

Reducing Energy Consumption of Methanol Production from Syngas by Modifying Heat Transfer Process

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Received: 19th December 2024; Revised: 28th December 2024; Accepted: 1st January 2025
Available online: 22th January 2025; Published regularly: June 2025



Abstract

Methanol is extensively used in the various products, including plastics, paints, adhesives, and synthetic fibers. With the many uses of methanol, its production must have high efficiency both in terms of energy and mass in order to obtain maximum profits. In this paper, we will explain how to reducing energy consumption and maximize methanol product yield by modifying the methanol synthesis. The process modification was carried out by adding heat exchanger, distillation column unit and recycle stream. Case study tools on chemical engineering software were used. Based on process modifications, an increase in methanol yield was obtained from 97.47% to 99%, the total energy savings from this process after adding distillation column is 57.618×10^6 kJ/h or 58.5% of the total initial energy, and with recycle stream the syngas produced reached 5.419 kgmol/h while without recycle stream, the syngas produced was only 4.157kgmol/h. The results of the case study indicate that the addition of heat exchanger, distillation column, and recycle stream is beneficial for the methanol production process by reducing the energy and increasing mass efficiency.

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Keywords: Methanol; Syngas; Energy Efficiency; Process Simulation; Reducing Energy Consumption

How to Cite: Fawwaz, A., Nugraha, A. P. A., Al Rasyid, M. P., & Muhammad, R. H. (2025). Reducing Energy Consumption of Methanol Production from Syngas by Modifying Heat Transfer Process. *Journal of Chemical Engineering Research Progress*, 2 (1), 122-131 (doi: 10.9767/jcerp.20307)

Permalink/DOI: <https://doi.org/10.9767/jcerp.20307>

Supporting Information (SI): <https://journal.bcrec.id/index.php/jcerp/article/downloadSuppFile/20307/5610>

1. Introduction

Methanol is a compound that currently plays an important role in the global economy as a versatile and important chemical, methanol serves as a base material for various products, including plastics, paints, adhesives, and synthetic fibers [1]. The global methanol market demand continues to grow with a CAGR (Compound Annually Growth Rate) of 5.2%, methanol consumption and production account for 10% of the total chemical sector. Global demand for methanol continues to increase, driven by its widespread use in various industrial applications and as a cleaner alternative fuel [2].

The formation of methanol from syngas consists of two reactions. The first reaction is the

process of forming syngas from natural gas (methane). Methane can be reacted with water vapor or oxygen to form syngas. The second reaction is the process of forming methanol from syngas. Syngas will be reacted in a reactor to form methanol at a certain pressure and temperature. The methanol mixture then undergoes a separation stage, namely distillation, to separate methanol from water and dissolved gases [3].

Based on previous studies, the gas reforming process used (SMR and ATR) produces hot gas that can be used for heating, but in the existing system, the use of heat exchangers for heat recovery may not be optimal. Some waste heat is still wasted without being fully utilized to reduce energy consumption in the next stage, such as in heating feed gas for reforming or the distillation process. Therefore, it is necessary to modify the process by adding a number of heat exchangers to

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recover heat from the hot gas stream coming out of certain processes, such as the reactor or reformer, and use it to heat the cooler stream [4]. By optimizing heat exchange between different process streams, heat exchangers can reduce the total energy requirements in the system, which will have a direct impact on reducing energy consumption and increasing efficiency [5]. In addition, a two-stage distillation column can reduce the energy consumption of the reboiler and condenser. In the first column (topping column), most of the volatile components (such as syngas or methane) are separated, while in the second column, methanol and water are separated more efficiently [6]. This reduces the amount of dilution and the need for external energy. The goal of this process modification is to minimize energy use and maximize methanol yield by modifying the methanol production process.

2. Material and Methods

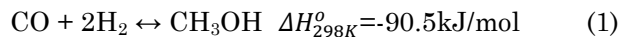
2.1 Partial Oxidation Process of Syngas Formation

Syngas is a mixed gas composed of carbon monoxide (CO), methane (CH₄), and hydrogen (H₂) [7]. Syngas can be further processed into various types of fuels and chemicals, one of which is methanol. Gasification is a thermochemical conversion method of solid fuel into syngas in a gasifier container by supplying gasification agents such as hot steam, air, and others [8]. The syngas production process uses partial oxidation where the reactions involved are exothermic and do not require energy to initiate. The syngas formation reaction is as follows. The syngas formation process involves reactants such as methane and air containing oxygen [9]. Both reactants are reacted in an equilibrium reactor in the gas phase. The selection of an equilibrium reactor is based on reactions that depend on equilibrium. This equilibrium is also highly influenced by pressure, temperature, and the molar ratio of reactants. Methane and air are introduced into the reactor with a molar ratio of 1:3. This molar ratio is chosen with an excess air composition so that more methane can be converted. The process is carried out at a pressure of 19.74 atm and a temperature of 1000 °C. Thermodynamically, the ATR process is conducted at a pressure of around 20 bar (19-20 atm) to resemble large-scale industrial processes [10]. Additionally, the reaction equilibrium will shift to the right with an increase in reaction temperature, resulting in more CO, CO₂, H₂, and H₂O. Therefore, a high temperature (1000 °C) is chosen for this syngas formation process to produce more syngas products [11].

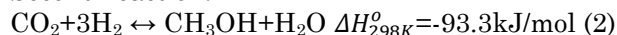
2.2 Chemical Reaction of Methanol Synthesis

Methanol synthesis is a complex process and typically involves catalysts to enhance the reaction rate. The chemical reaction for methanol synthesis is as follows:

First reaction:



Second reaction:

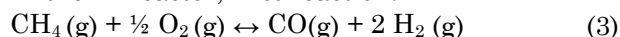


The production of methanol is highly influenced by thermodynamic equilibrium. This limits the process to low conversion, necessitating recycling of the outlet to achieve high conversion. The methanol synthesis reaction is notably exothermic, necessitating a cooling system to regulate the temperature within the reactor. To achieve complete conversion in a single pass, lower temperatures are essential. This requirement can be met through the low-temperature methanol synthesis (LTMS) process, which facilitates the conversion of nearly all syngas into methanol during each pass at temperatures ranging from 100 to 120 °C. The methanol production process utilizes syngas as a reactant, comprising CO, CO₂, and H₂. The reactants undergo reactions in a gas-phase equilibrium reactor, chosen specifically for reactions reliant on equilibrium conditions. The syngas generated from the prior process is directed into a second equilibrium reactor, where the operation occurs at a pressure of 19.4 atm and a temperature of 100 °C. These specific conditions are optimal because they enable the LTMS to convert almost all syngas into methanol per pass at these lower temperatures [12].

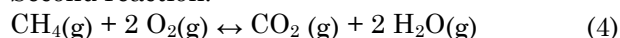
2.3 Thermodynamic Review

Thermodynamic review is used to determine the nature and direction of reactions, whether they are exothermic/endothermic and reversible / irreversible [13]. In the chosen process in this simulation, two reactors are used. Reactor 1 is the syngas formation reactor, and Reactor 2 is the methanol formation reactor.

In the 1st reactor, first reaction:



Second reaction:



The values of ΔH_f° for each component at a temperature of 298 K can be seen in Table 1. From the given data, the value of $\Delta H_{R, 298}^\circ$ values for the reactions are then obtained:

For the 1st reaction:

$$\begin{aligned} \text{CH}_4(\text{g}) + 1/2 \text{O}_2(\text{g}) &\leftrightarrow \text{CO}(\text{g}) + 2\text{H}_2(\text{g}) \\ \Delta H^\circ_{\text{R } 298} &= \Sigma \Delta H^\circ_{\text{f}(\text{product})} - \Sigma \Delta H^\circ_{\text{f}(\text{reactant})} \\ &= \Sigma ((\Delta H^\circ_{\text{f}} \text{CO} + 2 \Delta H^\circ_{\text{f}} \text{H}_2) - (\Delta H^\circ_{\text{f}} \text{CH}_4 + 1/2 \Delta H^\circ_{\text{f}} \text{O}_2)) \\ &= (-110.525 + 0) (74.5 + 1/2(0)) = -36.025 \text{ kJ/mol} \end{aligned}$$

For the 1st reaction 1, the $\Delta H^\circ_{\text{R } 298}$ value is -36.025 kJ/mol (negative), indicating that the reaction is exothermic. For the 2nd reaction:

$$\begin{aligned} \text{CH}_4(\text{g}) + 2\text{O}_2(\text{g}) &\leftrightarrow \text{CO}_2(\text{g}) + 2\text{H}_2\text{O}(\text{g}) \\ \Delta H^\circ_{\text{R } 298} &= \Sigma \Delta H^\circ_{\text{f}(\text{product})} - \Sigma \Delta H^\circ_{\text{f}(\text{reactant})} \\ &= \Sigma ((\Delta H^\circ_{\text{f}} \text{CO}_2 + 2 \Delta H^\circ_{\text{f}} \text{H}_2\text{O}) - (\Delta H^\circ_{\text{f}} \text{CH}_4 + 2 \Delta H^\circ_{\text{f}} \text{O}_2)) \\ &= (-393.509 + (-483.66)) - (-74.5 + 2(0)) = -802.7 \text{ kJ/mol} \end{aligned}$$

For the 2nd reaction 1, the $\Delta H^\circ_{\text{R } 298}$ value is -802.7 kJ/mol (negative), indicating that the reaction is exothermic. The values of $\Delta G^\circ_{\text{f}}$ for each component at a temperature of 298 K can be seen in Table 2.

For the 1st reaction:

$$\begin{aligned} \text{CH}_4(\text{g}) + 1/2 \text{O}_2(\text{g}) &\leftrightarrow \text{CO}(\text{g}) + 2\text{H}_2(\text{g}) \\ \Delta G^\circ_{\text{f } 298} &= \Sigma \Delta G^\circ_{\text{f}(\text{product})} - \Sigma \Delta G^\circ_{\text{f}(\text{reactant})} \\ &= \Sigma ((\Delta G^\circ_{\text{f}} \text{CO} + 2 \Delta G^\circ_{\text{f}} \text{H}_2) - (\Delta G^\circ_{\text{f}} \text{CH}_4 + 1/2 \Delta G^\circ_{\text{f}} \text{O}_2)) \\ &= (-137.168 + (-2(0))) - (-50.5 + 1/2(0)) = -86.668 \text{ kJ/mol} \end{aligned}$$

The K value of 1st reaction, Based on formula by Smith Van Ness [15]:

$$\begin{aligned} \ln K_{298} &= \left[\frac{-\Delta G^\circ_{\text{f}}}{TR} \right] \\ &= \left[\frac{-(-86.668) \left(\frac{\text{kJ}}{\text{mol}} \right)}{298 \text{ K} \times 8.314 \left(\frac{\text{kJ}}{\text{mol}} \right)} \right] \\ K_{298} &= 1.035 \end{aligned} \quad (5)$$

At a temperature of 1000 °C (1273.15 K), the equilibrium constant can be calculated as follows:

$$\ln \frac{K}{K_{298}} = \frac{-\Delta H^\circ_{\text{f}}}{R} \times \left(\frac{1}{T} - \frac{1}{T_{\text{ref}}} \right) \quad (6)$$

$$\begin{aligned} \ln \frac{K}{1.0236} &= \frac{-(-36.025)}{8.314} \times \left(\frac{1}{1273.15} - \frac{1}{298} \right) \\ K &= 1.0236 \end{aligned}$$

With an equilibrium constant value of 1.0236; it can be concluded that the 1st reaction occurs reversibly.

For the 2nd reaction:

$$\begin{aligned} \text{CH}_4(\text{g}) + 2 \text{O}_2(\text{g}) &\leftrightarrow \text{CO}_2(\text{g}) + 2\text{H}_2\text{O}(\text{g}) \\ \Delta G^\circ_{\text{f } 298} &= \Sigma \Delta G^\circ_{\text{f}(\text{product})} - \Sigma \Delta G^\circ_{\text{f}(\text{reactant})} \\ &= \Sigma ((\Delta G^\circ_{\text{f}} \text{CO}_2 + 2 \Delta G^\circ_{\text{f}} \text{H}_2\text{O}) - (\Delta G^\circ_{\text{f}} \text{CH}_4 + 2 \Delta G^\circ_{\text{f}} \text{O}_2)) \\ &= (-394.359 + (-2(-228.59))) - (-50.5 + 2(0)) = -801.039 \text{ kJ/mol} \end{aligned}$$

The K value of 1st reaction, based on formula by Smith Van Ness [15]:

$$\begin{aligned} \ln K_{298} &= \left[\frac{-\Delta G^\circ_{\text{f}}}{TR} \right] \\ &= \left[\frac{-(-801.039) \left(\frac{\text{kJ}}{\text{mol}} \right)}{298 \text{ K} \times 8.314 \left(\frac{\text{kJ}}{\text{mol}} \right)} \right] \\ K_{298} &= 1.381 \end{aligned}$$

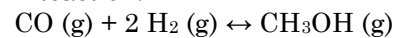
At a temperature of 1000 °C (1273.15 K), the equilibrium constant can be calculated as follows:

$$\begin{aligned} \ln \frac{K}{K_{298}} &= \frac{-\Delta H^\circ_{\text{f}}}{R} \times \left(\frac{1}{T} - \frac{1}{T_{\text{ref}}} \right) \\ \ln \frac{K}{1.0236} &= \frac{-(-802.669)}{8.314} \times \left(\frac{1}{1273.15} - \frac{1}{298} \right) \\ K &= 1.0776 \end{aligned}$$

With an equilibrium constant value of 1.0776; it can be concluded that the 2nd reaction occurs reversibly.

For the 2nd reactor, The reactions in reactor 2 are as follows:

1st Reaction:



2nd Reaction:

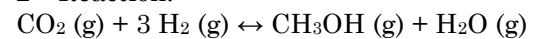


Table 1. Enthalpy of formation of components in 1st reactor [14].

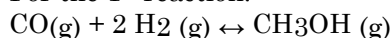
Compounds	Molecular Formulas	$\Delta H^\circ_{\text{f } 298}$ (kJ/mol)
Methane	CH ₄	-74.5
Oxygen	O ₂	0
Carbon Monoxide	CO	-110.525
Hydrogen	H ₂	0
Carbon Dioxide	CO ₂	-393.509
Water	H ₂ O	-241.83

Table 2. Gibbs' Energy of formation of components in 1st reactor [14].

Compounds	Molecular Formulas	$\Delta G^\circ_{\text{f } 298}$ (kJ/mol)
Methane	CH ₄	-50.5
Oxygen	O ₂	0
Carbon Monoxide	CO	-137.168
Hydrogen	H ₂	0
Carbon Dioxide	CO ₂	-394.359
Water	H ₂ O	-228.59

The values of ΔH_f° for each component at a temperature of 298 K can be seen in Table 1. From the given data, the value of $\Delta H_{R\ 298}^\circ$ is then obtained:

For the 1st reaction:



$$\begin{aligned}\Delta H_{R\ 298}^\circ &= \Sigma \Delta H_{f(\text{product})}^\circ - \Sigma \Delta H_{f(\text{reactant})}^\circ \\ &= \Sigma((\Delta H_f^\circ \text{CH}_3\text{OH}) - (\Delta H_f^\circ \text{CO} + 2 \Delta H_f^\circ \text{H}_2)) \\ &= (-201) - (-110.525 + 2(0)) = -90.475 \text{ kJ/mol}\end{aligned}$$

For the 1st reaction, the value of $\Delta H_{R\ 298}^\circ$ is -90.475 kJ/mol (negative), indicating that the reaction is exothermic.

For the 2nd reaction:

$$\begin{aligned}\text{CO}_{2(\text{g})} + 3\text{H}_{2(\text{g})} &\leftrightarrow \text{CH}_3\text{OH}_{(\text{g})} + \text{H}_2\text{O}_{(\text{g})} \\ \Delta H_{R\ 298}^\circ &= \Sigma \Delta H_{f(\text{product})}^\circ - \Sigma \Delta H_{f(\text{reactant})}^\circ = \Sigma((\Delta H_f^\circ \text{CH}_3\text{OH} + \Delta H_f^\circ \text{H}_2\text{O}) - (\Delta H_f^\circ \text{CO}_2 + 3 \Delta H_f^\circ \text{H}_2)) \\ &= (-201 + (-241.83)) - (393.509 + 3(0)) = -49.321 \text{ kJ/mol}\end{aligned}$$

For the 2nd reaction, the value of $\Delta H_{R\ 298}^\circ$ is -49.321 kJ/mol (negative), indicating that the reaction is exothermic. The values of ΔG_f° for each component at a temperature of 298 K can be seen in Table 2.

For the 1st reaction:

$$\begin{aligned}\text{CO(g)} + 2 \text{H}_2 \text{(g)} &\leftrightarrow \text{CH}_3\text{OH (g)} \\ \Delta G_{f\ 298}^\circ &= \Sigma \Delta G_{f(\text{product})}^\circ - \Sigma \Delta G_{f(\text{reactant})}^\circ \\ &= \Sigma((\Delta G_f^\circ \text{CH}_3\text{OH}) - (\Delta G_f^\circ \text{CO} + 2 \Delta G_f^\circ \text{H}_2)) \\ &= (-162.5) - (-137.168 + 2(0)) = -25.332 \text{ kJ/mol}\end{aligned}$$

The K value of 1st reaction, based on formula by Smith Van Ness [15]:

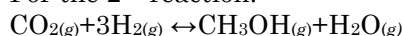
$$\begin{aligned}\ln K_{298} &= \left[\frac{-\Delta G^\circ F}{TR} \right] \\ &= \left[\frac{-(-25.332) \left(\frac{\text{kJ}}{\text{mol}} \right)}{298 \text{ K} \times 8.314 \left(\frac{\text{kJ}}{\text{mol}} \right)} \right] \\ K_{298} &= 1.010\end{aligned}$$

At a temperature of 100 °C (373.15 K), the equilibrium constant can be calculated as follows:

$$\begin{aligned}\ln \frac{K}{K_{298}} &= \frac{-\Delta H^\circ r}{R} \times \left(\frac{1}{T} - \frac{1}{T_{ref}} \right) \\ \ln \frac{K}{1.0236} &= \frac{-(-90.475)}{8.314} \times \left(\frac{1}{373.15} - \frac{1}{298} \right) \\ K &= 1.0174\end{aligned}$$

With an equilibrium constant value of 1.0174; it can be concluded that the 1st reaction occurs reversibly.

For the 2nd reaction:



$$\begin{aligned}\Delta G_{f\ 298}^\circ &= \Sigma \Delta G_{f(\text{product})}^\circ - \Sigma \Delta G_{f(\text{reactant})}^\circ \\ &= \Sigma((\Delta G_f^\circ \text{CH}_3\text{OH} + \Delta G_f^\circ \text{H}_2\text{O}) - (\Delta G_f^\circ \text{CO}_2 + 3 \Delta H_f^\circ \text{H}_2)) \\ &= (-162.5 + (-228.59)) - (-394.359 + 3(0)) = -3.269 \text{ kJ/mol}\end{aligned}$$

The K value of 2nd reaction, based on formula by Smith Van Ness[15]:

$$\begin{aligned}\ln K_{298} &= \left[\frac{-\Delta G^\circ F}{TR} \right] \\ &= \left[\frac{-(3.269) \left(\frac{\text{kJ}}{\text{mol}} \right)}{298 \text{ K} \times 8.314 \left(\frac{\text{kJ}}{\text{mol}} \right)} \right] \\ K_{298} &= 0.998\end{aligned}$$

At a temperature of 100 °C (373.15 K), the equilibrium constant can be calculated as follows:

$$\begin{aligned}\ln \frac{K}{K_{298}} &= \frac{-\Delta H^\circ r}{R} \times \left(\frac{1}{T} - \frac{1}{T_{ref}} \right) \\ \ln \frac{K}{1.0236} &= \frac{-(-49.321)}{8.314} \times \left(\frac{1}{373.15} - \frac{1}{298} \right) \\ K &= 0.9939\end{aligned}$$

With an equilibrium constant value of 0.9939; it can be concluded that the 2nd reaction occurs reversibly.

3. Result and Discussion

3.1 Comparison Between Basic and Modified Processes

The simulation of methanol production from syngas through partial oxidation for basic and modified process using Aspen HYSYS is depicted in Figure 1 to Figure 4. The basic process simulation is shown in Figure 1, while the PFD of the basic process is shown in Figure 3. In the basic process, there is no heat exchanger is used before entering the separator. Whereas the modified process simulation is shown in Figure 2, while the PFD is shown in Figure 4. In the modified process, a heat exchanger is added to cool the syngas from the ERV-100 unit and cool the exhaust gas in the T-100 unit. For the modified process, there is an additional distillation column which is used to separate the exhaust gas from reactor 2 in order to obtain methanol that enters the exhaust gas so that more methanol is produced and increases the yield. The addition of recycle flow is also added from the top of the X-100 unit to separate the methane still contained in the syngas from the reaction results in the ERV-100 unit and then mixed with the methane feed in the MIX-100 unit to increase the amount of feed and increase mass efficiency. The difference between those two is in the addition of a heat transfer, distillation

column, and recycle current which can be seen in the modified process.

3.2 Effect of Adding Heat Exchanger for Reducing Energy Consumption

The addition of heat exchangers aims to cool the syngas from the ERV-100 unit and cool the exhaust gas in the T-100 unit. At the top of the equilibrium reactor 1 ERV-100, syngas will be passed through a Heat Exchanger to cool the feed so that it is expected that the syngas feed before entering the separator already has a temperature close to 100 °C and reduces the energy load on the separator for cooling and energy used more efficiently. At the top of the T-100 separator, the syngas is passed through a heat exchanger to cool the flue gas so that the heat energy from the flue gas can be recovered.

Based on the calculation, the power requirement of the separator without heat exchanger is higher than the power requirement of the separator using heat exchanger. In the process without Heat Exchanger, the power requirement of the separator is 1.145×10^4 kJ/h and requires a cooler power of 8.772×10^8 kJ/h.

While the separator power requirement in the process with the Heat Exchanger is 2.515×10^6 kJ/h without the need for cooler. So, it can be concluded that the process with the addition of Heat Exchanger runs more efficiently [17].

3.3 Effect of Adding Distillation Column to Maximizing Methanol Product Yield

The addition of the T-100 distillation column is useful for separating residual gas from reactor 2 to obtain methanol that is included in the exhaust gas. The methanol produced from the T-100 unit will then be mixed with methanol from the ERV-101 unit. With this distillation column, more methanol will be produced and increase the production yield [18]. Based on the simulation with Aspen HYSYS V.12, the conversion of syngas to methanol product produced through the process with the addition of distillation column has a higher amount than the process without the addition of distillation column. Without the addition of a distillation column, the conversion only reached 97.47%. Whereas with the addition of a distillation column, the conversion can reach 99%. Thus, it can be concluded that the addition

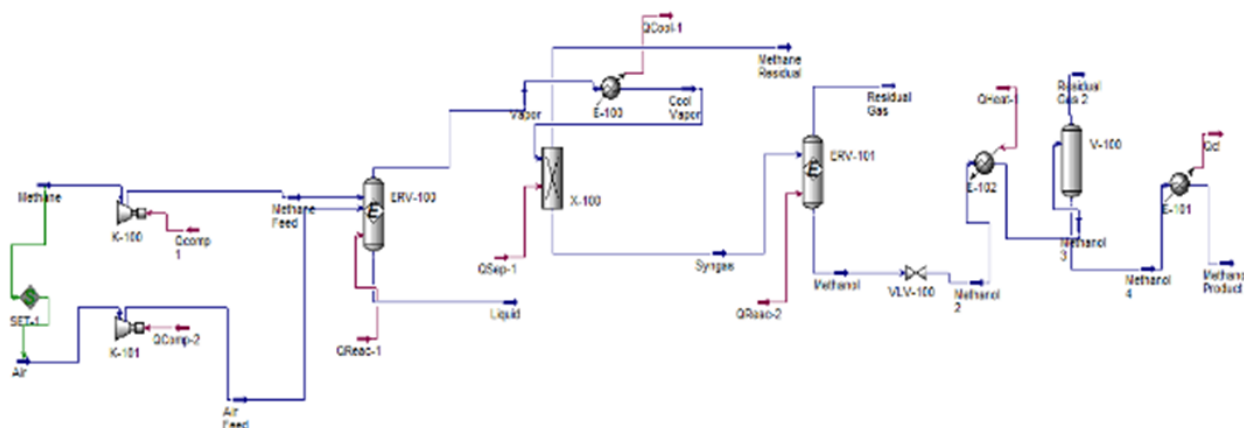


Figure 1. Simulation using Aspen HYSYS of basic (unmodified) process

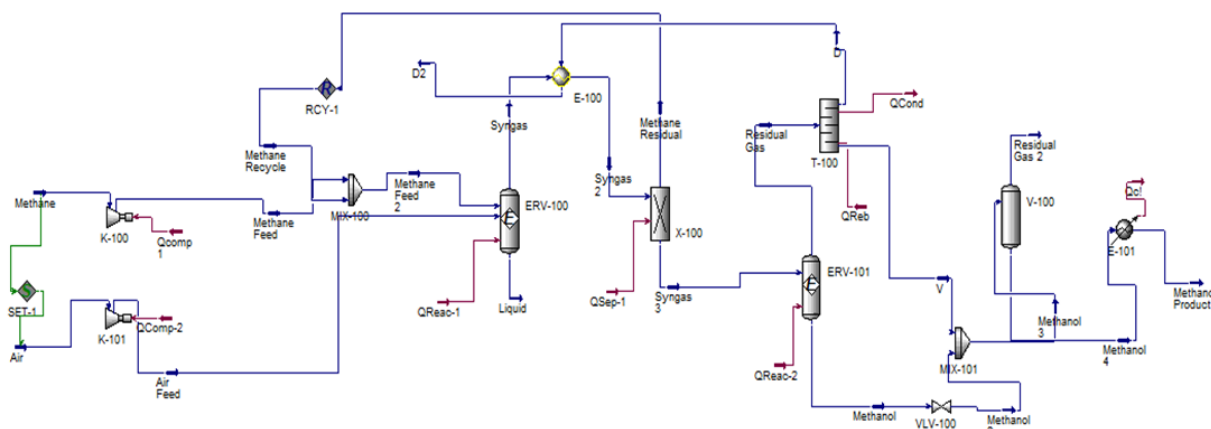


Figure 2. Simulation using Aspen HYSYS of modified process

4.157 kgmol/h. Thus, the addition of recycle stream leads to better and optimized mass efficiency [21].

3.5 Mass Balance and Energy Balance Results

The Table S1 (Supporting Information) are the results of the calculation of mass balance and energy balance in the process of methanol formation from syngas after the creation of the process. The data obtained is the result of process simulation using Aspen HYSYS V.12.

3.6 Calculation Results of Heating Media and Cooling Media Requirements

The Table 6 and Table 7 are the results of the calculation of heating and cooling media requirements cooling media in the process of forming methanol from syngas before after process creation. The data obtained is the result of simulation using Aspen HYSYS V.10.

4. Conclusions

The process creation can enhance process efficiency and conversion. The addition of a Heat Exchanger to cool the syngas before entering the separator can reduce the power demand of the separator and decrease the need for a cooler to cool the feed, making the energy required more efficient. The total energy savings from this process is 58.238×10^6 kJ/h or 58.5% of the total initial energy. Adding a distillation column to separate methanol from the exhaust gas can enhance conversion and the amount of methanol produced, thus improving mass efficiency. The introduction of a recycle stream from the overhead product of the separator, which produces methane, increases the amount of feed entering the first reactor. This leads to a higher production of syngas products because the equilibrium shifts to the right. With more syngas products, the production of methanol also increases, resulting in improved mass efficiency.

Through the research done within this paper, it can be seen that the addition of heat exchanger, distillation column, and recycle stream is beneficial for the methanol production process by reducing the energy and increasing mass efficiency.

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Tabel 6. Calculation results of heating media and cooling media requirements before process modification.

Energy Streams									
		Qcomp 1	QReac-1	QSep-1	QReac-2	Qc	QComp-2	QCool-1	QHeat-1
Heat Flow	kJ/h	1.371e+007	-5.419e+006	-1.211e+008	-6.779e+007	0.0000	4.623e+007	3.584e+007	0.0000

Tabel 7. Calculation results of heating media and cooling media requirements before process modification

Energy Streams									
		Qcomp 1	QReac-1	QSep-1	QReac-2	QCond	QReb	Qc	QComp-2
Heat Flow	kJ/h	1.371e+007	5.920e+006	-2.515e+006	-7.482e+007	-1.457e+008	1.131e+008	3.784e+006	4.623e+007

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