

An experimental investigation of Hydrogen production from Malaysia agricultural waste in a fluidized bed gasifier

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Abstract

Recently, hydrogen production from biomass is an attractive technology for power generation. The main objective pursued in this work is to investigate the hydrogen production potential from agricultural waste (coconut coir and palm kernel shell) by applying fluidized bed gasification technique. An experimental study was conducted using a bench-scale fluidized bed gasifier with 60 mm diameter and 425 mm height. During the experiments, the fuel properties and the effects of operating parameters such as gasification temperatures (800 to 900°C), fluidization ratio (2 to 3.33 m/s), static bed height (10 to 30 mm) and equivalence ratio (0.16 to 0.46) were analyzed and discussed. It was concluded that substantial amount of hydrogen gas (up to 67 mol%) could be produced utilizing agricultural residues as coconut and palm kernel shell by applying fluidization technique. For both samples, the rise of temperature till 900°C favored hydrocarbon further reactions and allowed an increase of almost 67 mol% in hydrogen release. However, other parameters such as fluidizing velocity and feed load showed only minor effects on hydrogen yield. As conclusion, agricultural waste can be assumed as an alternative renewable energy source to the fossil fuels, and the environmental pollution originating from the disposal of agricultural residues can be partially reduced.

Keywords: Hydrogen, agricultural waste, fluidized bed, palm kernel shell and coconut shell

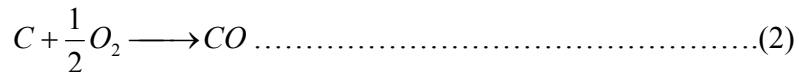
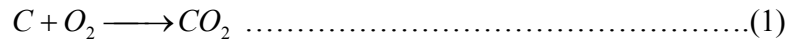
1. Introduction

Agricultural residues is potentially an attractive feedstock for producing energy as it use contributes little or no net carbon dioxide to the atmosphere. Agricultural production in Malaysia continued to record positive growth from 2000 to 2005 and expected to expand more in 2010 consistent to the government policies [1-3]. In Malaysia, the major agricultural products are palm oil, sawlogs, paddy and tropical fruits which more than 70 million tones collected annually. There are significant amount of wastes and residues also called as agricultural wastes produced from the post-processing of these products. In the past, agricultural wastes have been utilized as animal feed or organic fertilizers but are otherwise allowed to decay naturally in the fields, discarded or burnt which caused secondary pollution problems. Novel disposal technologies are in high demand by the industry and regulators to provide more energy efficient and environmentally and economically sound solutions.

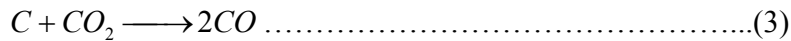
Thermochemical gasification of biomass is a well-known technology that seems to have a feasible application and has been developed attractively for industrial applications [4-7]. Air gasification process can convert these solid agricultural residues into a synthesis gas that is suitable for use in electricity production or for the manufacture of chemicals, hydrogen or transportation fuels. The producer gas from biomass gasification can be directly used as fuel in the internal combustion engines or gas turbines [5]. Alternatively, producer gas can also be used in fuel cells and as second generation of biofuel (such as BTL). Air gasification has advantage over other gasifying agent is to simplify the gasification process and reduce operating and maintenance cost because air separation to obtain oxygen is 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 dilute the fuel gas and leads to a low calorific value fuel. [6]. Although the feasibility of the air gasification process has been demonstrated by different feedstock, few studies have been conducted for pruning waste, plastic and other types of biomass utilization [5-12].

The transformation of biomass into hydrogen-rich gas is suitable source for energy production due to the advantages typical biomass (renewable character, net CO₂ emissions below zero, etc). The production of hydrogen from biomass was described as listed reactions below [10]:

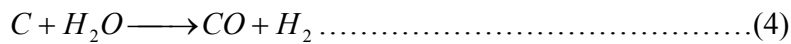
Oxidation reaction:



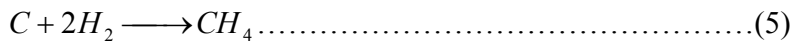
Boudouard



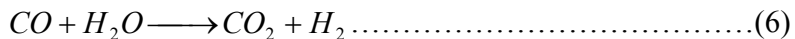
Water gas:



Methanation:



Water gas shift:



The gas producer quality (composition, production of CO, H₂, CO₂ and CH₄ and energy content) and the gasification performance (gas yield) depending upon feedstock origin, gasifier design and operating parameters such as temperatures, static bed height, fluidizing velocity, equivalence ratio, gasifying agent, catalyst and others which explain elsewhere [7, 9, 11 - 18]. Ragner Warnacke, 2000 demonstrated that the gas composition was a function of gasifier design where the same fuel may give different calorific value with different gasifier [13]. Among all designs, fluidized bed gasifier has been shown to be a versatile technology capable of burning practically any waste combination with high efficiency. The significant advantages of fluidized bed combustors over conventional combustors include their compact furnace, simple design, effective burning of a wide variety of fuels, relatively uniform temperature, and the ability to reduce emissions such as nitrogen oxide and sulphur dioxide gas [19].

In this work, the producer gas quality and the gasification performance (gas yield) obtained from the gasification of two different agricultural wastes (coconut shell and palm kernel shell)

in a atmospheric laboratory scale fluidized bed gasifier was investigate. The effect of gasification temperatures, fluidization ratio, static bed height and equivalence ratio(ER) on gas composition, gas yield and gas heating value were studied.

2. Material and methods

2.1 Material

The agricultural waste (palm kernel and coconut shell) from local palm mill and coconut processing were selected for this study. The samples were air dried for 2 to 3 days to remove moisture and to ease crushing. The proximate and ultimate analyses of the feed stock are reported in table 1.

Table 1

Fuel properties

	Palm kernel shell	Coconut shell
Moisture content (% wet basis)	7.96	4.89
Proximate analysis (% db)		
Volatile matter	72.47	30.62
Fixed carbon	18.56	26.40
Ash	8.97	42.98
Ultimate analysis (t% db)		
Carbon	51.67	45.29
Hydrogen	5.52	5.04
Nitrogen	1.89	1.46
Sulfur	<0.01	<0.01
Oxygen (by different)	40.91	48.2
Higher Heating Values (HHV) (MJ/kg)	22.97	16.07

2.2 Facility

The schematic diagram of the experimental facility used in this study is shown in Fig. 1. The gasifier was specially designed for operating under atmospheric conditions. The test facility consists of three main parts: the gasifier, the gas conditioning section and the gas sampling section. The fluidized bed gasifier, having an internal diameter of 60 mm and height of 425 mm, was made of heat resistant stainless steel and surrounded by two individually controlled electric heaters that supply heat for startup and counter heat loss during operation. Two K-type thermocouples (T1 and T2) were installed across the reactor: the probe T1 in the bed zone, defined as the gasification reaction zone (at the height of 10 mm above the distributor) and one probe in the freeboard (at the height of 300 mm above the distributor). Prior to gasification tests, all the fuel samples were grinded to less than 0.3 mm in order to ensure the homogeneity on the biomass composition. Air from compressor was used as the oxidant and fluidizing reagent in all gasification tests. The air flow rate was measured through a volumetric flow controller placed after the air regulation valve, while feeding rate was estimated as the weight difference of the fuel hopper prior and after the tests.

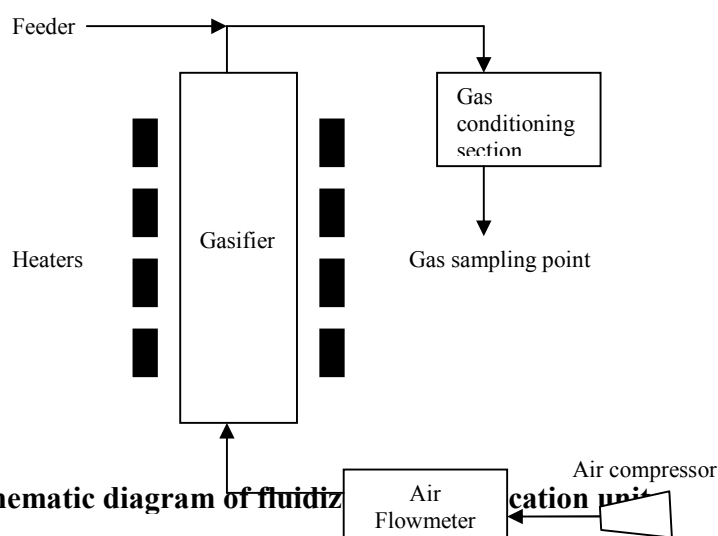


Fig. 1 – Schematic diagram of fluidized bed gasification unit

2.3 Experimental procedure and analytical methods

The experimental schedule has been performed in order to analyze the individual effect of the main parameters governing the producer gas quality (composition, production) and the gasification performance (gas yield, energy content), such as: a) varying the temperature (700 to 900°C); b) varying the fluidizing ratio (2.0 to 3.3 ms⁻¹); c) varying the average static bed height (10 to 30 mm) and d) varying the air to fuel ratio or equivalence ration (ER) (0.16 to 0.46).

At the start-up of each experimental run, the agricultural waste (coconut and palm kernel shell) was added to the hopper; the gasifier and furnace heaters for air gasification and the controllers were set at the selected operating temperatures. At the beginning of the experiment, the reactor was charged with 24g silica sand as bed material, which helped in stable fluidization and better heat transfer. After the bed temperature reached the desired level and remained steady, the air compressor was turned on to force the air through the pre-heater and air distributor into the reactor. When the bed temperature became steady, the samples were fed and the test began. Three samples were taken at an interval 3 min after the test ran in a stable state. Normally each experiment was repeated two times and the results had a good agreement.

A sample stream for permanent gas analysis was extracted from the process directly after the main filter housing. The gas stream was passes through a water cooled heat exchanger followed by an condenser trap and a coalescing filter. Grab samples of cool, clean and dry gas were collected and analyzed off-line for the permanent gases H₂, CO, CH₄ and CO₂ using a gas chromatography (GC)(Autosystem, Perkin Elmer,) equipped with a thermal conductivity detector (TCD). A GC calibration was performed using a certified standard gas prior each test.

3. Results and discussion

Average properties of the fuel used for the tests are shown in Table 1. The composition of the fuel is quite similar to the typical fuel used in the theoretical biomass types. Fuel moisture contents for palm kernel shell and coconut shell are 5 % and 8 %, respectively. Fuel higher heating value was determined to be 22 and 16 MJkg⁻¹, respectively.

Effect of reactor temperature

Results The test results are summarized in Table 2 and Fig. 2. Reactor temperature was increase from 700 to 900 °C in 50°C increments holding all other conditions constant. From Fig. 2, it can be seen that Hydrogen (H₂) concentration significantly increased from 43-67% for cocnut shell and 56-66% for palm kernel shell, based on dry inert-gas with temperature and the content of CH₄ showed an opposite trend. This is in accordance with Le Chatelier's principle; higher temperatures favor the reactants in exothermic reactions and favor the products in endothermic reactions. Therefore, the endothermic reactions of methane reforming reaction (Eqn 5) were verified this theory. The content of CO was mainly determined by Bourdard reaction (Eqn. 3) and it is an exothermic reaction. Higher temperature was not favorable for CO production, thus the content of CO decreased with temperature. However, in the present work the increase found in CO concentration at higher temperature obviously in the case of coconut shell. This may due on the one hand, to a higher partial gasification of carbon and on the other hand, to the endothermic character of Bourdouard equilibrium (Eqn. 3) which does also justify the decrease in CO₂. At a given pressure the equilibrium gas composition for this reaction is only a function of the temperature employed [8]. Table 2 showed that gas yield increased from 0.91 to 2.95 kg/kg biomass and 2.36-5.90 kg/kg biomass for coconut shell and palm kernel shell, respectively in the studied temperature range. The product gas Low heating value (LHV) showed a maximum value, 1174 KJ/Nm³ and 3711 KJ/Nm³, both at the temperature 850°C.

Table 2

Experimental results of different reactor temperature

Reactor temperature (°C)	700	750	800	850	900
Gas yield (mol/kg biomass)					
a) coconut shell	0.91	1.81	1.42	2.12	2.95
b) palm kernel shell	2.36	3.46	4.37	5.25	5.90
Gas LHV (kJ/Nm ³)					
a) coconut shell	930	1033	1174	1874	2384
b) palm kernel shell	2783	3276	3711	4451	3720

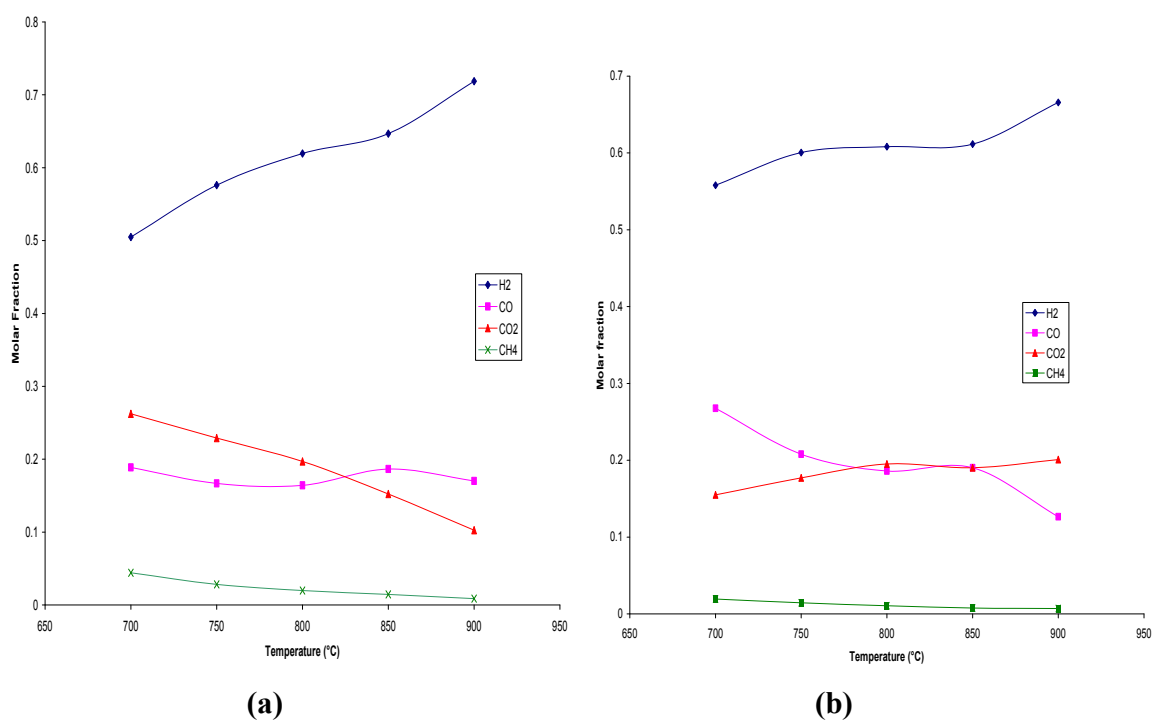


Fig. 2. Effect of temperature on gas composition a) coconut shell and b) PKS

Effect of fluidization ratio (U/U_{mf})

Fig. 3 presents the gas producers composition as a function of the fluidization ratio with fixed static bed height, temperature and equivalence ratio (ER). Increasing fluidization ratio from 2.0 to 3.3 m/s decreased the combustible gas components CO from 0.17 to 0.15 mol % and 0.32 to 0.13 mol% for coconut shell and palm kernel shell, respectively. However, different trend was observed for H₂ where the fraction decrease about 17% for coconut shell but increase up to 26% for palm kernel shell with increase fluidization ratio. This can be explained by the fact that palm kernel shell has higher particle density as compared with coconut shell. Thus, the higher fluidization ratio provides better mixing conditions for gasification of palm kernel shell. However, the CH₄ content was slightly increased. The content of CO₂ increased about 43% and 62% for coconut shell and palm kernel shell, respectively with the increase in fluidization velocity. The variations in the gas composition with fluidization velocity caused a decrease of the gas LHV (see table 3). R. Xiao et al, 2007[7] found the same trend, which can be attributed to rapid exothermic reactions and hence a higher CO₂ content at the expense of most combustible fractions. As fluidizing velocity increased, more carbon can be converted through reaction 1 and 6.

Table 3

Experimental results of different fluidizing velocity

Fluidization ratio (U/U_{mf})	2.00	2.70	3.33
Gas yield (mol/kg biomass)			
a) coconut shell	0.60	0.32	0.26
b) palm kernel shell	3.15	3.57	4.42
Gas LHV (kJ/Nm ³)			
a) coconut shell	484	292	279
b) palm kernel shell	4607	1756	1961

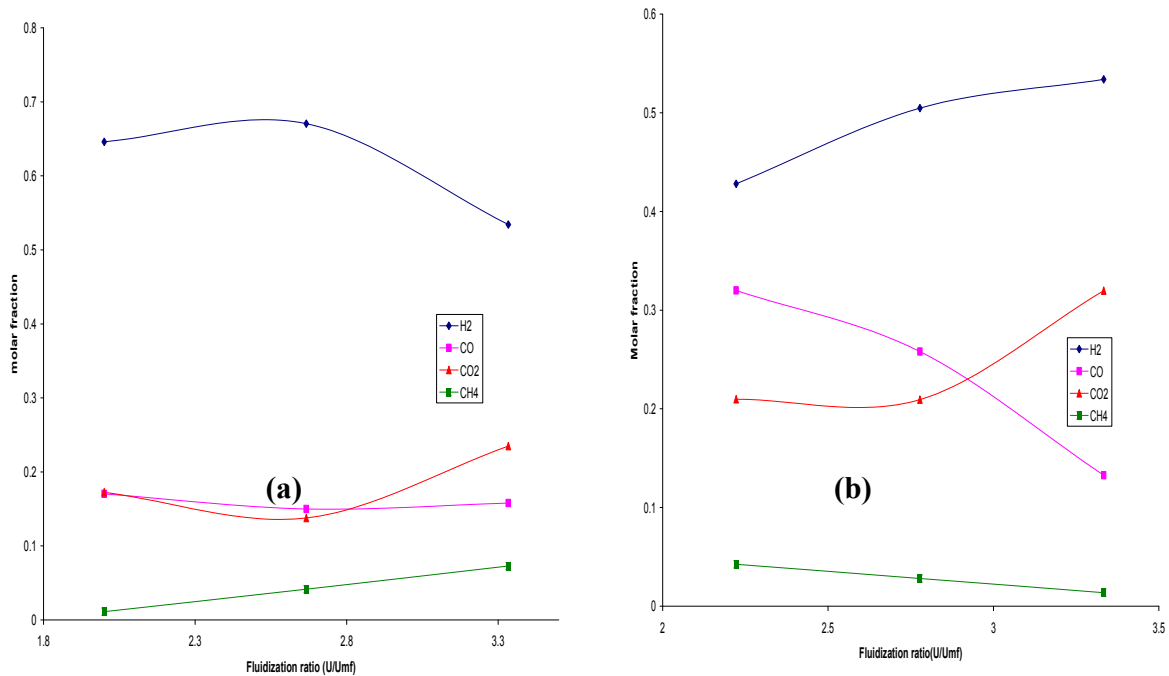


Fig. 3. Effect of fluidizing velocity on gas composition a) coconut shell and b) PKS

Effect of bed height

Table 4 and Figure 4 show the influence of static bed height on the results. For a fixed fluidizing velocity, increasing bed height would prolong the products residence time in the high temperature dense bed. This will favor secondary cracking reactions of tars and heavy hydrocarbons, and char gasification reactions, which will caused an increase in the gas yield [7]. As shown in Fig. 4, the H_2 and CO content increase from 0.68 to 0.70 mol gas/mol biomass and 0.06 to 0.17 mol gas/mol biomass for coconut shell but fro palm kernel shell H_2 decrease from 0.70 to 0.47 mol gas/mol biomass and CO increase from 0.19 to 0.25 mol gas/mol biomass. While CH_4 content slightly decreased from 0.04 to 0.011 mol gas/mol biomass for coconut shell but an increase CH_4 from 0.023 to 0.042 mol gas/mol biomass for palm kernel shell. However CO_2 content significantly increased in both cases with average 11 % and 38% for coconut shell and palm kernel shell, respectively. The gas yield was

decreased when the static bed height was increased to 30 mm. This indicated that there might an optimum bed height for a particular ER, at which the gas yield reached a maximum. This can be explained by the fact that a significant change of the hydrodynamic conditions due to the formation of larger bubbles in the bed height.

Table 4

Experimental results of different bed height

Bed Height (mm)	10	15	20	25	30
Gas yield (mol/kg biomass)					
a) coconut shell	0.49	0.54	1.21	1.38	1.19
b) palm kernel shell	3.43	3.41	3.57	3.95	3.97
Gas LHV (kJ/m ³)					
a) coconut shell	323	369	995	1204	1067
b) palm kernel shell	2877	3758	3427	5578	4887

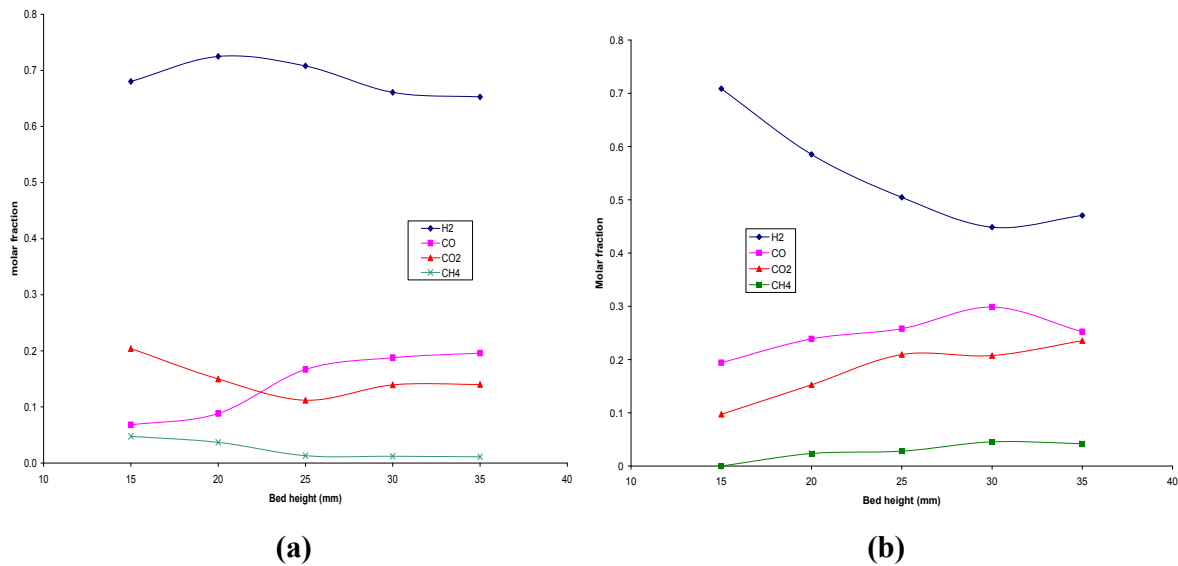


Fig. 5. Effect of static bed height on gas composition a) coconut shell and b) PKS
Effect of equivalence ratio (ER)

Equivalence ratio (ER) was varied from 0.16 to 0.46 through changing the mass flow rate and holding the other conditions constant. The tests results of varying ER were reported in Table 5 and Fig. 5 for all agricultural waste tested. As observed in these figure, the ER is the main parameter affecting the producer gas quality. Fig. 6 indicates that H₂ and CO content increased first and decreased as ER increased. As stated by P.M Lv et al, 2004 [9], ER not only represents the oxygen quantity introduced to the reactor but also affects the gasification temperature under the condition of auto thermal operation. On one side, higher ER will cause gas quality to other side, higher ER means higher gasification temperature, which can accelerate the gasification and improve the product quality to a certain limit. Therefore the gas composition is affected by the two contradictory factors of ER. Also, R. Xiao et al, 2007 [7] suggested that this increase in CO and H₂ content can be explained as due to thermal cracking of hydrocarbons and tars (reaction 2, 3, 4 and 6) at a higher temperature. While the decrease in CO and H₂ content with further increase ER due to partial combustion of different gaseous components which resulted a large increase in CO₂ concentration. Also, it was noted that a significant decrease in LHV from 117 and 2286 to 85 and 1482 kJ/m³ for coconut shell and palm kernel shell, respectively with increasing ER due to consumption of hydrocarbons by combustion. The results obtained agree fairly well with previous studies, no matter what was the nature of feed stock gasified [6-10].

Table 5

Experimental results of different equivalence ratio

Bed Height (mm)	0.15	0.20	0.25	0.30	0.45
Gas yield (mol/kg biomass)					
a) coconut shell	0.31	0.41	1.08	0.64	0.25
b) palm kernel shell	2.7	4.02	2.81	1.86	1.08
Gas LHV (kJ/m ³)					
a) coconut shell	117	190	473	261	85
b) palm kernel shell	2286	2863	3467	1812	1482

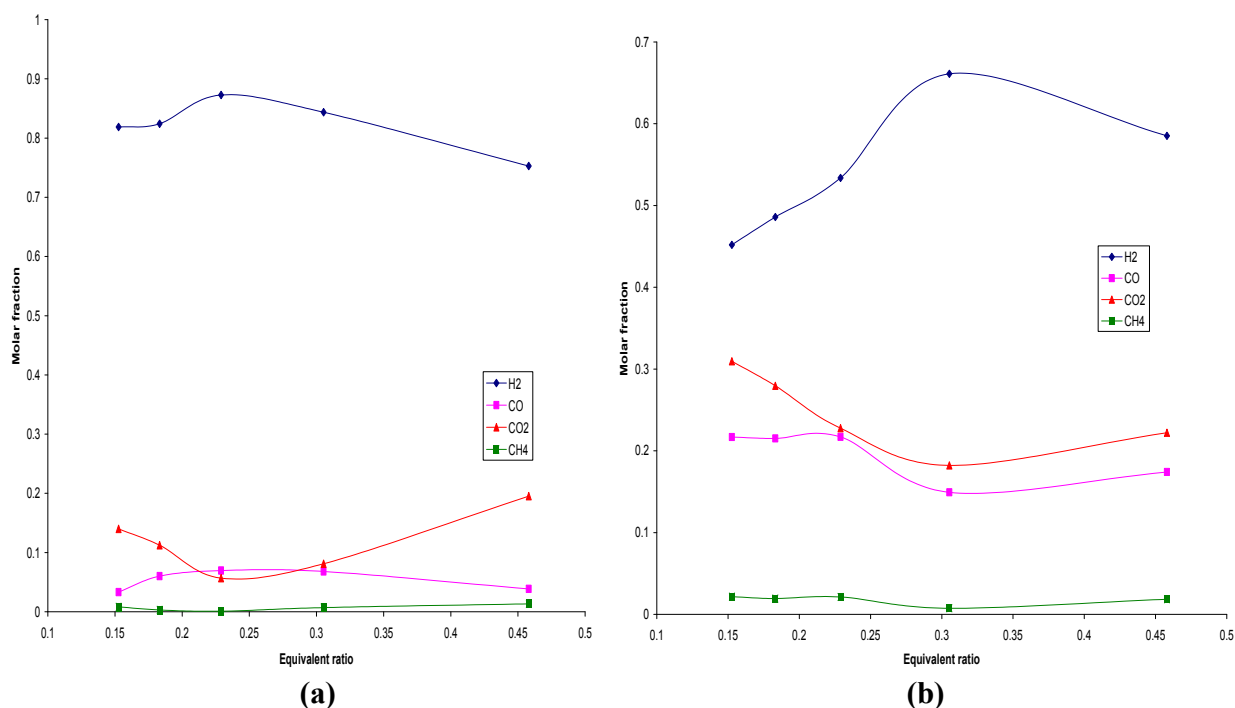


Fig. 5. Effect of equivalence ratio on gas composition a) coconut shell and b) PKS

4. Conclusion

In general, results show not very significant differences between the different agricultural wastes tested, which indicates that these wastes could be used in flexible fuel systems. However, producer gas quality and performance from palm kernel shell seems to be slightly more efficient than coconut shell. Parametric tests, varying the temperature, fluidization ratio, static bed height and equivalence ratio, have been performed to determine their effects on product gas composition, gas yield and gas LHV. Over the range of operating conditions tested, gas yield varied from 0.49 to 2.9 and 1.06 to 5.12; gas LHV ranged between 85 and 2384 kJ/Nm³ and 1482 and 5578 kJ/Nm³ for coconut shell and palm kernel shell, respectively.

Among the gasification parameters tested, temperature appeared to have the most pronounced effect on the gas yield, the gas LHV and the gas composition. Higher temperature favored hydrogen production and gas yield but did not always favor gas heating value. Too high a temperature lowered gas heating value. Then followed by equivalence ratio (ER) had complex effects on tests results and there existed an optimal value for this factor, which was different according to different operating parameters. The fluidizing velocity and static bed height would only show minor effect during the gasification process. The fluidization velocity was observed to have an influence on the gasification process to some extent because it will result in the carryover of fine chars from reactor. The bed height would affect the residence time of gases in the high temperature dense bed. Hence, the rise of the bed height favored the tars and hydrocarbon cracking reactions. However, too high bed height showed a negative effect due to large bubbles formation.

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