Effect of limestone catalyst on co-gasification of coconut fronds and wood chips

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Abstract. Biomass energy via gasification is an attractive substitute of fossil fuels. The distribution of biomass on the earth is scattered, so transportation and collection of biomass complicates the supply of biomass especially when the gasification rely on one type of biomass. Therefore, co-gasification of different biomass is proposed as a potential solution for interruption-free gasification. Beside, unwanted by-products such as tar that cause blockage in downstream equipment can be minimized through the use of catalyst in gasification to accelerate tar reforming process. In this study, catalytic co-gasification of blended feedstock of 70% wood chips and 30% coconut fronds was carried out in a downdraft gasifier using limestone as primary catalyst. The effects of catalyst loading (0%, 30%, 50%, and 70% w/w) on syngas composition, gas yield, carbon conversion efficiency and heating value of syngas were investigated. The results showed that at 50% biomass to catalyst ratio, maximum H₂ content of 11.39%, CO of 17.88%, carbon conversion efficiency of 69.49%, gas yield of 1.68 Nm³/kg and higher heating value of syngas 5.11 MJ/Nm³ were achieved. Higher catalyst loading (70%) blocked the air passage, which caused poor gasification. No more than 50% catalyst suggested for stable co-gasification operation.

1 Introduction

Energy demand has been growing for the past several decades due to rapid industrial and urban development, meanwhile fossil fuel reserves have been in decline. Biomass energy is a popular alternative to substitute fossil fuels [1-3]. Malaysia provides a huge quantity of biomass resources such as agricultural wastes, forest residues and urban wastes [4].

However, certain types of biomass are very sensitive to weather and temperature, thus required supply of feedstock throughout the year become a major problem for gasification operation specially when operation is dependent on only one type of biomass. Interruption of biomass supply, consequently affects the reliability of the gasification operation. Moreover, gasifiers are normally designed for one type of biomass gasification. In addition, not all

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biomass materials are suitable to gasify using downdraft gasifier due to their different physical nature such as density, particle size, shape etc. The combination of various forestry and agricultural wastes as feedstock for gasification provides higher operation flexibility and minimizes problem associated with the effect of seasonal supply variations [5]. Many researchers co-gasified coal and biomass at various blending ratios [6, 7]. However, the studies on co-gasification of two different biomass materials are very limited [8-11].

Biomass can be converted into synthesis gas via gasification. Biomass gasification is the partial combustion which converts the of solid biomass fuel into useful gases and chemical through a series of chemical reactions [12]. The syngas produced from biomass gasification are mainly consists of hydrogen, carbon monoxide, methane, carbon dioxide, water vapor and nitrogen. The yield of products of gasification is dependent on several parameters, which include temperature, biomass species, particle size, and reactor type as well as addition of catalyst [1-3, 10, 13, 14]. From the past studies, it is found that the gas yield, especially H₂ could be improved by adding catalyst together with, by improving the higher heating value of the synthesis gases [11, 15-17]. Many studies have been attempted to introduce the mineral catalyst in biomass gasification especially calcium based catalyst including CaO and CaCO₃ limestone and concrete waste [18, 19]. Limestone shows moderate performance, especially for heavy tar reforming. Past researches also indicated that the calcium based catalyst can facilitate the reaction rate, increase the syngas production and lower the by-product output so as to increase the economic feasibility of the biomass gasification [16]

In this study, air catalytic co-gasification of coconut fronds and wood chips was performed in downdraft gasifier. The effect of catalyst loading was investigated on syngas composition, gas yield, syngas heating value, and carbon conversion efficiency.

2 Materials and Methodology

2.1 Characterization and Preparation of Feedstock

Wood (*acacia mangium*) and coconut (*cocos nucifera*) fronds were considered potential feedstock for the present study. Wood logs were collected from the landscape area of Universiti Teknologi PETRONAS, Malaysia during trimming activities. These logs were chipped down using a wood shredder at the wood factory. The coconut fronds (CF) were collected from Teluk Intan, Perak, Malaysia. The fronds were cut down using a power circular saw. Ultimate and proximate analyses of feedstock were determined according to ASTM (American Society of Testing Materials) D3176-09 and ASTM E1755-01, respectively. Leco AC-350 bomb calorimeter was used for determined higher heating values of the feedstock according to ASTM D4809-00 standard procedure. The wood chips (WC) and CF were sieved to particle sizes ranging between 10-25 mm and were mixed at the blending ratio of 70% WC and 30% CF on mass basis. The moisture content of feedstock was determined according to ASTM E871-82 standard test method. The characterization results of the feedstock are presented in Table 1. Natural limestone was used as catalyst and it was collected from Ipoh, Perak, Malaysia. Elemental analysis of limestone are shown in Table 2 [20] Limestone has a high CaO up to 70.91%. Before using limestone as a catalyst, it was calcined at 900°C for 4 hours.

Table 1. Thermochemical properties of wood chips (WC) and coconut fronds (CF).

Biomass	Ultimate Analysis (wt. %)					Proximate Analysis (wt. %)				HHV (MJ/kg)
	C	H	O*	S	N	VM	FC	Ash	MC	
WC	43.54	3.59	51.71	0.16	1.00	88.07	10.77	1.16	11.40	18.09
CF	40.02	6.03	53.27	0.54	0.14	62.37	17.76	7.92	11.95	15.22

Table 2. Chemical composition of the selected limestone [20].

Content	Composition Percentage (wt. %)
CaO	70.91
MgO	16.80
SiO ₂	2.19
Al ₂ O ₃	4.43

2.2 Experimental Setup

A batch feed downdraft gasifier was used for catalytic co-gasification. The schematic of the experimental setup is shown in Figure 1. The gasifier consists of a cylindrical body with an internal diameter of 300 mm and a height of 1505 mm, with a capacity of 33.6 kW thermal output. The temperature of the gasifier was monitored by using six Type-K thermocouples, which were mounted at the wall of the gasifier with an interval of 215 mm. Thermocouples were plugged into a USB (universal serial bus) data logger and data readings were recorded at every 30 seconds intervals and were stored in the computer. Air was supply to the gasifier, with a commercial vortex blower of 360 L/min nominal flow rate and working pressure of 16 kPa was used. The flow rate of the inlet air was controlled by using rotameter. X-Stream X2GP gas analyzer was used for syngas analysis. A gas flare point was provided on the outlet piping in order to check the combustibility of the synthesis gases and to burn poisonous gases such as CO before being released to the environment.

2.3 Experiment Procedure

The catalyst was placed at the grate of the gasifier. The catalyst loading used, was varying as 30%, 50%, and 70% (w/w) of the feedstock. Then gasifier was filled with 14 kg of blended feedstock of WC and CF. Blending ratio and weight of batch were considered constant for all co-gasification experiments. Scrap paper and an ignition lighter were used to start ignition from ignition hole. After ignition, the hole was closed and the air was supplied the blower through the air inlet pipe. The airflow rate was adjusted and controlled by using a rotameter and valve. The temperature was measured using thermocouples and data was being recorded by a data logger and computer. For syngas analysis, the sample gas was tapped from the syngas outlet pipe and then was pumped to the gas cooling and cleaning system before it was supplied to the gas analyzer. The gas analyzer was connected to the computer with a local internet connection and the composition of the gas was recorded in the computer. The remaining syngas was flared at the flare points.

Once the feedstock was fully consumed, the process was stopped and the gasifier was allowed to cool down. Char and ash were collected from the ash box and on the grate. In order to determine the carbon conversion efficiency of the process, weight measurements of the ash and char were made by using Ohaus precision standard weighing balance.

Fig. 1. Schematic of experimental setup.

2.4 Data Interpretation and Calculation

The average syngas composition, higher heating value, gas yield and carbon conversion efficiency were applied to evaluate the performance of the catalytic co-gasification. The higher heating value of syngas was calculated by using empirical formula given by [12]:

$$HHV_{\text{Syngas}} = 12.63 \times X_{\text{CO}} + 12.74 \times X_{\text{H}_2} + 39.82 \times X_{\text{CH}_4} \quad (1)$$

where, X_{CO} , X_{H_2} and X_{CH_4} are volume percentage of gas field. Gas yield from the gasification was calculated by [21]:

$$Y = \frac{Q_a \times 79\%}{W_b (1 - X_{\text{ash}}) N_2\%} \text{ Nm}^3/\text{kg} \quad (2)$$

where, Y (Nm^3/kg) is the gas yield, Q_a is the flow rate of air (Nm^3/hr), W_b is the flow rate of biomass (kg/hr), X_{ash} is ash content in biomass (weight %) and $N_2\%$ is the volumetric percentage of nitrogen in the dry syngas. It was assumed that air composition consists of 21% oxygen and 79% nitrogen, furthermore all the oxygen in air reacts with feedstock during gasification process. In addition, syngas consists of CO , H_2 , CO_2 , CH_4 and N_2 , contents, where N_2 was determined by balanced.

Carbon conversion rate of each experiment was calculated by using the formula given by:

$$\eta_c = \frac{Y(\text{CO}\% + \text{CO}_2\% + \text{CH}_4\%) \times \text{Mc}}{22.4 \times C\%} \times 100 \quad (3)$$

where, $C\%$ is the mass percentage of carbon in the feedstock obtained from the ultimate analysis. Y is gas yield in Nm^3/kg , where, % denotes volumetric percentage of CO , CO_2 and CH_4 in syngas. Mc is the atomic mass of carbon atom in grams and 22.4 is the volume of one mole of an ideal gas in litres at standard temperature and pressure.

3 Results and Discussion

3.1 Steady Operation

Experimental results for batch fed operation of a downdraft gasifier are presented in this section. Figure 2. shows the gas composition on volume percent at different catalyst addition 0%, 30%, 50% and 70%. Unlike continuous feed operation, the results for syngas composition show variation with operation duration for the case of batch fed operation due to sharp increase during start-up and decrease near the end of the operation due to depletion of the feedstock. From Figure 2b, it can be seen that CO increased from 0% to 20% within the first 10th minutes. After the 10th minutes of operation, it is observed that the gas composition of the gasification is found to be stabilized until the 30th minutes. The decrease in gas composition after the 30th minutes of operation was caused due to the batch fed operation, whereby most of the feedstock was consumed near the end of the experimental run. Therefore, the time interval between 10th and 30th minutes of operation could be taken as the stable duration of operation.

Fig. 2. Gas composition at various catalyst loading (a) 0%, (b) 30%, (c) 50%, and (d) 70%.

3.2 Effect of Limestone Catalyst on Gas Composition and Gas Yield

In this work, limestone was used as catalyst for gasification. The effect of the catalyst was studied for the airflow rate at 360 liter per minute and air as the gasifying medium. It can be seen that each component of gas composition was higher when catalyst was used. This observation was in line with [13, 15, 18, 22]. Figure 3(a) indicates that the presence of limestone clearly encourage the production of H₂ and CO, while the content of CH₄ and CO₂ slightly decreased with increasing catalyst loading [23]. At 0% catalyst, H₂ was amounted to 7.32% and CO was about 9.35%. When 30% catalyst was introduced, H₂ content was amounted to 9.04% and CO was about 17.38%. The optimum amount of H₂ was produced

when 50% catalyst was added to the gasification, which amounted to 11.39%, and CO was 17.88%. At 70% catalyst, H₂ and CO content were low due to the improper airflow as too much of limestone had blocked the airflow passage. The H₂ content has increased 28.3% from that as in the non-catalytic gasification. The quantity of CO₂ was found to be higher than that obtained from the non-catalyzed case. With increased catalyst loading from 0% to 50%, higher content of CO₂ was obtained due to the release of CO₂ from limestone. The reforming reaction of tar on a limestone surface by capturing carbon to produce more H₂ and CO, according to reaction Eq.(4) [12]. Besides Figure 3(b) shows that addition of catalyst has favored the gas yield. Gas yield indicates the ability of feedstock converted into gaseous products. Generally, gas yield shows the amount of gas in normal cubic meter produced by one kilogram of biomass. At non-catalytic gasification gas yield was 1.38 Nm³/kg and has increased 21.78% up to 1.68 Nm³/kg at 50% of catalyst addition.

Fig.3. (a) Average syngas composition and (b) gas yield at various catalyst loading.

3.3 Effect of Limestone Catalyst Heating Value and Carbon Conversion Efficiency

The Figure 4a shows effect of catalyst loading on the syngas heating value. Syngas heating value was 2.82 MJ/Nm³ at non-catalytic gasification and the syngas heating value is between 1.96 MJ/Nm³ and 5.11 MJ/Nm³ at catalytic gasification and this value is similar to results of other studies (4-6 MJ/Nm³) derived from bamboo biomass gasification [24]. The syngas heating value has increased 81% at catalytic gasification. However, at 70% catalyst loading, syngas heating value was low due to the poor airflow rate, whereby the huge amount of catalyst has blocked the air passage. The syngas heating value was significantly enhanced due to addition of limestone, and this is related to the higher reaction rate of hydrocarbon reforming [25]. Furthermore, the chemical composition of feedstock could influence the syngas fraction and heating value in biomass gasification. That is because the H and O elements might have competed to react with C in biomass matrix to form the CH₄ or CO. In general, the CH₄ heating value is higher than CO. Therefore, the higher H₂ exhibits the higher syngas heating value.

Fig. 4. (a) Syngas higher heating value (b) carbon conversion efficiency at various catalyst loading.

Mass conversion efficiency is an important parameter that determines the amount of solid feed in percentage converted to permanent gases. High mass conversion efficiency means low generation of unwanted products such as tar, char and ash. Carbon conversion efficiencies indicates how much of the initial carbon content of fuel is converted to gaseous product, and thus affecting the amount of unconverted carbon in char and tar by products from gasification. Figure 4b shows the variation of carbon conversion efficiencies with different catalyst loadings. At non-catalytic gasification the carbon conversion efficiency was 35.20% and then increased sharply to 59.69% when 30% of catalyst was introduced to the gasification process. The maximum value of carbon conversion efficiencies of 69.49% was observed for the 50% of catalyst addition. The minimal value of carbon conversion efficiencies occurred at 70% catalyst loading, around 24.72% could be attributed to the blockage of catalyst at the gasifier's grate. Airflow rate was disrupted thus most of the feedstock were not burnt completely, therefore the conversion efficiency was low. These values of carbon conversion efficiencies obtained for WC and CF, were found to be comparable with the results for gasification of rice husk which were reported to be 55-81% [26]. The high mass and carbon conversion efficiency of WC and CF which could be mainly due to its low ash and fixed carbon composition which showed its high potential as a fuel for thermal conversion processes.

4 Conclusions

Catalytic co-gasification of blended feedstock of 70% wood chips and 30% coconut fronds was carried out in a downdraft gasifier using limestone as primary catalyst. The effect of catalyst loading was investigated on syngas composition, gas yield, and carbon conversion efficiency and heating value of syngas. The catalyst loading varies as 0%, 30%, 50% and 70%. The optimum catalyst loading (50%) showed that high syngas concentration of fuel components, H_2 , CO and CH_4 . The maximum H_2 , CO, syngas heating value, gas yield, carbon conversion efficiency were 11.39%, 17.88%, 5.11 MJ/Nm^3 , $1.68 \text{ Nm}^3/\text{kg}$ and 69.49% respectively. However, high catalyst loading (70%) is not suitable for downdraft gasifier. The results of current study were found to be comparable with results of past gasification studies of woody biomass. The results showed that WC and CF blend have a high potential as a fuel for gasification. It is proposed that in future different catalyst will be investigated for co-gasification of different biomass for better understanding of biomass to catalyst interaction during co-gasification.

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