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Simulation of Oxygen-steam Gasification with CO₂ Adsorption for Hydrogen Production from Empty Fruit Bunch

Murni M. Ahmad, Abrar Inayat, Suzana Yusup and Khalik M. Sabil
Center of Biofuel and Biochemical, Green Technology Mission Oriented Research,
Universiti Teknologi PETRONAS, Bandar Seri Iskandar, Tronoh 31750, Malaysia

Abstract: Current study focuses on the process development of hydrogen production via gasification of Empty fruit bunch (EFB) with *in-situ* adsorption of CO₂ based on equilibrium modeling approach. The process flowsheet simulation is performed using iCON, PETRONAS process simulation software. This work investigates the influence of the temperature within the range of 600 to 1000°C and steam/biomass ratio between 0.1 to 1.0 on the hydrogen yield and product gas composition. The importance of different reactions involved in the system is also discussed. Using the simulation, the optimal operating conditions are predicted to be at 800°C and steam/biomass ratio of 0.6. Hydrogen yield of 149g kg⁻¹ of EFB can be obtained at 1000°C. The preliminary economic potential per annum of the oxygen-steam gasification system coupled with *in situ* CO₂ adsorption is RM 6.64×10⁶ or approximately USD 2×10⁶.

Key words: Hydrogen, gasification, empty fruit bunch, simulation, CO₂ adsorption

INTRODUCTION

The world is facing a critical situation in which fossil fuel reservoir is depleting while the demand for energy is increasing worldwide. Scientists globally have shifted their effort towards developing alternative sustainable fuels and quite a number of technologies have been discovered. One potential alternative solution is to produce energy from hydrogen as its energy content per kilogram is three times larger than that of gasoline (Holladay *et al.*, 2009). The combustion of hydrogen produces water instead of greenhouse gasses, along with energy, making hydrogen even more attractive as a clean fuel. The current method to produce hydrogen via natural gas reforming is not favourable for long term implementation and large scale hydrogen production as natural gas itself is an energy source. Another potential method to produce hydrogen is via thermal conversion of biomass, i.e., pyrolysis and gasification (Kumar *et al.*, 2009). Pyrolysis converts biomass to bio-oil prior to hydrogen. On the other hand, gasification directly converts biomass to hydrogen (Kalinci *et al.*, 2009). Despite the current conclusion that pyrolysis has several advantages over gasification e.g., better transportability and potential production and recovery of higher value added co-products from bio-oil, enhanced gasification may offer economic and technical feasibility (Saxena *et al.*, 2008). Balat and Balat (2009) reported that hydrogen production via gasification

process is more economical than the pyrolysis process due to lower production cost. Moreover, different gasifying agent such as air, pure oxygen, steam or their combination, can affect the performance of the hydrogen production (Gonzalez, *et al.*, 2008; Jangsawang *et al.*, 2006). Furthermore, the hydrogen production via gasification can be further enhanced via integration with a CO₂ removal step. This concept has been proven experimentally by a few researchers (Guoxin and Hao, 2009; Mahishi and Goswami, 2007a; Marquard-Mollenstedt *et al.*, 2009): where the CO₂ adsorption step strongly favoured the forward water gas shift reaction that increased hydrogen yield. They however observed that high reactor temperature was not in favour of CO₂ adsorption.

For these purposes, simulation approach is implemented due to the exhaustive range of operating conditions and the higher cost to directly execute the process at plant scale or even lab scale. The potential for production of hydrogen from biomass in Malaysia is attractive due to the abundance of biomass in Malaysia as the world largest exporter of palm oil (Abdullah and Yusup, 2010; Muis *et al.*, 2010; Sumathi *et al.*, 2008). The availability of Empty Fruit Bunch (EFB) of palm oil in 2006 is recorded as 15.8 MT/year (Kelly-Yong *et al.*, 2007). Based on the conversion reported by Kelly-Yong *et al.* (2007), the amount of EFB alone can be converted to supply approximately 16% of the world hydrogen demand.

There are few modelling approaches reported for gasification process based on the kinetics (Corella and Sanz, 2005; Fiaschi and Michelini, 2001; Inayat *et al.*, 2010a; Inayat *et al.*, 2010b; Matsumoto *et al.*, 2009; Nikoo and Mahinpey, 2008; Petersen and Werther, 2005; Piatkowski and Steinfeld, 2010), equilibrium (Baratieri *et al.*, 2008; Brown *et al.*, 2009; Florin and Harris, 2007; Jarungthammachote and Dutta, 2008; Mahishi and Goswami, 2007b; Shen *et al.*, 2008) and fluid dynamics (Sadaka *et al.*, 2002; Wang and Yan, 2008) behaviours and based on using different type of reactors such as fixed bed, fluidized bed and dual fluidized bed reactor. Spath *et al.* (2005) reported a study performed using ASPEN PLUS on a low pressure indirectly heated steam gasification of hybrid poplar wood chips in a circulating interconnected fluidized bed reactor-char combustor system. Gas yield of 0.04 lb-mole dry gas/lb biomass is obtained at 870°C, 1.6 bar and steam-to-biomass ratio of 0.4. Furthermore, Shen *et al.* (2008) presented a simulation model in ASPEN PLUS on a similar system for straw gasification at temperatures between 750 and 800°C with steam-to-biomass ratio ranging from 0.6 to 0.7 and reported hydrogen yields of 54 to 63 g kg⁻¹ of biomass. Mahishi and Goswami (2007b) presented a thermodynamic equilibrium model for prediction of product gas composition and hydrogen efficiency in air-steam gasification process using Stanjan (v 3.93 L) software. They applied first law analysis on the gasifier and investigated the effect of temperature, steam/biomass ratio and equivalence ratio on hydrogen efficiency. They reported that hydrogen efficiency decreased by increasing of both temperature and steam/biomass ratio. Moreover, Nikoo and Mahinpey (2008) ran a simulation work on pine sawdust steam gasification in a fluidized bed reactor using ASPEN PLUS. They predicted that the hydrogen yield increased from 39 to 43% for temperature range of 700 to 900°C from 38 to 40% for steam/biomass ratio range of 0 to 4 and that the yield decreased from 40 to 38% for equivalence ratio value in the range of 0.19 to 0.27. In another work, a steam gasification process in a dual fluidized bed reactor system at temperatures from 850 to 900°C was simulated using IPSEpro by Proll and Hofbauer, (2008). Natural olivine was used as catalytically active bed material along with CaO/CaCO₃ for selective transport of CO₂, resulting in high hydrogen content in the produced syngas. Moreover Florin and Harris (2007) reported a thermodynamic equilibrium model for hydrogen production from biomass coupled with CO₂ capture step in a dual fluidized bed gasifier and investigated the influence of temperature, pressure, steam/biomass and sorbent/biomass ratios on hydrogen concentration. They

predicted that hydrogen concentration in the product gas could be increased from 50 to 80 vol% by using CaO as sorbent. In addition, Florin and Harris (2008) presented another equilibrium model based on steam gasification of biomass using CaO as sorbent, investigated the effect of temperature, pressure and steam/biomass ratio on the hydrogen production and predicted hydrogen yield more than 2 mole per mole of fuel.

The objectives of this work are to synthesize and develop a process to produce hydrogen from EFB via oxygen-steam gasification with *in situ* adsorption of CO₂ using CaO, to perform simulation for the developed flowsheet in iCON and to study the technical and economical feasibility of the gasification process at industrial scale by using a commercial process simulator developed by PETRONAS and Virtual Materials Group (VMG) inc., iCON.

MATERIALS AND METHODS

The current study is the part of Biohydrogen project carried out at Universiti Teknologi PETRONAS with collaboration with PETRONAS Research Sdn Bhd in 2010.

Process screening and development: To establish the process flow diagram, the comparison between the agents used is done based on results by Gonzalez *et al.* (2008). Generally, solid amount produced from steam gasification is significantly lower i.e., 28 to 6%, compared to 23 to 18% from air gasification for the temperature range between 700 and 900°C. In the same work by Gonzalez *et al.* (2008), the hydrogen yield for steam gasification was increased considerably from 8 to 33% compared to decrement observed for air gasification for temperature range of 850 to 900°C.

As illustrated in Fig. 1, the process has essentially two steps, i.e., EFB steam gasification in oxygen-enriched condition and CO₂ *in situ* adsorption using CaO. The overall process flow is the basis used to develop the simulation model in iCON, using Advanced Peng-Robinson thermodynamic package.

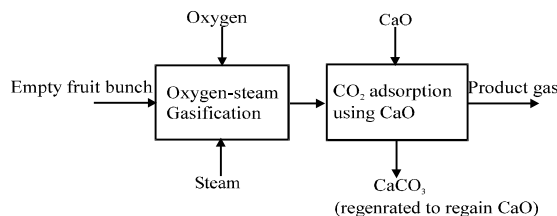


Fig. 1: Block diagram of the hydrogen production process via oxygen-steam gasification with CO₂ adsorption

Assumptions and chemical reaction kinetics: The simplifying assumptions are: EFB is represented as $C_{3.4}H_{4.1}O_{3.3}$ (Laohalidanond *et al.*, 2006), the process is at steady state, there are seven reactions occurring in the gasification process as listed in Table 1 and these reactions are simulated as a series of reactors. More specifically, the partial oxidation and carbonation reactions are assumed to be uni-directional and simulated as conversion reactors, other reactions are assumed to be at thermodynamic equilibrium and tar formation is assumed to be negligible. The kinetics of coal gasification adopted from literatures with respect to the reactions as listed in Table 1 (De Souza-Santos, 1989) are used in the simulation model. This approach is used instead, due to the limited availability of kinetics data for biomass gasification and that coal gasification kinetics can be applied to biomass gasification in modelling work (Nemtsov and Zabaniotou, 2008). The hydrogen yield is calculated using equation (Shen *et al.*, 2008).

$$\text{Hydrogen yield} = \frac{\text{Hydrogen produced in the gasifier (g)}}{\text{Biomass fed into the gasifier (kg)}}$$

Process simulation in iCON: The developed simulation model, as shown in Fig. 2, incorporates the reactions listed in Table 1 and hence configured to consist of a partial oxidation reactor, a gasifier, a methane reformer, a

Water Gas Shift (WGS) reactor, a carbonation reactor and a desorber. Pretreated EFB, oxygen and steam are fed into the gasifier. The first reaction involves EFB oxidation into CO_2 and H_2O and this is modeled as a partial oxidation reactor. The products and unreacted EFB next undergo steam gasification in which EFB is reacted with steam under constant pressure and temperature. This reaction produces H_2 and CO and is simulated as an equilibrium reaction. Parallel to this reaction, Boudouard reaction also occurs in the same reactor. Methanation is assumed to happen subsequently and this reaction that occurs between EFB and H_2 produces CH_4 and H_2O . CH_4 is next cracked in a steam-assisted environment to produce hydrogen and CO as by-product. The equilibrium reaction of Water Gas Shift (WGS) between CO and steam next produces H_2 and CO_2 . The final reaction is the carbonation; the adsorption of CO_2 using CaO that is fed into the system. This *in situ* CO_2 removal shifts the Water Gas Shift (WGS) reaction forward thus resulting in higher hydrogen content in the product gas. The hydrogen-rich product gas is to be further run through a separator which conceptually represents a pressure, swing adsorption unit for hydrogen purification. The molar flow rate of EFB and oxygen are kept constant at $100 \text{ kg mole h}^{-1}$. The operating conditions used in the simulation and the mass balances for the hydrogen production process are given in Table 2.

Table 1: Reaction schemes and parameters (De Souza-Santos, 1989)

Reaction	Reaction scheme	Conversion (%)	Equilibrium constant (K_C)
Partial oxidation	$C_{3.4}H_{4.1}O_{3.3} + 2.775O_2 \rightarrow 3.4CO_2 + 2.05H_2O$	97.00	-
Gasification	$C_{3.4}H_{4.1}O_{3.3} + 0.1H_2O \rightarrow 2.15H_2 + 3.4CO$	-	3.139×10^{12}
Boudouard	$C_{3.4}H_{4.1}O_{3.3} + CO_2 \rightarrow 4.4CO + 0.9H_2O + 1.15H_2$	-	1.238×10^{10}
Methanation	$C_{3.4}H_{4.1}O_{3.3} + 8.05H_2 \rightarrow 3.4CH_4 + 3.3H_2O$	-	1.435×10^{11}
Methane reforming	$CH_4 + H_2O \rightarrow CO + 3H_2$	90.00	-
Water gas shift	$CO + H_2O \rightarrow H_2 + CO_2$	39.97	-
Carbonation	$CaO + CO_2 \rightarrow CaCO_3$	99.00	-

Table 2: The operating conditions and mass balances for the simulation of the hydrogen production process

Name	EFB	O2	PG	S1	S2	S3	S4	S5	S6	S7	S8	SW	Steam	CaO
VF	0	1	1	0.59457	1	1	1	1	1	0	0	0	1	0
T [C]	150	150	800	149.9	800	800	800	800	800	1124.1	800	800	150	150
P [kPa]	300	300	300	300	300	300	300	300	300	300	300	300	300	300
Mole flow [kgmole/h]	100	100	161.27	280	340.36	351.12	351.22	352.63	352.63	602.63	472.86	311.59	80	250
Mass flow (Fraction)														
EFB	1	0	0	0.67795	0.43357	0.41723	0.41751	0.41751	0.41751	0.21163	0.21163	0.23369	0	0
O ₂	0	1	0	0.22204	0	0	0	0	0	0	0	0	0	0
H ₂	0	0	0.01217	0	0	0.0006	0.00064	0.00094	0.00227	0.00115	0.00115	0	0	0
CO	0	0	0.00388	0	0	0.0178	0.01917	0.00072	0.00037	0.00037	0.00037	0	0	0
CO ₂	0	0	0.02149	0	0.3741	0.37127	0.37127	0.37127	0.40026	0.20289	0.00203	0	0	0
CH ₄	0	0	0	0	0	0.00094	0.00078	0	0	0	0	0	0	0
H ₂ O	0	0	0.96246	0.10001	0.19233	0.19216	0.19199	0.19111	0.17924	0.09086	0.09086	0	1	0
CaO	0	0	0	0	0	0	0	0	0	0.49311	0.23715	0.26187	0	1
CaCO ₃	0	0	0	0	0	0	0	0	0	0	0.45682	0.50444	0	0
Total	1	1	1	1	1	1	1	1	1	1	1	1	1	1

S: Stream line, SW: Solid waste, VF: Vapor fraction, T: Temperature, P: Pressure, PG: Product gas

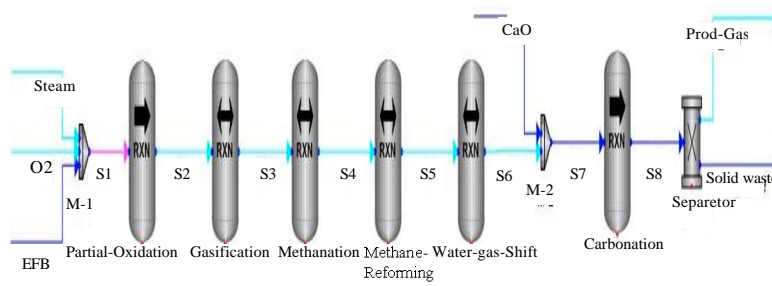


Fig. 2: Simulation model of the hydrogen production process in iCON

RESULTS AND DISCUSSION

Effect of temperature: Figure 3 shows the effect of temperature on the product gas composition within the temperature range of 600 to 1000°C. At 600 K, the mole percentage of H₂, CO and CO₂ are 87, 2 and 12% respectively. At a high gasification temperature, a gas mixture richer in H₂ is produced. For instance, at 1000°C, the product gas contains H₂ with the mole percentage of 89.5, CO, 9.5 and CO₂, 9.5. By increasing temperature in the gasification process, the amount of EFB, CH₄ and carbon decreases and meanwhile the H₂ amount and total gas amount increase. The plot shows that H₂ increases from 87 to 90.5 mole% in the product gas within the temperature range of 600 to 800°C. Beyond 800°C, the mole percentage of H₂ in the product gas starts decreasing. These results can be explained by the Le Chatelier’s principle (Liu *et al.*, 1996) on the endothermic reforming reactions of EFB and CH₄ that are promoted by the increasing temperature (Mahishi and Goswami, 2007b).

Meanwhile, the amount of CO in the product gas is predicted to be increasing with the increase in temperature, due to exothermic and reversible behavior of water gas shift reaction. Figure 3 also shows that initially the amount of CO₂ in the product gas is higher and then starts decreasing by increasing temperature due to CO₂ reaction with EFB, represented by the Boudouard reaction (Yergey and Lampe, 1974) and consumption in the carbonation reaction (Inayat *et al.*, 2010b). Figure 3 plots profiles similar to those published by Khadse *et al.* (2006) who employed an equilibrium model to predict the product compositions of the gasification process for saw dust, bagasse, subabul and rice husk. The similarity arises from the same consideration of reactions occurring, except carbonation. Despite the qualitative similarity, the values differ quite dramatically. This is due to the fact that the reaction equilibrium constants used are obtained for coal and not particularly for biomass. The steam/oxygen ratio is also another contributing factor to the difference in the results.

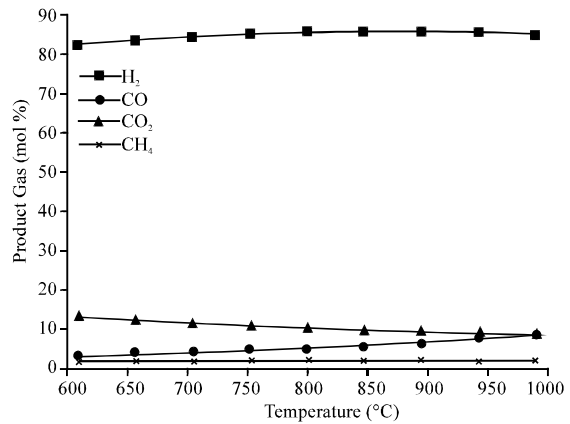


Fig. 3: Effect of temperature on product gas; H₂, CO, CO₂ and CH₄

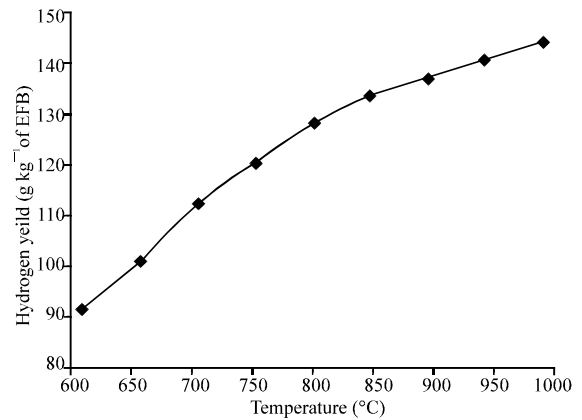


Fig. 4: Effect of temperature on hydrogen yield

Moreover, this process is integrated with CO₂ absorption, resulting in the deviation in the final compositions of the syngas, especially for CO₂.

Figure 4 shows the changes in hydrogen yield with respect to temperature. The plot shows that hydrogen yield increases rapidly with temperature. It is predicted that at a high temperature of 1000°C, hydrogen yield as high as 149 g kg⁻¹ of EFB can be obtained. By increasing the gasification temperature, the hydrogen yield increases

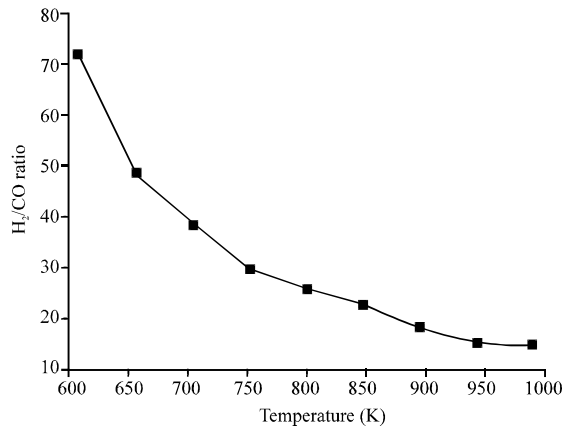


Fig. 5: Effect of temperature on H₂/CO ratio

due to endothermic behavior of gasification, methane reforming and CO₂ adsorption.

H₂/CO ratio is another indicator on the performance of the gasification process, especially when CO₂ adsorption technique is also incorporated in the process. H₂/CO ratio trend is a good representation for the Water Gas Shift (WGS) reaction which is the most important reaction in the steam gasification process. Figure 5 shows the effect of temperature on the H₂/CO ratio: the ratio is higher at lower temperature and lower at higher temperature. This observation is explained by the increase of CO production at high temperature that is caused by the promotion of the reverse reaction of the endothermic Water Gas Shift (WGS) reaction. This further indicates the insuitability of performing the steam gasification process at high temperature for hydrogen production, due to the increase of CO amount in the product gas.

Effect of steam/biomass ratio: In Fig. 6, it is observed that the hydrogen composition in the product gas increases with the increase in steam/biomass ratio. Meanwhile, the composition of CH₄ and CO decreases. The observation is due to the enhanced CH₄ steam reforming and Water Gas Shift (WGS) reactions, which are highly dependent on steam feed rate. CO₂ decreases due to the carbonation reaction with CaO. This simultaneous removal of CO₂ shifts the equilibrium of Water Gas Shift (WGS) reaction to produce more hydrogen, resulting in hydrogen rich gas stream. The presence of steam and the removal of CO₂ promote the forward reaction of Water Gas Shift (WGS) reaction, causing the increase in the consumption of CO.

Figure 7 plots the curve for H₂/CO ratio as affected by the steam/biomass ratio. The profile shown here i.e., that the H₂/CO ratio is increasing rapidly when the steam fed is increased, illustrates an opposite effect

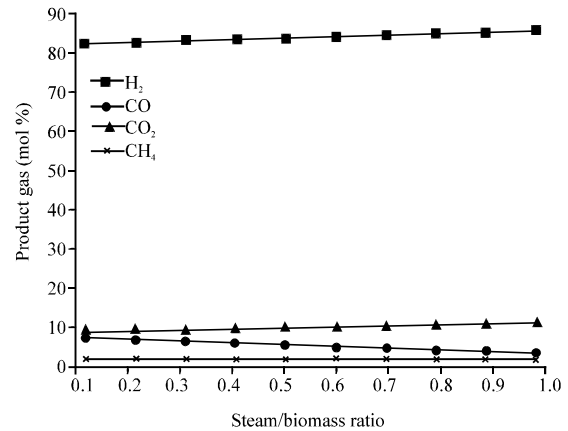


Fig. 6: Effect of steam/biomass ratio on product gas H₂, CO, CO₂ and CH₄

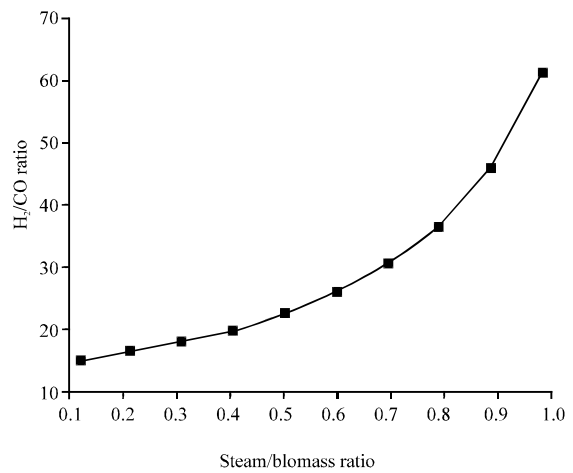


Fig. 7: Effect of steam/biomass ratio on H₂/CO ratio

compared to by the temperature increase. The rapid increase of hydrogen and the decrease of CO are caused by the escalation of CH₄ reforming and Water Gas Shift (WGS) forward reaction.

Economic potential: A simplified economic feasibility of the process is reported based on the economic potential level 1, i.e., hydrogen value minus the cost of EFB, steam and oxygen consumption. The current prices of steam, oxygen and hydrogen are RM 345, RM 3200 and RM 4217, respectively (MOX-LINDE GASES SDN BHD (MOX), Malaysia) and EFB is considered free. The preliminary economic potential per annum of the oxygen-steam gasification system coupled with the CO₂ removal step is RM 6.64×10⁶ or approximately USD 2×10⁶. The positive value indicates the economic feasibility of hydrogen production from EFB via the gasification system; however no further analysis can be deducted thus far.

Table 3: Comparison of hydrogen yield and syngas compositions

Basis	This work	Shen <i>et al.</i> (2008)	Mahishi and Goswami (2007b)	Salleh <i>et al.</i> (2010)	Mohammad <i>et al.</i> (2009)
Biomass	EFB	Straw	Wood	EFB	EFB
Method	Equilibrium modelling and simulation	Equilibrium modelling	Equilibrium modelling	Experimental	Experimental
Gasifying agent	O ₂ -Steam	O ₂ -Steam	O ₂ -Steam	Air	Air
CO ₂ removal step	Yes	No	No	No	No
Temperature (°C)	600-1000	600-920	700-800	500-900	700-1000
Pressure (atm)	1	1	1	1	1
Steam/biomass ratio	0.6	0.6	1.9	-	-
H ₂ (vol%)	85-90	40-60	58-59	10-56	10-38
CO (vol%)	1-8	1-20	22-27	4-7	21-36
CO ₂ (vol%)	8-12	20-40	28-25	29-25	62-10
CH ₄ (vol%)	0-1	0-20	1-0	5-45	5-14
Hydrogen yield (g kg ⁻¹ biomass)	88-145	30-62	-	-	-

Comparison with literature: A comparison has been carried out with literatures reporting results for similar operating conditions, as shown in Table 3. Comparing current results to the findings by Shen *et al.* (2008) who investigated straw gasification without CO₂ adsorption, the predicted hydrogen yield is approximately doubled. This may be due to the use of different biomass and the CO₂ removal step. In addition, the profiles shown in Fig. 3 for the compositions of CO and CH₄ observed for the same gasification temperature interval are in match to a certain extent to those reported by Shen *et al.* (2008). However, the compositions of hydrogen and CO₂ differ significantly because of the *in situ* CO₂ adsorption step using CaO that reduces CO₂ amount and shifts the Water Gas Shift (WGS) reaction forward and increases the hydrogen production.

The comparison in Table 3 also shows that our simulation work predicts higher amount of hydrogen compared to the results reported by Mahishi and Goswami (2007b) who used an equilibrium modelling approach. In addition, the results have also been compared with the experimental work of EFB gasification reported by Salleh *et al.* (2010) and Mohammed *et al.* (2009) who used different gasifying agents. The higher value of hydrogen indicates that the oxygen-steam gasification of EFB with *in situ* CO₂ capture is able to produce more hydrogen than the conventional gasification.

CONCLUSIONS

A simulation approach has been employed to predict the hydrogen production via oxygen-steam gasification of EFB. The iCON simulation model is able to profile the product gas compositions comparable to refereed literatures. The results are further extended to include hydrogen yield and H₂/CO ratio. It is predicted that the integration of oxygen-steam gasification with CO₂

adsorption using CaO yields higher hydrogen content in the product gas. Water gas shift reaction and methane steam reforming reactions are mostly affected by the process main variables i.e., temperature and steam/biomass ratio and can be considered as the main reactions for the hydrogen production. Hydrogen production is increased by increasing temperature initially. However, at high temperature, hydrogen amount in the product gas decreases due to the exothermic and reversible behavior of water gas shift reaction. Steam/biomass ratio is also very important in the steam gasification process because hydrogen yield increases when steam/biomass ratio is increased. Meanwhile, CO and CH₄ amounts in product gas decrease as both methane reforming and water gas shift reactions are highly dependent on the steam feed rate. The optimum operating conditions identified for the gasification process is 800°C with the steam/biomass ratio of 0.6. The maximum H₂/CO ratio is achieved at low temperature and high steam/biomass ratio due to the high consumption of CO in the enhanced forward water gas shift reaction. Based on the preliminary economic potential study, the process has positive economic feasibility.

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