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Process Analysis of an Integrated Gasification and Methanol Synthesis Process for Bio-Methanol Production from Untreated and Torrefied Biomass

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The integrated biomass gasification and methanol synthesis process is investigated in this study. The different types of biomass i.e., the untreated and torrefied biomass at 250 $^{\circ}$ C (TB250) and torrefied biomass at 300 $^{\circ}$ C (TB300) are considered feedstock. The influence of torrefying temperature on the yield and composition of raw syngas derived gasifier is investigated. The biomass processed torrefaction leads to an increase in syngas and methanol yields. Moreover, the bio-methanol production process using torrefied biomass releases lower amount of CO₂ than the raw one. An energy analysis is also performed using overall energy consumption and cold gas efficiency (CGE) of the integrated process as the indicators. The TB300 offers better performance in methanol production and CO₂ emission. However, it requires high energy for methanol synthesis unit and offers low CGE.

1. Introduction

Energy production from biomass has been attracting a considerable amount of attention due to concern in energy security and global climate change. Methanol is an important chemical that can be used as a clean burning fuel for replacing the liquid fossil fuel without changing the existing infrastructures. It can also be used as intermediate for the synthesis of numerous chemicals. Generally, methanol is produced from syngas derived fossil fuels, i.e., partial oxidation of methane, steam reforming of natural gas, or gasification of coal (Zhang et al., 2010), which causes high CO₂ emission during the process. Therefore, the bio-methanol production from syngas through biomass gasification has attracted crescent interest, especially, in agricultural countries. However, the use of biomass for gasification requires large amount of biomass because it has low energy density which sequentially results in high transportation costs. The high moisture content of raw biomass is another a topic of concern because a high energy demand is required to evaporate the contained water (Siew Ng et al., 2011). To overcome these limitations, torrefaction, a thermal pretreatment process improving the energy density, gridability and hydrophobicity of biomass, is introduced. Torrefaction is carried out in absence of oxygen at a temperature range of 200-300 °C with a residence time from few minutes to several hours. The derived products are known as torgas, consisting of light gas (i.e., CO, CO₂, CH₄, and H₂) and other organic condensable compounds, and torrefied biomass which is the residual solid (Basu, 2013). Previously, most studies focused on the gas production from the gasification of raw biomass and torrefied biomass. Jamin et al. (2020) studied and compared the gasification performance of raw and torrefied wood wasted. They reported that the syngas yield, higher heating value (HHV), CGE and carbon conversion (CC) increased for both feedstocks when gasifying temperature increased. Tapasvi et al. (2015) found that the torrefied biomass gave higher H₂ and CO contents and higher cold gas energy and exergy efficiencies than untreated biomass. Kuo et al. (2014) compared the syngas production from raw and torrefied bamboo. They found that the syngas yield increased with torrefying temperature and the torrefied bamboo at 250 °C was the most feasible fuel. Muslim et al. (2017) studied the effect of gasifying condition on the syngas production from

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raw empty fruit branch (EFB) and torrefied EFB. They summarized that the gasification of torrefied biomass at high temperature could enhance the H_2 yield.

However, the study of bio-methanol production via an integrated gasification and methanol synthesis process using torrefied biomass is limited only on untreated biomass. Bio-methanol synthesis from syngas derived from pine biomass gasification was studied over different operating conditions. The offgas separated from methanol reactor was utilized as fuel for the combustion unit to reduce the requirement of a carbon source at the gasifier (Puig-Gamero et al., 2018). Im-orb et al. (2020) performed thermodynamic analysis to investigate the performance of bio-methanol production from untreated oil palm biomass residues (i.e., trunk, frond, and EFB). They found that the maximum yield of methanol was achieved at gasifying temperature of 750 °C and equivalent ratio (ER) of 0.25 when the trunk was a feedstock. Therefore, the potential of bio-methanol from torrefied biomass is examined in this study using a process model developed in Aspen Plus V.8.8. The untreated and torrefied bamboo at different torrefying temperatures of 250 °C (TB250) and 300 °C (TB300) are used as biomass model compound. The effect of torrefying temperature on the yield of raw syngas produced from gasifier and methanol as well as the CO₂ emission is firstly investigated. The energy analysis using overall energy consumption and cold gas efficiency (CGE) are also investigated.

2. Process modelling description

The integrated biomass gasification and methanol synthesis process for bio-methanol production consists of three main sections (i.e., gasification, syngas cleaning and conditioning, and methanol synthesis) as shown in Figure 1. Modelling of the integrated process was done in Aspen Plus V8.8 using the untreated biomass, TB250 and TB300 as biomass model compound which their ultimate and proximate analyses are presented in Table 1 (Kuo et al., 2014). The Aspen plus model flowsheet is shown in Figure 2.

2.1 Biomass gasification process

The biomass gasification model is a thermodynamic equilibrium model simulated based on the main assumptions as: (1) the process is performed under isothermal and steady state conditions, (2) pyrolysis is instantaneous, (3) char consists of only carbon, (4) ash is a non-reactive compound, and (5) tar consists of toluene, naphthalene, phenol, and pyrene. In the simulation, biomass was defined as a non-conventional component and the HCOALGEN and DCOALGEN models were used to determine the enthalpy and density of the solid biomass. Biomass was firstly converted to conventional component in RYIELD reactor (DECOMP) by identifying the yield distribution in the calculator block according to its ultimate and proximate analyses. The tar yield which was assumed to contain 65 wt% toluene, 20 wt% naphthalene, 10 wt% phenol, and 5 wt% pyrene (Sharma et al., 2017) was specified in RYIELD reactor (R-TAR). Oxygen-rich air was used as gasifying agent because it provided high concentration of major components in methanol synthesis. The gasification reactions were simulated using RGIBBS reactor (GASIF), in which the syngas composition was estimated using Gibbs free energy minimization method. The results of the developed gasification model were validated with those of published experiment (Lan et al., 2019) at the same operating conditions and the model results were match well with experimental data with a root mean square error (RMSE) of approximately 2.55% (Imorb et al., 2020). In this study, the operating conditions of gasification were controlled at gasifying temperature of 900 °C to prevent the operational problems from tar formation (Berry et al., 2017), the ER and biomass feed rate were set at 0.25 and 0.74 kg/h, respectively.

Biomass	Ultimate Analysis (%wt dry biomass)				Proximate Analysis (%wt dry biomass)				
	С	Ν	Н	0	S	FC	VM	Ash	HHV
Raw	48.64	0.52	5.64	44.09	0.03	15.28	83.57	1.15	18.94
TB 250	56.58	0.52	5.55	35.90	0.02	25.05	70.20	1.43	20.99
TB 300	69.56	0.12	4.77	23.6	0.00	48.47	49.52	2.01	27.23

Table 1: Ultimate and	proximate anal	vses of untreated	biomass. T	B250 and TB300



Figure 1: Simplified diagram of bio-methanol production process



Figure 2: Aspen plus model flowsheet of the Integrated gasification and methanol synthesis process

2.2 Syngas cleaning and conditioning process

2.2.1 The catalytic partial oxidation (CPOX) process

The CPOX process was used to reform methane and tars to syngas (H_2 and CO) via partial oxidations (Berry et al., 2017). The RTOIC reactor (REFORM) was used to simulate the CPOX process, and its operating condition was set to be similar to gasifying condition. The oxygen enriched air was supplied to the reactor until methane and tars were completely reformed.

2.2.2 Pressure swing adsorption (PSA) process

The PSA process was used to remove some amount of CO_2 until the ratio of syngas was achieved the feed gas specification of methanol synthesis (Eqs(1)-(2)) (Hernandez et al., 2016). PSA process consisted of three sub- units which simulated using SEP blocks (PSA1, PSA2 and PSA3) based on the real operating conditions of 35 °C, 3.03 MPa. The syngas was adjusted to these operating conditions through the multistage compressors (COMP1, COMP2 and COMP3). The separation percentage of considered components was specified based on actual plant data (Puig-Gamero et al., 2018). The pressure of syngas satisfying methanol synthesis specification from PSA process was increased through compressor (COMP4) to achieve the operating pressure of 5.07 MPa for methanol synthesis.

$$\frac{H_2 - CO_2}{CO + CO_2} \approx 2.1\tag{1}$$

$$\frac{H_2}{CO} = 2.4 - 2.5 \tag{2}$$

2.3 Bio-methanol synthesis process

The methanol reactor containing CuO-ZnO-AlO-based catalyst was simulated using REQUIL reactor (MEOHSYN). The derived product was calculated based on the chemical equilibriums of the reactions given in Eqs(3)-(5). It was noted that the accuracy of the model should be improved by using reaction kinetics for industrial scale up purpose. The operating condition was maintained at 220 °C and 50 atm to ensure that the catalysts were active, and the heat of reaction was effectively used (Rafael et al., 2018). The product from MEOHSYN reactor was cooled down to 25 °C via COOLER1 and the generated offgas was separated from raw bio-methanol at METSEP. The separated offgas was recycle to MEOHSYN to enhance the methanol yield.

$$CO + 2H_2 \leftrightarrow CH_3OH$$
 (3)

$$CO_2 + H_2 \leftrightarrow CO + H_2O$$
 (4)

 $CO_2 + 3H_2 \leftrightarrow CH_3OH + H_2O$

3. Energy analysis

The energy requirement of the overall methanol production process as well as individual unit using different types of biomass was investigated. The overall energy consumption was determined by sum of energy demanded in every single unit. The CGE of the system reveals the conversion efficiency of biomass to raw methanol was defined as a ratio of the LHV of the methanol product and biomass input (Eq(6)) Muslim et al. (2017).

$$CGE = \frac{m_{MEOH}LHV_{MEOH}}{m_{Biomass}LHV_{Biomass}}$$

(6)

4. Results and discussions

4.1 Effect of torrefying temperature on raw syngas production

Figure 3 shows the concentration of each component, yield and H_2/CO ratio of the raw syngas produced from gasification of raw biomass, TB250 and TB300. It indicates that syngas yield increased as torrefying temperature increased. The concentration of H_2 of raw syngas deriving from each biomass was not different, whereas that of CO from TB300 showed the highest value followed by TB250 and untreated biomass due to its high carbon content. As a result, the H_2/CO ratio of raw TB300-syngas exhibited the highest value. Moreover, the concentration of CO_2 was found to decrease as torrefying temperature increases. However, the yield of TB300-syngas after cleaning and conditioning to satisfy the specification of methanol synthesis decreased and showed the lowest value compared to others (Figure 4). The concentration of H_2 and CO manifested the same trend as those of raw syngas.

4.2 Performance comparison of methanol production from untreated and torrefied biomass

Methanol production, CO_2 emission, overall energy consumption and CGE of the integrated biomass gasification and methanol synthesis process is summarized in Table 2. Although the TB300 offered the lowest amount of feed gas for methanol synthesis, it provided the highest yield of bio-methanol due to the high concentration of H₂ and CO in feed gas. Moreover, the use of TB300 also had the least environmental impact based on CO_2 emission. Regarding the energy analysis, the integrated biomass gasification and methanol synthesis process was an exothermic process. The use of TB250 led to the highest energy consumption because it produced largest amount of feed gas for bio-methanol synthesis, hence, the highest energy was required at compressor 4 and methanol reactor (Table 3). For the CGE, TB300 had a higher HHV than TB250 and untreated biomass, therefore, it had the lowest CGE of approximately 28 % followed by TB250 and untreated biomass, respectively.



Figure 3: Composition, yield, and H_2/CO ratio of syngas derived from gasifier



Figure 4: Composition, yield, and H_2 , CO_2 and CO ratio of methanol synthesis feed gas derived from gas cleaning and conditioning processes

Table 2: Performance comparison of methanol production from untreated and torrefied biomass

Performance indicator	Raw	TB250	TB300
Bio-methanol production (kmol/h)	0.00702	0.00781	0.00843
CO ₂ emission (kmol/h)	0.00686	0.00475	0.00057
Overall energy consumption (KW)	3.25863	3.66957	3.50999
CGE (%)	35.06	34.60	27.99

Table 3: Energy consumption of each operating unit in methanol production process from untreated and torrefied biomass

Performance indicator	Raw	TB250	TB300
Gasifying temperature (°C)	900	900	900
Equivalent ratio (ER)	0.25	0.25	0.25
Biomass feed rate (kg/h)	0.74	0.74	0.74
Methanol reactor temperature (°C)	220	220	220
Methanol reactor pressure (MPa)	1.013	1.013 1.013	
Energy consumption (KW)			
Gasifier	0.1574	0.2405	0.1771
CPOX	-1.4168	-1.2103	-0.8331
Compressor 1	0.5816	0.6388	0.6813
PSA1	0.0014	0.0013	0.0010
Compressor 2	0.1106	0.1329	0.1577
PSA2	0.0005	0.0004	0.0000
PSA3	0.0002	0.0002	0.0001
Compressor 3	0.0865	0.1184	0.1504
Compressor 4	0.3811	0.4242	0.2890
MeOH reactor	0.4604	0.5126	0.4468
Cooler1	0.1804	0.2006	0.1534
Heater1	-0.0018	-0.0018	-0.0014

5. Conclusions

The performance of the integrated biomass gasification and methanol synthesis process for bio-methanol production was investigated using the model developed in Aspen Plus V.8.8. The untreated biomass, TB250, and TB300 were used as feedstock. The yield of raw syngas leaving gasifier which controlled the gasifying

condition at 900 °C, 1.013 MPa and ER at 2.50 increased as torrefying temperature increased. The TB300 offers the highest yield raw syngas but the opposite trend was observed when the raw syngas was cleaned and conditioned to meet the specification of methanol synthesis. However, the use of TB300 still offered the highest yield of methanol due to the high concentration of H₂ and CO in the feed gas. The energy analysis indicated that the use of TB250 required the largest energy and the use of TB300 offered the lowest CGE. Regarding an environmental impact, the bio-methanol production from TB300 showed the best performance.

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