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Process simulation of hydrogen production through biomass gasification: Introduction of torrefaction pre-treatment

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HIGHLIGHTS

- A process model for biomass torrefaction and gasification was developed.
- Introduction of steam into gasifier increased H_2 concentration by 10.1%.
- The H_2/CO ratio was higher with lower gasifier temperature, lower equivalence ratio and higher steam/biomass ratio.
- Optimum conditions for hydrogen production have been identified.

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ABSTRACT

Torrefaction is a pretreatment method that converts biomass to a fuel-like substance that can replace coal for sustainable power generation. In this work, a thermodynamic-based process simulation model was developed to simulate the gasification of empty fruit bunch (EFB), with torrefaction as pretreatment, to determine the optimum conditions; equivalence ratio, reactor temperature, torrefaction medium concentration, steam-to-biomass (S/B) ratio and system configuration were studied to determine their influence on hydrogen concentration, higher heating value (HHV), syngas ratio and cold gas efficiency (CGE). The highest hydrogen yield was obtained at an S/B ratio of 1.3 at 800 °C, with a syngas ratio of 2.5 and a CGE of 84%. Concentration of torrefaction medium showed no effect on hydrogen concentration due to the simplicity of the model used, but work is in progress in this direction. Therefore, steam gasification is more suitable than air gasification in hydrogen production.

Introduction

Malaysia, as one of the main oil palm plantation countries, generates about 55.73 million tonnes of oil palm waste per annum, which includes empty fruit bunch (EFB), oil palm frond (OPF), palm kernel shell (PKS), oil palm trunk (OPT) and mesocarp fibre (MF) from the activities of palm oil production and oil palm cultivation [1,2]. Among these wastes, MF and PKS contribute to the sustainability of the oil palm mill operation by being fed into a combustor in order to produce electricity and steam. On the other hand, OPT and OPF are returned to plantations as mulch, erosion control and long-term nutrient recycling [3,4]. This means that only small amount of biomass, including OPF, is utilised for bioenergy, or

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fibre board, paper or fertiliser production, while the rest is merely piled at the roadside [1]; therefore, it should be fully utilised as a renewable energy source.

Biomass as a renewable energy source is better than other sources in some respects, such as low sulphur content, all-year availability and high versatility, therefore increasing its deployment in industry [5,6]. Biomass helps to reduce greenhouse gas emissions and mitigate waste management and pollution problems while reducing dependency on fossil fuels [7–9]. There are several methods of processing biomass: thermochemical and biochemical methods. The currently available biomass thermochemical conversion processes are liquefaction, carbonisation, combustion, pyrolysis and gasification [10,11]. Gasification aims to gasify biomass into gaseous products and focuses on the production of methane (CH_4), hydrogen (H_2), carbon dioxide (CO_2) and carbon monoxide (CO) [12]. Raw syngas that comprises of H_2 and CO can be used for downstream processes like heat and power generation or liquid fuels and bulk chemical productions [13–15]. Gasification involves several stages: drying (<150 °C), pyrolysis (150–700 °C), oxidation (700–1500 °C) and reduction (800–1100 °C) [16]. The gasifying agents often used include oxygen, air and steam, whereby their optimum concentrations promote higher carbon conversion. H_2 gas has high efficiency since it achieves 'zero-carbon' emissions during the generation of power and is environmental friendly, making it one of the best energy carriers [17]. Steam gasification increases the H_2 concentration in product gas and provides higher thermal cold gas efficiency (CGE), therefore is given higher attention [18,19]. However, raw biomass usage in gasification has exposed some problems, as raw oil palm waste often has a high moisture content that reduces the heating value of the product, increases storage and transportation costs, causes incomplete burning that causes carbon burnout, increases biological degradation risks and increases the O_2 content of the product. The increase in storage and transportation costs is due to the low bulk and energy density of raw biomass, while poor grindability increases the energy required for grinding. Its hygroscopic nature, on the other hand, increases biological degradation risks and may cause moisture uptake during storage. This has caused the industry to lose interest in the use of biomass as an energy source [20–24].

As a result, in order to overcome the aforementioned circumstances, torrefaction as a pretreatment method can be a useful option, in that moisture content can be reduced effectively while increasing the calorific value of the biomass [25]. It is a mild thermal heating process at 200–300 °C using an external fuel source in the absence of air or O_2 to produce a charred product with improved fuel performance and physical transport properties [26–29]. Torrefied biomass has higher heating values, improved grindability, lower capacity to uptake moisture and lower oxygen/carbon and hydrogen/carbon ratios [30]. These properties are similar to coal and thus are suitable for gasification [5].

Hydrogen is a clean energy source that can substitute for fossil fuels for direct combustion in an engine or fuel cell and produces only water as a by-product [31]. Its high energy density characteristics makes it more effective in its transportation and use compared to other fossil fuels; it could become a primary source, leading the global energy system in the future [32]. Hydrogen production processes can be separated into two major categories: conventional processes that utilise non-renewable energy sources such as fossil fuels and renewable technologies that utilise biomass as feedstock. For example, coal has been used to produce H_2 through the water gas shift (WGS) reaction and purification in conventional processes [33–35]. In the production of H_2 using biomass, two major routes are available: biological processes, such as biophotolysis and dark fermentation, and thermochemical processes, such as electrolysis and thermolysis [36]. Thermochemical methods have higher efficiency and are generally more cost-effective than biochemical methods [37,38].

Gasification, one of the thermochemical technologies, gives higher H_2 production without the emission of polluting by-products and process conditions are more easily achieved compared to combustion and liquefaction [39]. It produces clean hydrogen energy efficiently on a large scale from several types of biomass, and it is the most economical process for bio-hydrogen production [8,40,41]. Steam gasification is able to produce H_2 in yields far higher than fast pyrolysis with an overall efficiency of up to 52% providing an effective method of renewable H_2 production [42,43]. With the aforementioned advantages, many studies have been conducted related to gasification using biomass. Prins et al. conducted a study on gasification by using torrefied and raw wood and found that the efficiency of torrefied wood in air-blown gasification is promising and the performance and efficiency are comparable to those of raw wood [44]. Muslim et al. simulated gasification using raw and torrefied EFB, where an increase in gasifier temperature increased the amount of H_2 produced, while an increase in equivalence ratio (ER) reduced the amount of H_2 produced. The use of torrefied biomass contributed to higher lower heating value (LHV) and gasification efficiency [45]. Therefore, ER is often lower than 1.0 [46]. A study has also shown that the gas produced by gasification using torrefied biomass has better energy and exergy efficiencies and higher H_2 and CO content compared to raw biomass [47]. Bach et al. simulated torrefaction combined with gasification and CO_2 capture for spruce wood branches, where torrefaction increased the LHV and CGE of the product [5]. Few experimental gasification works with torrefaction pretreatment have been conducted for biomasses such as olive kernel [48] and sewage sludge [49] while for EFB, combination experimental work has been done but at a torrefaction temperature of 300 °C and steam gasification only [50]. To the best of our knowledge there has been no work yet done on the combination of torrefaction and gasification using EFB and the comparison between the air gasification and steam gasification of EFB. So, the aim of this work was to develop and simulate both torrefaction and gasification process of EFB to produce a high yield of H_2 in the product gas.

Methodology

Description of the model

A model for torrefaction and gasification was constructed as shown in Fig. 1. The biomass feedstock in this research was adopted from other work. The fuel properties of the raw biomass including moisture content (MC), fixed carbon (FC),

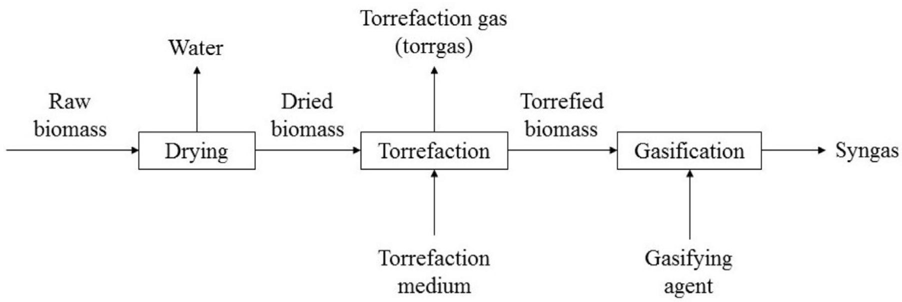


Fig. 1 – Process flow diagram of the overall process of this work.

volatile matter (VM), ash, carbon (C), hydrogen (H), nitrogen (N) and oxygen (O) are shown in Table 1, which includes the higher heating value (HHV). The raw EFB was dried to minimise the moisture content of the oil palm waste prior to entering the torrefaction reactor and to improve its efficiency. The dried EFB was torrefied in the torrefaction reactor in the presence of a medium, i.e., nitrogen (N_2), oxygen (O_2) or carbon dioxide (CO_2) to produce torrefied EFB and gaseous product. The solid product of torrefaction was fed into the gasifier to react with the gasifying agent, i.e., air and steam, to produce syngas that contained H_2, CO, CO_2 and CH_4. Fig. 2 shows the Base Case of this study in the Aspen Plus model.

The biomass was pretreated using the Rstoich DRYER to reduce the moisture content to 1%. Then the RYield YIELD reactor was used to obtain the mass yield of the solid product and the volatile matter composition. The temperature of the YIELD reactor was controlled at 250 °C under atmospheric condition with different types of torrefaction medium added at 10 mL/min. The RYield DECOMPOS reactor was used to convert the feed into carbon, hydrogen, oxygen, sulphur, nitrogen atoms and ash. The decomposed feed and different types of gasifying agent then entered the RGibbs GASIFIER, operated at temperature of 800 °C at atmospheric pressure. Gasifying agents were added in various ratios in order to study the effect of concentration of gasifying agent on the concentration of H_2 generated.

Model assumptions

The assumptions made for the simulation were:

- The model used MIXNC stream class, since both conventional and nonconventional solids are present,
- The properties method selected is Redlich-Kwong-Soave–Boston-Mathias (RKS-BM) cubic equation,
- All pressure drops are neglected with operation under atmospheric pressure,
- A steady state is assumed in all calculations,
- The ambient temperature is 25 °C, and
- Air is assumed to be a combination of 79% N_2 and 21% O_2 on a molar basis.

In order to determine the accuracy of the developed model, the sum-squared deviation method was used, as shown in Equations (1)–(3) [52]:

$$RSS = \sum_{i=1}^{N} \left(\frac{y_{ie} - y_{ip}}{y_{ie}} \right)^2 \tag{1}$$

$$MRSS = \frac{RSS}{N} \tag{2}$$

$$RMSE = \sqrt{MRSS} \tag{3}$$

where RSS is the residual sum of squares, MRSS is the mean residual sum of squares, N is the number of data, y_{ie} is the estimated value, y_{ip} is projected value, and RMSE is the root mean square error. The amount of heat released by the dry gas after gasification for combustible species, known as the higher heating value was calculated using Equation (4) [53]:

$$HHV(MJ/Nm^3) = \frac{12.76(vol\%H_2) + 12.63(vol\%CO) + 39.76(vol\%CH_4)}{100} \tag{4}$$

For the solid fuel, the HHV was estimated using Equation (5) [54]:

$$HHV(MJ/kg) = 0.3491C + 1.1783H + 0.1005S - 0.1034O - 0.0015N \tag{5}$$

where C, H, S, O and N are the carbon, hydrogen, sulphur, oxygen and nitrogen contents (%) in the biomass.

Another important aspect that shows the performance of the syngas produced is the CGE, as shown in Equation (6) [55]:

$$CGE(\%) = \frac{v_g HHV_g}{\dot{m}_f HHV_f} \times 100 \tag{6}$$

where v_g is the gas flow rate (m^3/hr), $\dot{m}_f$ is the fuel mass flow rate (kg/s), and HHV_g and HHV_f are the values of gas (MJ/Nm^3) and fuel (MJ/kg) respectively. Syngas ratio was calculated by Equation (7):

$$Syngas\ ratio = \frac{H_2\ flow\ rate}{CO\ flow\ rate} \tag{7}$$

Case study description

In this work, several studies were conducted for the optimisation of the production of H_2 from EFB using pretreatment of torrefaction.

Table 1 – Ultimate and proximate analysis of raw EFB [1,51].

	Proximate analysis (wt % dry)				Ultimate analysis (wt % dry ash free)				HHV (MJ/kg)
	MC	FC	VM	Ash	C	H	N	O	
Raw EFB	6.55	10.23	80.11	3.11	42.26	6.25	0.73	50.76	16.94

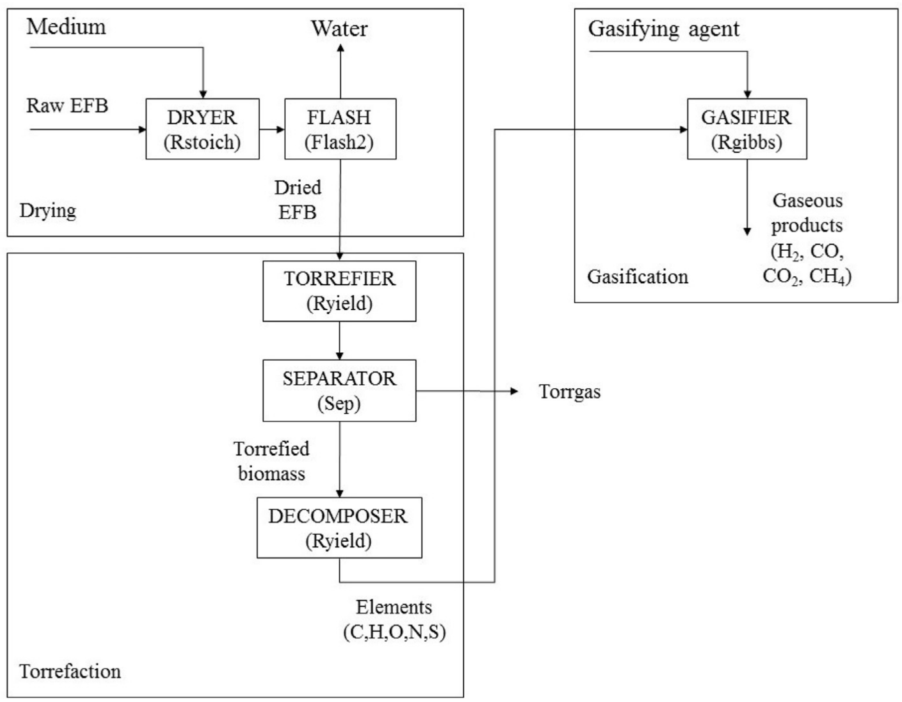


Fig. 2 – Base Case.

Base Case

The H_2 production in terms of molar composition was observed using torrefaction under inert atmosphere, where the product gas was purged and the torrefied biomass reacted with O_2 during gasification. Sensitivity analysis was conducted to detect the effects of gasifier temperature and ER on the production of H_2.

Case 2: Effect of torrefaction medium

Oxidative torrefaction was conducted where O_2 and CO_2 were introduced into the torrefaction reactor to test the effect on H_2 production, as oxidative torrefaction can produce biomass with higher HHV and higher carbon content.

Case 3: Effect of gasification medium

Steam was fed into the gasifier, as steam gasification can produce a higher H_2 concentration in syngas. Therefore, sensitivity analysis of the steam-to-biomass (S/B) ratio was conducted to attain the optimum S/B ratio for highest H_2 concentration produced.

Case 4: Effect of torrgas recycling

Torrgas was recycled into the gasifier instead of being purged to determine whether this could improve the production of H_2 in the syngas. Fig. 3 shows the configuration of Case 4.

The case studies summary is shown in Table 2.

Results and discussion

Validation of model

The simulated results from this work were compared with those of previous experimental works from the literature [44,51] in order to validate the model's accuracy, as shown in Tables 3 and 4. The final simulation results were very similar to the literature results. The RMSE is low, which shows that the model is reliable and validated.

Effects of gasifier temperature and ER

The effect of ER and temperature were investigated to identify the optimum ER and temperature to produce highest H_2 concentration, HHV and CGE. Fig. 4 shows the effects of ER on syngas production in the Base Case. When ER increased from 0.2 to 0.4, the amount of H_2 gas produced decreased slightly. This is because oxidation is faster than cracking reactions in the presence of air, therefore enhancing the formation of more CO_2 while reducing the concentrations of H_2 [56]. Fig. 5 shows the effect of ER on HHV, syngas ratio and CGE. HHV decreased with ER due to the reduction in amount of H_2 produced. A maximum syngas ratio of 1.03 was obtained at an ER of 0.2. Syngas ratio is important, as further utilisation of the

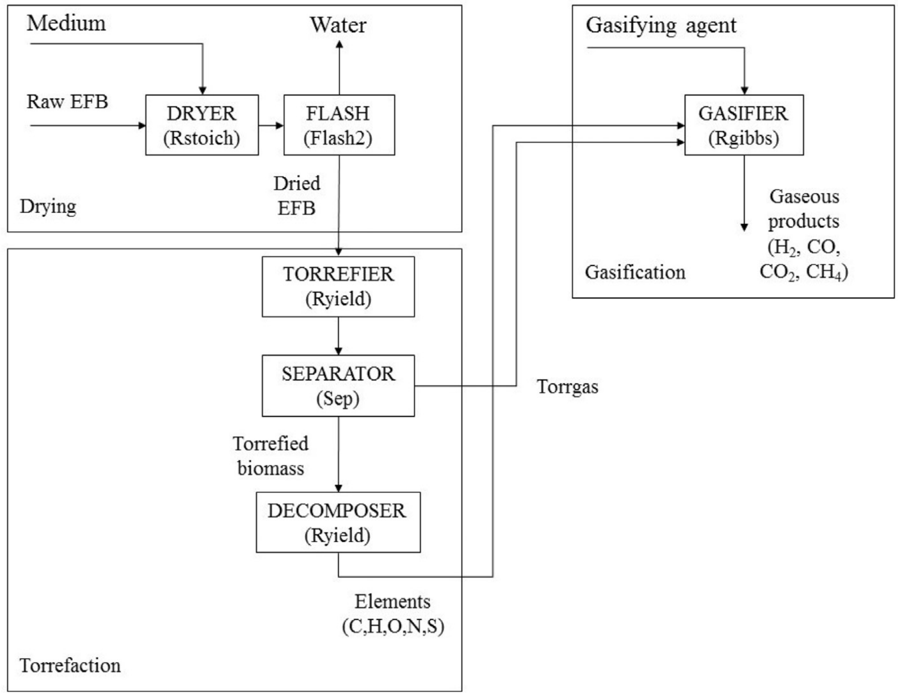


Fig. 3 – Configuration of Case 4.

Table 2 – Case studies summary.

Case study	Torrefaction medium	Gasifying agent	Torrgas
1 (Base Case)	N_2	O_2	Purged
2	O_2, CO_2	O_2	Purged
3	N_2	Steam	Purged
4	N_2	O_2	Recycled

syngas in the production of many chemicals, such as Fischer-Tropsch liquids and methanol, depends on the composition of the syngas [57]. CGE on the other hand increased when ER increased, similar to the trend obtained by Liu et al. [58].

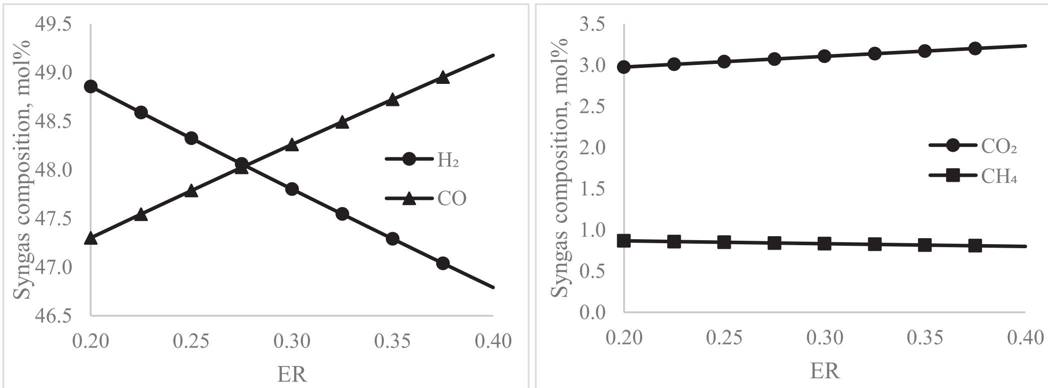
Fig. 6 shows the effect of gasifier temperature on syngas composition. As shown, H_2 content showed a slight decrease of 0.3 mol % as temperature increased from 800 to 1000 °C. CO on the other hand increased significantly with temperature This is due to the occurrence of secondary reactions such as the shifting reaction [59–61]. The CO_2 decrease with temperature might be due to steam reforming and water gas reactions where CO_2 is decomposed into CO [62]. The CH_4 decrease in the composition is due to the cracking reaction into H_2 and CO_2 at higher temperatures [63]. Fig. 7 shows the effect of temperature on HHV, CGE and syngas ratio. The increase in temperature resulted in a significant increase in

Table 4 – Syngas product yield comparison between literature [50] and this work.

Components	Actual yield (vol %)	Simulated yield (vol %; this work)	Difference in yield
H_2	63.52	57.39	6.13
CO	27.22	28.00	0.78
CO_2	9.07	14.54	5.47
CH_4	0.18	0.07	0.11
RMSE, %			4.13

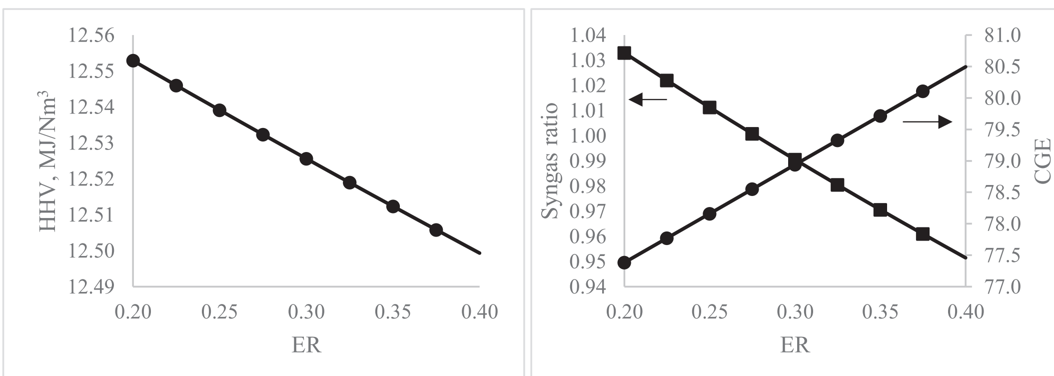
Table 3 – Product yield comparison between literature data [44,51] for the mass yield and volatile product respectively and this work.

Components	Actual yield (wt. %)	Simulated yield (wt. %; this work)	Difference in yield
Torrefied biomass mass yield	86.4514	86.4515	0.0001
Water	1.5814	1.5814	0
Acetic acid	7.3800	7.3800	0
Formic acid	0.5271	0.5271	0
Methanol	0.4744	0.4744	0
Lactic acid	0.1054	0.1054	0
Furfural	0.0011	0.0011	0
Carbon dioxide	3.0574	3.0574	0
Carbon monoxide	0.4217	0.4217	0
RMSE, %			0



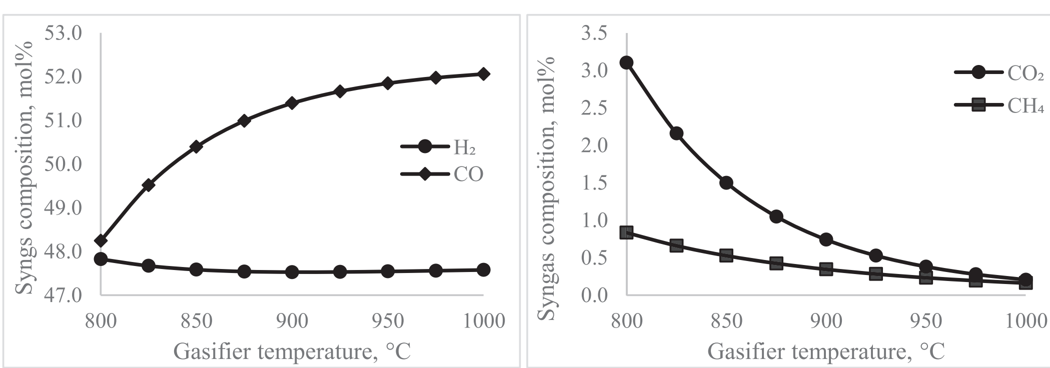
(a) (b)

Fig. 4 – Effects of ER on molar composition of (a) H_2 and CO and (b) CO_2 and CH_4.



(a) (b)

Fig. 5 – Effects of ER on (a) HHV of syngas and (b) syngas ratio and CGE.



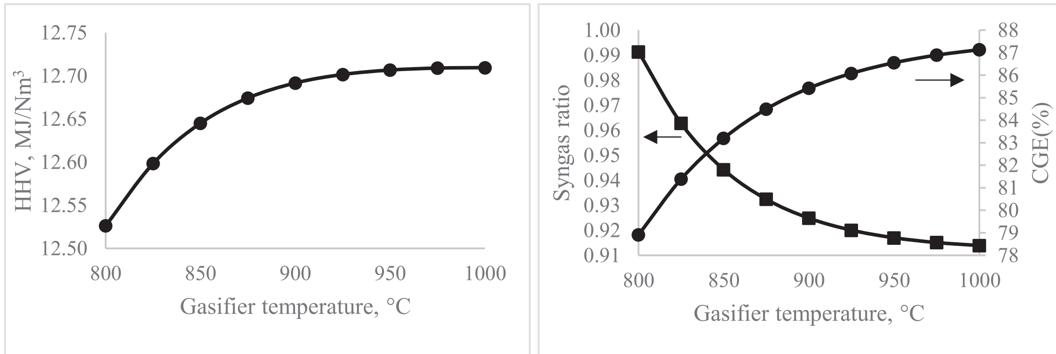
(a) (b)

Fig. 6 – Effect of gasifier temperature on molar composition of (a) H_2 and CO and (b) CO_2 and CH_4.

HHV due to the reduction of CO_2 and increases in CO and CH_4 in the product gas [5]. The syngas ratio reduced with increasing temperature, with a maximum of 0.99 at 800 °C. CGE increased with increased gasifier temperature. This shows that high temperature favours the gasification process but needs to be controlled due to the technical limitations of the gasifier [64].

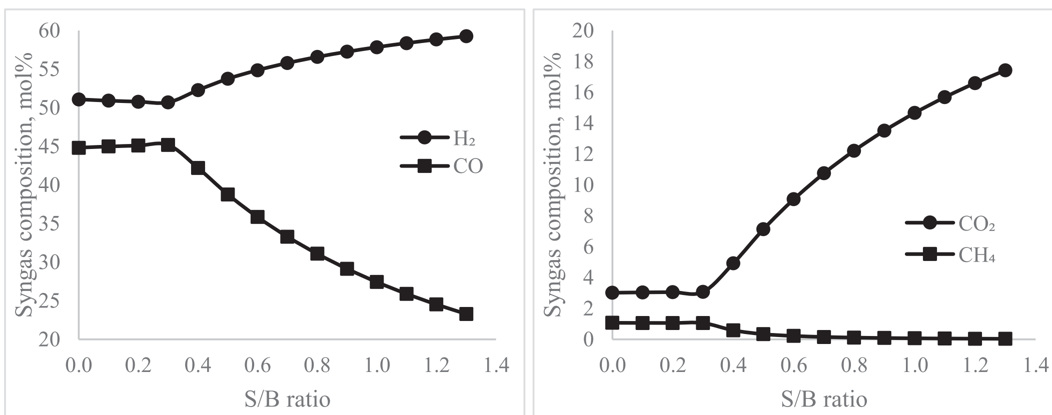
Effects of torrefaction medium concentration

The effect of torrefaction medium concentration on HHV and syngas composition was investigated, as oxidation during torrefaction would increase the HHV of the solid product, therefore increasing the HHV of the syngas. However, there was no effect of torrefaction medium compared to the yield



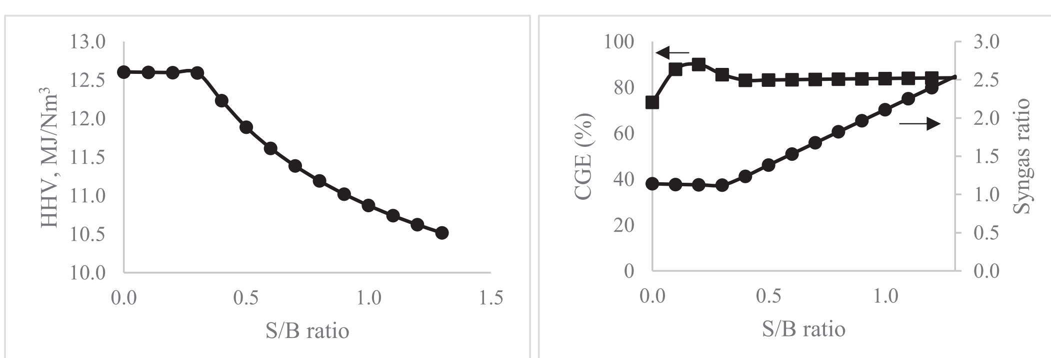
(a) (b)

Fig. 7 – Effect of gasifier temperature on (a) HHV and (b) syngas ratio and CGE.



(a) (b)

Fig. 8 – Effects of S/B ratio on molar composition of (a) H_2 and CO and (b) CO_2 and CH_4.



(a) (b)

Fig. 9 – Effects of S/B ratio on (a) HHV and (b) CGE and syngas ratio.

distribution in the Base Case. This contradicted the results of Sellappah et al., where torrefied EFB in CO_2 resulted in lower H_2 content than in N_2 due to devolatilisation and dehydration during torrefaction [65]. A similar decreasing trend in H_2 content was observed by Adnan et al., who conducted torrefaction in O_2 [66]. This may due to the simplicity of the model meaning that it was unable to predict the changes in the yield when O_2 and CO_2 were introduced into the system, as oxidation was not considered in the model. Therefore, the kinetics of oxidative torrefaction should be introduced into the system to identify further the effect of torrefaction medium on HHV and H_2 concentration in syngas.

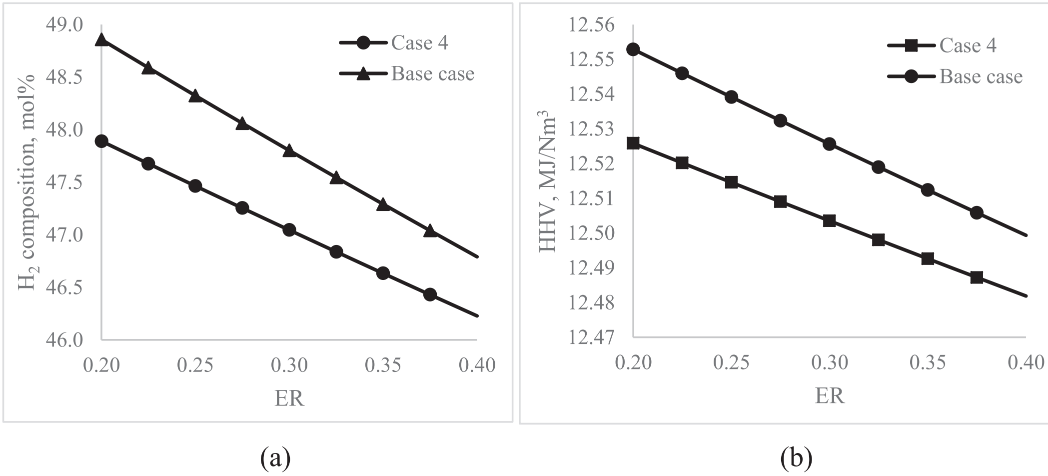


Fig. 10 – Effects of configurations on (a) H_2 concentration and (b) HHV.

Effects of steam as gasifying agent

In order to determine the influence of steam in producing the highest H_2 concentration and HHV, it was used to replace air as gasifying agent in the gasifier, as steam gasification increases H_2 production [67]. Fig. 8 shows the effect of the S/B ratio on syngas composition. It can be observed that there is a range where the concentration of the gases was almost constant up to an S/B ratio of 0.3. H_2 and CO_2 concentrations increased with increasing S/B ratio, while CO and CH_4 concentrations decreased. Similar trends were reported by Pala et al. [68]. The addition of water enhances water gas and methane reforming reactions, which increase the H_2 concentration. Reduced CO concentration is due to the WGS reaction where CO reacts with steam to form H_2 and CO_2 [69]. Fig. 9 shows the effects of S/B ratio on HHV, CGE and syngas ratio. It is worth noting that syngas ratio increased with S/B ratio, reaching a maximum of 2.5 at an S/B ratio of 1.3. However, CGE increased up to an S/B ratio of 0.4 then decreased with increasing S/B ratio because the increase in the amount of steam introduced into the system lowered the concentration of reactants but increased the concentration of products, thereby decreasing the reaction rates and the efficiency of the process [40].

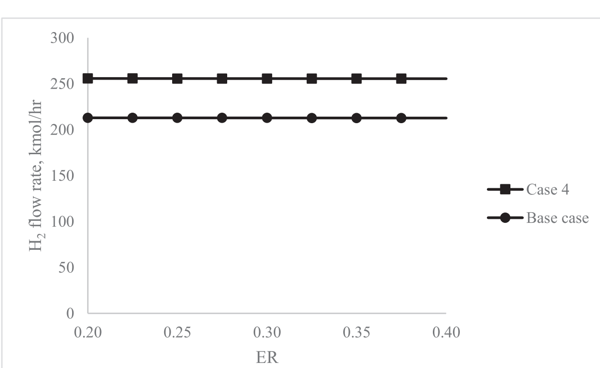


Fig. 11 – Effects of configuration on H_2 flow rate.

Table 5 – Comparison of findings of the current study with the literature.

Aspects	Current work	[70]	[50]	[63]	[71]
Feedstock	EFB				
Temperature (°C)	800	770 ± 20	780	800	800
HHV (MJ/kg)	12.55	7.2	N/A	N/A	N/A
CGE (%)	77.4	40	N/A	N/A	N/A
H_2 concentration (vol %)	59.25	N/A	N/A	53	17.23
H_2/CO ratio	2.05	N/A	8.4	2.52	0.52

Effects of configuration

The gas product stream after torrefaction was recycled to the gasifier to study the effect on H_2 concentration in the syngas, as mentioned in section Case 4: Effect of torrgas recycling. Fig. 10 shows the comparison of the H_2 concentration produced between the Base Case and Case 4, while Fig. 11 shows the effect of recycled torrgas on H_2 flow rate. It can be seen that the recycled gaseous product of torrefaction (Case 4) had lower H_2 concentration than the purged gas product. However, from Fig. 11, Case 4 showed a higher H_2 flow rate than the Base Case. This is due to the increased concentration of other gases in Case 4, which lowered the concentration of H_2. Therefore, recycling the torrefaction gas product produced higher amounts of H_2 but in lower concentration. In terms of HHV, the Base Case had higher HHV than Case 4, which might be due to the lower concentration of H_2 in Case 4, so the Base Case was better, as it resulted in higher HHV and H_2 concentration in the syngas compared to Case 4. Table 5 shows the comparison of the findings of this work with those of other studies, where this work managed to achieve high HHV, H_2 concentration and CGE.

Conclusions

In conclusion, a model of combined torrefaction and gasification using Aspen Plus for EFB was simulated. H_2 concentration increased with a decrease in ER and an increase in gasifier temperature in the Base Case, an increase in S/B ratio in Case 3 and without recycling of torrgas in Case 4. Of the four cases, the best was Case 3, which applied steam into the gasification reactor. It achieved the highest concentration of H_2 gas, of 59.25 mol % at 800 °C, the highest syngas ratio of 2.5 at the same temperature, and a maximum CGE of 84%. However, the highest HHV of 12.60 MJ/Nm^3 was achieved at an S/B ratio of 0.1. Further optimisation is required to obtain a balance between HHV, H_2 concentration, syngas ratio and CGE. Furthermore, for Case 2, a kinetic model of torrefaction should be introduced in order to investigate the effect of torrefaction medium concentration on H_2 concentration and the HHV of syngas. Work is in progress in this direction. The use of torrefaction in gasification for hydrogen production increases the efficiency of the system.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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