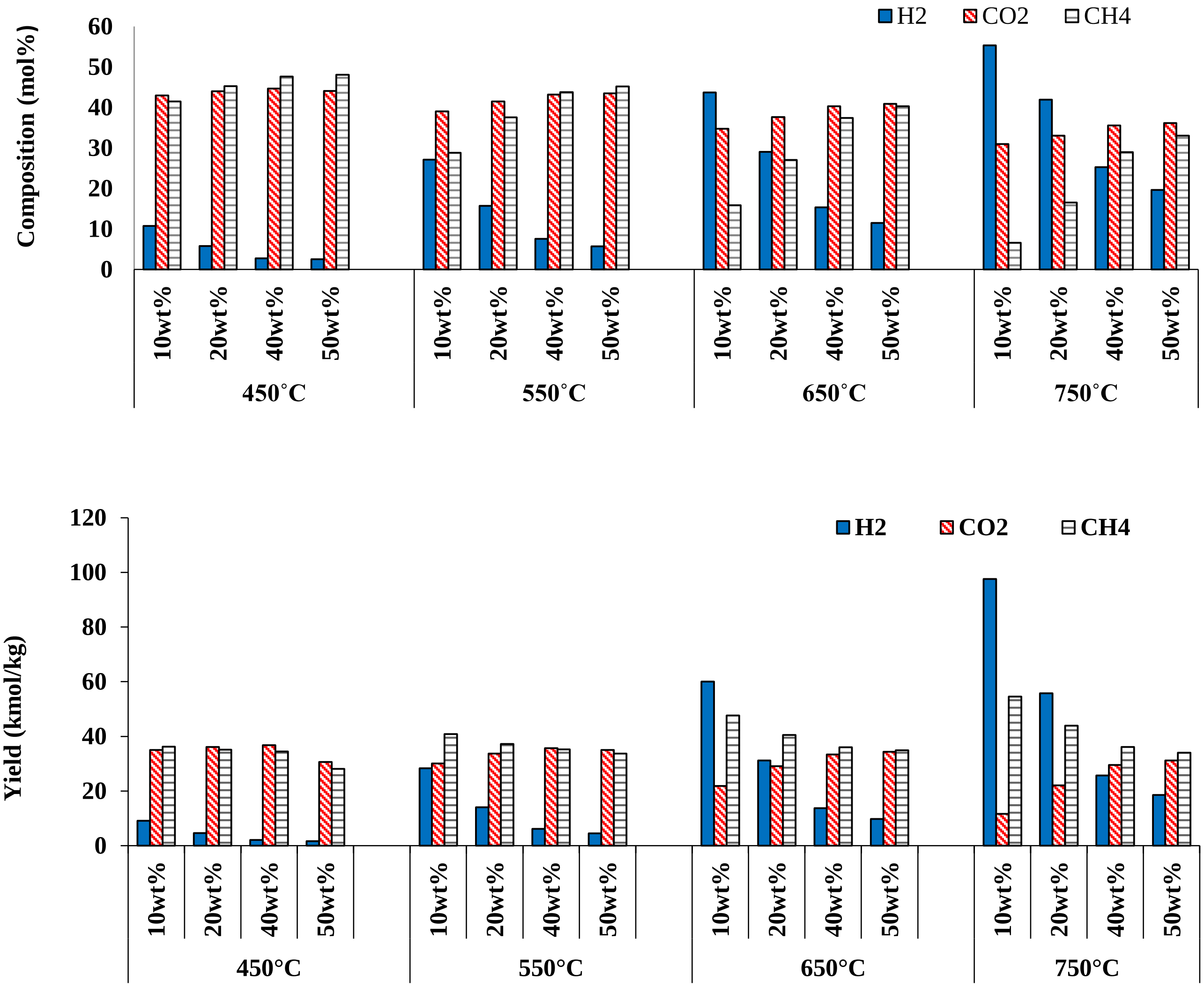

Figure 5: Effect of biomass concentration at temperatures 450˚C, 550˚C, 650˚C and 750˚C respectively on the yield and composition of H_2, CO_2 and CH_4 obtained from hydrothermal gasification of sugarcane bagasse at 28MPa.

corresponding yield of 29.96 kmol/kg. The mole fraction of CH_4 increased from 42.10 - 48.23 mol% as the biomass concentration, increased from 10 - 50 wt.% and the yield increased slightly from 35.21 to 36.82 kmol/kg. The hindrance in the production of H_2 is as a result of the methanation reaction favoring the CH_4 gas production at the expense of H_2 gas.

D. *Effect of Influencing Parameters on Net Calorific Value*

The net calorific value for the gas products was calculated using the equation reported for lower heating value by Mustapha *et al* (2021). The resulting composition data for H_2, CH_4 and CO with CH_4 composition contributing a significant impact on the end value of lower heating value (LHV). The effect of biomass concentration on net calorific value at varying temperatures is shown in Figure 6. Due to the considerable effect of lower temperatures on the improved generation of methane gas; the peak value was attained at 450°C with minimal difference as the biomass content elevated under this same temperature condition. LHV obtained at 10, 20, 40, and 50 wt% were 16.16, 16.92, 17.39 and 17.55 MJ/kg respectively, indicating LHV of the produced gas is favored at increasing biomass concentrations with low temperatures. The high hydrogen production of sugarcane bagasse indicates that hydrothermal gasification is a yardstick in upgrading such low-value biomass waste to high-value energy carriers, and 450°C may be the most effective temperature for high energy recovery of sugarcane bagasse.

IV. CONCLUSION

The modeling of supercritical water gasification of sugarcane bagasse has been studied using Aspen plus V10 simulation environment. The research focused on the composition, yield, and lower heating value (LHV) of the product gases with significant attention to the hydrogen gas at varying temperature, pressure, and biomass concentration. In comparison to pressure, temperature has the greatest impact on the gasification reaction. High temperature (750°C) and the least biomass concentration (i.e. 10 wt%) produced the highest H_2 yield and mole fraction of H_2 gas while low temperature (450°C) and high biomass concentration (i.e. 50 wt%) yielded the lowest H_2 yield and H_2 mole fraction. On the contrary, the maximum CH_4 mole fraction with a modest increase in produced CO_2 was attained at the least temperature (450°C) and highest biomass concentration (50 wt%). The greatest LHV of 17.55 MJ/kg was achieved at 450°C with a biomass content of 50 wt%. The results obtained from this study show that the resulting CO_2 is at significantly elevated mole fractions. Therefore, there is a need to minimize the CO_2 emitted from the process either by the introduction of CO_2-absorber that can reduce the CO_2 concentration towards minimization of potential greenhouse effects or CO_2 recycling option, which can enable the production of minimum CO_2 emission by serving as a gasifying agent in the biomass gasification. This information is essential for the development of initiatives that use biomass as a renewable energy source.

AUTHOR CONTRIBUTIONS

S. I. Mustapha: Conceptualization, Resources, Supervision and Methodology. **I. A. Mohammed:** Visualization, Formal analysis, Methodology. **F. A. Aderibigbe:** Resources, Supervision and Methodology. **T. L. Adewoye:** Resources, Supervision and Methodology. **F. O. Omoarukhe:** Methodology, Investigation, Validation, Writing - original draft. **A. O. Sowole:** Methodology, Investigation, Validation, Writing.

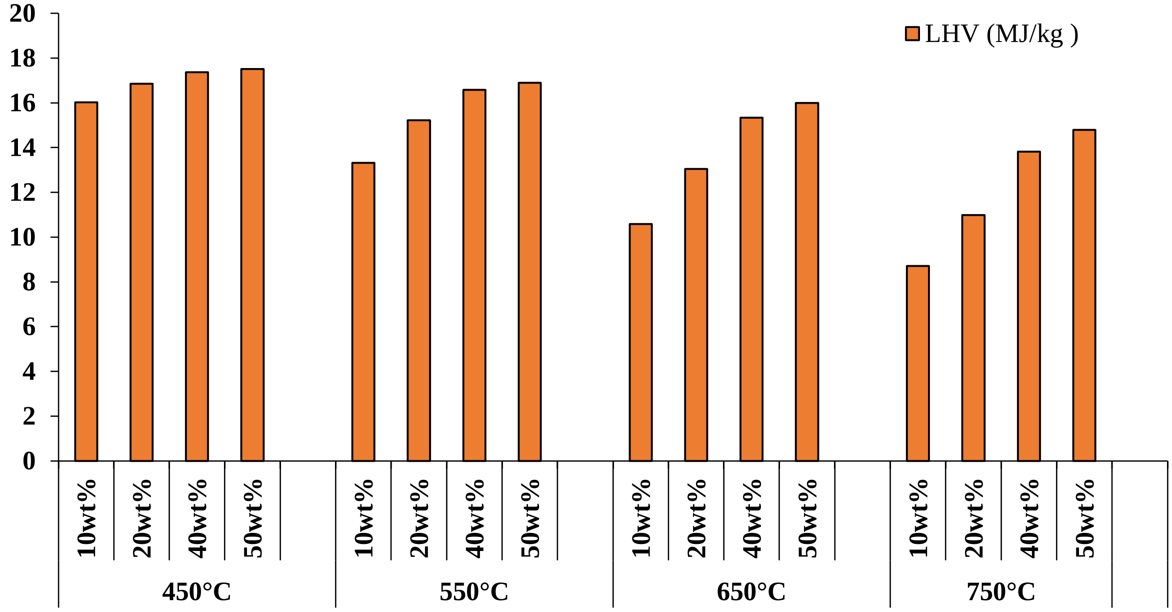

Figure 6: Effect of biomass concentration at temperatures 450°C, 550°C, 650°C and 750°C respectively on net calorific value obtained from hydrothermal gasification of sugarcane bagasse at 28MPa.

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