

Simulation of Enhanced Biomass Gasification for Hydrogen Production using iCON

Mohd K. Yunus, Murni M. Ahmad, Abrar Inayat, and Suzana Yusup

Abstract—Due to the environmental and price issues of current energy crisis, scientists and technologists around the globe are intensively searching for new environmentally less-impact form of clean energy that will reduce the high dependency on fossil fuel. Particularly hydrogen can be produced from biomass via thermo-chemical processes including pyrolysis and gasification due to the economic advantage and can be further enhanced through in-situ carbon dioxide removal using calcium oxide. This work focuses on the synthesis and development of the flowsheet for the enhanced biomass gasification process in PETRONAS's iCON process simulation software. This hydrogen prediction model is conducted at operating temperature between 600 to 1000°C at atmospheric pressure. Effects of temperature, steam-to-biomass ratio and adsorbent-to-biomass ratio were studied and 0.85 mol fraction of hydrogen is predicted in the product gas. Comparisons of the results are also made with experimental data from literature. The preliminary economic potential of developed system is RM 12.57 x 10⁶ which equivalent to USD 3.77 x 10⁶ annually shows economic viability of this process.

Keywords—Biomass, Gasification, Hydrogen, iCON.

I. INTRODUCTION

INCREASING current demand of energy and depleting fossil fuel reserves has driven nations around the globe to extensively search for new sustainable energy sources. One of the main focuses is to utilize green energy such as biomass due to its large potential as renewable energy source which can covers 14% of total energy demand [1]. Biomass including forestry waste, wood-based materials and agricultural residue is considered CO₂ neutral which net CO₂ intake during photosynthesis by plants and CO₂ release during its natural decomposition or via utilization of biomass is zero.

Biomass can be converted to useful products including bio-synthesis gas [2] and hydrogen [3-5]. With respect to hydrogen production, gasification is more economically attractive as compared to pyrolysis [6]. As a promising technology of biomass thermo-chemical gasification process, gaseous products can be obtained including hydrogen, carbon monoxide, carbon dioxide, methane and higher hydrocarbons as well as tar and char. Hydrogen specifically is one of the

attractive energy carriers that can be utilized as an alternative fuel, which can be a major market drive to replace fossil fuel in automotive industries. Moreover power generation sector and chemical industries also demand hydrogen as raw material to generate energy and chemical value-added products.

In gasification process of biomass, various gasification agents such as air, pure oxygen and pure steam were utilized by previous researchers. However, steam gasification shows superior result in term of producing medium calorific value gases [7]. Dupont et al. [8] conducted modeling of biomass steam gasification of Sylverster pine and spruce using kinetic data from Chemkin software to estimate the total gas yield and composition. They reported that hydrogen in product gas is estimated at 42 mol% (dry basis) of methane steam reforming (methane is assumed present in volatile compound in the raw material) and water gas shift at atmospheric pressure. Limitation of this work is that there is no consideration of removing unwanted product gas such as carbon dioxide from product gas flow.

On the other hand, Nikoo et al. [9] reported simulation of biomass gasification in fluidized bed gasifier using ASPEN PLUS software. They reported that the product gas increased as the temperature increased, and maximum hydrogen obtained from his simulation is at 45 mol% of product gas composition. They studied other effects of the system such as steam-to-biomass ratio and biomass particle sizes at atmospheric pressure system. Reported optimum operating condition of biomass gasification is at steam-to-biomass ratio of 2.5 and the particle size of biomass of 0.25-0.75 mm.

Mahishi et al. [10] simulated equilibrium model of biomass gasification system using Stanjan (v 3.93L) software to predict product gas composition. They studied the thermodynamics efficiency of the atmospheric gasification system which optimum at temperature of 1000K and steam-to-biomass ratio of 3. 60 vol% of hydrogen was produced from the woody biomass.

In term of economic analysis of biomass gasification, Lv et al. [11] reported that hydrogen supplied at the cost of USD 2.34 x 10⁵ annually of 6.4 ton per day of biomass gasification plant. They analyzed the biomass residue as raw material in downdraft gasifier producing hydrogen at 56.3 vol% using steam and air as gasifying agents.

This work focuses to investigate the technical and economic feasibility for hydrogen production via biomass gasification with in-situ carbon dioxide removal using modeling and simulation approaches. The objectives of this work are to screen process routes of hydrogen production from biomass gasification in steam-assisted gasifier with in-situ carbon

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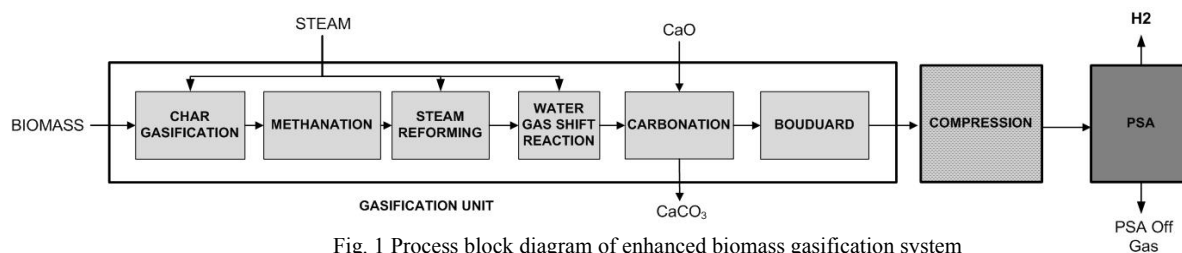


Fig. 1 Process block diagram of enhanced biomass gasification system

dioxide removal using calcium oxide, to synthesize and develop process flowsheet of hydrogen production from biomass via steam gasification, to perform simulation of developed flowsheet using PETRONAS process simulation software which is iCON [12], and to determine the optimum operating parameters and economic viability of the system.

II. METHODOLOGY

A. Process Screening and Reaction Scheme

Careful consideration on reaction scheme has been done according to related literature such Mann et al. [13] and Florin et al. [14]. Since Mann only consider biomass gasification using steam without consideration of carbon dioxide removal in his study, the work done by Florin is added into this study to incorporate the in-situ removal of carbon dioxide using calcium oxide. The reactions considered in this process are carbon steam gasification (R1), methanation (R2), methane steam reforming (R3), water gas shift (R4), carbonation (R5) and bouduard (R6).

TABLE I
REACTIONS OF ENHANCED BIOMASS STEAM GASIFICATION

Reaction	Reaction Scheme	ΔH (kJ/mol)
R1	$C + H_2O \rightarrow H_2 + CO$ where $r=k C_c$	118.9
R2	$C + 2H_2 \rightarrow CH_4$ where $r=k C_c$	-74.8
R3	$CH_4 + H_2O \rightarrow CO + 3 H_2$ where $r=k C_{CH_4}$	222.35
R4	$CO + H_2O \leftrightarrow H_2 + CO_2$ where $r=k C_{CO}C_{H_2O}$	-42
R5	$CaO + CO_2 \rightarrow CaCO_3$ where $r=k C_{CO_2}$	-170.5
R6	$C + 2CO_2 \rightarrow 2CO$ where $r=k C_c$	172

Reactions listed above assumed to occur simultaneously, but the reaction is simulated to happen in sequence which from R1 to R6 sequentially.

B. Reaction Kinetics

The kinetics parameters for the reactions assumed in the gasification process are listed in Table II. First order kinetic model of reactions have been considered for simplicity and availability in literature. Kinetic constants listed in Table I are found to suit well with biomass gasification system. Reaction

kinetics reported by Corella et al.[15] in R1, R3, and R4 adopted some correction factors to suit his modeling of woody biomass in circulating fluidized bed gasifier. Kinetic constants reported by various researchers [16, 20-24] were varies in term of biomass types and gasification systems hence explained the correction factors adopted by Corella et al.[15]. However, R2 and R6 were originated from coal reaction scheme (which is considered in this work) of carbon reactions in solid form as reported by Mann et al.[13]. On the other hand, enhancement of biomass gasification system (which is in-situ removal of carbon dioxide) suggested by Florin et al.[14] was without kinetic constant. Hence kinetic constant of R5 is considered into this work on the basis of the carbon dioxide capture by calcium oxide will behave similarly in biomass gasification system due to limited literature specifically on reporting the kinetic constants.

C. Process Development

The whole process is assumed to consist of reactions (R.1 to R.6) happening in sequence which are char gasification, methanation, methane steam reforming, water-gas shift, carbonation and bouduard. The limiting factor of this reaction sequence is the result of some gases is needed to be produced first before it can be used as reactants of following reaction. For example, methane is involved in R2 and R3 where methane is the product in R2 and reactant in R3. Hence, R2 should occur before R3. Same analysis is conducted for all reactants and products which are carbon, carbon dioxide, carbon monoxide, and hydrogen.

TABLE II
KINETICS CONSTANTS USED IN ENHANCED BIOMASS GASIFICATION
SIMULATION

Reaction	Kinetic constant	Basis	Ref.
R1	$2.0 \times 10^5 \exp(-600/T)$	Biomass	[15, 16]
R2	$0.12 \times 10^3 \exp(-1792/T)$	Coal	[18, 19]
R3	$3.1005 \exp(-1500/T)$	Biomass	[15, 20, 21, 22]
R4	$10^6 \exp(-6370/T)$	Biomass	[15,16, 23,24]
R5	$10.20 \times 10^6 (-44.5/T)$	Pure CO ₂	[25]
		Biomass	[26]
R6	$4.4 \times 10^3 \exp(-1.62 \times 10^3/T)$	Coal	[18]
		Biomass	[27]

The basic flow diagram for the above process can be represented in Fig. 1. For this simulation, 1 ton/h biomass is fed into the gasifier of and steam will be injected into the gasifier according to mass ratio of steam-over-biomass. Amount of adsorbent inside the gasifier is in accordance to mass ratio of adsorbent-over-biomass, and product gas of the gasifier will be compressed before entering the purification section. Hydrogen is obtained after the Pressure Swing Adsorption (PSA) unit and the remaining gases separated at the bottom of PSA unit called PSA off-gas.

D. Process Assumptions

The assumptions made regarding the biomass gasification are listed below:

1. Biomass is represented as carbon, C
2. Gasification unit is operated isothermally and under steady state condition
3. Ash is considered as inert and will not participate in the reaction
4. Carbonation reaction (R5) is assumed to be a forward reaction and reactions other than R5 are assumed to be at equilibrium
5. Product gases of biomass gasification are hydrogen, carbon monoxide, carbon dioxide and methane.

E. Process Simulation using iCON

This work uses iCON, PETRONAS process simulation software developed by PETRONAS and Virtual Materials Group (VMG) Incorporation using SIM42 open-source simulation frame. iCON runs on built-in thermodynamics package of VMG Thermo, and this work uses gasification package that caters gasification properties with solid support.

III. RESULTS AND DISCUSSIONS

A. Process Description

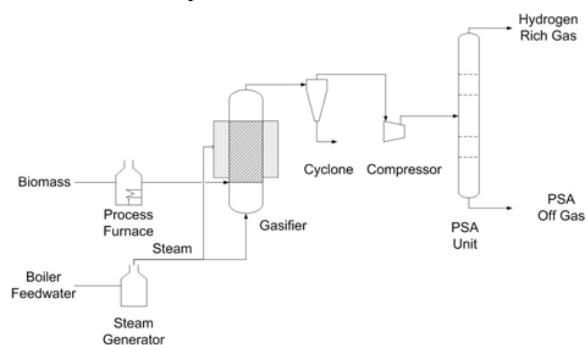


Fig. 2 Process flow diagram of enhanced biomass gasification system

Process flow diagram shown in Fig. 2 developed with considerations of reaction sequence (shown in Fig. 1) and reaction schemes (R1 to R6). Gasifier is a steam-jacketed vessel with biomass and steam as raw materials and calcium oxide as adsorbent inside the gasifier. The gasifier operates at 600-1000°C at atmospheric pressure. Compressor downstream of the gasifier is to increase the product gas pressure up to 6 bar so that hydrogen rich gas can be obtained as PSA unit separates the hydrogen from other product gases.

The simulated flowsheet of enhanced biomass gasification is shown in Fig. 3. The assumptions taken in this simulation is that the gasification which consists of a series of reactions occurring in separate reactors. The operating condition of reactors all reactor is kept constant reflecting one physical reactor.

Biomass is fed into the gasifier along with steam as the gasifying agent. Reaction occurs between biomass and steam produces hydrogen and carbon monoxide in R1. Remaining of biomass reacts with hydrogen produced in R1 to form methane in R2. The reaction between methane and steam occurs to produce carbon monoxide and hydrogen in R3.

Due to the present of carbon monoxide and steam in the system, the water gas shift reaction in R4 takes place next to form hydrogen and carbon dioxide. This carbon dioxide reacts with calcium oxide to form calcium carbonate in R5. This in-situ carbon dioxide removal shifts the water gas shift reaction forward to produce more hydrogen according to Le Chatelier's Principle. Hydrogen reacts with the remaining biomass to form carbon monoxide in R6.

Solid waste of carbonation process which is calcium carbonate can be generated in the separate vessel that will regenerate to produce calcium oxide and carbon dioxide. Calcium oxide can be fed back into the main reactor and carbon dioxide needs to be captured by appropriate means. However, regeneration process of calcium carbonate is not included in this system since it is not parts of the focus of this work.

The simulation snapshot and material balance table of enhanced biomass gasification system are as shown in Fig. 3 and Table III. Operating parameters of simulation are as shown in Table II.

TABLE II
OPERATING PARAMETERS OF BIOMASS GASIFICATION SYSTEM

Parameter	Value	Unit
Temperature	600 – 1000	°C
Pressure	1 (Atmospheric)	Bar
Biomass Flow	1	ton/h
Steam Flow	2.4	ton/h
Adsorbent Flow	3.5	ton/h

TABLE III
OPERATING PARAMETERS AND MASS FRACTION OF COMPONENTS IN ENHANCED BIOMASS GASIFICATION SYSTEM

Name	water	LPS	IFB	S1	SL1	S2	S3	SL2	S4	S5	SL3	S6	S7	CaO	S8	S9	S10	S11	S12	S13	SL4	S15	Solid_Waste	Hydrogen	PSA_Off_Gas
T [C]	25	850	300	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850	850
P [kPa]	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	101.325	600.00
MASS FLOW [kg/h]	2400	2400	1000	1000	1050	2050	2050	300	2050	2350	1050	2350	3400	3500	3400	6900	6900	6900	599	599	599	0	6301	266	333
CARBON	0.0000	0.0000	1.0000	0.0000	0.0000	0.4878	0.1463	0.0000	0.0000	0.0578	0.0000	0.0591	0.0000	0.0000	0.0056	0.0027	0.0027	0.0027	0.0027	0.0027	0.0027	0.0000	0.0000	0.0000	0.0000
WATER	1.0000	1.0000	0.0000	0.0000	1.0000	0.5122	0.0000	1.0000	0.0000	0.0000	0.0000	0.0249	0.3360	0.0000	0.0056	0.0027	0.0027	0.0027	0.0027	0.0027	0.0027	0.0000	0.0000	0.0000	0.0570
HYDROGEN	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0615	0.0425	0.0000	0.0784	0.0386	0.0386	0.0386	0.0386	0.0386	0.0386	0.0000	0.0000	1.0000	0.0000
CARBON MONOXIDE	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.8544	0.5906	0.0000	0.0923	0.0455	0.0455	0.0455	0.0455	0.0455	0.0455	0.0000	0.0000	0.0000	0.9406
CARBON DIOXIDE	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
METHANE	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
CALCIUM OXIDE	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1.0000	0.0000	0.5072	0.0157	0.0157	0.0157	0.0157	0.0157	0.0000	0.0172	0.0000	0.0000
CALCIUM CARBONATE	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.8773	0.8773	0.8773	0.8773	0.8773	0.0000	0.9607	0.0000	0.0000

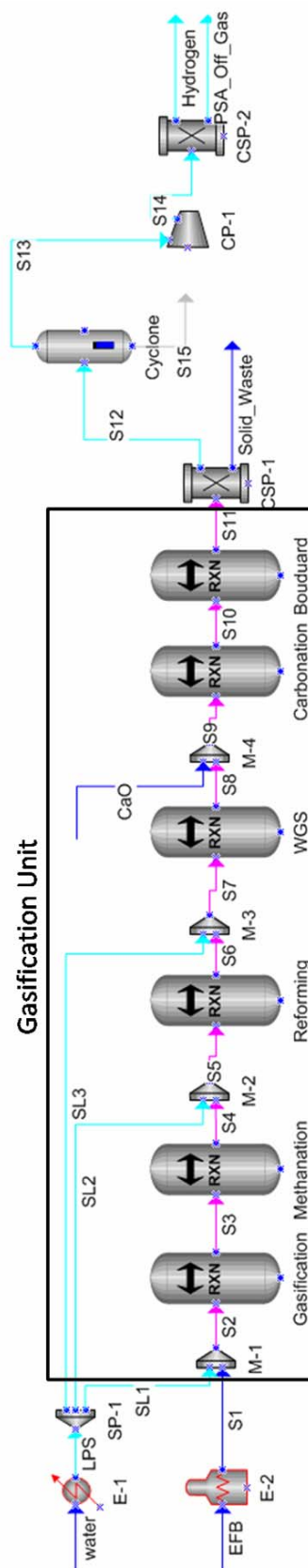


Fig. 3 Simulation snapshot of enhanced biomass gasification unit

B. Effect of Steam/Biomass Ratio

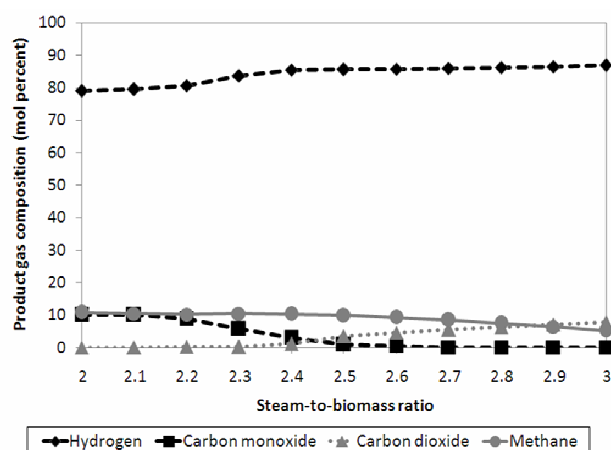


Fig. 4 Effect of steam-to-biomass ratio on product gas composition

Fig. 4 shows that the effect of steam-to-biomass ratio from 2 to 3 at a specific temperature of 850°C and adsorbent-to-biomass ratio of 3.5. The plot shows that the production of hydrogen as main product gas increases from 79% to 85% and almost constant after that point. Trends for methane and carbon monoxide are also decreases over the range of the ratio while carbon dioxide shows increasing trend. Methane production is decreased due to more steam is available to reform the methane hence decreasing the amount of methane in the system. Carbon monoxide and carbon dioxide plots were inverted to each other explaining the effect of water gas shift reaction which is more steam is available to convert carbon monoxide to carbon dioxide and hydrogen. The optimum steam-to-biomass ratio that can be predicted from plot in Fig. 3 is 2.4 that also match the theoretical-stoichiometric ratio. Mahishi et al. [10] and Gao et al. [28] reported that suitable steam-to-biomass ratio of biomass gasification is 3 and 2.05 respectively, which is close to the finding of this study i.e. 2.4.

C. Effects of Temperature

i- Effect of Temperature on Product Gas Composition

Figure 5 shows that effect of temperature ranging from 600 to 1000°C on the product gas composition at a specific condition of steam-to-biomass ratio of 2.4 and adsorbent-to-biomass ratio of 3.5. The plot shows production of hydrogen is almost constant throughout the temperature range studied. However production of other product gases like methane and carbon dioxide shows increases trend, but carbon monoxide production decreases over an increased temperature.

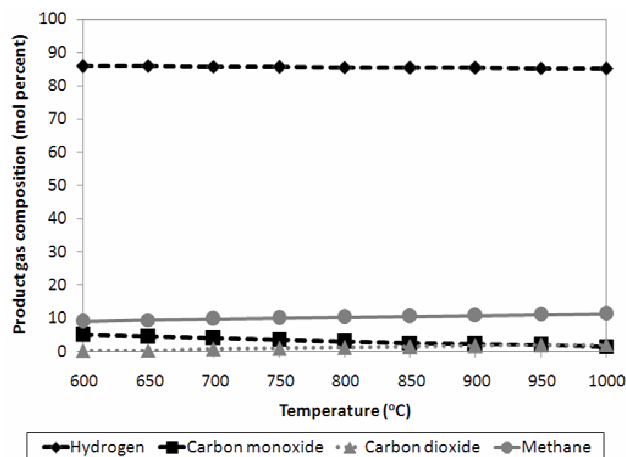


Fig. 5 Effect of temperature on product gas composition

Although there is not much difference on overall trends of the plot, consistent mol fraction of gases is observed at 850°C and onwards. This is due to the amount of carbon monoxide and carbon dioxide is almost stable, hence the improvement can be done at this point via manipulating other effects such as to increase steam-to-biomass ratio to be converted to more carbon monoxide to carbon dioxide and hydrogen, and also to increase adsorbent-to-biomass ratio to capture more carbon dioxide to drive the overall reaction scheme to produce more hydrogen. Mahishi et al. [10] also reported that the optimum operating temperature of his system is at 1030 K (857°C) which is close to the optimum temperature of this system which is 850°C.

ii- Effect of Temperature on Hydrogen Yield

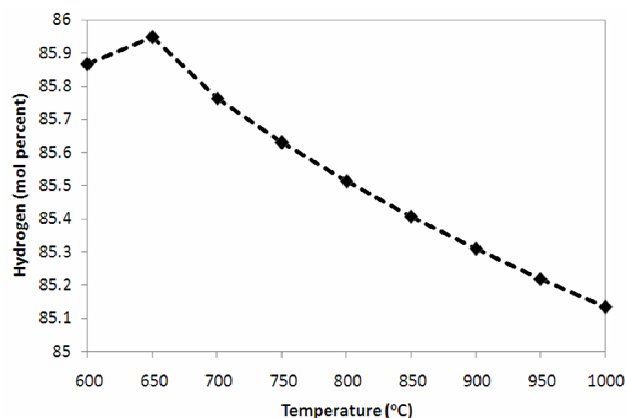


Fig. 6 Temperature effect on hydrogen yield

Fig. 6 shows hydrogen yield over a temperature range of 600 to 1000°C at steam-to-biomass ratio of 2.4 and adsorbent-to-biomass ratio of 3.5. Hydrogen yield increases between 600 to 650°C and decreases after 650°C. This is due to more hydrogen is consumed to produce methane at higher temperature hence reducing the quantity of hydrogen in the product gas. Also, water gas shift reaction is favored at lower

temperature resulting less hydrogen is produced from this reaction at high temperature. This trend of this finding is consistent with trend reported by Mahishi et al. [10] that hydrogen yields decreases over increasing temperature. However, only 82.2 mol% of hydrogen due to the higher amount of CO₂ present is reported by Mahishi et al. [10] as compared to this finding which is 85.95 mol%.

iii- Effect of Temperature on H₂/CO Ratio

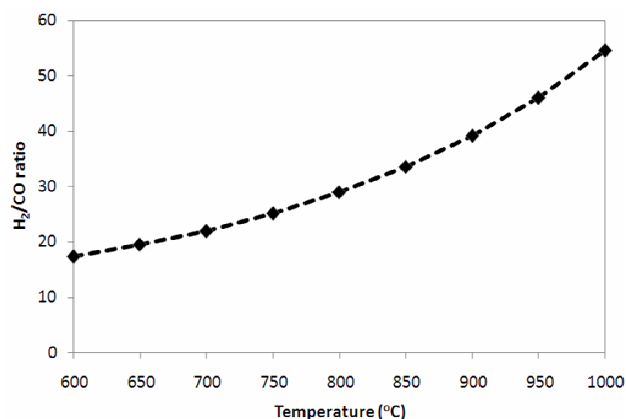


Fig. 7 Temperature effect on hydrogen/carbon monoxide ratio

Fig. 7 shows the effect of temperature between 600 to 1000°C on hydrogen over carbon monoxide ratio at steam-to-biomass ratio of 2.4 and adsorbent-to-biomass ratio of 3.5. The trend shows an increase of the ratio over increases temperature, which indicates that more hydrogen being produced and more carbon monoxide being converted in the system. This explains the effect of water gas shift reaction which consumes carbon monoxide to produce more hydrogen. This values of hydrogen-to-carbon monoxide ratio is important to evaluate the product (hydrogen) over unwanted product (particularly in this case is carbon monoxide). It is important to indicate this ratio since carbon monoxide can be converted to carbon dioxide via water gas shift reaction, hence produce more hydrogen. Trend of increasing ratio of hydrogen-over-carbon monoxide of this work is also reflective to same ratio reported by Mahishi et al. [10].

D. Effect of Adsorbent/Biomass Ratio

Fig. 8 shows the effect of adsorbent-to-biomass ratio of 1.5 to 4.5 on the product gas composition at the temperature of 850°C and steam-to-biomass ratio of 2.4. Hydrogen production increases with increasing ratio up to 3.5 where the trend becomes constant at 85 mol percent. Similar trend can be observed also on methane production where it also stops increasing at the ratio of 3.5. Carbon monoxide production, on the other hand, shows no change as it has reached equilibrium. However carbon dioxide production decreases constantly until the ratio reaches 3.5 and the trend reaches almost 0 above this ratio. It can be concluded that the optimum adsorbent-to-biomass ratio for this system is 3.5. Adsorbent-to-biomass molar ratio of this work found to be optimum at 0.75 which is

much lower (which is one) reported by Mahishi et al. [10].

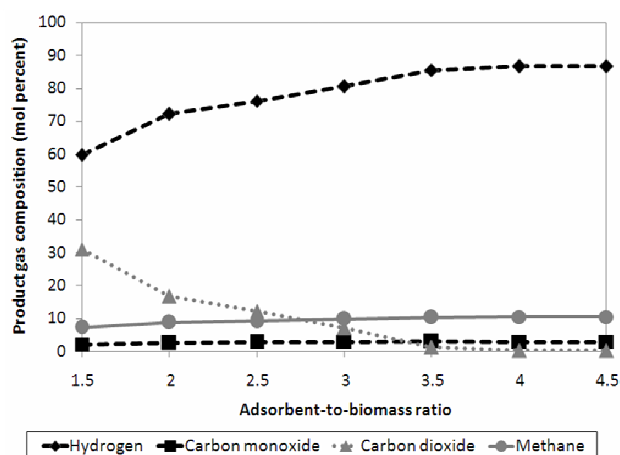


Fig. 8 Adsorbent-to-biomass ratio effect on product gas composition

E. Economic Analysis

The preliminary economic potential of this system is determined to evaluate the viability of this process from the economic point of view. Table IV shows that the analysis on the economic potential level 1 of the enhanced biomass gasification system.

TABLE IV
PRODUCT SELLING PRICE AND RAW MATERIALS COST [29]

Material	Selling price (RM per kg/h)	Cost (RM per kg/h)
Hydrogen	RM 10 per kg/h	-
Steam	-	RM 0.38 per kg/h

Preliminary annual economic potential of industrial scale of biomass gasification is economic potential 1 is as below:

$$\begin{aligned}
 &= \text{Revenue} - \text{Raw material cost} \\
 &= \text{Hydrogen produced} - \text{steam consumed} \\
 &= \text{RM } 19,800,000 - \text{RM } 7,223,040 \\
 &= \text{RM } 12,576,960 \\
 &= \text{USD } 3.77 \times 10^6
 \end{aligned}$$

(Currency exchange rate is RM 3.34 as of 15 October 2009)

TABLE V
ECONOMIC POTENTIAL OF DIFFERENT BIOMASS GASIFICATION SYSTEM FOR HYDROGEN PRODUCTION

Preliminary Economic Potential		Remarks
(RM/year)	(USD/year)	
7.83×10^5	2.34×10^5	Estimated from [11]
12.57×10^6	3.77×10^6	This study

For comparison, Table V below shows that the economic potential level 1 of another biomass gasification plant reported by Lv et al. [11], which adopting oxygen-enriched air and steam as gasifying agent. It shows that greater value of economic potential shows that this system i.e. enhanced biomass-steam gasification is economically more attractive

than gasification system [11].

IV. CONCLUSION AND RECOMMENDATIONS

The prediction model of enhanced biomass gasification in steam-assisted gasifier is successfully performed using iCON simulation software. The results obtained show good agreement and follow trends in referred literatures. Optimum condition of biomass gasification is at 850°C, steam-to-biomass ratio of 2.4 and adsorbent-to-biomass ratio of 3.5. The preliminary economic analysis shows that proposed flowsheet is positively viable. This work can be a platform to further develop the process of more detailed design of commercial hydrogen production.

However some recommendations that can further improve this simulation of biomass gasification. Methods of flowsheet development can be improved to increase the quality of the results. Experiments can be conducted to generate more valuable reaction kinetics that will give higher impact on the simulation work. Single unit reactor in simulation can be developed to be more presentable to the actual physical reactor. Exact chemical formula of biomass should be used to reflect the experimental conditions and improve simulation results.

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