Exergy Analysis of Hydrogen-Rich Syngas Production System Utilizing Palm Oil Empty Fruit Bunch using Pyrolysis Connected with Steam Methane Reformer

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ARTICLE INFO	ABSTRACT
       	Fossil fuel consumption on a large scale continues to increase yearly, causing a shortage of energy sources. Hydrogen can help to overcome the shortage of energy sources. It can be produced from renewable energy sources, such as oil palm Empty Fruit Bunch (EFB) as biomass using pyrolysis processes equipped with steam methane reformer and water gas shift reactions. This research was conducted to determine the amount of hydrogen that can be produced and to analyze the exergy of the existing systems in the hydrogen production process. Aspen Plus has been used to design and analyze pyrolysis processes, steam methane reform, and water gas shift reactions. The results showed that when 6.54 kg Empty Fruit Bunch was used as raw material in a pyrolysis reactor, hydrogen could be produced in the amounts of 0.435 kg. The exergy values of raw materials and fuels are relatively high because the chemical exergy values of the compounds contained therein are also high. Based on the calculations, it is found that the combustion process is the process that experiences the most significant exergy destruction. The exergy efficiency of this hydrogen production process system is 22.77%.

1. Introduction

Fossil fuels contributed 33.05% of world energy consumption in 2019, followed by natural gas and coal with 24.2% and 27.3%, respectively. With consumption of up to 97.6 million barrels per day in 2018 and a projected consumption increase of 112.2 million barrels per day in 2035, fossil fuels and their derivatives continue to be the world's most exploited source of energy [1]. Meanwhile, with the development of industrialization, fossil fuel consumption has massively increased yearly, causing a shortage of traditional energy sources [2]. In addition, fossil fuel-based energy impacts the environment due to greenhouse gas emissions. So, there is an urgent need to develop carbon-neutral alternative fuels [3]. Decarbonization will require large amounts of renewable liquid fuels and gases

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to meet energy needs. In recent years, the demand for renewable fuel has demonstrated the need to increase global renewable fuel production capacity [4].

Hydrogen, produced from low-emission feedstocks, can be vital in decarbonizing the energy sector [5]. Hydrogen can be used in transportation, households, commerce, chemicals, heavy industry, and electricity [6]. Hydrogen production from renewable sources is highly recommended because the process releases carbon dioxide gas, which plants use for photosynthesis [1]. When compared to hydrogen produced from fossil fuels, hydrogen produced from biomass is more environmentally friendly [7]. Biomass is gaining popularity because it is a renewable, abundantly available, and carbon-neutral energy source. In addition, environmental problems caused by the disposal of agricultural waste can be managed efficiently through biomass utilization. Thus, this biomass is quite promising to be used as a raw material resource for hydrogen production [8,9].

Indonesia has a potential biomass supply of around 146.70 Mt/year, including lignocellulosic biomass (such as rice straw, sugarcane stalks, and palm oil residue), municipal solid waste, and industrial waste. This biomass has an energy potential of 31,461 MWe, of which 12,660 MWe comes from oil palm biomass [10]. This large biomass from the palm oil mill waste is considered a potential renewable energy resource and can be converted into new alternative energy resources through pyrolysis [11-16]. Biomass waste from palm oil processing plants is still a problem for the environment and public health due to insufficient utilization [17]. One way to handle biomass from palm oil mill waste is to convert it into power, heat, and fuel by the thermochemical conversion process, such as pyrolysis [18]. Biomass pyrolysis, typically operating at temperatures of 300 – 500 °C, can be used to produce syngas and hydrogen due to its carbon-free feature [19]. Depending on the facility, the cost of producing H_2 from biomass pyrolysis is between US\$8.86 and \$15.52 per GJ. Pyrolysis-based technology offers the best economics due to the possibility of co-products. An additional benefit of biomass as a renewable fuel source is that it is not sporadic but can be used to generate H_2 as needed [20-22]. The palm oil wasters fixed bed pyrolysis reactor with a working temperature of 500 °C – 900 °C through catalyst presence of K_2CO_3, Na_2CO_3, $CaMg(CO_3)_2$, Ni, γ-Al_2O_3, La/Al_2O_3, and Fe_2O_3 produced H_2-rich gas from 3.56 to 33.49 vol% (equivalent to 10.40 g/Kg). The presence of 5 wt% of Ni catalyst contributes to an increase in the gas yield of up to 29.78 g/Kg [23,24].

As the authors know, few researchers have reported on a biomass pyrolysis system integrated directly with a steam methane reformer and water gas shift reactor to convert biomass into hydrogen-rich syngas. Arregi *et al.,* [25] analyzed and proved that pyrolysis with in-line steam reforming conducted in a conical spouted bed-fluidized bed configuration is suitable and flexible for producing H_2 from agroforestry residues. Tabat *et al.,* [26] analyzed a proposed conceptual mobile auto thermal methane pyrolysis unit for onsite hydrogen production with a levelized cost of hydrogen (LCOH) range from 1.3 to 1.47 US\$/kg, and the net present value (NPV) was in the range of 3.76-4.35 M.US\$. Due to the enormous potential of biomass from palm oil mill waste in Indonesia, it is necessary to develop H_2 production by utilizing the palm oil empty fruit bunch (EFB) using pyrolysis connected with a steam methane reformer and water gas shift reactor. The literature review conducted by the authors found that there is still limited information related to the thermodynamic performance analysis of the hydrogen production system through biomass pyrolysis. This study aims to develop processes using Aspen Plus simulation and analyze the system performance by involving exergy analysis during converting EFB into hydrogen-rich syngas.

2. System Simulation Description

Aspen Plus contains a collection of mathematical models for various types of chemical process equipment, such as heat exchangers, pumps, compressors, turbines, distillation units, absorbers, and

chemical reactors [27,28]. In this simulation, Peng-Robinson methods (PENG-ROB), typically used in hydrocarbon processing applications, have been used to calculate all model properties [29,30].

The biomass pyrolysis in line with SMR and WGS model flowsheet to convert EFB hydrogen-rich syngas is shown in Figure 1. The system process flow consists of a biomass combustion unit, pyrolysis reactor (YLD-PYR, RC-PYRLY, and HRSG), steam methane reformer (SMR), water gas shift reactor (WGS), condenser, and steam generator for steam methane reaction. Generally, the process will start by burning solid biomass from dry wood as fuel in the combustion chamber (COMB-CHA). Air for combustion is supplied by a blower (BLOWER). The air required for combustion will be analyzed using the air-fuel ratio formula. The heat released by this combustion (stream 4) will be directly supplied to the closed pyrolysis reactor (RC-PYRLY), which only contains EFB. Syngas produced from the pyrolysis reactor (stream 9) will go to the steam methane reformer (SMR) before entering the water gas shift reactor (WGS). The hydrogen-rich syngas (stream 13) will enter the condenser (CNDSR) and then be separated into noncondensable hydrogen-rich syngas (GAS) and bio-oil (LIQUID). The steam that will be used for the SMR process is obtained from combustion chamber heat by considering that the steam-carbon ratio is equal to 2.
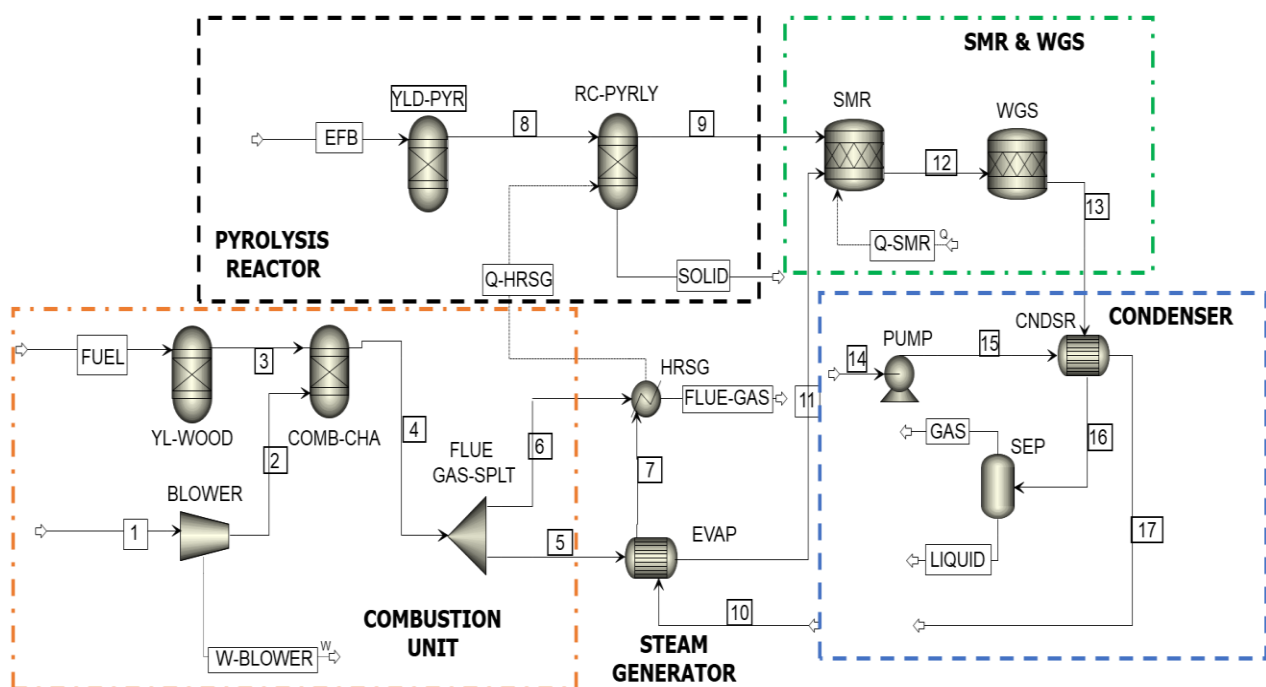

Fig. 1. The biomass pyrolysis in line with SMR and WGS flowsheet to convert EFB into hydrogen-rich syngas

2.1 Combustion Processes

The combustion process consists of three components (YL-WOOD, COMB-CHA, and BLOWER). Wood is defined as a non-conventional component. The wood is first sent to the RYield reactor (YL-WOOD) to decompose into conventional elements (C, H, O, N, S, Ash, and Moisture). RGibbs reactor (COMB-CHA) is used to simulate the combustion of wood. The decomposed element will react with air in the RGibbs reactor (COMB-CHA), producing a certain amount of heat (stream 4). The air-fuel ratio for the combustion processes (stream 1) can be obtained using Eq. (1) [31].

$$\left(A/F\right) = 11.5Y_C + 34.48Y_H + 4.31Y_S + 9.85Y_N - 4.31Y_O \tag{1}$$

where, Y_C, Y_H, Y_S, Y_N, and Y_O are the mass fractions of carbon, hydrogen, sulfur, nitrogen, and oxygen in the fuel, respectively. The lower heating value of solid fuel (LHV) can be analyzed using Eq. (2) [32].

$$LHV = HHV - h_g \left(\frac{9H}{100} - \frac{M}{100} \right) \qquad (2)$$

where HHV is the higher heating value (kJ/kg), h_g is the latent heat of water evaporation (2260 kJ/kg), H is the percentage of hydrogen (%), and M is moisture percentage (%).

2.2 Steam Methane Reforming (SMR) and Water Gas Shift Reaction (WGS)

The SMR will be simulated using the Rstoic component (SMR). The syngas from pyrolysis, which contains many elements such as methane, carbon monoxide, and carbon dioxide, will be reacted with steam with the help of a catalyst in SMR and water-gas shift reaction (WGS) by following reactions as shown in Eq. (3) and Eq. (4) [31,33,34]. In this study, only the occurring reactions are used without involving detail on specific catalysts, with fractional conversion assumed to be 1.

Steam Methane Reforming: $CH_4 + H_2O \leftrightarrow CO + 3H_2$ $\qquad$ (3)

Water Gas Shift: $CO + H_2O \leftrightarrow CO_2 + H_2$ $\qquad$ (4)
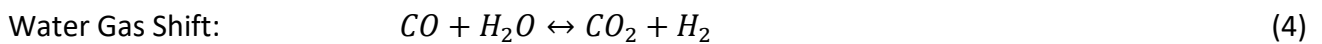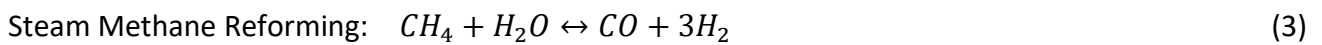
3. Methodology
3.1 Data Input

The materials used in the simulation are the local palm oil mill in North Sumatra, Indonesia, which has the properties seen in Table 1.

Table 1
Properties of EFB, Biochar, and Wood

	EFB	Biochar	Wood [13]
Proximate Analysis (%)			
Volatile Matter	67.58	12.72	63.46
Ash	7.26	17.20	10.76
Moisture	8.66	12.47	9.36
Fixed Carbon	16.5	57.61	16.43
Ultimate Analysis (%)			
Carbon	44.08	61.15	42.43
Hydrogen	6.40	3.74	5.49
Oxygen	41.08	16.86	51.1
Nitrogen	1.03	0.93	0.87
Sulfur	0.15	0.12	0.11
Ash	7.26	17.20	10.76
Higher Heating Value (kJ/kg)	17,777.82	22,945.06	16,151.20
Lower Heating Value (kJ/kg)*	16,671.78	22,466.17	15,246.07

*Using Eq. (2) to determine LHV

The operating parameters for the combustion chamber, blower, water gas shift (WGS) unit, and condenser are obtained from experimental research in a laboratory. Meanwhile, the operating parameters, especially for the SMR and WGS working temperature, are obtained from a literature review. The data that has been collected from both experimental and literature reviews are

combined. The operating parameters and other main assumptions for this simulation can be seen in Table 2. The dead state temperature in this simulation is 32°C with a pressure of 1 atm.

Table 2
Operational Conditions and Assumption for Design Simulation

Combustion Chamber (COMB-CHA)	
Temperature (°C)	1,052
Pressure (bar)	1.015
Wood flow rate (fuel) (kg/hr)	25
Air flow rate (kg/hr)	116.5
Blower (BLOWER)	
Discharge Pressure (bar)	1.015
Pyrolysis Reactor (RC-PYRLY)	
Temperature (°C)	500
Pressure (bar)	1.61
EFB (raw material) (kg/hr)	6.54
Steam Methane Reform Reactor (SMR)	
Temperature (°C) [28]	350
Steam flow rate (kmol/hr)	0.08
Water Shift Reactor (WGS)	
Temperature (°C) [28]	200
Condensor (CNDSR)	
Cooling Water (Shell)	
Temperature (°C)	26.3
Pressure (bar)	2.01
Mass Flow (kg/hr)	150
Cooled Syngas Temperature (Tube) (°C)	32.3

3.2 The Exergy Analysis

Exergy is the maximum amount of work a system can produce to undergo a reversible process from a specified initial state to its environmental state [35,36]. The exergy can determine the plants' magnitudes, locations, and causes of irreversible. Therefore, it can be a helpful tool in designing, evaluating, optimizing, and improving heat exergy, which can be determined using Eq. (5).

$$X_{heat} = \left(1 - \frac{T_0}{T}\right)\dot{Q} \tag{5}$$

where T_0 is the dead state temperature of the system (°C), T is the operating condition temperature (°C), and $\dot{Q}$ is the heat flow rate (kW). The work exergy can be determined by using Eq. (6).

$$X_{work} = W_u \tag{6}$$

where W_u is the actual amount of work produced by the system (kW). The physical exergy can be determined by using Eq. (7).

$$X_{mass} = \dot{m}[(h - h_0) + T_0(s - s_0)] \tag{7}$$

where $\dot{m}$ is the mass flow rate (kg/hr), h_0 is dead state enthalpy of a system (kJ/kg), h is the enthalpy of a stream (kJ/kg), s_0 is dead state entropy of the system (kJ/kg.K), and s is the entropy of the stream (kJ/kg.K) [35].

The chemical exergy can be determined by using Eq. (8).

$$X_{CH} = \bar{m}\left(\sum_{i=1}^{f} \bar{y_i}\bar{e_i}^{CH} + RT_0 \sum_{i=1}^{f} \bar{y_i} \ln \bar{y_i}\right) \tag{8}$$

where $\bar{m}$ is molar flow rate (kmol/hr), $\bar{y_i}$ is the molar fraction of the component, $\bar{e_i}^{CH}$ is the chemical exergy of the component shown in Table 3, and R is the ideal gas constant (8.3145 kJ/kmol K) [37].

Table 3
Chemical Exergy of Component at 25°C, 1 atm

Component	$\bar{e}^{CH}$ (kJ/mole)
N_2	720
O_2	3,970
CO_2	19,870
H_2O	900
C	410,260
H_2	236,100
S	609,600
CO	275,100
CH_4	831,650

For solid fuel (Coal, Biomass), the chemical exergy can be determined using Eq. (9).

$$X_{Solid} = m_{Solid} \times LHV \times \left(1.0064 + 0.1519\frac{Y_H}{Y_C} + 0.616\frac{Y_O}{Y_C} + 0.0429\frac{Y_N}{Y_C}\right) \tag{9}$$

where m_{Solid} is fuel mass flow (kg/hr), LHV is lower heating value (kJ/kg) [38].

The amount of exergy destroyed in the system can be determined by using Eq. (10).

$$X_{destroyed} = X_{in} - X_{out} \tag{10}$$

where X_{in} is the total amount of exergy entering the system, and X_{out} is the total amount of exergy leaving the system.

The Exergy Efficiency of the entire process can be obtained using Eq. (11).

$$\eta_{II} = \frac{Exergy\ input}{Exergy\ output} \tag{11}$$

where exergy recovered is the total amount of exergy leaving the process, and exergy expended is the total amount of exergy entering the process [33,34].

4. Results
4.1 Syngas Composition

The process flow in the biomass pyrolysis system connected to SMR and WGS, as shown in Figure 1, is analyzed. Syngas produced from the pyrolysis process at a working temperature of 500 °C contains components in the form of 38.17% H_2O, 22.47% H_2, 0.94% N_2, 0.12% S, 16.85% CH_4, 19.26% CO_2, and 2.19% CO as shown in Stream 9 Table 4. Then, the syngas from the pyrolysis reactor undergoes a reaction with H_2O (steam) in the SMR reactor and produces H_2 at a higher level, thereby increasing the molar composition of H_2 in the syngas to 43.06%, as seen from stream 12. The

difference in composition of the syngas after having pyrolysis, SMR, and WGS processes is shown in Figure 2. The amount of H_2 increased to 54.29% of the total syngas composition after leaving the WGS unit, while the remainder was 22.5% H_2O, 22.6% CO_2, 0.1% S, and 0.6% N_2. A further application is needed to separate H_2 from other syngas components, or it can be used directly as fuel in internal combustion engines, which is not discussed in this paper.

Table 4
Temperature, Pressure, and Molar Composition of Stream

Stream	T	P	ṁ	Molar Composition (%)								
	(°C)	(bar)	Kg/hr	H_2O	C	H_2	N_2	S	O_2	CH_4	CO_2	CO
1	32	1.013	116.5	0	0	0	79	0	21	0	0	0
2	32.2	1.015	116.5	0	0	0	79	0	21	0	0	0
3	32	1.015	25	Wood, as shown in Table 1								
4	1052	1.015	141.5	15.41	0	0	65.97	0.02	2.08	0	16.52	0
5	1052	1.015	70.75	15.41	0	0	65.97	0.02	2.08	0	16.52	0
6	1052	1.015	70.75	15.41	0	0	65.97	0.02	2.08	0	16.52	0
7	1010	1.015	70.75	15.41	0	0	65.97	0.02	2.08	0	16.52	0
8	32	1.613	6.54	EFB, as shown in Table 1								
9	500	1.613	4.55	38.17	0	22.47	0.94	0.12	0	16.85	19.26	2.19
10	35.1	2.013	1.515	100	0	0	0	0	0	0	0	0
11	122	2.013	1.515	100	0	0	0	0	0	0	0	0
12	350	1.613	6.07	33.73	0	43.06	0.55	0.07	0	0	11.35	11.23
13	200	1.613	6.07	22.51	0	54.29	0.55	0.07	0	0	22.58	0
14	26.3	1.013	150	100	0	0	0	0	0	0	0	0
15	26.4	2.013	150	100	0	0	0	0	0	0	0	0
16	32.3	1.613	6.07	22.51	0	54.29	0.55	0.07	0	0	22.58	0
17	35.1	2.013	150	100	0	0	0	0	0	0	0	0
FLUE-GAS	147.3	1.015	141.5	15.38	0	0	67.77	0.02	0.36	0	16.48	0
GAS	32.3	1.613	4.59	2.51	0	68.36	0.70	0	0	0	28.43	0
LIQUID	32.3	1.613	1.48	99.66	0	0	0	0.34	0	0	0	0
SOLID	500	1.613	1.99	0	100	0	0	0	0	0	0	0

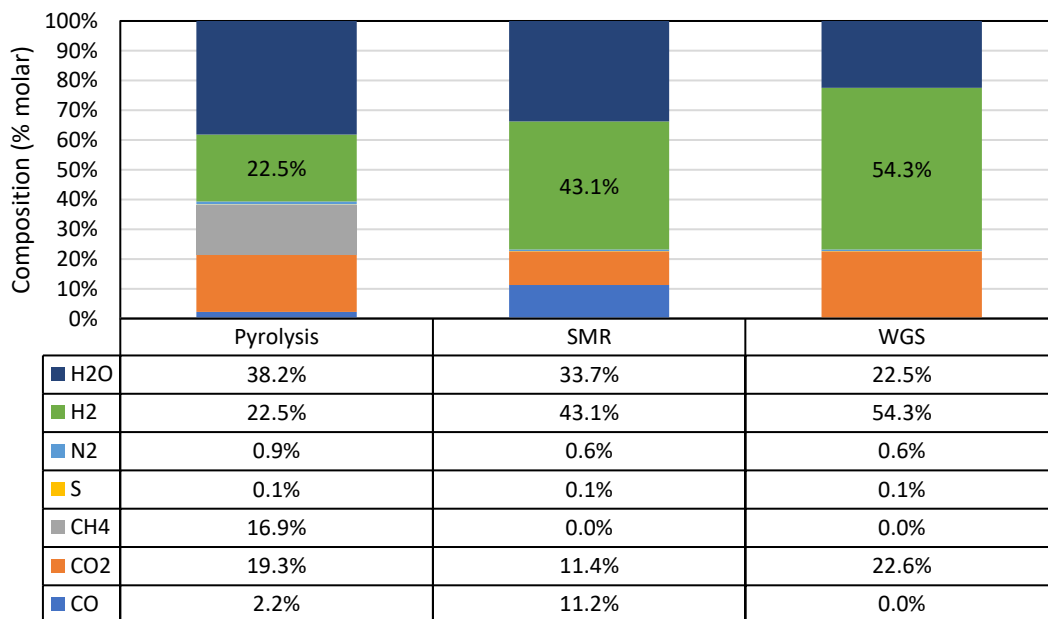|	Pyrolysis	SMR	WGS
■ H2O	38.2%	33.7%	22.5%
■ H2	22.5%	43.1%	54.3%
■ N2	0.9%	0.6%	0.6%
■ S	0.1%	0.1%	0.1%
■ CH4	16.9%	0.0%	0.0%
■ CO2	19.3%	11.4%	22.6%
■ CO	2.2%	11.2%	0.0%

Fig. 2. Syngas composition (% molar) out from pyrolysis reactor, SMR, and WGS units

4.2 Exergy of Stream

Table 5 shows the flow exergy based on Aspen Plus simulation results. In the combustion process, air (stream 1) enters with an exergy of 0.111 kW and exits the blower with an exergy of 0.114 kW (stream 2), where the compressor uses a working exergy of 0.0052 kW (W-BLOWER flow). The physical exergy of stream 1 is 0 kW because the temperature of the air entering through the blower is the same as the ambient temperature. Biomass fuel in the form of wood, which is used to produce heat through combustion (stream 3), enters with an exergy of 116.5817 kW and produces flue gas with an exergy of 30.8681 kW (stream 4). Meanwhile, the EFB has an exergy value of 32.9179 kW and is put into the pyrolysis reactor. Stream 9 and SOLID are the outputs of the pyrolysis process, with exergy values for each stream of 13.697 kW and 14.877 kW. The details of the exergy values for each flow can be seen in Table 5.

Table 5
Exergy value of stream

Stream of Material			
Stream	Physical exergy (kW)	Chemical Exergy (kW)	Total exergy (kW)
1	0.0000	0.1105	0.1105
2	0.0038	0.1105	0.1142
3	-	116.5817	116.5817
4	28.5983	2.2698	30.8681
5	14.2991	1.1349	15.4341
6	14.2991	1.1349	15.4341
7	13.4356	1.1349	14.5705
8	-	32.9179	32.9179
9	0.6495	13.0472	13.6968
10	0.0001	0.0210	0.0211
11	0.2401	0.0210	0.2612
12	0.6346	14.6118	15.2464
13	0.3211	14.4247	14.7459
14	0.0102	2.0816	2.0917
15	0.0149	2.0816	2.0965
16	0.1041	14.4247	14.5288
17	0.0078	2.0816	2.0894
FLUE-GAS	1.1129	2.2698	3.3828
GAS	0.1034	14.4819	14.5853
LIQUID	0.0000	0.0664	0.0664
SOLID	0.1206	14.7568	14.8774
Stream of Heat			
Stream		Heat Exergy (kW)	
Q-HRSG		40.4039	
Q-SMR		1.9830	
Stream of Work			
Stream		Work Exergy (kW)	
W-BLOWER		0.0052	

In the SMR process, streams 9 and 11 enter the SMR reactor with 13.697 kW and 0.261 kW, respectively. The process was then accompanied by the influx of exergy heat indicated by the stream Q-SMR flow with a value of 1.9830 kW. Stream 12 is an SMR product with an exergy of 15.25 kW, where this stream will also enter the WGS reactor. Stream 13 output from WGS has an exergy of 14.7459 kW. In the syngas cooling process, the hydrogen-rich syngas, indicated by stream 13, enters the condenser. Then, it will come out of the condenser (stream 16) with an exergy of 14.53 kW. The

condenser cooling water in flow 15 has an exergy of 2.096 kW, and the cooling water coming out of the condenser in flow 17 has an exergy of 2.089 kW.

4.3 Exergy Destroyed in Production Process

Based on the analysis, the exergy destroyed in the biomass combustion process in the combustion chamber (COMB-CHA) is 85.8278 kW, resulting in an exergy efficiency of 26.5%. The total exergy entering the combustion chamber is 116.6959 kW, the sum of the exergy in biomass wood (stream 3) and air for combustion (stream 2). Furthermore, the outflow is in the form of combustion flue gas (stream 4) with an exergy of 30.8681 kW. Meanwhile, in distributing combustion air through a blower unit (BLOWER), the destruction exergy value was 0.0014 kW, resulting in an exergy efficiency of 98.7%. On the other hand, the incoming exergy is 0.1157 kW, which is obtained from input (stream 1) and the work required by the boiler (W-BLOWER). Stream 2 is the flow leaving the blower with an exergy of 0.1142 kW.

During the pyrolysis process in the reactor (RC-PYRLY), the exergy destroyed was 44.7476 kW with an exergy efficiency of 39.0%. The total exergy entering the pyrolysis reactor is 73.3218 kW from the stream of 8 and the heat flow (stream of Q-HRSG). The total exergy that comes out of the reactor is 28.5741 kW, which comes from the sum of the exergy in the stream of 9 and stream of SOLID.

The exergy destroyed in the steam methane reform process (SMR reactor) is 0.6946 kW, resulting in an exergy efficiency of 95.6% for the unit. Meanwhile, the exergy destroyed during the water gas shift (WGS) process is 0.5005 kW. Thus, the WGS unit's exergy efficiency is 96.7%. The condensation process in the condenser unit (CNDSR) destroys 0.2241 kW of exergy, resulting in an exergy efficiency of around 98.7%.
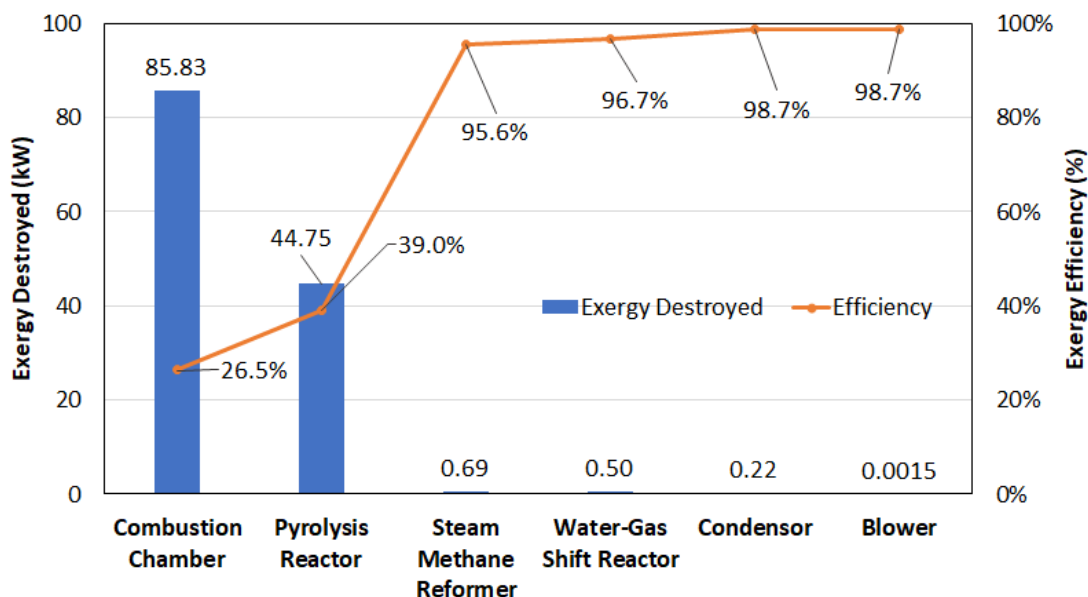

Fig. 3. Exergy Destroyed Value and Efficiency of Every Component

Based on the analysis, it was found that the most significant exergy destruction of the system occurred in the biomass burning process, followed later by the process that occurred in the pyrolysis unit, as shown in Figure 3. Utilizing wasted heat through a flue gas during biomass combustion is necessary to reduce destroyed exergy. Overall, the exergy efficiency of the system is in the range of 22.77%, so it can be said that it is still inefficient. Several things are needed related to the utilization of exhaust gas temperature or waste heat from the combustion chamber into electrical energy

through an organic Rankine cycle scheme or conversion of heat energy into energy in other forms to increase the exergy efficiency value of the system. Utilizing waste heat will increase system efficiency and minimize energy waste in the form of waste heat from the system.

5. Conclusions

The process of converting EFB biomass into hydrogen-rich syngas using pyrolysis techniques was carried out using the Aspen Plus simulation. The conversion process also applies exergy analysis during the conversion of EFB to hydrogen-rich syngas to get an idea of the steps that must be taken in the future to improve system performance. In general, based on the simulation results, it is known that 6.54 kg of EFB can be converted into 4.55 kg of syngas by pyrolysis, and with the help of the SMR and WGS processes, 6.07 kg of hydrogen-rich syngas is produced. By applying the second law of thermodynamics, namely exergy analysis, it was found that the most significant exergy destruction occurred in the combustion process, followed by the pyrolysis process, the SMR process, the WGS process, and the condensation process. The exergy efficiency of this EFB biomass conversion system is around 22.77%. Utilizing waste heat will increase system efficiency and minimize energy waste in the form of waste heat from the system. One of the system efficiency improvements that is being implemented is building an ORC unit design that is integrated with the hydrogen-rich syngas production system.

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