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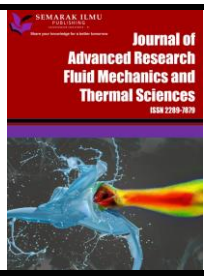

The Impact of Temperature on the Production of High Calorific Value Syngas using Cogasification Technology

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ABSTRACT

Cogasification represents a thermochemical reaction employed to transform by combining biomass or fossil carbonaceous materials into combustible matters. Widely acknowledged as the most appealing approach among various combustible material to useful energy. This method offers significant potential for environmentally friendly energy production, boasting low carbon emissions. This study conducted gasification tests utilizing an updraft gasifier, incorporating parameters variable at 650°C to 850°C. The materials utilized consisted of a blend of Municipal-Solid-Waste (MSW) and coconut shells, maintaining a steam to biomass at 1.3. Optimum temperature at 750°C, the syngas revealed 41.30% mol CO, 20.90 mol% CO_2 37.25 mol% H_2, and 0.55 mol% CH_4. Notably. The highest H_2 gas production was achieved at this temperature. Furthermore, the net caloric value at this temperature, surpassed other variations, reaching 374.67 kJ/mol, accompanied by the generation of 11.38% of tar and 21.1% char.

1. Introduction

Efficient waste management plays a crucial role in diminishing waste volume and mitigating pollution. One effective approach involves harnessing waste for energy purposes. Presently, the conversion of waste into energy remains relatively modest at 18.8% [1]. Biomass is one of the largest renewable energies that contributes to energy resources, besides solar and wind energy in recent years [2-4]. The utilization of a mix of renewable energy sources with fossil fuels has been increasing due to its significant contribution to reducing greenhouse gas emissions from fossil fuel consumption [5]. The energy derived from waste holds significant promise for bolstering renewable energy sources and providing a viable solution for waste reduction [6]. Among various waste-to-energy technologies, gasification technology stands out as the most appealing method. This technology not only has the

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potential to generate clean energy but also contributes to the improving energy efficiency, adopting sustainable practices in industries, and promoting reforestation to absorb and store carbon [7,8]. Gasification is employed to transform carbon sources like biomass into syngas, encompassing H_2, CO, CH_4, and CO_2 [9]. When gas products are contaminated with diluents such as N_2 and CO_2, they are typically referred to as producer gas. Meanwhile, pure CO, H_2, and CH_4 gas are known as syngas [10]. Typically, this syngas can serve as a direct means for energy generation.

Moreover, the extracted H_2 and CH_4 from the product can be utilized as chemical raw materials or fuel. Beyond its role in generating syngas for renewable energy, gasification also addresses the pressing issue of waste reduction, offering a solution to the significant challenges associated with waste management today. The chemical reaction occurring in the gasification process is outlined as follows:

(i) *Drying Process*

Primary Feedstock + Heat → Dry Feedstock + H_2O

Primary feedstock undergoes a reaction with heat, resulting in the formation of dry feedstock and water (H_2O).

(ii) *Pyrolysis Process*

Dry Feedstock + Heat → Char + Volatiles matter

The process involves dry feedstock reacting with heat, leading to the formation of char and volatiles.

(iii) *Reduction*

Boudouard Reaction

$C + CO_2 \longleftrightarrow 2CO$ (-164,9 MJ / kg mol) (1)

Steam - Carbon Reaction

$C + H_2O \longleftrightarrow CO + H_2$ (-122,6 MJ/kg mol) (2)

Water - Gas Shift Reaction

$CO + H_2O \text{ ==== } CO_2 + H_2$ (+42,3 MJ/kg mol) (3)

Methanation

$C + 2\,H_2 \longleftrightarrow CH_4$ (+75 MJ/kg mol) (4)

$CO + 3H_2 \longleftrightarrow CH_4 + H_2O$ (-205,9 MJ/kg mol) (5)

$CO_2 + 4H_2 \longleftrightarrow CH_4 + 2H_2O$ (-165 MJ/kg mol) (6)

$CO + 2H_2 \longleftrightarrow CH_4 + CO_2$ (-165 MJ/kg mol) (7)

(iv) *Partial Oxidation*

$C + O_2 \longleftrightarrow CO_2$ (+393 MJ/kg mol) (8)

$C + 0.5\ O_2 \longleftrightarrow CO$ (+393 MJ/kg mol) (9)

$2H_2 + O_2 \longleftrightarrow 2H_2O$ (+242 MJ/kg mol) (10)

(v) *Tar Cracking*

$$C_pH_q(tar) \rightarrow C_xH_y + C + H_2 + CO \quad (11)$$

The presence of a significant amount of tar in the gasification process poses challenges for the production of high-qualitygas yealds. In cogasification, achieving the correct combining and composing of feedstocks can yield positive effects on product co-conversion [11]. This manifests as a synergistic effect, resulting in increased reactivity of materials during process is influenced by various factors, including temperature, pressure, residence time. Its reactivity surpasses the average yield of individual products obtained from each biomass [12].

The gasification syngas product was influenced by several factors, including biomass characteristics, temperature, equivalent ratio (ER), steam to biomass ratio (SBR), gasification agent and gasification reactor [13]. Gasification agents consist of air, oxygen or steam. The high production of hydrogen can be attributed to the use of steam, oxygen and air as gasification medium. The highest sensible energy efficiency value was obtained for air as a gasification medium, while the optimal value was obtained for steam as a gasification medium [14,15].

The effect of reaction temperature is known to increase the H_2 product in syngas [16,17]. In research conducted by Almeida, it was shown that the gasification of olive dregs particles used air as a gasification agent with varied reaction temperatures. The higher the operating temperature, the gas production and other gasification performance parameters can increase [18].

This study aimed to investigate the impact of incorporating coconut shells into Municipal Solid Waste (MSW). Coconut shells, abundant in tropical climates, especially in Indonesia, the world's leading coconut-producing country [19]. were chosen as a biomass product. The combination of coconut shells, known for their higher lignin content, with MSW was anticipated to induce a synergistic reaction, yielding syngas with an elevated heating value. A higher (hemicellulose + cellulose)/lignin ratio is expected to enhance the syngas content [20]. The intriguing aspect lies in combining MSW, rich in cellulose and hemicellulose elements. Raw materials derived from Municipal Solid Waste (MSW) in the certain region which generally have typically exhibit notable amounts of organic and plastic components, introducing an intriguing aspect to this research. The objective of the study is to assess how the operational temperature influences the generation of syngas with a high calorific value, employing MSW and coconut shells as the fuel source. Additionally, the research aims to scrutinize the resulting products, including charcoal and tar. The gasification procedure is implemented within a temperature range spanning at a specified temperature range, utilizing an updraft-type reactor featuring induction heating and operating without the use of a catalyst.

2. Methodology

2.1 Sample Preparation

The first step of this study involved the preparation of samples comprising Municipal Solid Waste (MSW) and discarded coconut shells. The MSW was sourced from the Final Waste Disposal Site in the Cilowong area of Serang City, Banten Province, Indonesia. Various waste categories are utilized in this study, including 50% food waste (bones, rice, and vegetables), 8% paper and cardboard, 4% wood and garden waste, 0.75% cloth, 0.50% rubber, 35% plastic, 0.5% metal, 0.25% glass, and 1.00% other.

Following collection, the material underwent drying at 105°C for a period of 6 hours using an electric heater. The samples were then crushed until achieving an average size of 0.5 × 0.5 cm. The composition of the raw material was established at 50% MSW and 50% coconut shell, with a total mass of fifty grams. The results of the proximate and ultimate analysis of the feedstock are presented in Table 1.

Table 1
Characterization of MSW

	Proximate Analysis (%)				Ultimate Analysis (%)				
	Moisture	Ash	Volatile	Fixed Carbon	C	H	O	N	S
Municipal Solid Waste	6,27	3,98	77,33	12,42	49,07	6,05	39,79	0,95	0,17
Coconut Shell	8,62	0,48	72,78	18,12	47,63	6,29	45,42	0,13	0,046

2.2 Experimental Procedure

As illustrated in Figure 1, the Updraft type biomass gasifier is used because it is believed that this design offers a simpler mechanism and the advantage of effectively processing lower-quality biomass. The biomass is introduced into a cup lined with thin kwool. To initiate the gasification process, the engine is activated, and the temperature on the control panel is initially set to 200℃. The temperature is then incrementally raised by 100℃ every 15 minutes until reaching 400℃. Subsequently, the temperature is further increased to 650℃ after 30 minutes, followed by the introduction of steam into the gasifier. The temperature is then elevated to 750℃ and maintained. Approximately 15 minutes before reaching the target temperature, steam is introduced into the reactor, with a consistent steam-to-biomass ratio of 1.3. Steam settings use a water heater whose electric current can be adjusted using a dimmer.

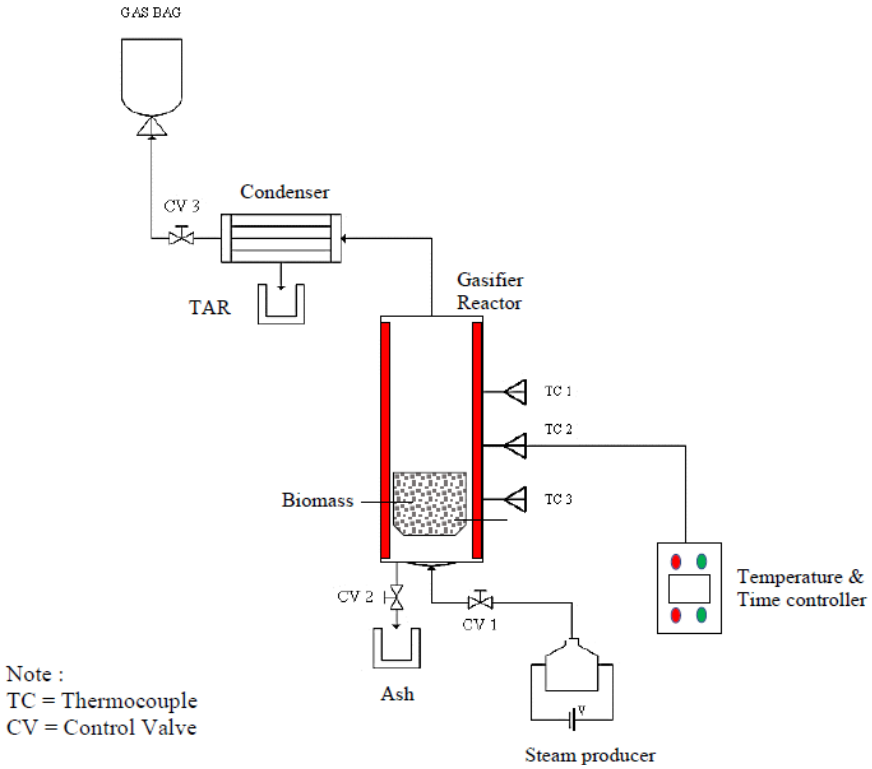

Fig. 1. Diagram illustrating the experimental gasification reactor process

During the data collection in every stage, the predetermined temperature set point is systematically adjusted within the specified range of 650°C to 850°C. The collected data during testing encompasses the composition of the syngas, encompassing carbon monoxide, carbon dioxide, hydrogen, and methane. Additionally, other gathered information includes the quantities of tar and char produced in the residual stages of the gasification process. Following the gasification tests, the obtained syngas samples underwent composition testing using Gas Chromatography (GC) Agilent-7890 equipped with a Thermal Conductivity Detector (TCD). Moleculae Sieve, DC200, UCW, and Haysep columns were employed according to the GPA 2261:2020 standard. Helium gas served as the carrier gas. The heating temperature was initially set at 60°C for a duration of 60 seconds, followed by a gradual increase at a rate of 30°C per minute until reaching a final temperature of 120°C. The gas sample is introduced using a hermetic syringe, and the gas composition results, encompassing CO, CH_4, H_2, and CO_2, were obtained through this process. The char resulting from gasification was tested using X-Ray diffraction (XRD) to determine the crystalline phase of the material. Tests were carried out at a diffraction angle of 20 (10-70^o). XRD testing on char was carried out on the variant coconut shell (CS) to municipal solid waste (MSW) CS:MSW 50:50.

3. Results

3.1 Gas Yeald

As the temperature rose to 750°C, the concentration of carbon monoxide gas declined slightly from 41.35% mol to 41.30% mol. Additionally, with a further temperature increase to 850°C, there was an observed rise in CO gas content, reaching 43.45% mol as Figure 2. The observed concentration of the carbon monoxide in this study exceeds that of previous research, attributed to differences in reactor design, biomass type, and the gasifying agent employed. Elevated temperatures tend to stimulate increased CO production, primarily due to the influence of the Boudouard reaction (refer to Eq. (1)) and the steam and carbon reaction (refer to Eq. (2)). These reactions contribute to

heightened CO and H_2 gas yields. The substantial production of CO gas, as indicated in the figure, suggests that these reactions occur concurrently during the gasification process.

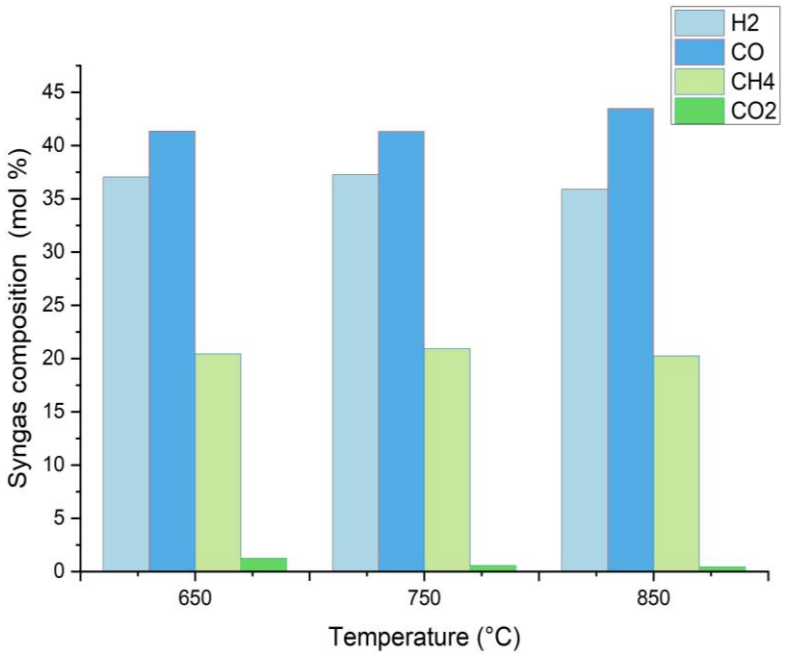

Fig. 2. Gas yield at varied gasification temperatures

The findings regarding CO gas align with Almeida *et al.,* [18], showing an increase in CO gas yields with rising temperatures. However, their study reported a decline in CO gas yield at temperatures between 850°C and 900°C, attributed to a water-gas shift reaction (refer Eq. (3)) causing a reaction between CO and H_2O, thereby reducing CO concentration. Similar observations were made in the studies by Sulaiman *et al.,* [17], indicating a decrease in CO concentration with temperature increases due to reactions with H_2O, resulting in the production of CO_2 and H_2 gases.

The syngas exhibiting the highest hydrogen content is observed at a temperature of 750°C, registering a mole concentration of 37.25%, indicating an increase compared to previous temperature. However, sat 850°C, the H_2 concentration decreased to 35.89%. Elevated temperatures in the reactor enhance hydrogen production by providing energy for the endothermic reaction. Increased hydrogen production due to the water gas shift reaction (Eq. (2)) contributes more significantly than the Boudouard reaction (Eq. (1)), further increasing H_2 concentration in the resultant gas.

Studies by Syarif *et al.,* [21], have explored the increase in H_2 gas yield, demonstrating that gasification at higher temperatures enhances hydrogen production. However, the results presented in Figure 2 do not fully align with existing theory, potentially due to mismatches between high temperatures and the mass of fuel used, leading to fuel depletion as gasifier temperature rises.

The composition of CH_4 gas remained relatively constant across temperature variations. At 650°C, methane gas yeald was 20.41% mol, and at 750°C and 850°C, concentrations were rise up to 20.90% mol and go down 20.23% mol, respectively. The stability was credited to the utilization of steam as the gasifying agent, leading to water gas shift reactions (as per Eq. (3)) and methanation (as per Eq. (4) to Eq. (7)). As temperatures increase, this reforming of CH_4 gas yeald tends to occur, reducing methane concentration and increasing mole persentace of H_2 and CO gas yeald. The findings regarding methane (CH_4) in this investigation surpassed those of Almeida *et al.,* [18], who reported a constant CH_4 value of around 6% across all temperature ranges tested, highlighting differences in reactor design and biomass type.

CO_2 concentration also decreased with each temperature range tested. The highest concentration was at 650°C, measuring 1.22% mol, followed by 0.55% mol at 750°C, and 0.43% mol at 850°C. The use of air as a gasifying agent typically produces high CO_2 concentrations, but steam as a gasifying agent, conversely, reduces CO_2 content in the resulting gas. This reduction is attributed to methane reformation at high temperatures, overlapping between reactions (5) to (7), Impacts the decline in the production rate of CO_2 at elevated temperatures. The liberated CO_2 can engage in reaction (6), tar splitting, and the Boudouard reaction (1), thereby contributing to the decrease in CO_2 content at the maximum test temperature.

To investigate the influence of MSW and coconut shell mixture, gasification tests were also conducted at the optimum temperature of 750°C as depicted in Figure 3. The highest hydrogen production occurred with a mixture of municipal solid waste (MSW) and coconut shell (CS) in a 50:50 ratio, reaching 37.27%. When compared to pure MSW biomass and pure coconut shell biomass, a higher hydrogen content was observed with pure MSW. Similarly, this trend was observed in CO production.

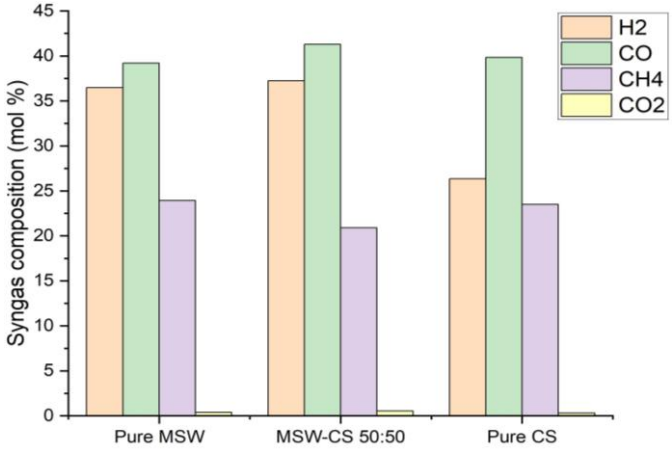

Fig. 3. Gas yield at varied biomass composition

3.2 Analysis of Heating Value

The syngas composition obtained has an impact on the total lower heating value (LHV) or Net caloric value (NCV) generated by gas yields. In accordance with the ASTM 3588 standard, quantify the amount of energy released during combustion or other relevant processes and is measured under specific conditions. The LHV for CO, CH_4, and H_2 within syngas is established at 282.9 kJ/mol, 802.71 kJ/mol, and 241.79 kJ/mol, respectively. The formula employed for computing NCV in syngas is outlined as follows:

$$NCVV_{syngas} = \sum_{i=1}^{n} Y_i \cdot NCV_i \quad (12)$$

Y_i = Gas Yield persentace
NCV_i = NCV = Net caloric value of combustible gas

The Lower Heating Value (LHV) of the syngas exhibits an increasing at 750°C, followed by a decrease at 850°C. This rise is driven through the alteration of temperature, the gas composition is influenced. The peak LHV value is observed at 750°C, reaching 374.67 kJ/mol (refer to Figure 4), whereas the lowest LHV value is recorded at 650°C, measuring 370.35 kJ/mol. In the course of the study, the LHV value is predominantly influenced by the gas composition of CO and H_2, with a

relatively smaller impact from the composition of CH_4. However, it is noteworthy that the overall temperature does not significantly impact the augmentation of LHV values, potentially attributed to the stochastic nature of combustion kinetic reactions.

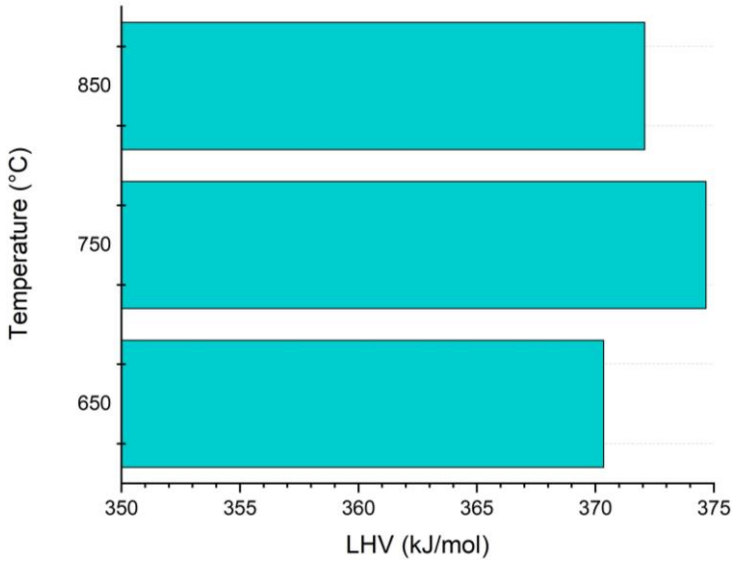

Fig. 4. Lower heating value at temperature variations

3.3 Analysis of Char and Tar

The primary output of gasification is syngas, yet there are accompanying by-products such as ash, char, and tar. The quantity of these by-products generated can differ based on the type of gasification reactor used. As indicated by a study by Rivas [22], the gasification process employing an updraft reactor will yield by-products in the form of tar and char.

The gasification of coconut shell and MSW at 650°C yielded 12.15 grams of char, constituting 24.4% of the total mass, and 1.89 g of tar, representing 3.78%. At 750°C, it produced 10.35 grams of char (20.7%) and 5.69 g of tar (11.38%). When the temperature was increased to 850°C, the production resulted in 10.55 g of char (21.1%) and 10.20 g of tar (20.4%) (refer to Table 2). Char, as a byproduct of cogasification, represents the mass that remains in the reactor after undergoing multiple stages of gasification. The residual charcoal mass in this study was comparatively high, attributed to insufficient heating time within the reactor, ranging from 20.7% to 24.4% of the total fuel used. A comparison with the characteristic analysis of coconut shell and MSW revealed that the char content was approximately six times higher than the results of the characteristic analysis. This increased char mass could be attributed to uneven temperature distribution in the biomass cup, causing incomplete combustion and leaving a significant portion of the fuel unburned. Tar, described as a viscous black liquid formed during the pyrolysis process, resulted from the mixture of complex hydrocarbons [23].

Table 2
Tar and Char pasca gasification process

Temperature (°C)	*Char* (g)	Tar (g)
650	12,15	1,89
750	10,35	5,69
850	10,55	10,20

$$C_nH_mO_p \text{ (biomass)} + \text{Heat} \longrightarrow C_aH_bO_c \text{ (tar)} + C_xH_yO_z \text{ (gas)} + H_2O + \text{char} \quad (13)$$

In gasification using an updraft reactor, the tar production typically falls within the range of 5-20%, with the maximum tar production reaching 20% of the biomass raw material mass. The highest tar production in the gasification of coconut shells and MSW was observed at 850°C, amounting to 10.20 grams (20.4%). The elevated tar production can be attributed to the design of the reactor and the reaction operation parameter [24]. At temperatures below 500°C, tar production tends to increase, and gas production decreases as the reactor temperature approaches 500°C. This is due to tar cracking occurring as the temperature nears 500°C, resulting in the production of CO, CO_2 gas, and steam (H_2O) [25]. In this study, the introduction of steam 15 minutes before reaching the set point temperature led to the generation of significant condensed hydrocarbons, contributing to the increase in tar production.

Design of the gasifier employed in this investigation has the disadvantage of producing elevated tar levels and displaying a sluggish initial combustion process. This prolongs the time taken for temperature distribution from the combustion zone to other regions. The reactor's heigth also influences tar formation, as taller reactors require more time to distribute heat. The prolonged heat distribution time leads to a delayed attainment of the target temperature, contributing to an increase in tar formation [24].

In XRD testing, according to Figure 5, the XRD pattern shows the presence of polycrystalline calcite. This is identified at phase angle (104) 2θ=29.81; (110) 2θ=35.78; (113) 2θ=39.22; 2θ=40.32; 2θ=42.97; (116) 2θ=48.33; (10 10) 2θ=57.25 [26,27]. This element is thought to increase the reactivity of biomass gasification. With the presence of this natural catalyst, it was found that there was an increase in hydrogen production.

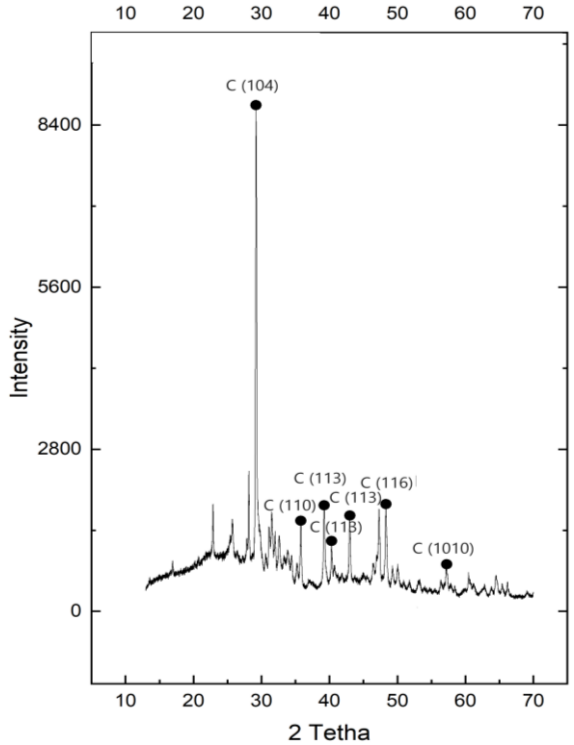

Fig. 5. XRD analysis and (b) gas composition in variations of coconut shells and MSW

4. Conclusions

Cogasification testing of the updraft system was conducted using an electric induction heater. The biomass utilized was a blend of Municipal Solid Waste (MSW) and coconut shells, aiming to enhance gas space reactivity throughout the process. The most favorable outcomes, including syngas with the highest hydrogen (H_2) content and Lower Heating Value (LHV), were achieved at an operational temperature of 750°C. With a further increase in temperature, there was a decline in the proportion of H_2 gas and the calorific value of the resultant LHV. This decline can be attributed to the presence of a natural catalyst (calcite), obtained from organic materials, particularly chicken bones, which undergo agglomeration at elevated temperatures. Consequently, at the highest temperatures, carbon monoxide dominates the gas composition. The by-products generated post-gasification include charcoal and tar. At a temperature of 750°C, the biomass fuel undergoes complete decomposition, evidenced by the lowest percentage of char and tar. This indicates effective material decomposition at this temperature, resulting in an ideal gas yealds concentration and impacting the heating value.

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