

Process Simulation Fischer Tropsch Syntehsis Gas-to-Liquid (GtL) from Sales Gas Using Aspen HYSYS

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Abstract. The production of fuels, such as gasoline, diesel, and jet fuel, the use of sales gas through GTL-FT (Gas to Liquid Fischer Tropsch) has received increasing attention over the last few years. In this research, the Gas to Liquid process to produce jet fuel from Sales Gas is simulated. Kinetic data and simulation basis was taken from published journal papers. Simulation was initially validated based on reference and optimized based on several aspects: ATR and FT operating temperature, hydrocarbon-steam-ratio, and staging ratio while looking into several objective parameters: the H₂/CO ratio in syngas produce, reactor conversion, and jet fuel (C₆-C₁₆) product mol fraction. An increase in the steam-gas ratio and an increase in operating temperature on the ATR will result in an increase in the resulting H₂/CO syngas ratio. The staging effect on ATR will also increase the value of the H₂/CO ratio of the resulting syngas. The increase in operating temperature on the Fischer Tropsch will reduce the liquid fuel gain and mole fraction yield to be obtained. Furthermore, staging on the reactor fischer tropsch will increase the actual conversion and mole fraction yield of jet fuel.

Keywords: Autothermal reformer, Fischer tropsch, Gas to liquid, Jet fuel.

1 Introduction

In 2016, energy consumption in the transportation sector reached 331.7 million SBM (Oil Barrel Equivalent) with a fuel mix consisting of 55.3% gasoline; 14.0% diesel oil; 22.3% biosolar; 0.04% fuel oil, 0.07% natural gas; 0.005% avgas, 8.15% avtur and 0.04% electricity. Meanwhile, in 2014, CO₂ gas emissions from motor vehicles reached 126.56 million tons, consisting of 69.63 million tons of CO₂ emissions from the use of gasoline and 56.92 million tons of CO₂ emissions from diesel [1].

Growing concerns over the environmental impact of the aviation industry have also sparked the potential to look for renewable fuels that could reduce the industry's current dependence on Aviation fuel Jet-A1 fuel which is still based on kerosene. Alternative energy sources that have the potential to be developed include coal, natural gas, and biomass. These three sources can be converted into synthesis gases (syngas), a mixture of CO and H₂. Syngas can be converted into hydrocarbons equivalent to fossil fuels such as gasoline, kerosene, and diesel. The conversion of syngas into fuel is usually carried out through the synthesis of Fischer-Tropsch [2]. Fischer-Tropsch synthesis, subsequent hydrocracking, or hydro-purification steps to further upgrade candles are needed to increase selectivity to liquid fuels [3].

The production of renewable fuels, such as gasoline, diesel, and jet fuel, using gas sales through GTL-FT (Gas to Liquid Fischer Tropsch) has also gained increased attention over the past few years. Renewable fuels from GTL-FT are much cleaner and environmentally friendly and contain little or no sulfur and other contaminant compounds [4]. In addition, products of fischer tropsch synthesis have a low aromatic content, and lower emissions of hydrocarbons, CO, NOx, and particulates in the combustion process, when compared to other traditional fuels [5].

The synthesis of liquid fuel with the main jet fuel product using Gas to Liquid through Fischer–Tropsch Synthesis (GTL-FT) will provide a breakthrough in liquid fuel production in Indonesia. In addition, this will also increase the availability of liquid fuel needs in the country and become an export opportunity for Indonesia to improve the economy.

2 Method

2.1 Research Variables

Research variables consist of free variables, bound variables, and fixed variables. Free variables are variables that are unbound and made free. Bound variables are variables that undergo changes due to changes in the value of free variables. Fixed variables are variables that are made constant and are not affected by other variables. In this research, the variable details are provided in Table 1.

Table 1. Research variables

Fixed Variables	Free Variables	Bounded Variables
Sales gas flow rate and component fractions	Steam-Hydrocarbon (Sales gas feed rate) ratio ATR operating temperature Fischer Tropsch Reactor operating temperature	CO/H2 ratio in syngas Conversion and product mole fractions Jet fuel product yield

2.2 Simulation Basis

Components used in the process simulation is provided in Table 2. In the ASPEN HYSYS model setup, the fluid package used is Peng-Robinson (Equation of State), as this EOS model can correctly calculate all the properties of the gas sales as well as most hydrocarbon compounds according to the composition of the compounds used in this simulation (**Figure 1**)

Table 2. Components used in the process simulation

Components				
Methane	n-Pentane	n-Decane	n-C15	n-C20
Ethane	n-hexane	n-C11	n-C16	n-C30
Propane	n-heptane	n-C12	n-C17	H2
n-Butane	n-octane	n-C13	n-C18	CO

i-Butane n-nonane n-C14 n-C19 CO2

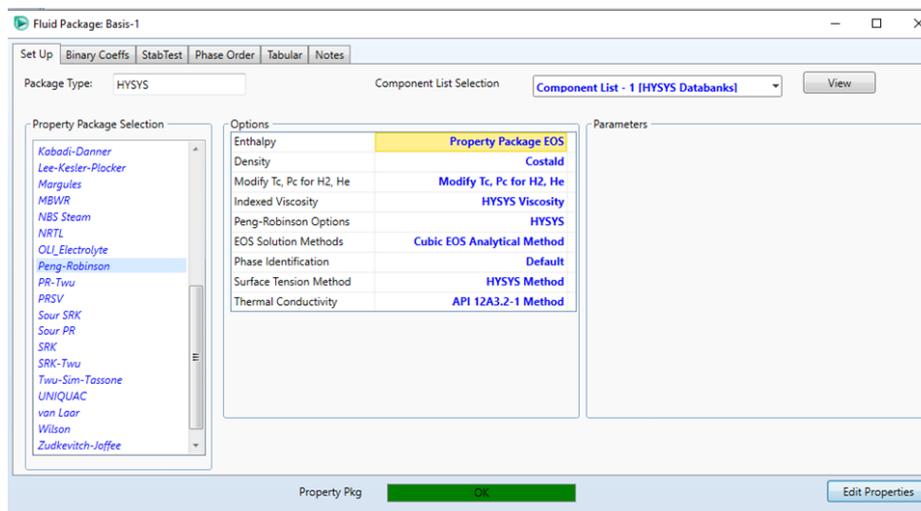


Fig. 1. Simulation Setup Property Package

2.3 Reaction Setup and Kinetics

In the Gas to Liquid (GtL) simulation via Fischer Tropsch synthesis using feed in the form of sales gas, involving 3 stage of reactions that occur in pre-reformer, Autothermal Reactor (ATR), and Fischer Tropsch synthesis. The reaction simulation setup are as follows:

a. Pre-reformer

The pre-reformer is simulated in ASPEN HYSYS using an equilibrium reactor, where all temperatures in the reactor are high, and all hydrocarbons will be converted completely. The input reactor equilibrium data used is as follows: in an atmospheric 450 °C muffle furnace for 2 hours. Details on reaction equations included in equilibrium pre-reformer system are provided in Table 3.

Table 3. Equilibrium Data Pre-Reformer [6]

Reaction	$\Delta_{rxn}H^{0}_{298}$ (kJ/mol)
$C_2H_6 + 2H_2O \leftrightarrow 5H_2 + 2CO$	350
$C_3H_8 + 3H_2O \leftrightarrow 7H_2 + 3CO$	500
$C_4H_{10} + 4H_2O \leftrightarrow 9H_2 + 4CO$	650
$CO + 3H_2 \leftrightarrow CH_4 + H_2O$	-210
$CO + H_2O \leftrightarrow CO_2 + H_2$	-41

b. Autothermal Reactor

ATR is simulated with ASPEN HYSYS as a kinetic reactor with heterogeneous catalyst. All reactions that occur in the reformer are modeled as kinetic reactions. Table 4 provides kinetics data for pre-reformer reactions.

$$R_1 = \frac{k_1 p_{CH_4} p_{O_2}^{\frac{1}{2}}}{(1 + K_{CH_4}^c p_{CH_4} + K_{O_2}^c p_{O_2}^{\frac{1}{2}})^2} \quad (1)$$

$$R_2 = \frac{k_2}{p_{H_2}^{2.5}} (p_{CH_4} p_{H_2O} - \frac{p_{H_2}^3 p_{CO}}{K_e^2}) * \frac{1}{Q_r^2} \quad (2)$$

$$R_3 = \frac{k_2}{p_{H_2}} (p_{CO} p_{H_2O} - \frac{p_{H_2} p_{CO_2}}{K_e^3}) * \frac{1}{Q_r^2} \quad (3)$$

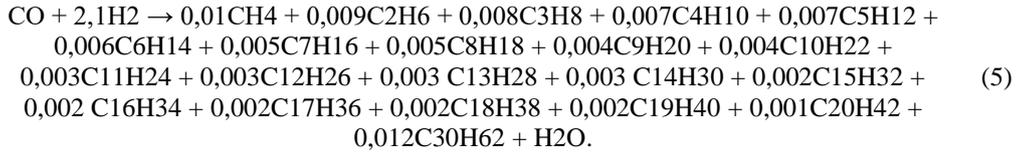
$$Q_r = 1 + K_{CO} p_{CO} + K_{H_2} p_{H_2} + K_{CH_4} p_{CH_4} + \frac{K_{H_2O} p_{H_2O}}{p_{H_2}} \quad (4)$$

Table 4. Kinetics Data Pre-Reformer [6].

Reaction	K_{oj} ($\frac{kmol}{kg \text{ cat } h}$)	E_j ($\frac{kJ}{kmol}$)
$CH_4 + 2O_2 \leftrightarrow CO_2 + 2 H_2O$	$5.852 * 10^{17} \text{ bar}^{-1.5}$	204
$CH_4 + H_2O \leftrightarrow CO + 3 H_2$	$54.225 * 10^{15} \text{ bar}^{0.5}$	240,1
$CO + H_2O \leftrightarrow CO_2 + H_2$	$1.955 * 10^6 \text{ bar}^{-1}$	67,13

c. Fischer Tropsch Reactor

The Fischer Tropsch reaction is modeled as a plug flow reactor (PFR) in ASPEN HYSYS, where the reaction that occurs is modeled as a heterogeneous catalyst reaction with a cobalt catalyst.



Stoichiometric coefficient for Fischer Tropsch reaction based ASF distribution and kinetic data implemented using Iglesias rate of reactions, where reaction rate, A, E, and n formulated as: [5]. Table 5 summarized kinetics data of Fischer Tropsch.

$$r_{CH_4} = \frac{k_1 p_{H_2} p_{CO}^{0,65}}{1 + (K_1 p_{CO})} \quad (6)$$

$$r_{CH_4} = \frac{k_2 p_{H_2}^{0,6} p_{CO}^{0,65}}{1 + (K_1 p_{CO})} \quad (7)$$

Table 5. Kinetic Data Fischer Tropsch [5].

Reaction	A	-E	Exponent PH2	Exponent PCO		N
				Numerator	Denominator	
r_{CH4}	8,8 E-6					
k1	1,1 E-12	37326	1	0,05	-	-
k1		-68401,5	-	-	1	1
r_{CO}						
K2	1,6E-5	37326	0,6	0,65	-	-
k1	1,1E-12	-68401,5	-	-	1	1

2.4 Gas-to-liquid Process Description

Gas to Liquid process modeling involves 3 main processes, namely the manufacture of syngas, the conversion of syngas through Fischer Tropsch reactions, and the separation of the main product. Simulations were performed using ASPEN HYSYS 8.6, with equilibrium and kinetic reaction approaches. The feed used is gas sales, with a constant rate of 22,000 kmol/h or 15.35 MMSCFD at a temperature of 40°C and 3000 kPa, and the composition of the feed is describe in Table 6.

Table 6. The Composition Of Feed Sales Gas

Components	Mole Fraction
Methane	0.95
Ethane	0.02
Propane	0.015
i-Pentane	0.005
i-Butane	0.01

Before entering the pre-reformer, the feed sales gas is preheated to a temperature of 455 C. Then, the heated feed sales gas is mixed with steam at a temperature of 455 C before entering the preheater. Pre reformer serves to break down the bonds of heavy hydrocarbons into H₂ and CO before entering the ATR, thus preventing the formation of side reactions and carbon in the ATR [8]. In pre-reformers, CH₄ has not reacted because the temperature in the pre-reformer has not been high enough to be able to rebrand methane, so that only compounds with a higher content in feed sales gases such as propane and ethane are converted. Pre reformer simulated as an equilibrium reactor, with isothermal condition. Out pre reformer at 423.9°C and 2900 kPa. Performance of pre-reformer reaction is described in Table 7 with outlet reactor compositions are given in Table 8.

Table 7. Pre-Reformer Reaction Conversion Details

Reaction	Actual % Conversion	Base Component
$C_2H_6 + 2H_2O \leftrightarrow 5H_2 + 2CO$	100	Ethane
$C_3H_8 + 3H_2O \leftrightarrow 7H_2 + 3CO$	100	Propane
$C_4H_{10} + 4H_2O \leftrightarrow 9H_2 + 4CO$	0	Butane
$CO + 3H_2 \leftrightarrow CH_4 + H_2O$	0	CO
$CO + H_2O \leftrightarrow CO_2 + H_2$	0	CO

Table 8. Composition Detail Outlet Pre-Reformer

Components	Mole Fraction
Methane	0.687
Ethane	0
Propane	0
i-Pentane	0.0034
i-Butane	0.00068
CO	0.0004
CO ₂	0.0206
H ₂	0.0498
H ₂ O	0.2319

The pre-reformer output will then enter the heater to heat the stream to a temperature of 580°C. Then, from the heater, it will enter the autothermal reactor. In ATR reactors, a baseline is used in the form of reactor equilibrium. The other input on the ATR is oxygen with conditions of 200°C, 3000 kPa. The ATR output will produce syngas at a temperature of 1047°C and a pressure of 2900°C. Performance of pre-reformer reaction is described in Table 9.

Table 9. ATR Reaction Conversion Details

Reaction	Actual % Conversion	Base Component
$CH_4 + 2O \leftrightarrow CO_2 + 2 H_2O$	10.07	Methane
$CH_4 + H_2O \leftrightarrow CO + 3 H_2$	53.9	Methane
$CO + H_2O \leftrightarrow CO_2 + H_2$	4.651e-06	CO

The ATR output syngas then enters the heat exchanger and cooler to be lowered to 25°C or room conditions. This aims to separate the water content contained in the syngas. After the moisture content on the syngas is separated, then the dry syngas will be adjusted to its condition before entering the Fischer Tropsch reactor. The Fischer Tropsch reactor will operate at an inlet temperature of 210 °C, and a pressure of 2465 kPa. The catalyst used in this process is a cobalt catalyst with a feed fraction value of 0.4. Output Fischer Tropsch Reactor

at a temperature of 200 °C and a pressure of 2265 kPa, with detailed composition of reaction conversion and output given in Table 10.

Table 10. FTR Reaction Conversion Details

Reaction	Actual % Conversion	Base Component
$\text{CO} + \text{UH}_2 + n\text{CO} \rightarrow \sum^{20} (\text{ASF Coef.})_i \text{C}_i\text{H}_{2i+2}$ $+ (\text{ASF Coef.})_{30} \text{C}_{30}\text{H}_{62} + \text{H}_2\text{O}$	18.34	CO
$\text{CO} + 3\text{H}_2 \leftrightarrow \text{CH}_4 + \text{H}_2\text{O}$	27.8	CO

The output product of Fischer Tropsch reactor will enter the cooler and 3 phase separator for purification and separation of main products and by-products. Furthermore, the hydrocarbon liquid formed will be mixed and become the output of this system [8].

3 Results and Discussion

To validate the results of the simulation that has been carried out, several important parameters are used to compare the simulation results with reference data, namely the resulting H₂ / CO syngas fraction value, actual conversion of ATR and FTR reactor, and mole fraction yield value of C₉-C₁₆ (jet fuel) at the output of the Fischer Tropsch reactor. Simulation PFD in HYSYS is given in **Figure 2**.

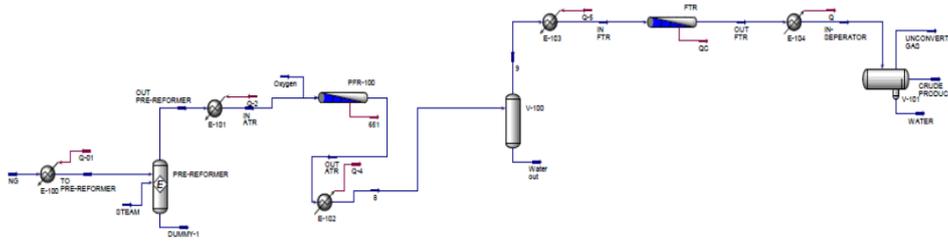


Fig. 2. Simulation Setup Property Package

The comparison between baseline simulation and the reference is provided in Table 11.

Table 11. Comparison baseline simulation and reference

Parameter	Baseline Simulation	Reference	Error (%)
Fraksi H ₂ /CO syngas output ATR	3.8	3.85	1.3
Actual Conversion ATR (%)	60.86	60.86	0

<i>Actual Conversion Fischer Tropsch Reactor (%)</i>	21.156	21.15	0
Mole Fraction C ₉ -C ₁₆	0.13	0.135	0

From the results of the simulation comparison and the results on the reference [10]. It can be seen from several parameters that the resulting value is similar and close to the reference value and with a small error value. So that gas to liquid simulation modeling using equilibrium and kinetics data input is valid and can provide an accurate representation of the process.

Variations in the Ratio of Steam / Sales Gas to the ratio of H₂ / CO of the resulting syngas. The feed sales gas used in this simulation is 22000kmol/h or 15.35 MMSCFD. Varied steam inlet based on the ratio to inlet feed sales gas. The variation in the steam / sales gas ratio used is 0;0.2;0.4;0.6;0.8;1. Of the five ratio variations, the results in shortcut and rigorous simulation were obtained and provided in Table 12.

Table 12. Results Of Variations in The Ratio Of Steam / Sales Gas To The Ratio Of H₂ / CO Syngas

Sales Gas (MMSCFD)	Steam (kmol/h)	Steam/Sales Gas Ratio	H ₂ /CO Ratio in Syngas
15.35	0	0	0
15.35	4400	0.2	2.02
15.35	8800	0.4	2.15
15.35	1320	0.6	2.28
15.35	1760	0.8	2.42
15.35	2200	1	2.56

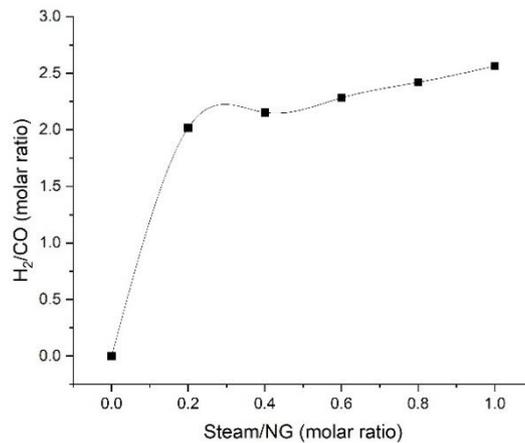


Fig. 3. Relationship of Steam/SG Ratio to H₂/CO Ratio of ATR Outlets

From the results of the variation in the ratio to feed sales gas as seen in **Figure 3**, that the higher the steam / sales gas ratio, the higher the ratio of H₂ / CO syngas produced [11]. This

can occur due to the influence of reaction equilibrium on the Pre reformer and Autothermal Reactor. When the ratio and amount of steam are getting higher, it shows that there is an increase in reactants. Based on Le Chatelier's principle, when there is an addition of reactants, the reaction that occurs will shift to the right, so that the products obtained tend to be more. In the reaction in Pre-reformer 1- 3, it is seen that the stoichiometric coefficient of H₂ is greater than that of CO, so that as the amount of steam increases, the reaction will shift to the right, the amount of H₂ produced will also increase compared to CO and the resulting H₂/CO ratio will also increase.



In the ATR reaction, there are two reactions involving steam as a reactant, namely reactions 8 and 9. An increase in the amount of steam will trigger an equilibrium reaction to shift to the right, so that the amount of CO and H₂ produced is more. However, because the stoichiometric coefficient of H₂ is greater than CO, the resulting ratio of H₂ / CO is also greater. In addition, some of the CO formed in reaction 8 will also react again with steam to form H₂, so the amount of H₂ will also increase.

In accordance with the purpose in chapter 1, to optimize the acquisition of jet fuel requires an H₂ / CO ratio of about 2.15. From the simulation results, the Steam / Sales Gas ratio of 0.4 in the simulation results gives the H₂ / CO ratio value which is closest to 2.15. Thus, the optimum ratio of Steam / Sales Gas used in this simulation is 0.4, with a total steam of 8800 kmol / h.

Variation of Operating Temperature ATR to the ratio of H₂ / CO syngas produced. In the previous optimum steam conditions, namely, 8800 kmol/h steam, the ATR operating temperature was varied to see the effect of operating temperature on the resulting H₂ / CO syngas ratio [12]. The temperature variation used is based on the reference that has been obtained, where the operating temperature of the Autothermal reactor is at 500-700 °C. The results are summarized in Table 13 and plotted in **Figure 4**.

Table 13. Results Of Variations In The Ratio Of Steam / Sales Gas To The Ratio Of H₂ / CO Syngas

ATR Operating Temperature (°C)	H ₂ /CO
500	2.17
540	2.17
580	2.16
620	2.15
660	2.15
700	2.14

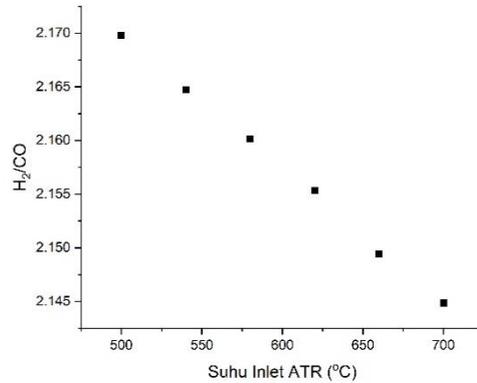
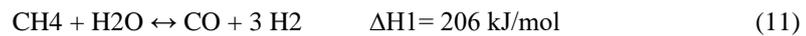


Fig. 4. Variation of Operating Temperature ATR to Ratio of H₂ / CO syngas

From the results of shortcuts and rigorous simulation on the optimum steam input, which is 8800 kmol / h, it can be seen in the picture above that the higher the operating temperature of the ATR, the lower the ratio of H₂ / CO syngas produced. This can occur due to the influence of reaction equilibrium on the ATR. The reactions that occur in ATR are:



The 10th reaction is an endothermic reaction. Where, based on the Principle of Le Chatelier, when the temperature is raised, the reaction will shift to the right. This will trigger an increase in the amount of CO and H₂ produced. However, reaction 10, which is a follow-up reaction of CO with steam, is an exothermic reaction, where when the temperature is raised, the equilibrium reaction will shift to the left. So, it will predominantly provide a high amount of CO compared to the amount of H₂. A high amount of CO will result in a low ratio of H₂/CO produced. Where this is according to what is obtained on the chart. Thus, the optimum operating temperature used in this simulation is 660 °C.

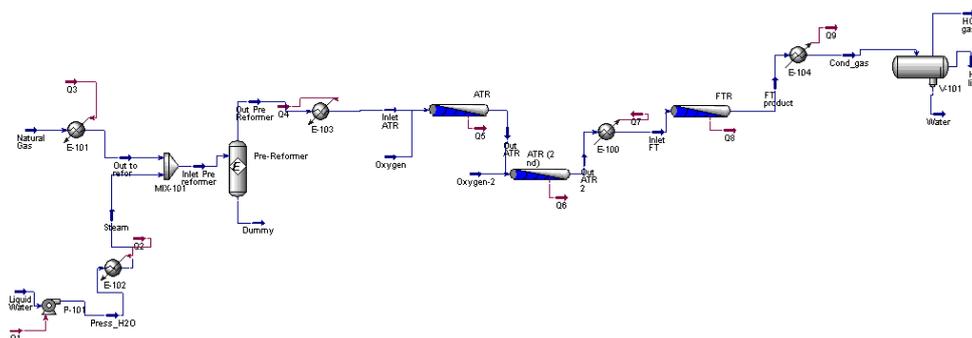
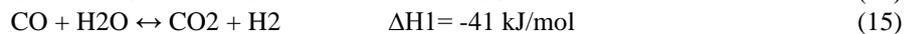
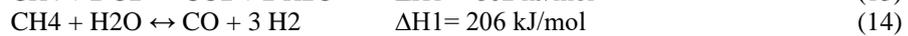
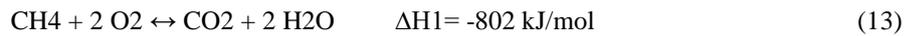


Fig. 5. Double Stage ATR Simulation Results

From the simulation results on the steam inlet of 8800 kmol/h and the operating temperature of the reactor at 660°C, by comparing single stage and double stage on the ATR (**Figure 5**),

the results were obtained that the double stage will increase the ratio of H₂/CO to more than 3. This can happen because of the equilibrium of the reaction that occurs in the ATR. In the first ATR, the resulting reaction conversion is rudimentary so that there is still residual reactants that have not yet reacted. When the reaction is continued in the second reactor, it will trigger a follow-up reaction to methane oxidation. Where the addition of oxygen to the exothermic reaction of burning methane will cause the equilibrium reaction to shift to the left so that methane will tend to form CO and H₂ as the 12th reaction.



The formation of CO and H₂ will cause an over ratio in the resulting H₂ / CO, so that although the double stage increases the reaction conversion, the ratio at the resulting double stage does not meet the desired H₂ / CO ratio specifications. This will affect the condition of the resulting syngas specification, affect and enlarging the water gas shift reaction on the Fischer Tropsch, so that it will also affect the distribution of the product to be obtained. Therefore, the use of single stage is more recommended in this process compared to double stage.

Effect of Feed Operating Temperature on mole fraction yield jet fuel produced. From the results of the syngas specification produced in the previous process, the effect of operating temperature variations on the reactor Fischer Tropsch will be reviewed. Fischer Tropsch reactor LTFT (Low Temperature Fischer Tropsch) operates at a temperature of 200-240 °C. In this simulation, the operating temperature of the Fischer Tropsch reactor varies to 200, 210, 220, 230, and 240 °C. From the results of the operating temperature variation, the mole fraction of the jet fuel product will be seen in **Figure 5**.

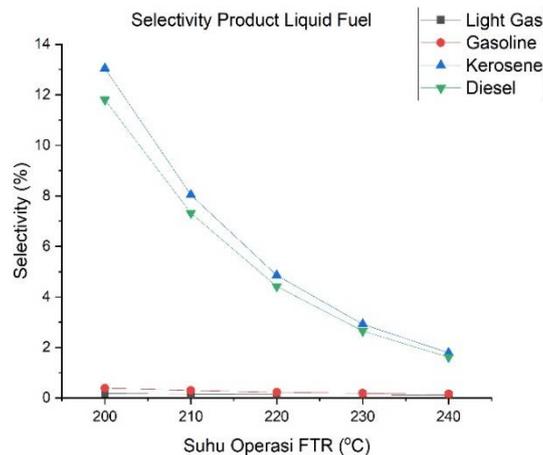


Fig. 5. Distilled Liquid Fuel Fischer Tropsch Products at Operating Temperatures Of 200-240°C

From the results of simulations that have been carried out, it was found that moles fraction liquid fuel is dominant in C₁₀-C₁₆ and C₃₀ or high olefin compounds and waxes. From the

results of the temperature variations carried out, it can be concluded that the C₁₀-C₁₆ fuel jet, the higher the operating temperature, the lower the acquisition of C₁₀-C₁₆ and vice versa will increase the acquisition of waxes. In accordance with the objectives that have been proposed at the beginning, to maximize the acquisition of jet fuel with the majority of high olefin compounds (C₁₀-C₁₆), the operating temperature of 200 °C provides a higher yield fraction value of C₁₀-C₁₆. This can happen because the reaction that occurs is exothermic, so according to the principle of equilibrium, when there is a rise in temperature, the reaction will shift to the left. This causes the rate of product formation to be further reduced as the temperature increases.

Table 14. Relationship Of Operating Temperature with Actual Conversion and Liquid Fuel FTR Production

Temperature (C)	Actual Conv 1 (%)	Actual Conv 2 (%)	Mass crude liquid fuel (kg/h)
200	18.34	12.78	12180
210	15.01	10.4	9431
220	12.24	8.402	7236
230	8.095	5.439	5356
240	9.952	6.764	3595

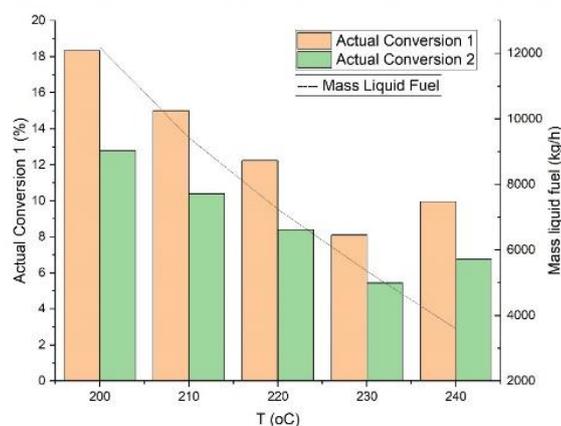


Fig. 6. Graph of the Relationship of Operating Temperature with Actual Conversion and Liquid Fuel Period

In addition, when comparing the actual conversion and mass flow of liquid fuel produced, the optimum temperature that provides the highest actual conversion and the most mass flow liquid fuel is at an operating temperature of 200 °C. Thus, the temperature of 200°C in this simulation process is the optimal temperature to obtain many products in the form of jet fuel. Effect of Staging FTR (simulated as **Figure 8**) on mole fraction yield crude jet fuel produced summarized in Table 15.

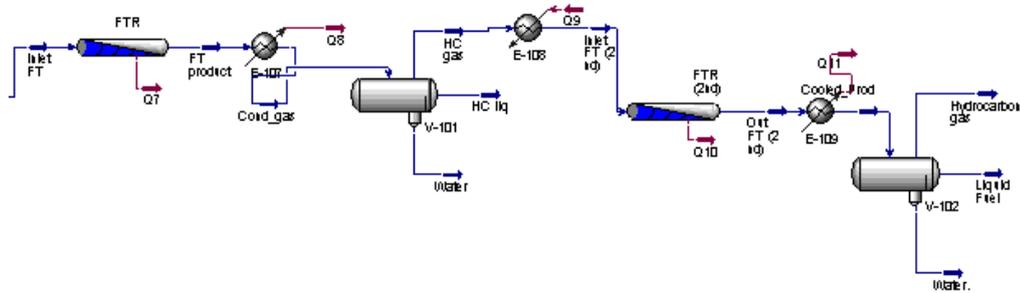


Fig. 7. Effect of vapor product staging (FT staging) on mole fraction yield

Table 15. Comparison Single and Double Stage FTR Reactor

Parameter	Single Stage	Double Stage
Total Actual Conv 1 (%)	18.34	24.52
Total Actual Conv 2 (%)	12.78	16.31
Mole fraction yield total (C ₉ -C ₁₆)	0.115	0.151
Masa C ₉ -C ₁₆ (kg/h)	1694.24	1897.11

By staging the output of 3 phase separators, an increase in reaction conversion and mole fraction yield was obtained, as well as the total mass of the C₉-C₁₆ fraction obtained. This can happen because the small reaction conversion in the first reactor of the Fischer Tropsch results in many syngas that have not yet reacted. When reacted again at the second stage, there will be a follow-up reaction in the Fischer Tropsch reactor and the mass of liquid fuel produced will increase.

In addition, with the same total reactor volume, unreacted syngas output Fischer Tropsch reactor will experience an increase in residence time. Residence time will increase the frequency of collisions between reactants, so that it will increase the conversion of reactors at the second stage as well as the mole fraction of the desired product.

4 Conclusions

The simulation results of the Gas to Liquid process using ASPEN HYSYS with reaction kinetics data taken from the references that have been obtained provide valid simulation results, where the resulting parameter output is the same and has a very small error or error value <1%. An increase in the steam-gas ratio and an increase in operating temperature on the ATR will result in an increase in the resulting H₂/CO syngas ratio. The staging effect on ATR will also increase the value of the H₂/CO ratio of the resulting syngas. The increase in

operating temperature on the Fischer Tropsch will reduce the liquid fuel gain and mole fraction yield to be obtained. Furthermore, staging on the reactor Fischer Tropsch will increase the actual conversion and mole fraction yield C9-C16.

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