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Hydrogen Production Enhancement and Tar Reduction using New Three-Layer Annular Downdraft Air-Steam Biomass Gasifier

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ABSTRACT

This study presents experimental analysis of biomass air-steam gasification for the enhancement of H₂ production in producer gas (PG). Wood pellets which are widely available in Malaysia are used in this investigation. A pilot scale downdraft gasifier based on new three-layers annular reactor design was implemented to produce hydrogen enriched PG. The reactor is initially heated using hot flue gas from LPG combustion which is switched to PG when the reactor reaches stable operation. The effect of steam to biomass (S/B) ratio from 1.1 to 1.5 on PG quality in terms of gas composition, HHV and tar contamination is studied. Maximum H₂ production of 27.7% was achieved at optimum air flow rate of 75 LPM and S/B ratio of 1.2. At the optimum air flow rate, HHV and tar contamination in PG where in the range of 5.06 - 6.08 MJ/Nm³ and 187 - 30mg/m³, respectively.

1. Introduction

Due to the rapid growth of the world's population, energy demand is increasing every day. Most of the world's primary energy demand is fulfilled through the combustion of fossil fuels like gas, oil, and coal. The extensive use of such fuels produces greenhouse gas emissions, requiring the investigation of alternative energy sources. New ways have been proposed to utilize the traditional renewable technologies such as PV, not only for power, but for thermal output residential applications as well [1]. Improvements and new designs have been also proposed to improve the other renewable energy such as residential power generation through low-wind-speed wind turbine designs [2]

Biomass is a promising renewable energy resource due to its wide availability and many environmental advantages [3]. Biomass can be obtained from agricultural, industrial and urban wastes, but a consistent mass production in large quantities can be obtained from microalgae cultivation for liquid biofuels production [4]. Thermochemical conversion of biomass into energy by processes like pyrolysis, gasification, and combustion has been widely investigated in recent years [5,6]. However, the primary method for power generation is still depending on direct biomass

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combustion in steam boilers, which suffers from the lower efficiency and incomplete biomass combustion especially for low-grade biomass types [7].

Biomass gasification, on the other hand, facilitates the utilization of solid biomass more efficiently as it converts it into attainable gaseous fuel form known as producer gas (PG) that can be used directly in IC engine and gas turbine applications. The gasification performance and PG quality are strongly dependent on temperature, gasification agents (air, steam, CO₂ and oxygen), feedstock, and types of gasifiers [8]. Wide range of feedstocks such as hazelnut shells, wood waste from furniture, wood chips and charcoal were investigated in literatures [9-11]. Downdraft gasifiers produce less tar compared to other reactors which makes it suitable for power generation applications [12-14]. As for the gasification agents, air is one of the most common agents since its available free of cost, but it suffers from the presence of nitrogen content. When using air as the gasification agent, PG contains 16-17 % hydrogen (H₂), 10-12% carbon dioxide (CO₂), 17-18% carbon monoxide (CO), 2-3% Methane (CH₄), 45-50% Nitrogen and some higher hydrocarbons and tar [15,16]. Using oxygen as agent increases the higher heating value and quality of PG due to the absence of nitrogen, but it is very expensive [17]. Therefore, steam is proposed as it is more affordable and does not cause N₂ dilution in PG while increasing gas quality and higher heating value. The main challenge, however, is that steam requires an external heating source to maintain the reactor temperature as the reaction is heavenly endothermic in nature [8]. For small lab-scale experimental steam gasification, heat is commonly supplied from heater bands surrounding tube reactors with small diameters to ensure efficient heat supply for the reaction [8]. To further enhance heat transfer from the heater jacket to the fuel, molten blast furnace slag was tested as the heat carrier and shown good heat transfer which enhanced H₂ production from the steam gasifier [18]. Transferring the heat internally through the circulation of hot sand as heat carrier between two fluidized bed (FB) gasification-combustion reactors was also demonstrated experimentally [19]. Other heat sources have been proposed and simulated including solar external heating and the heat from steam itself by using high-temperature steam source to provide the heat as well as acting as the gasification agent [20]. As the difficulty of transferring the heat efficiently to the fuel presents a techno-economic challenge, hindering the upscaling and commercializing steam gasifiers. Thus, providing some of the heat internally has been proposed as a compromise. Burning some of the fuel internally using limited supply if air can provide the needed heat for the reaction in dual gasification agent configuration (i.e., air and steam). The amount of N₂ dilution can be controlled by the amount of air supply. On the other hand, air-steam gasification was shown to be an economically feasible approach to enhance PG quality using existing downdraft gasifiers with minor modification [21-23]. Several published investigations on air-steam gasification with different reactors designs and biomass types are shown in Table 1.

Hydrogen is considered by many researchers as the fuel of the future as it does not emit carbon to the atmosphere during combustion. Other common renewable fuels suffer from combustion stability issues [24] and high emissions especially in reciprocating engines where combustion residence time is very limited [25]. However, hydrogen is considered an ideal fuel for such engines as it does not emit pollutants such as CO, while NO_x emission can be eliminated by removing nitrogen sources [26]. The main downside for hydrogen is its extremely low density, resulting in high pressure and volume requirements for storage. For mass hydrogen storage, underground storage utilizing the natural underground caverns has been proposed [27].

Table 1

Comparison of the results of different types of air-steam gasifiers with biomass

Reactor types	Biomass	H ₂ (%)	CO (%)	CO ₂ (%)	CH ₄ (%)	S/B	ER	HHV (MJ/Nm ³)	Ref
FB Externally heated /727°C	Wood pellets	16.2	11.5	18.6	5.9	0.28	0.19	5.3	[28]
FB Externally heated/800°C	Pine sawdust	32.1	32.1	18.6	7.5	2.7	0.22	na	[29]
FB externally heated	a-cellulose	18.6	8.3	27.4	3.0	1	0.27	7.2	[30]
Integrated gasifier and externally heated /950°C	Coconut shell	21.0	64.0	6.0	9.0	1.95	0.24	13.0	[31]
Updraft External heater	Diary biomass	18.6	11.9	16.1	0.2	0.7	1.53	na	[32]
Updraft preheated feed	Wood pellets	21.8	24.4	16.1	1.7	2.53	na	8.9	[33]
Downdraft preheated feed	EucalyptusWood chip	22.3	18.9	16.2	2.0	0.17	0.12	6.2	[34]
Two-stage Downdraft self-heated	Eucalyptus	19.4	12.2	na	1.6	0.47	0.40	4.2	[35]

It can be noticed that H₂% is heavily influenced by two main factors: the steam-to-biomass (S/B) ratio and external heating. The increase of S/B ratio is directly proportioned to H₂%. However, as the addition of steam require additional external heat supply to the system, heat supply can be in the form of direct reactor heating through heating jacket [28-32], preheating the feed flow (i.e., air and steam inputs) [33,34] or by providing the heat internally through the combustion zone in two-stage reaction [35]. Commonly, the higher heating value (HHV) of PG is below 10 MJ/Nm³, unless a limited amount of air is used, which will increase the amount of external heat supply needed for the steam gasification reactions.

In the current study, air-steam gasification of wood pellets is investigated in a new annular reactor design geometry with three concentric tubes. Hot flue gas is passed through the outer annular tube to supply heat to the reactor while steam is passed through the inner tube and biomass is placed in between. The performance of the reactor is evaluated in terms of reactor temperature, S/B ratio, PG composition, HHV and tar contamination in PG.

2. Materials and Methods

2.1 Materials

Wood pellets are used as biomass feedstock for the experiment as it is available in abundance in Malaysia at a low cost. The average length and diameter of wood pellets are about 20mm and 5mm, respectively, as shown in Figure 1.



Fig. 1. Biomass wood pellets feedstock

Ultimate analysis was done to determine the elemental mass % of carbon, hydrogen, nitrogen, and sulphur in feedstock using CHNS analyzer. The proximate analysis was also done to determine moisture content, fixed carbon, volatile matter and ash content in the feedstock using TGA (Leco 701). The heating value of feedstock was obtained using bomb calorimeter. Table 2 shows the proximate, Ultimate analysis, and HHV of wood pellets feedstock.

Table 2
Ultimate, proximate analysis and
HHV of wood pellets

Proximate analysis	Percentage (%)
Fixed carbon	6.82
Moisture	7.84
Volatile matter	84.34
Ash	0.95
Ultimate analysis	
C	46.12
H	6.97
N	0.16
S	0.29
O ^a	46.46
HHV (MJ/Nm ³)	16.20

^a by difference

2.2 Experimental Set-Up

The experimental set-up consists of the annular reactor, LPG stove, steam boiler, air blower, flare port and gas & tar sampling trains. The reactor is based on a new annular downdraft gasifier design along with external heating jacket as shown in Figure 2. Steam is passed through the inner annular tube to provide some of the internal pre-heat of the reactor. Steam is injected to the intermediate annular tube through a row of 2mm injection holes at the top of the reactor where steam is pushed downwards through biomass which is placed inside the intermediate annular tube. Air is injected at the bottom of the intermediate annular tube above the grate where combustion zone is created. The top of the reactor is sealed, so PG is forced to go downwards through the combustion zone and the grate. Ash and large particles fall to the ash bin by gravity while PG exit port is on the side wall of the reactor. The outer annular tube is used as a heating jacket to provide the needed heat for the steam gasification reaction externally, where hot combustion flue gas is passed upwards. An LPG stove was installed at the bottom of the heating jacket for the external heating of the reactor. K- type thermocouples shown as a, b, c and d in Figure 2, are used to measure the temperature, where they are connected to the data logger. A steam boiler supplies steam at a constant steam flow rate of 30 g/min. An air blower with a rotameter is used to supply air at different flow rates of 25, 50, 75, 100 LPM for the characterization of the reactor. The Mass of wood pellets put inside the reactor was Fixed at 500 g for all the test runs.

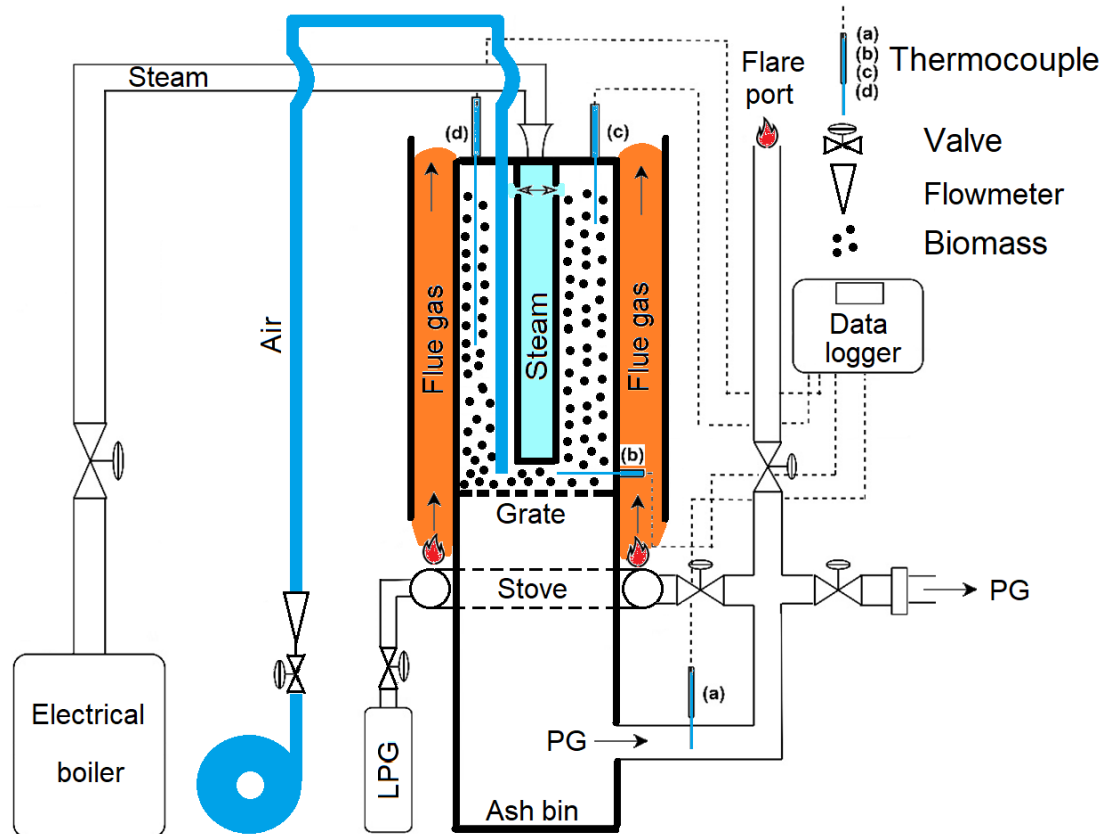
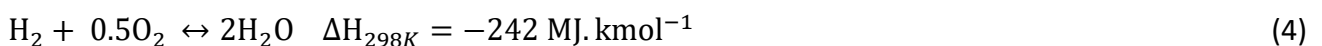
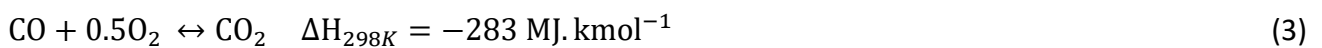
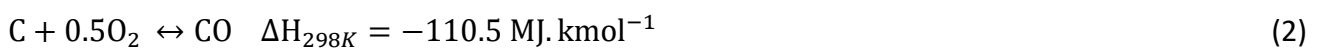
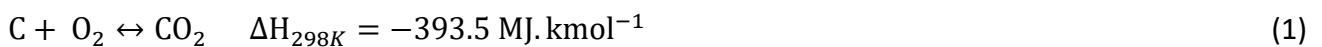


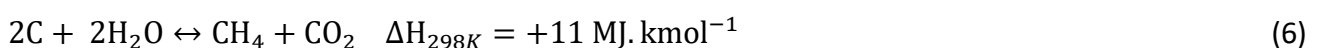
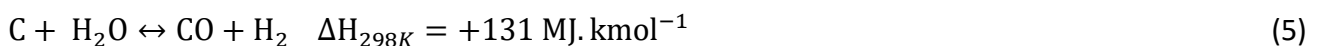
Fig. 2. The Schematic diagram of experimental setup

2.3 Analysis and Sampling

In the air-steam gasification process, endothermic and exothermic reactions occur simultaneously as shown in Eq. (1) to Eq. (11) [36]. The heat generated from the exothermic reactions is commonly limited and not adequate to supply the needed heat for the endothermic reaction as the supply of air is limited. Therefore, external heat supply is still needed to avoid significant PG dilution with N_2 that can drop its quality. Heat is supplied internally through the exothermic oxidation and partial oxidation reactions shown in Eq. (1) to Eq. (4)

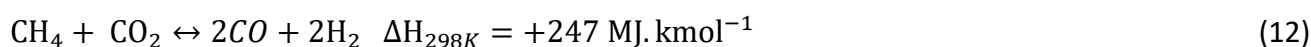
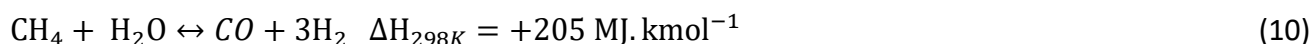
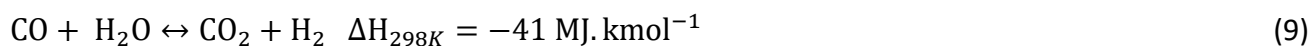


Solid carbon conversion (gasification) reactions, however, mostly require heat supply as shown in Eq. (5) to Eq. (8)





Gas with steam and also interactions among the PG compositions requires heat supply, starting with the water gas shift reaction (WGSR) in Eq. (9), followed by methane reforming reactions in Eq. (10) to Eq. (12)



For this new design annular pilot-scale downdraft gasifier, air is injected at the reactor bottom while steam is injected at the reactor top. Air is in direct contact with biomass above the grate where reactions (Eq. (1) to Eq. (4)) occur which generates heat that is transferred to the upper layers of the reactor. Simultaneously, heat is also supplied from the outer jacket using hot flue gas from the stove, and highest temperature is at the reactor bottom, then temperature is gradually reduced as heat is absorbed by biomass causing the flue gas to cooldown.

In order to analyse the characteristics of the reactor, the main performance indicating parameters are the temperature profile across the reactor height, S/B ratio and PG quality. For PG quality evaluation, gas composition especially H₂%, HHV of PG and tar contamination in the gas are analysed. While the manipulative variable for the experiments is the air supply flow where four flow rates of 25, 50, 75, and 100 LPM at a constant steam flow rate of 30 g/min were tested.

Two sampling trains are connected at the sampling port of the reactor for PG and tar sampling to clean the contamination of gas. The PG sampling train consists of two bottles interconnected in series, as shown in Figure 3(a), to ensure that all the gas was cleaned and dried before being collected into sampling bags. The first bottle was filled with 500 ml water and another with 200g of silica gel. Both bottles are kept in salt-ice-bath at a low temperature. At the end of these bottles, a small pump was used to help the flow of PG into the PG sampling train. PG gas is collected in three sampling bags. The gas composition of these bags was analyzed by GC (Gas chromatography).

The tar sampling train Contains only one bottle filled with 50 ml of isopropanol, as shown in Figure 3(b). This bottle was immersed in salt-ice-bath at a temperature of around -20°C, sufficient to condense the tar compound. A Two-way Electric air pump was used to increase PG flow into the tar sampling train. At the end of this tar sampling train, a flowmeter was installed to measure PG gas flow. After completing all runs, tar was recovered by evaporating isopropanol with the help of a magnetic stirrer at 100°C. After evaporation of isopropanol, tar settles down in the sampling bottle. These tar yields are extracted from the sampling bottle and weighted.

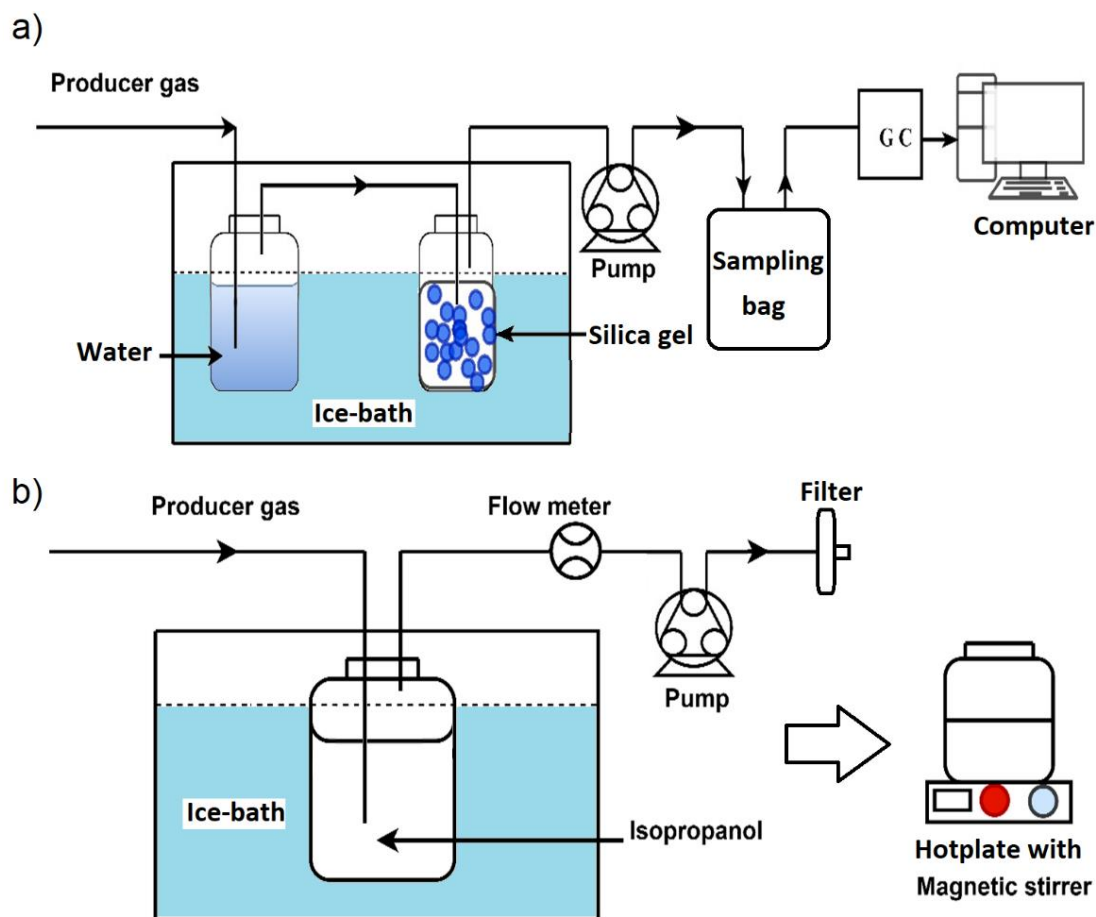


Fig. 3. Schematic diagram of sampling trains of a) PG b) tar

2.3 Experimental Procedure

When wood pellets are placed in the reactor, heat is supplied externally first using LPG fuel in the stove with a continuous flow of air to start up the combustion zone above the grate in air gasification mode (without steam) for 20 min. After that, steam is injected at the reactor top, and heat is supplied internally and externally to activate steam reactions. The reactor is then operated in air-steam gasification mode until the end of the run. LPG flow rate is fixed at constant flow rate of 5 LPM to ensure consistent external heating of the reactor. Temperature profiles for all thermocouples are monitored for the start-up and reactor heating until temperature is stabilized. After that, the fuel of the heating stove is switched from LPG to PG to test the flame stability, and PG and tar samples are collected.

3. Results and Discussion

The experimental study of air steam gasification was conducted, and the influence of the main performance indicating parameters will be discussed in this section. This will include the analysis of temperature profiles, S/B ratio and PG quality in terms of gas composition, HHV and tar contamination at different air flow rates.

3.1 Pressure Distribution

The temperature profiles at reactor bottom, middle, top and outlet are recorded for the different air flow rates during the start-up period until the temperature stabilizes as shown in Figure 4. For the initial 20 minutes, gasifier start-up depends on air flow rate as the external heat supply is constant for all cases. At lower air flow of 25 LPM, the combustion zone temperature above the grate was low of about 195°C and elevated gradually with the increase in air flow reaching 300°C at 100 LPM flow. The temperature dropped slightly once steam was injected due to the instant cooling effect, causing disturbance in combustion. However, temperature elevation was resumed after that, and it was also affected mainly by air flow rate. Some of the excess air, especially at higher flow rates, can drift upwards and oxidize H₂, CO and CH₄ in PG. This will elevate the temperature along the reactor height due to the additional internal heating, but at the expense of the drop in PG quality. This effect was clear at the highest flow of 100LPM with minimal temperature difference between grate and reactor top at 1015°C and 980°C, respectively. On the other hand, the lowest air flow of 25LPM provided limited internal heat supply where grate temperature stabilized at 755°C, which dropped considerably down to 620°C at the reactor top where most of the heat is supplied externally through the outer jacket. As the temperature is crucial for the activation of most of the gasification exothermic reactions, at such low reactor temperatures, the activation of many reactions will be limited which is expected to reduce hydrogen production from steam gasification considerably. The WGSR (Eq. (9)) to oxidize CO from PG into CO₂ using steam was found to be heavily dependent on the reaction temperature as well as the nature of the surrounding materials that can act as a catalyst. The reaction is shifted to the left (i.e., reducing CO reactivity with water) based on Le Chatelier's principle at elevated temperatures [37]. On the other hand, increasing the temperature above 900°C creates favourable condition for the highly endothermic Boudard reaction (Eq. (7)) which competes with the WGSR [38].

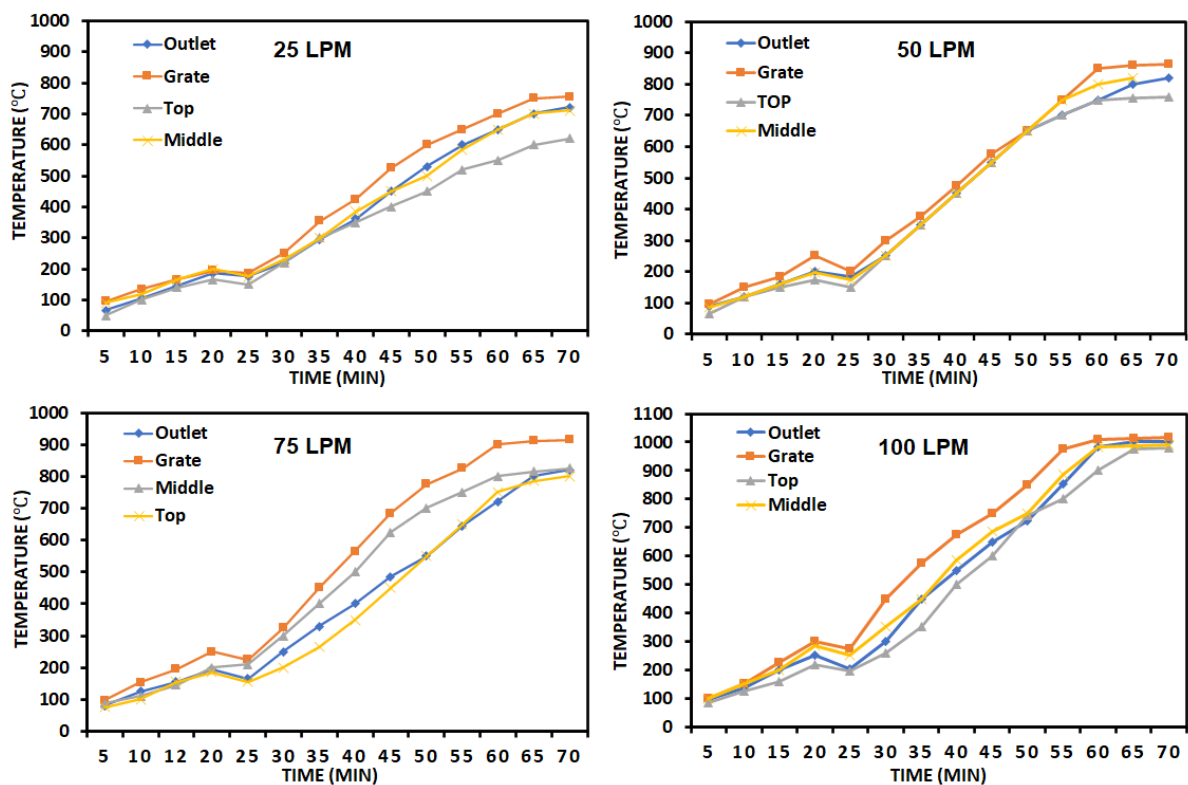


Fig. 4. Temperature profiles at different air flow rates of 25, 50, 75 and 100 LPM

3.2 Steam to Biomass (S/B) Ratio

As the steam flow was kept as a constant in this study, S/B ratio is fully dependent on biomass consumption rate. In the current setup, the consumption of biomass can be contributed to: reaction with air, reaction with steam and thermal decomposition (pyrolysis). As the air flow rate is increased, the contribution of pyrolysis reduces significantly as air and steam spreads to the majority of the reactor height. Biomass consumption is directly proportional to the flow of air as the increase of air flow increases the combustion zone size and accelerates biomass consumption. On the other hand, biomass consumption is inversely proportional to the S/B ratio as steam flow is constant for all setups. Therefore, air flow rate is also directly proportional to S/B ratio as shown in Figure 5. At the moderate steam flow rate of 30 g/min, the highest S/B was 1.5 at 25 LPM air flow rate. This S/B value is still within the acceptable range for effective steam gasification [39]. Also, from the temperatures profile shown earlier, the amount of external heating, despite having low internal heating at this air flow, was adequate for this S/B ratio which did not cause severe cooling effect.

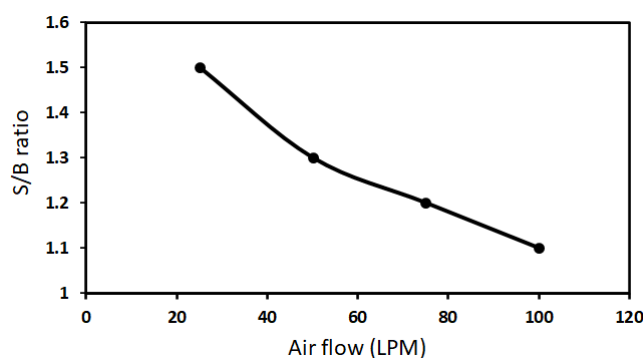


Fig. 5. Effect of air flow rate on S/B ratio

3.3 Producer Gas Composition

The gas composition of producer gas obtained from air-steam gasification is shown in Figure 6 at different air flow rates in the range of 25-100LPM. H_2 production was mainly governed by the change in temperature since steam flow supply is fixed during the experiments. As the external heat supply from the stove is fixed, the change in reaction temperature was controlled by the air flow rate as discussed in the previous section. Increasing air flow increases the size of the combustion zone above the grate and elevates the temperature throughout the height of the reactor. The devolatilization of biomass is accelerated at elevated temperatures which converts biomass into char which can directly react with steam to produce H_2 and CO as shown earlier in Eq. (5). At the lowest air flow of 25LPM, the production of H_2 was low of about 15% due to the low reactor temperature especially at the reactor top which was about $600^\circ C$ where steam is injected. However, H_2 production increased considerably up to 27.7% when the reactor top temperature exceeded $800^\circ C$ at air flow rate of 75LPM. But with the increase of temperature, CO is converted into CO_2 through the water gas shift reaction shown earlier in Eq. (9), which also produces H_2 . This dropped CO concentration down to about 12% while increasing CO_2 concentration up to 14.6%. Further increase in air flow up to 100LPM further increased CO_2 concentration not only through water gas shift reaction but also by the direct oxidation of C and CO using the excess air supply following Eq. (1) and Eq. (3). This in turn affected the other gas concentrations as PG is becoming more contaminated with CO_2 and N_2 . It was reported in literatures [38] that at elevated temperatures above $900^\circ C$, the Boudard reaction shown in Eq. (7),

is fully activated. This caused slight elevation in CO production despite the sharp drop in other PG compositions at maximum air flow rate.

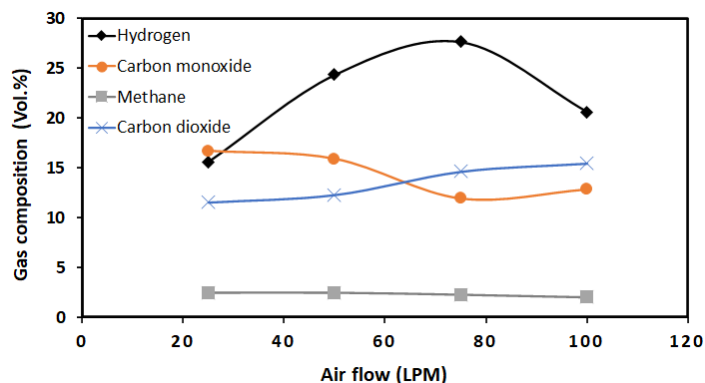


Fig. 6. Effect of air flow rate on PG composition

3.4 PG Higher Heating Value

The heating value of PG is mainly contributed by H₂ and CO contents where both gases have similar heating value while CH₄ contribution is insignificant. The effect of air flow supply on the HHV of PG is shown in Figure 7. H₂ content showed considerable effect on HHV as the trends for HHV and H₂ concentration showed close resemblance. However, maximum HHV of 6.1 MJ/m³ was achieved at air flow of 50LPM instead of 75 LPM at which H₂ concentration is maximum. This was mainly caused by the effect of CO concentration of 15.9% at 50LPM air flow compared to 12% at 75LPM which caused the accumulated value of HHV to slightly drop down to 5.9 MJ/m³. At maximum air flow of 100LPM, the significant drop of H₂ content as well as the drop in CH₄ content caused a considerable drop in HHV down to 5.1 MJ/m³. Having an excessive amount of air will cause a degradation in PG quality as the contamination with CO₂ and N₂ increases, which is also in a good agreement with the experimental findings of other researchers [40].

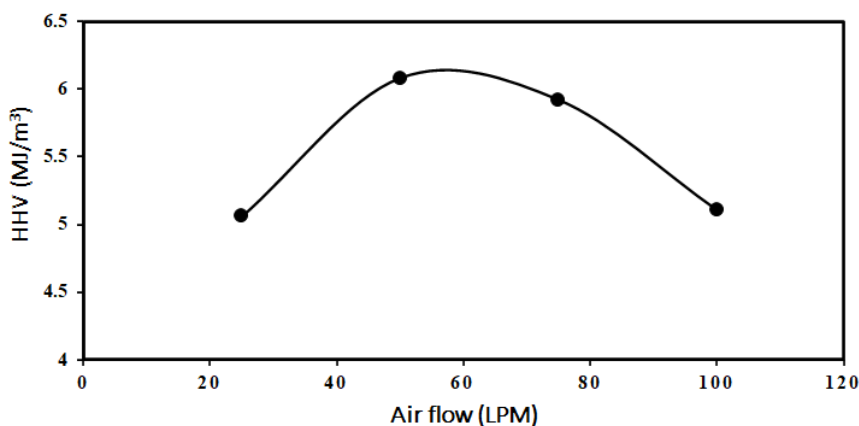


Fig. 7. Effect of air flow rate on PG HHV

3.5 PG Tar Contamination

One of the downsides of biomass gasification is tar contamination in PG which causes serious blocking and fouling of the surfaces and pipelines of the engine when PG is used as a fuel which reduces the engine lifetime. Removing tar by cleaning PG after the gasifier elevates the maintenance

cost of the system. Therefore, investigating the tar generation and the ways to reduce it inside the reactor is of an utmost importance from the techno-economic point of view especially when scaling up the system for power generation. Tar contamination was obtained from the tar sampling train as described earlier. The effect of air flow rate on tar contamination in PG is shown in Figure 8. Tar contamination was found to be inversely proportional to the reaction temperature due to the acceleration in tar cracking when temperature is increased [41,42]. At low air flow rate, biomass in the middle section of the reactor is exposed to a mild temperature level near the reactor wall while the steam supply from the reactor top and air supply from the reactor bottom is very limited at this zone. This creates a pyrolysis dominant zone where biomass is decomposed thermally without any reacting agent. The thermal decomposition of lignin, cellulose and hemicellulose at relatively lower temperatures below 600°C causes the formation of primary tar [43]. With the limited supply of air of 25LPM, the combustion zone size is limited around the air inlet nozzle which accelerated the formation of primary tar above this zone through the carbonization and pyrolysis of biomass. Moreover, the size of the combustion zone is not wide enough to cover the whole width of the reactor above the grate which allow tar to pass through the cold zones without cracking which elevated tar contamination to a high level of 187 mg/m³. Increasing air flow rate reduced the size of the pyrolysis dominant zone at the middle of the reactor which reduced the sources of tar generation. Also, increasing the size of the combustion zone increased the tar cracking when PG passes through this zone in a downdraft manner, and with the increase in the combustion zone height, the residence time for tar cracking is increased. At the highest flow of 100LPM, tar contamination in PG was reduced down to 30 g/m³ which is comparable to the amount of tar generated from fluidised bed gasifiers [44].

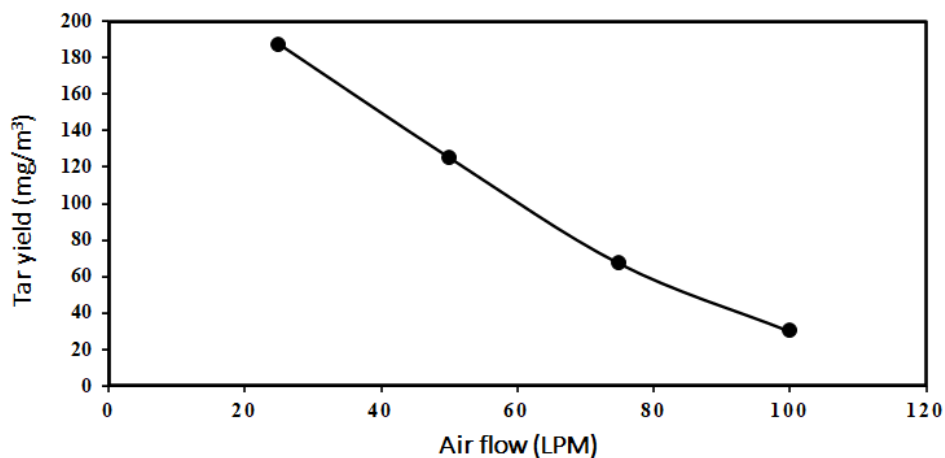


Fig. 8. Effect of air flow rate on tar contamination in PG

3.6 Sustainability Test of Reactor

As the air-steam gasification can provide some of the required heat internally, the necessity for external heating become less demanding. In order to test the sustainability of the reactor when using PG as the source for the external heating (through the stove), the fuel source was switched from LPG to PG after reaching steady state temperature. The sustainability test was performed for about 10 minutes with the different air flow rates and the flame stability was observed. At both ends of the air flow scale (i.e., 25LPM and 100LPM), frequent flame blow-off was observed indicating the low stability of PG combustion, which can be explained by the lower heating values at low and high flow ranges. On the other hand, the flame at 50 LPM was the most consistent and stable without any

flame blow-off. The low CO laminar flame velocity can cause instability in the stove diffusion flame. The dilution with N₂ and CO₂ could also affect the flame stability negatively as it compromises the air-fuel mixing quality which is already limited by the diffusion flame nature with natural air draft upwards by heat. Thus, the added flame stability could be attributed to the positive effect of H₂ fuel enrichment which increases the reactivity rate and laminar flame velocity in PG.

4. Conclusions

Air-steam gasification of wood pellets was carried out on a new design pilot-scale annular downdraft gasifier. The reactor was successfully characterized with wood pellets as the fuel, and air flow range of 25-100LPM. It was shown that the reactor temperature is directly proportional to the air flow rate, which can be explained by the expansion of combustion zone by air increments, resulting in more internal heat supply. Consequently, the reactivity of steam-carbon and steam-CO reactions was enhanced by increasing air flow but to a certain extent. Increasing air flow from 25LPM to 75 LPM has increased H₂ production from 15% to around 28%. However, excessive air supply of 100LPM has shown negative effects on PG quality as it was heavily contaminated with CO₂ and N₂. Additionally, the elevation in temperature above 900°C at 100LPM air flow resulted in the activation of highly endothermic reactions such as Boudard reaction which competes with steam reactions and block the active sites on the char surface. As for PG heating value, highest HHV value was not directly associated with the highest H₂ production. Thus, maximum HHV of around 6 MJ/m³ was achieved when both CO and H₂ concentrations were high. It was also shown that the addition of low air flow rates increased tar contamination significantly as it caused rapid pyrolysis reaction above combustion zone due to the elevation in temperature which converted volatile matter in wood into tar. While the limited size of the combustion zone at low flow did not provide adequate tar cracking. On the other hand, increasing air flow has resolved this issue as the reactor was able to act like a downdraft gasifier where PG is passed through a large combustion zone before it is released from the reactor which effectively reduced tar contamination.

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