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Hydrogen-Rich Gas Production from Palm Kernel Shell by Applying Air Gasification in Fluidized Bed Reactor

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ABSTRACT: An experimental study of gasification of palm kernel shell residues from the palm oil industry, as a potential hydrogen feedstock, is investigated. The gasification is conducted in a bench-scale fluidized bed gasifier with 55 mm diameter and 850 mm height. The operating conditions were studied in the following ranges: reaction temperature in gasification zone (750–1100 °C), feeding rate (0.20–1.21 kg/h), feedstock particle size (0.1–5 mm), and equivalence ratio (ER) (0.23–0.27). The increasing temperature and equivalence ratio was found to significantly influence the gas yield (up to 90 wt % conversion on the raw biomass) at 1000 °C and hence hydrogen composition (40 vol %). The feedstock particle size and feeding rate only showed minor influence. In conclusion, palm kernel shell is a potential candidate for hydrogen production.
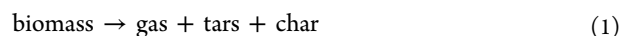
1. INTRODUCTION

Hydrogen production from biomass has become an attractive alternative in energy generation. Hydrogen is not only considered as renewable energy for future but the best supersede for fossil fuels with the high heating value (HHV) for transportation, home usage, and heating. In Malaysia, agricultural waste is foreseen as the best biomass feedstock for hydrogen production due to the rapid growth in the agriculture sector over the years. The utilization of agricultural waste as hydrogen feedstock will anticipate both future shortage in petroleum supply and landfill disposal problem of the agricultural residues.

Palm kernel shell is one of the best potential biomass feedstock available. In Malaysia in 2000, the palm kernel shell generated reached up to 471 thousand tones, and potential power generation from the utilization of palm kernel shell was about 77.65 MW. The oil-palm solid wastes (including shell, fiber, and empty fruit bunch) are cheap and abandoned materials produced during the palm oil milling process. For every ton of oil-palm fruit bunch being fed to the palm oil refining process, about 0.07 tons of palm shell, 0.146 tons of palm fiber, and 0.2 tons of empty fruit bunch are produced as the solid wastes.[1]
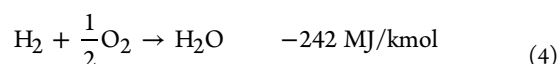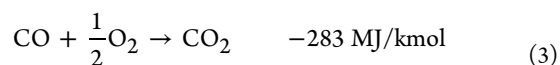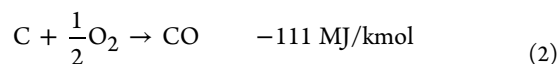
Thermochemical gasification of biomass is well-known technology that seems to have a feasible application and has been developed attractively for industrial applications. As a result of the presence of non-oxidation conditions, the pollutant and green house gases emissions are much lower.[2−7] An air gasification process can convert solid agricultural residues into a synthesis gas that is suitable for use in electricity production or for the manufacture of chemicals, hydrogen, or transportation fuels. The producer gas from biomass gasification can be directly used as fuel in internal combustion engines or gas turbines.[8] Air gasification has advantages over other gasifying agents, simplifying the gasification process and reducing operation and maintenance costs, because air separation to

obtain oxygen is a more complex and expensive process. On the other hand, in the case of air gasification, its nitrogen content remains as an inert component in the producer gas that dilutes the fuel gas and leads to a low calorific value fuel.[5]
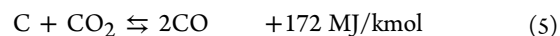
According to Xiao and co-workers,[5] it is generally reported that biomass gasification occurs through the main three steps. At the first step in the initial pyrolysis, it is at high bed temperature in the first part of reactor that the major amount of gas is produced. The second step is the tar-cracking step that favors high temperature reactions, and more light hydrocarbons, gases such as H_2, CO, CO_2, and CH_4, are produced at this step. The third step is the char gasification step that is enhanced by the Boudouard reaction. The important gasification mechanisms of the biomass particles are described by the following reactions:

$$biomass \rightarrow gas + tars + char \tag{1}$$

The combustion reactions:

$$C + \frac{1}{2}O_2 \rightarrow CO \qquad -111 \text{ MJ/kmol} \tag{2}$$

$$CO + \frac{1}{2}O_2 \rightarrow CO_2 \qquad -283 \text{ MJ/kmol} \tag{3}$$

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \qquad -242 \text{ MJ/kmol} \tag{4}$$

The Boudouard reaction:

$$C + CO_2 \leftrightarrows 2CO \qquad +172 \text{ MJ/kmol} \tag{5}$$

The water−gas reaction:

$$C + H_2O \leftrightarrows CO + H_2 \qquad +131 \text{ MJ/kmol} \tag{6}$$

The methanation reaction:

$$C + 2H_2 \leftrightarrows CH_4 \qquad -75 \text{ MJ/kmol} \qquad (7)$$
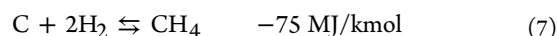
The water−gas shift (CO shift) reaction:

$$CO + H_2O \rightleftarrows CO_2 + H_2 \qquad -41 \text{ MJ/kmol} \qquad (8)$$
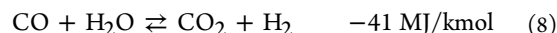
The gas producer quality (composition, production of CO, H_2, CO_2, and CH_4, and energy content) and the gasification performance (gas yield) depend upon feedstock origin, gasifier design, and operating parameters such as temperatures, static bed height, fluidizing velocity, equivalence ratio, gasifying agent, catalyst, and others, which are explained elsewhere.[7,9,11−17] Cao and co-workers[8] studied the air gasification process of wood sawdust using the lab scale model gasifier combined with two individual regions of pyrolysis, gasification, and combustion. They reported that the system carbon conversion and gasification efficiency reach above 87.1% and 56.9%, respectively, and reached a lower heating value (LHV) of 5000 kJ/Nm³. Pengmei and co-workers[9] investigated the air gasification of pine wood block in a down draft gasifier. They observed a fuel gas yield of 0.82−0.94 Nm³/kg biomass, a hydrogen percentage of 21.18−35.39 mol %, a LHV of 4.76−5.44 MJ/Nm³, and a hydrogen yield of 21.18−29.70 g/kg biomass. They proposed that higher temperature contributes to more hydrogen production. Midilli and co-workers[11] applied air-blown gasification to hydrogen production from hazelnut shell, and they observed that the moisture content of the fuel affects the amount of combustible gases and can cause an increase in the amount of hydrogen gas and other hydrocarbons components. In their study, they obtained a hydrogen yield of 2.4 kg/h from 100 kg of wet hazelnut shells. Zhao and co-workers[11] proposed a two-step process that started with biomass pyrolysis followed by secondary decomposition. In optimized conditions, they achieved 60% and 65 g/kg biomass, hydrogen content and hydrogen yield, respectively. Among all the designs, the fluidized bed gasifier has been shown to be a versatile technology capable of burning any waste combination with high efficiency. The significant advantages of the fluidized bed include compact furnaces, simple design, effective burning of a wide variety of feedstock, relatively uniform temperature, and ability to reduce emissions.

Despite extensive research on biomass gasification, less emphasis has been given to optimization of pyrolysis and gasification processes integration for optimal hydrogen production, which is the focus of this paper. In this work, producer gas quality and gasification performance were evaluated and optimum conditions for hydrogen production were determined.

2. EXPERIMENTAL SECTION

2.1. Materials. Palm kernel shell used in this study is obtained from a local factory. The sample is air-dried for 2−3 days to remove moisture and to ease crushing. The particle size of the feedstock was 0.2−0.3 mm; the proximate and ultimate analyses of the biomass are reported in Table 1. The heating media used in the fluidized bed is glass bead with a mean particle size of 0.2−0.8 mm, 1470 kg/m³ density, and approximately 69.3% SiO_2, 26.5% CaO, and 1.7% Fe_2O_3 with the balance being trace amounts of other oxides.

2.2. Facility and Experimental Setup. The schematic diagram of the experimental facility used in this study is shown in Figure 1. The test facility consists of three main parts: the gasifier, the gas conditioning section, and the gas sampling section. The reactor is made of stainless steel pipe and is externally heated by two electrical furnaces. The total height is 850 mm, with a bed diameter of 55 mm

Table 1. Proximate and Ultimate Analysis of Palm Kernel Shell by Elemental Analysis

	palm kernel shell
Proximate Analysis (wt %, wet basis)	
volatile matter	30.53
fixed carbon	48.5
ash	8.97
moisture	12
Ultimate Analysis (wt %, dry basis)	
hydrogen	5.52
carbon	51.63
oxygen	40.91
nitrogen	1.89
sulfur	0.05
cellulose	20.8
hemicellulose	22.7
lignin	50.7
apparent density (kg/m³)	733
HHV (MJ/kg)	24.97

and a freeboard diameter of 60 mm. Two K-type thermocouples (T1 and T2) were installed across the total height of the reactor for temperature detection. Below the reactor, one perforated air distributor with 3 mm thickness is installed for better air distribution. The biomass is fed into the reactor through a feeder that is connected at the top of the gasifier (overbed feeding system). Air is used as the fluidizing agent and comes from the air compressor. The air is preheated to 65 °C before entering the reactor. The produced gas flow exits the reactor and passes through a gas conditioning section. The cleaned gas is collected using gas sampling bag to be analyzed using gas chromatography (GC).

2.3. Experimental Procedures and Product Characterizations. Prior to each experiment, 0.20−1.21 kg/h of biomass mixed with 20 g of silica beads were put in the gasifier. The solenoid valve (SV) was turned on, and a preheated air flow passed through the bed and the reactor when the temperatures in the bed (pyrolysis zone) reached the desired temperature (500 °C). The feeder was turned on once the temperatures in these two parts stabilized. Typically, each test took about 20 to 25 min to stabilize, and measurements were taken at 2 min intervals. During each experiment, the air stream and the biomass feedstock were introduced from bottom and top of the gasifier, respectively. The cool, dry, clean gas was sampled using gas bags and was analyzed on a dry gas chromatograph (Model HP6890N, Agilent Technology, U.S.A.), which is fitted with a GS carbon plot column, flame ionization, detector (FID), and thermal conductivity (TCD), and standard gas mixtures were used for quantitative calibration. The tar sampling line is shown in Figure 1. Dichloromethane cooled to approximately −10 °C is used to condense and collect the tar. A gas chromatography−mass spectrometry instrument (Agilent, 7890A) is used to analyze the tar sample. The operating conditions are as follows: capillary column, 30 m × 0.25 mm × 0.25 μm; carrier gas, N_2; temperature program, 75 °C (hold 5 min) to 285 °C at 3 °C/min (hold (40 min); injector.

3. RESULTS AND DISCUSSION

A series of gasification experiments was conducted to determine the effect of the reaction temperature, equivalence ratio (ER), particle size, and feeding rate that governing the producer gas quality (composition, production) and the gasification performance (gas yield, energy content). For the temperature effect, 100 g of air-dried feedstock was continuously fed to the gasifier at feeding rate of 10 g/min. The gasifier temperature was examined in the range 750−1000 °C at 50 °C increments. For the ER effect, ER was varied from 0.23 to 0.27 by changing the air flow rate at a reaction
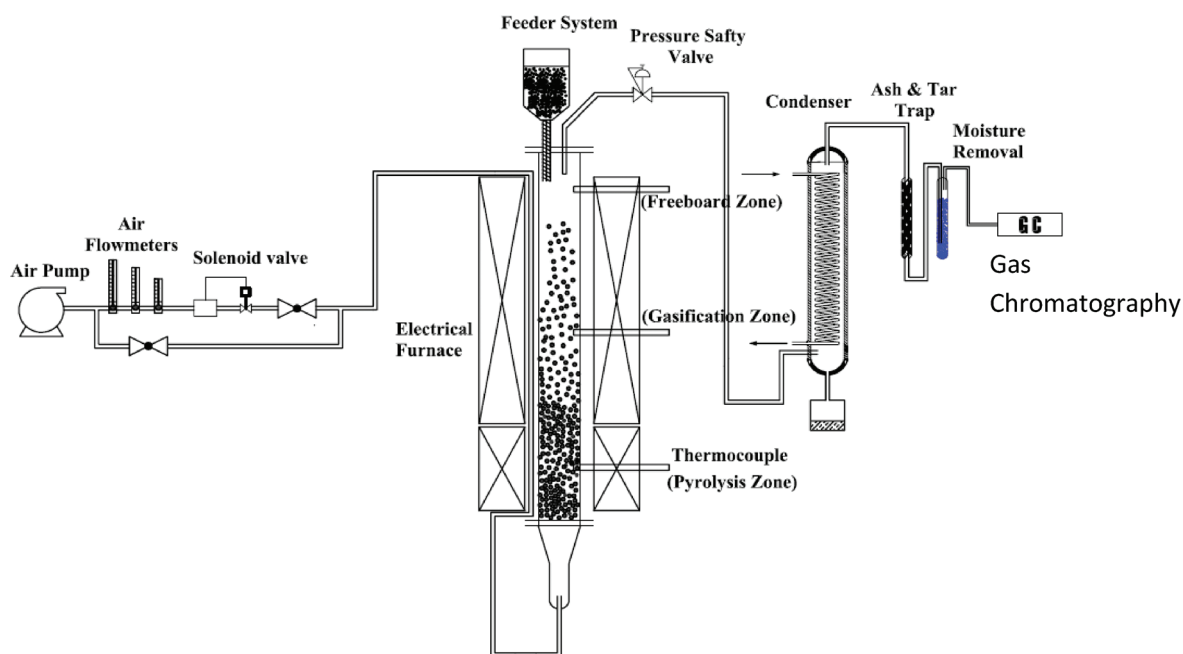
Figure 1. Schematic diagram of biomass air gasification in fluidized bed reactor.

Table 2. Summary of Results for the Application of Different Temperatures in Palm Kernel Shell Gasification

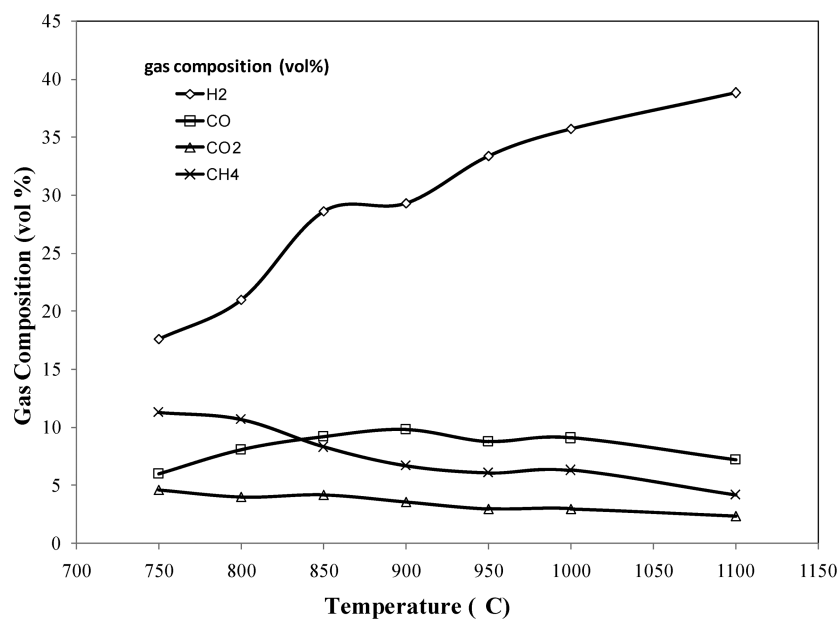| operating conditions							
air (Nm^3/h)	0.84						
feeding rate (kg/h)	0.78						
ER (kg/kg)	0.26						
reactor temp. (°C)	750	800	850	900	950	1000	1100
H_2 yield (g/kg biomass)	14.08	16.8	22.9	23.4	26.7	28.9	31.0
HHV (MJ/kg)	25.8	29.9	25.5	24.9	24.4	21.3	18.3
ash yield (g/kg biomass)	0.17	0.16	0.14	0.14	0.12	0.11	0.10
oil yield (g/kg biomass)	0.29	0.16	0.13	0.12	0.10	0.096	0.08

Figure 2. Effect of temperature on gas composition for palm kernel shell at ER (0.26 w/w).

temperature of 900 °C and a feeding rate of 0.75 kg/h. The effects of the feeding rate and the particle size were evaluated by performing the runs in the range 0.30−1.21 kg/h and with

three different particles sizes ranges, namely, 0.1 mm, 2 mm,

and 5 mm, respectively. Detailed calculations for determining the parameters are given by eqs 9−10.

$$ER = \frac{actual\ wt\ air\ (oxygen)/total\ wt\ feedstock}{stoichiometric\ air\ (oxygen)/total\ wt\ feedstock} \quad (9)$$

$$H_2\ yield = H_2\ obtained\ from\ exptl\ (wt)/biomass\ fed\ (wt) \quad (10)$$

3.1. Effect of Different Bed Reaction Temperatures.
The experimental results are presented in Table 2 and Figure 2. The results showed that H_2 yield exhibits an increasing trend with temperature, ranging from 14 to 31 g H_2/kg biomass. As expected, the increase in temperature led to higher gas yields and a reduction in the amounts of ash and tar formed, which could be due to further cracking of liquids and enhanced char reaction with the gasifying medium (air). This increase in gas yield with temperature could be due to various reasons, such as (i) increased production of gases in the initial pyrolysis step, which has a faster rate at higher temperatures,[18−20] (ii) the production of gas through the endothermal char gasification reactions, which are favorable at elevated temperatures, and (iii) the increase in the gas yield resulting from the stream reforming and cracking of heavier hydrocarbons and tars.

The effect of temperature on the gas composition was also studied and plotted in Figure 2. The rise in temperature was found to increase H_2 formation from 18 to 39 vol %, while the content of CH_4 decreased from 10.7 to 4.2 vol %. This is in accordance with Le Chatelier's principle: higher temperatures favor the reactants in exothermic reactions and favor the products in endothermic reactions. The H_2 formation is favored by the increasing of the gasifier temperature, which assumed to be due to the combination effect of exothermal character of the water−gas shift reaction (eq 8), which occurs and predominates between 500 and 600 °C, and the water−gas reaction (eq 6), which becomes significant at temperatures from 1000 to 1100 °C and upward.[10] The water shift reaction always occurrs in any gasification process as a result of the presence of water in fuel and water vapor in air. Water vapor and carbon dioxide promote hydrogen production in the biomass gasification process.[8] The content of CO first increases and decreases with temperature, which indicates that the reactions in eqs 5−8 happen simultaneously in the process, and no significant changes were detected in CO_2 formation, which was around 3−5%.

3.2. Effect of Equivalence Ratio (ER).
ER is defined as the ratio of the amount of oxygen (air) supplied and the amount of oxygen (air) needed for the stoichiometric combustion of the fuel. Table 3 shows that the H_2 yield increased first and then

Table 3. Summary of Results for the Application of Different Equivalence Ratios in Palm Kernel Shell Gasification

	operating conditions				
air (Nm³/h)	1.10				
feeding rate (kg/h)	0.75				
reactor temp. (°C)	900				
ER (kg/kg)	0.23	0.24	0.25	0.26	0.27
H_2 yield (g/kg biomass)	20.5	22.0	26.0	23.4	20.0

decreased as ER increased. A similar trend is observed for H_2 gas composition in Figure 3. As stated by Pengmei Lv and co-workers,[9] ER not only represents the oxygen quantity introduced to the reactor but also affects the gasification

temperature under the condition of autothermal operation.[12] On one hand, higher ER will cause higher gasification temperatures, which can accelerate the oxidation reaction and hence improve the product quality to a certain limit. On other hand, lower ER will cause less oxygen to be available to complete the gasification reactions, which is not favorable for the process. Therefore, the gas composition is affected by the two contradictory factors of ER (see Figure 3). Furthermore, Xiao and co-workers[5] suggested that this increase in CO and H_2 content can be explained as being due to the thermal cracking of hydrocarbons and tars (eqs 5−8) at a higher temperature. The decrease in CO and H_2 contents with further increase of ER due to the partial combustion of different gaseous components resulted a large increase in CO_2 concentration.

3.3. Effect of Feeding Rate.
Various feeding rates ranging from 0.20 to 1.21 kg/h were tested for palm kernel shell gasification to investigate the effect on hydrogen yield. At the condition of air gasification, keeping ER at a constant value of 0.26, the biomass feeding rate is changed from 0.20 to 1.21 kg/h to conduct the tests. From Table 4 and Figure 4, it can be found that, at lower feeding rates, the temperature increases with feeding rate, whereas the hydrogen yield shows an opposite trend whereby the hydrogen content increased and reached to the maximum value of 29.1%. This is explained by the fact that the higher feeding rate attributed to less residence time per volume of air, which caused less oxygen to be in contact with the biomass particles.[18] Thus, the decrease in temperature at pyrolysis and, consequently, a decrease in the gasification process occurred, and hence, the biomass samples remained raw or partially gasified. This proves that excessively high feeding rates are unbeneficial for biomass gasification cracking and reforming reactions, leading to a reduction of hydrogen content in the gases.

3.4. Effect of Biomass Particle Size.
Table 5 and Figure 5 summarize the hydrogen production performance for palm kernel shell at difference particle sizes (0.1, 2, and 5 mm). It was observed that, with decreasing the particles size, the produced hydrogen and hydrogen yield decreased, with the maximum value of 22.2% belonging to the smallest particle size. Lv and co-workers[19] reported that the pyrolysis process of small particles is mainly controlled by reaction kinetics. Thus, as the size of biomass particles increases, the production gas resultant inside the particles is more difficult to diffuse out and the process is mainly controlled by gas diffusion. On other hand, larger particles are not only difficult to be entrained by fluidizing gas but also produce fewer smaller particles after gasification reaction. This results in a reduction in fine particle entrainment and hence decreases the amount of volatile matter and unburned char.[20]

3.5. Bio-Oil Characterization.
The oil ratio obtained at different gasification temperatures ranged from 0.10 to 0.29%. This bio-oil has the potential to substitute for conventional fossil liquid fuels[13,14] and to be upgraded to fine chemicals, which can be regularly used in the pharmaceutical industries. Table 6 lists components found in these products at selected gasification temperatures. The major components that were common and recognized by GC-MS were acetic acid, propanoic acid, phenol, 4-methylphenol, 2-methylphenol, 2,2-oxybisethanol, and acetamide. However, as the temperature increases, the percentage of the components also changes, depending on types of feedstock studied. For example, acetic acid and phenol had higher percentages, compared with the
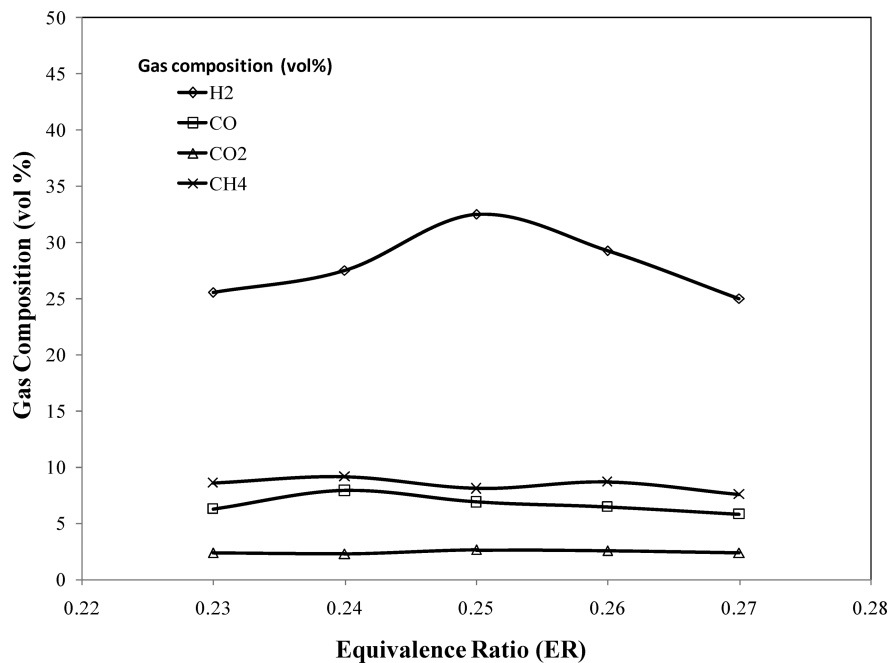
Figure 3. Effect of equivalence ratio (ER) on gas composition for palm kernel shell gasification at 900 °C.

Table 4. Summary of Results for the Application of Different Feeding Rates in Palm Kernel Shell Gasification

operating conditions					
air (Nm³/h)	0.54				
ER (kg/kg)	0.26				
reactor temp. (°C)	900	870	840	800	798
feeding rate (kg/h)	0.30	0.60	0.70	0.90	1.2
H₂ yield (g/kg biomass)	14.0	16.3	22.0	19.0	14.6
ash yield (g/kg biomass)	0.07	0.11	0.14	0.16	0.22
tar yield (g/kg biomass)	0.05	0.10	0.11	0.12	0.15

Table 5. Summary of Results for the Application of Different Particle Sizes in Palm Kernel Shell Gasification

operating conditions		
air (Nm³/h)	0.84	
ER (kg/kg)	0.26	
reactor temp. (°C)	900	
particle size (mm)	bulk (>4)	granule (2−4)	powder (0.1−0.3)
H₂ yield (g/kg biomass)	8.16	13	17.8

results of researchers that worked at a low pyrolysis temperatures.[14−17] Thus, it is clear that the presence of oxygen

in the high temperature gasifier had a positive effect on phenol and phenolic compound formation. On the other hand,
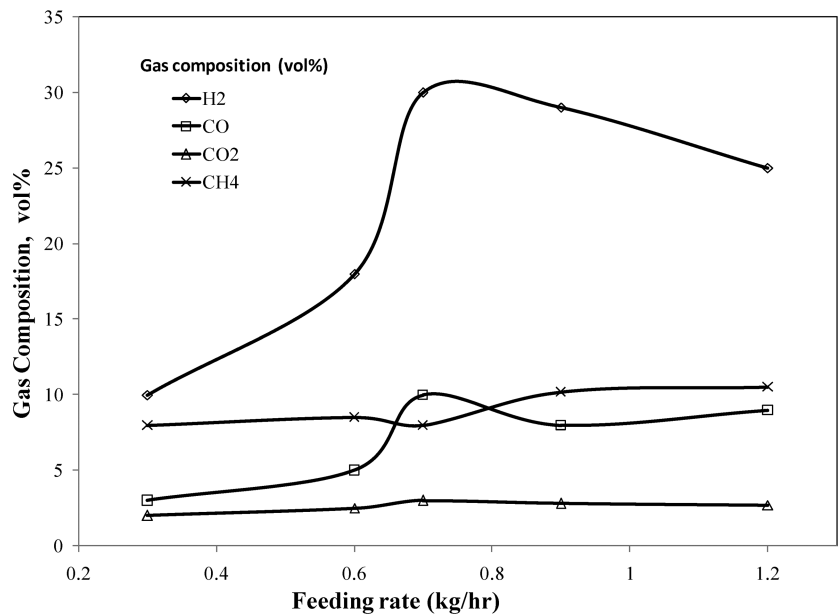

Figure 4. Effect of feeding rate on gas composition at optimized condition of palm kernel shell gasification at constant ER (0.26).
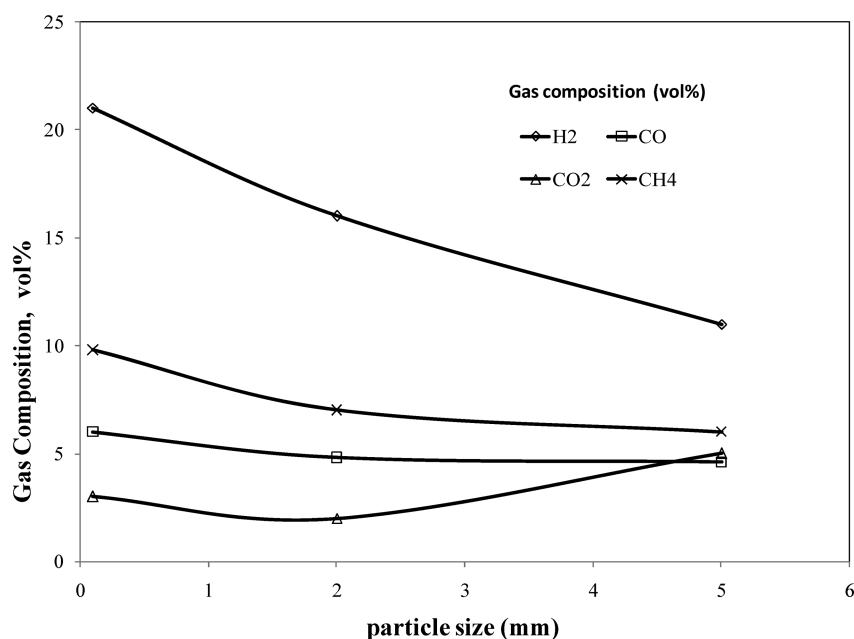
Figure 5. Effect of particle size on gas composition for palm kernel shell gasification at constant ER (0.26) and gasification temperature (900 °C).

Table 6. Properties of Bio-Oils Obtained at Different Temperatures in Palm Kernel Shell Gasification

no.	name	formula	750 °C	800 °C	850 °C	900 °C	950 °C	1000 °C
1	acetic acid	$C_2H_4O_2$	18.53	22.31	25.78	26.63	25.71	21.08
2	propanoic acid	$C_3H_6O_2$	0.98	1.97	1.95	1.18	1.15	1
3	3-methylbutanoic acid	$C_4H_8O_2$		1.39		1.26		
4	acetamide	C_2H_5NO	1.43	4.53	10.28	7.44	4.40	8.15
5	propanamide	C_3H_7NO	0.25	0.77	1.02	0.82	0.52	0.85
6	phenol	C_6H_6O	47.72	47.75	43.24	49.33	52.33	54.57
7	4-methylphenol	C_7H_8O	4.49	4.37	1.52	1.35	1.37	1.07
8	2-methylphenol	C_7H_8O	3.32	3.21	1.6	1.53		0.57
9	2,2-oxybisethanol	$C_4H_{10}O_3$	1.04	0.42	1.96	3.30	2.71	4.86
10	3-furan methanol	$C_5H_6O_2$	1.67	1.27	1.77	0.4	0.8	1.09
11	benzonitrile	C_7H_5N				1.21	2.45	1.30
12	butyrolacone	$C_4H_6O_2$	1.15					1.50
13	1-methylimidazolidin-2-one	$C_4H_8N_2O$	1.2					
14	5-methyl-2,4-Imidazolidinedion,	$C_4H_6N_2O_2$		1.12				
15	1,3-diazine	$C_4H_4N_2$			1.8			
16	4-ethyl-2,5-dimethyl isoxazolidine,	$C_7H_{15}NO$			1.63			
17	isoquinoline	C_9H_7N		0.36	0.6	0.61	1.45	1
18	butyrolacone	$C_4H_6O_2$						
19	formic acid	CH_2O_2			0.74	1		0.64

increasing the temperature caused a decrease in acetic compounds and caused an increase in the phenol and phenolic compounds. This generally is in agreement with the results reported by Butt.[14]

4. CONCLUSION

Air gasification of agricultural wastes was successfully performed in a lab scale fluidized bed gasifier, producing producer gas with low tar content. Among the gasification parameters tested, the equivalence ratio appeared to have the most pronounced effect on the reactor temperature, product yield distribution, and the gas composition, particularly hydrogen. The influence of the equivalence ratio on the performance of gasifier could be regarded as the effect of reactor temperature because the reactor temperature was found to be ER dependent. The feeding rate and biomass particle size

would only show minor effects during the gasification process. The overall bio-oil obtained from gasification had stability in percentage and component in all temperatures applied. The obtained results led to the conclusion that agricultural wastes are a potential candidate for hydrogen production as an alternative renewable energy source.

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