Exergy Analysis of Microalgae Thermochemical Conversion using Aspen Plus Simulation

by Arif Hidayat

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Abstract

Microalgae is known as the future bioenergy resources due to its unlimited potential and availability. One of the numerous paths to acquire an energy source is gasification, which produce syngas and methane as a hydrocarbon fuel or feedstock product. To set up an efficient gasification plant, several essential information is needed including the effect of oxidizing agent and steam to carbon (S/C) ratio to energy efficiency on certain biomass properties. This paper aims to study the highest exergy possibility on microalgae gasification process by examining the effect of steam and air flowrate independently via ASPEN Plus simulation. The result was validated with experimental data to verify the simulation reliability. It was found that the thermodynamic based simulation is suitable to predict the reactor behavior and acquire an optimum operating condition.

INTRODUCTION
Thermochemical conversion

The existence of renewable energy was claimed to succesfully improve the world energy growth within the last few decades. In the future prediction, renewable energy will cause a massive emission reduction, has a rapid market shares growth, and succesfully reduce the global atmospheric temperature (Gielen *et al.,* 2019). However, fossil fuel still offers much attractivenes currently since it is cheaper and has less compatibility issues. Renewable energy still has to face the conversion efficiency problem in order to reach a competitive point (Hassan and Kalam, 2013). This challenge especially lies on biomass thermochemical processing such as direct combustion, pyrolysis, gasification, and liquefaction which produce gaseous fuel, liquid hydrocarbon, and carbon solid fuel.

Biomass wet thermochemical conversion or commonly known as gasification is a process which converts hydrocarbons contained feed to primarily syngas, small amount of biofuel and solid char in the presence of water as gasification agent (Ruan *et al.,* 2019). During the gasification process, several reactions occurred, starting with drying, devolatilization, cracking, reforming and oxidation. Drying process takes place until the temperature is slightly higher than water evaporation temperature due

to the diffusion resistance. Then, it is followed by devolatilization process of solid biomass which includes the conversion and the release of volatile matters. Later, high temperature process embark the steam-methane reforming (SMR), water-gas shift (WGS) reaction and cracking by cutting long carbon chain and shifting carbon element. The heat required for the previous process was supplied from homogeneous and heterogeneus oxidation involving all flammable gases and solid carbon (Speight, 2015, Glassman and Yetter, 2008).

These reactions take place from early heating process until final gasification step which usually performed on moderate temperature range around 600-900°C to provide better gaseous product quality and quantity. Hence, the gasification process consumes large amount of energy. Moreover, water consumption become serious issue lately so that the process with high efficiency should be achieved and make it more profitable. Several economic studies already examined that profitable prices in steam gasification would be obtained from a high temperature biorefinery plant operated above 1,000°C, and proper operating control that favors minimal tar formation (Brown et al., 2009, Swanson et al., 2010).

Microalgae gasification

Algae became an interesting topic to be discovered in many research areas. Beside containing valuable compounds, the availablity and high growth rate become a major reason to be explored. It can be used for an alternative resource of food, pharmacy, pigment and renewable fuel (Plaza et al., 2008). Algae based fuel could be a promising product with a right processing path, starting with fine chemical extraction and later using the thermochemical processing to convert the solid residue for fuels.

One of suitable method to convert the algae to fuel is gasification process. Algae gasification is considered as a combination of second and third bioenergy generation, i.e. thermochemical process and algae utilization (Lee et al., 2019). Until now, algae gasification process has been widely investigated to identify the suitable operating condition and catalyst. The gasification process involving Fe catalyst succesfully enhanced hydrogen production and tar degradation up to 80-100% for macro algae and 53-70% for micro algae (Duman et al., 2014). A supercritical water gasification of several algae gives a better hydrogen production with the presence of alkali. On other hand, an addition of nickle catalyst succesfully cut down the production of tar which contains stable aromatic compounds and heterocyclic nitrogen compounds (Onwudili et al., 2013). A low temperature catalytic gasification on micro algae was proposed to be combined with nitrogen cycle in form of ammonia as a nutrient source for algae cultivation process (Tsukahara and Sawayama, 2005). However, no certain catalyst that was stated able to make the overall gasification process become more economic.

Microalgae gasification plant is classified as an unstable process since the product quality and quantity are highly affected by reactor operating conditions including heating rate, temperature approach, pressure, feed composition, catalyst presence and reactor design. Herewith, a method to determine the basic feasibility should be provided.

Exergy analysis

Exergy was introduced as a concept to evaluate the system efficiency based on practical thermodynamics variables (Taheri et al., 2014). Several studies use the exergy analysis to assest the performance of thermochemical reactor. For example, gasification of sawdust wood in a single model reactor at temperature range of 1,000-1,500 K produce 200-700 MW exergy and 100-450 MW destroyed exergy with 51-63% yield of H_2 (Abuadala et al., 2010). Another exergy analysis was also performed on rice hulk torrefaction in a bubbling fluidize bed giving a maximum 30% exergy efficiency at 250°C operating temperature (Manatura et al., 2017). In other hand, 70% exergy efficiency succesfully achieved from combined gasification and syngas chemical looping (SCL) of 350 ton/h black liquor at 800°C gasification and 930-1,000°C chemical looping temperature (Darmawan et al., 2018). However, the study of exergy analysis on micro algae is still limited.

In this study, the exergy of product and reactant were calculated from several variables generated by ASPEN simulation to determine the exergy efficiency. By addressing the exergy properties of product, the gasification of micro algae can be evaluated, so that the reactor performance can be improved. This research is important to determine the input parameter which affecting the product distribution and final exergy efficiency.

Exergy efficiency (η_ε) was defined as a comparison between exergy of output stream ($\varepsilon_{(out)}$, kJ/s) and input stream ($\varepsilon_{(in)}$, kJ/s) of the whole process (Eq. 1) (Demirel, 2014). By giving the assumption of minimal potential and kinetics exergy, the exergy contained at the stream ($\varepsilon_{(st)}$, kJ/s) is defined as an internal exergy calculated from combination of physical exergy ($\varepsilon_{(ph)}$, kJ/mol) and chemical exergy ($\varepsilon_{(ch)}$, kJ/mol), multiplied by flowrate (F, mol/s) (Eq. 2). The physical exergy is defined from thermodynamic properties by differentiating the enthalpy (H, kJ/mol) and entropy (S, kJ/mol.K) to the temperature reference point (H^0, T^0, S^0) (Eq. 3) (Chen et al., 2018). While chemical exergy represents the sigma of molar speciess exergy i ($\varepsilon_{(sp,i)}$, kJ/mol) and mol fraction (y_i, %) added with gas constant (R, kJ/mol.K) adjustment (Eq. 4) (Szargut et al., 1987). Speciess chemical exergy of algae biomass and bio oil is predicted by calculating a hydrocarbon constant (β) (Eq. 6 & Eq. 7) from the fraction of carbon (x_C, %), hydrogen (x_H, %), oxygen (x_O, %) and nitrogen (x_N, %) with A-J is equation constants studied from the reference, deducting the approximation of low heating

value (LHV) (Eq. 5) (Rahbari *et al.,* 2018, Stepanov, 1995).

$$\eta_\varepsilon = \frac{\sum \varepsilon_{(out)}}{\sum \varepsilon_{(in)}} \qquad (1)$$

$$\varepsilon_{(st)} = \left(\varepsilon_{(ph)} + \varepsilon_{(ch)}\right)F \qquad (2)$$

$$\varepsilon_{(ph)} = (H - H_0) - T_0(S - S_0) \qquad (3)$$

$$\varepsilon_{(ch)} = \sum y_i \varepsilon_{(sp,i)} + RT_0 \sum y_i \ln(y_i) \qquad (4)$$

$$\varepsilon_{(sp,hydrocarbon)} = \beta_i . LHV_{hydrocarbon} \qquad (5)$$

where

$$\beta_{biomass} = \frac{A + B\left(\frac{x_C}{x_H}\right) - C\left(\frac{x_O}{x_H}\right)\left[1 + D\left(\frac{x_H}{x_C}\right)\right] + E\left(\frac{x_N}{x_C}\right)}{1 - F\left(\frac{x_O}{x_C}\right)} \qquad (6)$$

$$\beta_{bio\,oil} = G + J\left(\frac{x_H}{x_C}\right) + I\left(\frac{x_O}{x_C}\right) \qquad (7)$$

Model Development

Simulation provides an advantage in reducing the laboratory trials for difficult experiments. Thus, a model was proposed to assess the feasibility studies of algae gasification using ASPEN Plus software. A simplified thermodynamic modelling which expressed by equilibrium physicochemical reactor was likely to be used to quickly simulate the biomass gasification. A three stages gasification has been designed as a standard to accommodate previously mentioned gasification step including drying, devolatilization, cracking and reforming.

First stage of the simulation has a function to extract the water (moisture) from biomass. The second stage has a function to decompose the biomass to hydrocarbon element (C, H, O, N) and other impurities such as sulphur or silica. The third stage has a function to convert the element to product substance using Gibbs free energy minimization approach (Ramzan *et al.,* 2011, Deng *et al.,* 2019). The Spirulina microalgae was selected as the feed. Liquid product, known as tar, was modelled as a single nitrogenated compound. The model was validated by a comparation against experimental data to verify its accuracy.

RESEARCH METHOD

Gasification process of micro algae was simulated on ASPEN Plus with configuration as shown at Figure 1. Each of block on the simulation was not represent an actual arrangement of real equipment, yet as a single system inside the reactor which express drying, devolatilozation, cracking, and combustion.

The simulation consists of several processes. First, a wet spirulina biomass at 10,000 kg/s basis, which has been analyzed by other research as seen in Table 1, was fed to the DRYER as the early stage of heating and drying in real gasification reactor. The moisture content was separated and flew out to the exhaust. Steam and air (21% O_2 and 79% N_2) was used as gasification agent and oxidizer. The dried microalgae biomass was later fed to Ryield reactor named DECOMP, which decomposed the hydrocarbon substance to carbon (C), hydrogen (H), oxygen (O), nitrogen (N), and sulphur (S) elements.
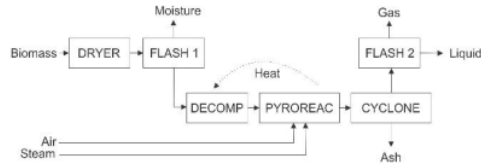

Figure 1. Simulation flow sheet of micro algae pyrolysis and gasification

Table 1. Ultimate analysis and Proximate analysis of Spirulina (Hong *et al.,* 2017)

Proximate Analysis Component	Composition (%w/w)
Moisture content	6.7 %
Volatile matter	73.5 %
Fixed carbon	13.2 %
Ash content	6.6 %
Ultimate Analysis Component	Composition (%w/w)
C	49.8 %
H	6.6 %
N	11 %
O	31.9 %
S	0.7 %

Table 2. Block condition

Block name	Block type	Function
DRYER	Heater	Removing moisture content in biomass.
DECOMP	RYield	Decomposing the substance into element by converting non-conventional stream to conventional stream as yield defined reactor.
PYROREAC	RGibs	React the substance into gasification product and providing heat from partial oxidation reaction as gibbs free energy reactor.
CYCLONE	SSplit	Separating solid from gas oil vapor.
FLASH1	Flash2	Separating moisture from dry biomass.
FLASH2	Flash2	Separating gas from condensed oil.

These elements were fed in RGibs reactor named PYROREAC to be reacted as gasification product in phase of gas, liquid, and solid (ash). Later, each product was separated to analyze the yield and quality. The heat of reaction in DECOMP was obtained from partial oxidation in PYROREAC. The detailed configuration of the equipments is shown in Table 2.

In order to succesfully run the system at Figure 1, several basic data was required such as component spesification for nonconventional stream, properties of streams and blocks, input and output units, valid operating phase, and proper operating condition. Property model used for the simulation was Advanced NC-Props form. The overall process was operated in atmospheric pressure (1 atm). Biomass was classified as non-conventional substance, so that advanced NC-Props form should be enabled and feed data such as mass component were required to specify the biomass characteristic. In other hand, non-conventional enthalpy and density should be defined from HCOALGEN DCOALGEN since it cannot participate in chemical or phase equilibrium automatically. Reactor performance was assessed by proceeding the sensitivity analysis in certain range of partial combustion and steam to carbon ratio (S/C) toward the product distribution.

RESULTS AND DISCUSSION
Model Validation

Thermochemical process of biomass generates three type of product phase. Gas phase containing methane (CH_4), carbon monoxide (CO), hydrogen (H_2) and other non-flammable gas including carbondioxide (CO_2) and unreacted nitrogen (N_2) was formed during sequential process of solid carbonaceous devolatilization, tar cracking, methane reforming and water-gas shift reaction. While liquid phase (biofuel) which usually consist of several type of compounds including aromatics, phenols and nitrogenated compound was formed from condensed volatile tar as the product of devolatilization and primary tar cracking. The solid residue (bio char) left by thermochemical process consist of unreacted carbon and several minerals, depend on what type of biomass was used (Hong *et al.,* 2017).

Gasification of spirulina microalgae via ASPEN Plus software was simulated by defining the ultimate and proximate analysis of spirulina as the biomass feed properties. Two simplifications were taken by assuming liquid product as nitrogenated compound i.e. Indole (C_8H_7N) based on liquid major fraction of algae feed, and solid product which defined as a mixture of ash with solid carbon and solid oxygen based on residue analysis of algae char. Simulation result of product phase distribution and major gases component were validated with the experiment data shown in Table 3. Within the experiment that was conducted in atmospheric pressure with no presence of air and steam, the simulation result gave a reasonable deviation. Thus, reactor performance can be studied further. However, this model produced a lower accuracy due to simplified reaction and neglected multiphase reaction. This error also occured in several researches which applied similar reactor model (Ramzan *et al.,* 2011, Deng *et al.,* 2019).

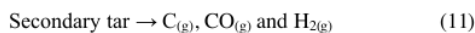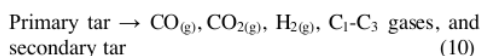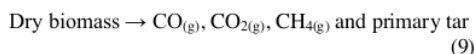Table 3. Yield distribution of experiment and simulation

Compound	Experiment (Hong, Chen et al. 2017)	Simulation	Relative Error
Product Distribution (wt.%)			
Gas	84	85	1%
Liquid	7	8	14 %
Solid	9	7	22 %
Gaseous product composition (vol.%)			
H_2	32	32	0 %
CO	40	42	5 %
CH_4	12	18	50 %

System Performance

The first stages of gasification process is drying. During the drying, 6.7 %w/w moisture content stated from proximate analysis is extracted from solid body and flowing out before the system reach devolatilization temperature, leaving dry biomass.

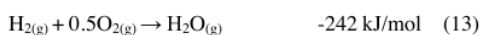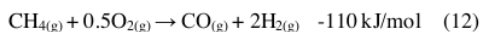$$\text{Wet biomass} \rightarrow \text{Dry biomass} + H_2O(g) \quad (8)$$
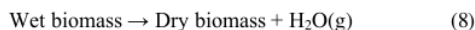
When the system reach early thermal reduction stage, dry biomass decomposed to carbon gaseous and long chain molecules as primary tar, followed by tar cracking which producing lower carbon chain and syngas, as seen in Eq. 9-11.

$$\text{Dry biomass} \rightarrow CO_{(g)}, CO_{2(g)}, CH_{4(g)} \text{ and primary tar} \quad (9)$$

$$\text{Primary tar} \rightarrow CO_{(g)}, CO_{2(g)}, H_{2(g)}, C_1\text{-}C_3 \text{ gases, and secondary tar} \quad (10)$$

$$\text{Secondary tar} \rightarrow C_{(g)}, CO_{(g)} \text{ and } H_{2(g)} \quad (11)$$

The effect of air flow (21% O_2 and 79% N_2) at ambient temperature and atmospheric pressure which varied from 0 to 1 kg/s was studied. As seen in Figure 2, the increased air flow escalated the gaseous phase product from 2.12 kg/s to 3.28 kg/s and enhanced tar decomposition from product yield of 0.21 kg/s to 0.04 kg/s, identical with other pyrolysis research trendline (Saleh *et al.,* 2019). At this stage, combustion reaction took place, reacting O_2 and reducing flammable gases including H_2, CO and CH_4 with the differential amount of 2.1%, 3.7% and 7.2% respectively, leaving CO_2 and unreacted N_2 as seen in Figure 3.

During the combustion, the only compound that purely formed as product is CO_2, which produced both in homogenous oxidation (Eq. 12-14) and heterogeneous oxidation (Eq. 15-16). CO become an intermediate product as it also oxidized, while H_2O

will be consumed during SMR reaction. However, when the supply of oxidator is limited, the overall oxidation reaction tend to choose the path which has a higher enthalpy of reaction (ΔH_R) due to exothermic properties at high temperature environment. In this case, the oxidation of CO and C which produce CO_2 has a higher exothermic properties (lower ΔH_R) than others, so that the amount of CO is larger from the initial condition until certain addition of air supply. In other hand, all of oxidation process produce the heat required for all endhotermic reaction including tar decomposition. As the air supply increased, the reactor temperature also increased which lead more tar decomposition reaction.

$$CH_{4(g)} + 0.5O_{2(g)} \rightarrow CO_{(g)} + 2H_{2(g)} \quad -110\,kJ/mol \quad (12)$$

$$H_{2(g)} + 0.5O_{2(g)} \rightarrow H_2O_{(g)} \quad -242\,kJ/mol \quad (13)$$

$$CO_{(g)} + 0.5O_{2(g)} \rightarrow CO_{2(g)} \quad -283\,kJ/mol \quad (14)$$

$$C_{(s)} + O_{2(g)} \rightarrow CO_{2(g)} \quad -393\,kJ/mol \quad (15)$$

$$C_{(s)} + 0.5O_{2(g)} \rightarrow CO_{(g)} \quad -123\,kJ/mol \quad (16)$$

Along with the process, solid product mass flow was relatively stable, not affected by the differentiated air supply eventhough the solid carbon was already stated as the product. It was guessed that fixed carbon was totally reacted from the beginning, leaving ash as the solid product.
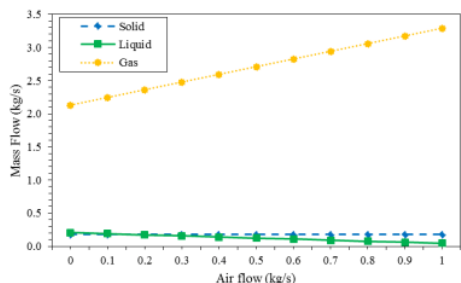

Figure 2. Effect of air injection to reactor product phase distribution
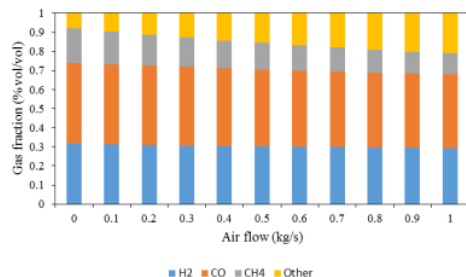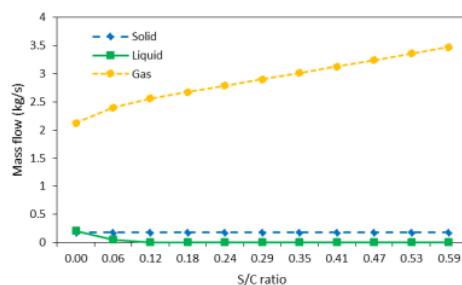

Figure 3. Effect of air injection to gas component yield

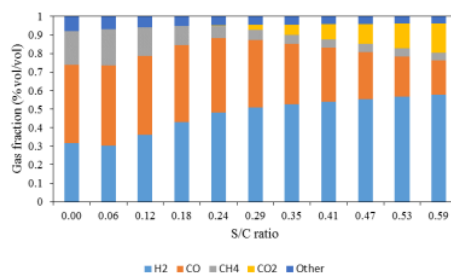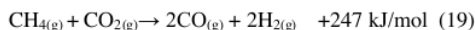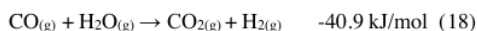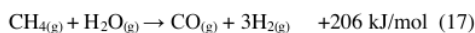Figure 4. Effect of S/C ratio to reactor product phase distribution

Figure 5. Effect of S/C ratio to gas component yield

Steam at varied flowrate of 0-1.41 kg/s which gave steam to carbon ratio (S/C) of 0-0.59, favored an increasing of gas product yield from 2.12 kg/s to 3.47 kg/s as seen in Figure 4. It also effectively enhanced tar decomposition at S/C above 0.12 with 8.2% reduction. The presence of water triggered the steam-methane reforming and water-gas-shift reaction, followed by dry reforming as CO_2 appeared in Eq. 17-19 below:

$$CH_{4(g)} + H_2O_{(g)} \rightarrow CO_{(g)} + 3H_{2(g)} \quad +206\,kJ/mol \quad (17)$$

$$CO_{(g)} + H_2O_{(g)} \rightarrow CO_{2(g)} + H_{2(g)} \quad -40.9\,kJ/mol \quad (18)$$

$$CH_{4(g)} + CO_{2(g)} \rightarrow 2CO_{(g)} + 2H_{2(g)} \quad +247\,kJ/mol \quad (19)$$

When the steam injection increased, the conversion of CH_4 and CO to CO_2 and H_2 also increased with the differential fraction amount of -13.9%, -23.9%, 15.7% and 26.3% respectively as presented in Figure 5. According to the simulation, CO_2 is found in the excess product sstarting at S/C 0.24, and the fraction of CO is always larger than CO_2 before S/C 0.59. It was guessed as water present at the system, the steam-methane reforming and dry reforming reactions have a higher tendency to proceed due to endothermic properties in high temperature environment. Therefore, the appearance of CO_2 begins when large amount of CH_4 was consumed and increase the water-gas-shift reaction probability. Similar with air supply treatment, solid product mass flow was relatively stable due to highly reacted fixed carbon.

This trendline has an identical behavior with other gasification research (Gil *et al.,* 1999, Song *et al.,* 2015).

Exergy Assesment

The exergy assessment was carried out by entering stream data on ASPEN simulation result including stream composition, enthalpy, entropy and pseudo cold stream to equation 1-7. Based on equation 5 and 6, spirulina microalgae massflow at 10,000 kg/h and room temperature provide exergy result of 63,900 kJ/s which is used as the reference to calculate the exergy efficiency (η_ε). Since ash has very low species chemical exergy and mass flowrate, the exergy of solid product which has no other compound was neglected. Both of partial combustion and S/C ratio showed that gaseous product dominated the exergy amount of product as seen in Figure 6 and 7. Regarding to product distribution, it was clear that gaseous phase dominated the product and kept rising as the air flow increased from 0 to 1 kg/s, providing exergy amount of 45,000 kJ/s to 49,700 kJ/s for gas product and declining exergy amount of 8,200 kJ/s to 1,800 kJ/s for liquid product. While the same trend line was also shown from increased S/C ratio, providing exergy amount of 45,000 kJ/s to 53,700 kJ/s for gas product and declining exergy amount of 8,200 kJ/s to 0 kJ/s for liquid product at S/C ratio of 0 to 0.12.

The overall exergy efficiency (η_ε) derived from physical and chemical exergy calculation showed different pattern between air massflow and S/C ratio variable effect. The increase of partial combustion produced a slight linear declining of total exergy efficiency of the system, while the S/C ratio produced an oscilatory graph as seen in Figure 8 and Figure 9.
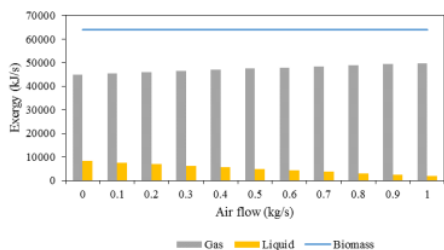

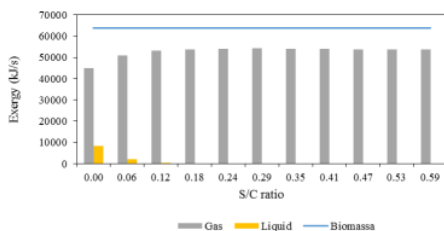Figure 6. Effect of air injection on stream's exergy

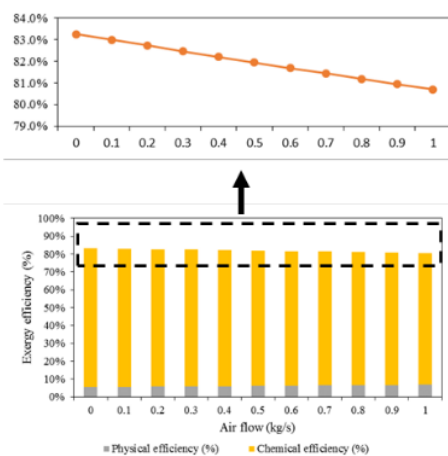Figure 7. Effect of S/C ratio to stream's exergy

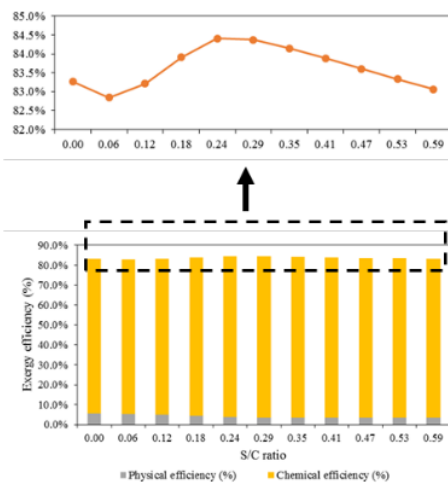Figure 8. Effect of air injection to total exergy efficiency

Figure 9. Effect S/C ratio to total exergy efficiency

Both efficiency calculation was dominated by chemical exergy at range of 73.9% to 79.5% portion rather than physical exergy at range of 3.6% to 6.8% portion. Hence, the energy related properties of the product became an important target to be stated in a system design since the chemical speciess exergy take the major role. The exergy efficiency obtained by another simulation research is about 69.5–71.8%. (Saidur *et al.,* 2012)

The increasing air supply to the reactor reduced total exergy efficiency from 83.3% to 80.7%. It was guessed that by large amount of oxygen provided, the declining slope of exergy calculated from tar decomposition was larger than exergy calculated from increased gas product. The S/C ratio has a minimum turning point at 0.06 and maximum at 0.24 which giving 82.8% and 84.4% exergy efficiency

respectively. Before the minimum point, a large tar yield provided high exergy ammount. Along with the increased steam flowrate, tar decomposition was proceeded until almost zero point. Here, the total exergy efficiency was declining to the minimum point. Later, the exergy efficiency increased to the maximum point due to increased gaseous product quality and quantity indicated by high hydrogen (H_2) and carbon monoxide (CO) yield at 48% and 40% respectively. After the maximum point, the increased S/C ratio decreased the exergy efficiency due to water-shift-gas reaction which produced high amount of CO_2. It was analyzed from chemical exergy efficiency of gaseous product which reduced from 54.1% to 53.7%.

CONCLUSION

The proposed simulation model was suitable to predict the thermochemical conversion behavior of spirulina with acceptable accuracy. Spirulina microlagae gasification provided favorable product due to high exergy efficiency. Partial combustion enhanced gas product mass flow and tar decomposition but reduced total exergy efficiency. Thus, the air supply should be minimized to reduce excess oxidation and maintain product quality. The presence of water caused a reduction of CH_4 and CO, but it increases the H_2 yield. The exergy assessment of steam effect to the system performance give both the minimum and maximum point which highly affected by H_2 and CO and the absence of CO_2. Finally, High exergy efficiency would be acquired if the system succesfully produced high-quality product in term of energy content.

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