

Optimization of Energy Consumption in Formaldehyde Production Process Using Reboiled Absorption Process

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Abstract

Formaldehyde is a crucial chemical building block in various industries, and its production often involves energy-intensive processes. This study focuses on optimizing energy consumption in formaldehyde production, explicitly employing the reboiled absorption process with a production capacity of 27,000 tons per year. The objective of this article is to develop a more energy-efficient process of formaldehyde synthesis by addition or refrigerant cycle stream to preserve the energy, reducing energy consumption and improving the sustainability of the process. The reboiled absorption process involves the absorption of formaldehyde gas into a liquid absorbent, followed by reboiling to release the absorbed formaldehyde. A comprehensive analysis of the entire production system compares unmodified and modified process simulations, heat integration, and energy analysis. Beside the energy consumption of the process, the number of stages within the absorption process contributes to the product mass flow rate of the overall process by increasing the surface area which mass transfer can occur. However, adding too many stages to the process may negatively impact the energy efficiency of the process. Therefore, optimizing energy consumption and absorption processes in formaldehyde production is essential to improve the sustainability of the process and increase the overall profitability of the production process. The results show that the proposed method dramatically improves the sustainability of CH₂O synthesis by reducing overall energy consumption and emissions by 93.978%, reducing energy consumption from 153,735,360.4 kJ/h to 9,256,646.618 kJ/h.

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1. Introduction

The production of formaldehyde, a crucial material in various industries [1], is a resource-intensive process that generates significant amounts of waste and contributes to environmental pollution [2]. One of the ways to

address these concerns is by optimizing the energy consumption in the reboiled absorption process, which has a capacity of 27,000 tons/year. This process has been the subject of numerous studies [3,4], which have explored various aspects such as mathematical modeling, process control, and waste reduction. However, most of these studies have focused on improving the methane steam reforming [4] and loss in the formaldehyde

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synthesis process [3] to reduce costs without considering the entire production process. This paper aims to fill this gap by providing a comprehensive analysis of the energy consumption optimization in the formaldehyde production process using the reboiled absorption process capacity of 27,000 tons/year. By reducing the energy consumed by the production process, less emission will be released into the atmosphere and profits can be increased by reducing cost associated with energy.

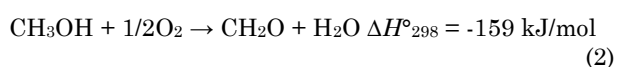
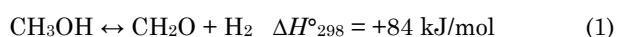
The objectives of this study are to identify the best methods of reducing the overall energy consumption of formaldehyde production process and the optimal stages of the absorption process to increase profitability and reduce emissions by increasing energy efficiency.

2. Methods

2.1. Basic Chemical Process Description of Formaldehyde Production

Formaldehyde is one of the simplest molecules in organic chemistry and is widely used in the industrial world. It is used as an additive to tanning agents in the leather industry, a biocide of latex in the rubber industry, a preservative in the lumber industry, and many more. Formaldehyde is also a key component in synthesizing industrial resins, such as melamine-formaldehyde, urea-formaldehyde, and phenol-formaldehyde, which are widely used. Thus, the demand for formaldehyde steadily increases as industrial complexes grow around it. As the demand for it increases, so does the need to fulfill it, making industrial production of formaldehyde even more critical [5].

Formaldehyde can be synthesized through various means with several methods and reaction types. The general sequence of formaldehyde production is as follows: production of synthesis gas is done through steam reforming of natural gas, which is converted to methanol via hydrogenation of carbon monoxide, and eventually, partial oxidation of methanol that has been produced leads to the production of formaldehyde. Formaldehyde production is done through two main methods: the dehydrogenation of methanol and the partial oxidation of methanol through the following reaction:



The most common methanol oxidation production methods are the BASF method and the Formox method [5–7]. The BASF process uses a silver catalyst supported by alumina to increase

and facilitate the oxidation process of methanol to formaldehyde under mild conditions. The methanol feed is processed with silver contact, dehydrogenation, and partial oxidation reactions to produce formaldehyde. The BASF process was widely used in large-scale industrial formaldehyde production because of its higher conversion rate. The Formox process, on the other hand, utilizes a molybdenum-iron (Mo-Fe) catalyst [8]. The Formox used a process in which only partial methanol oxidation occurs. This results in better stability and reliability caused by a two-step reaction, including methanol oxidation to formaldehyde and subsequent oxidation to formic acid. The Formox reaction also generally run at higher temperature and pressure [9].

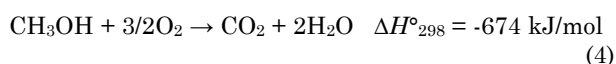
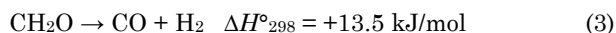
The BASF method is one of the first industrially viable methods of producing formaldehyde on a large scale [4]. Developed in 1923, it initially used gauze made from silver wire as a catalyst. Now, the BASF method is almost exclusively run with the help of a shallow bed of silver crystal as its catalyst [3]. On the basic level, the BASF method is a single pass, almost complete conversion of methanol method. Typically, it is run at a temperature of 600 to 700 °C, and formaldehyde yields were as high as 87.5 %. The diagram of the BASF process can be seen in Figure 1.

As can be seen from the diagram above, a fresh mixture of methanol and water enters an evaporation chamber along with air. In this stage, recycled off-gas from the absorption column is also mixed with the fresh feed so that part of the leftover heat from the absorption column can be used to help the evaporation process. After that, the evaporated mixture goes through a superheating process before filtering and fed into the reactor. As explained before, the catalyst used in this process is a fixed bed of silver crystals placed vertically within the reactor walls. The top product of the reactor is then recycled back into the superheater and the bottom product goes into the first absorption column. It is possible to use a cooler right after the reactor to stop any further reaction that may cause unwanted side products.

To increase the selectivity of the reaction and the production of formaldehyde, some variables can be adjusted. This includes the adjustment of the reaction temperature [6]. This can be done in several ways, one of them is through the control of the evaporation temperature but this may affect the composition of the feed and have very little impact after the superheating process. Another way to increase the selectivity of the process is that the ratio of water to methanol can be increased. This way, an optimal temperature of the reaction can be maintained alongside the increase of the reaction's selectivity [7]. This may increase the operating cost of the entire system;

thus, a balance must be made so that water can be added to the mixture without drastically affecting the final product. It is suggested that a mass ratio of water to methanol of 2:3 can be used as the optimal ratio [8].

The use of silver catalyst and the optimization of temperature and feed ratio is done to increase the selectivity of the catalyst. This means suppressing the side reactions attributed to the direct dehydrogenation of methanol. The reactions that may occur include in Equations (3-5).



As can be seen from the reaction above, carbon monoxide, carbon dioxide, water, and hydrogen are common side products of the dehydrogenation of methanol. Methyl formate and formic acid can also be produced through certain reactions. Although the products of these reactions can be separated and the resulting formaldehyde purified, the presence of these by-products can decrease both the conversion and the speed at which formaldehyde can form [3].

The Formox process or sometimes called metal oxide process, utilizes an iron molybdate catalyst (FeMo) unlike the BASF method that uses a silver catalyst [4]. The Formox process uses endothermic dehydrogenation and oxidative dehydrogenation of methanol to produce formaldehyde products. Formox process occurs

under atmospheric pressure with the temperature of 250-400 °C [3]. To promote the production of formaldehyde, air is fed in excess amount into the reaction process. Under these conditions, MeOH can produce formaldehyde with around 99% conversion and 88-92% yield [5]. The process diagram of the Formox method can be seen in Figure 2. In this figure, the methanol feed passes a steam evaporator and enters the reactor. The excess air is used to cool the product of the reactor as part of its pre-treatment stage before entering and mixing with the methanol in the steam evaporator mentioned above. As stated before in the BASF process, the cooling of reactor product is done to stop further reactions that may produce unwanted by-products. This is because the excess of air within the Formox method can cause excess amount of carbon monoxide to form as a product of side reaction. This treatment reduces conversion rate and the formaldehyde yield within the process. To control this phenomenon, variables and system operation must be adjusted and optimized accordingly.

One method is by controlling feed temperature within the reactor. Similar to the BASF method, increasing the temperature of the reaction may increase its selectivity. Other than unwanted deterioration and calcination of feed components, the presence of excess air within the Formox method made it overly sensitive to temperature increase. This is why generally, the temperature of the Formox process does not exceed 743.15 K. A balance of acceptable temperature that promotes improvements of reactions kinetics while keeping the selectivity of

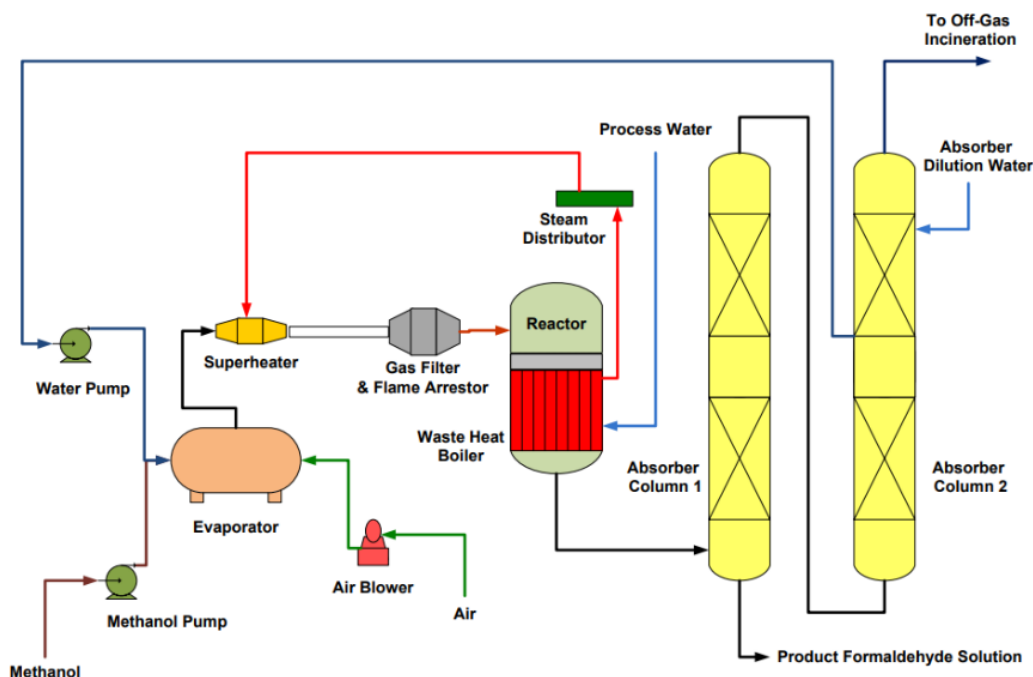


Figure 1. BASF Method Process

reaction high must be struck and thus the Formox process operates at a temperature of 270-400 °C [3].

The lower temperature and pressure may reduce the need for maintenance of the system [9]. This treatment reduces the capital need for maintenance operation and increase the overall yield of the production because of the continuous operation of the system. The lower temperature may also benefit the health of the catalyst used, this is because lower pressure and temperature leads to a more stable and predictable reaction which benefits the regeneration of the catalyst used in the system [2]. This treatment reduces the need to exchange the catalyst for a new unused one, reducing operational cost.

2.2. Energy Optimization through Process Modification

In an industrial area, energy is one of its more important components. This is because virtually every single one of its operations requires some sort of energy. It is imperative that an industrial operation has access to an energy source of some sort. This may include energy in the form of electricity or its other latent form, this includes fuel, coal, charcoal, firewood, or others [9]. This method help ensuring production runs smoothly and continuously. The presence of energy also ensures the ability to use and utilize

more advanced modes of processing which may need more and more stable energy sources [4–6,10]. This treatment increases the economic viability of a production line by increasing the number of products that can be produced in a given time [6]. Alongside the growth of the world’s industrial sector, the need for energy grows alongside it, which proves to be a challenge both economically and ecologically [2].

One way energy can be conserved is through optimization of the production process itself [5]. Optimization can be achieved in several ways, one of them is to increase the efficiency of the equipment used in the process itself. Even then, there are some challenges attributed to it, one of them is that efficiency can only go so far to decrease the energy consumed by the production process [10]. Other than that, a production company or facility usually prefers to increase the rate of production whilst keeping energy consumption the same thus increasing profits that can be earned. The other way energy can be conserved is through creating a process in which energy can be conserved in the process itself [3]. This can be achieved by creating efficient cycles of both products and other utilities that use their own unwanted byproducts to power or feed to another process within the system to increase its efficiency. This way, production rate can be increased while energy consumption is decreased [11].

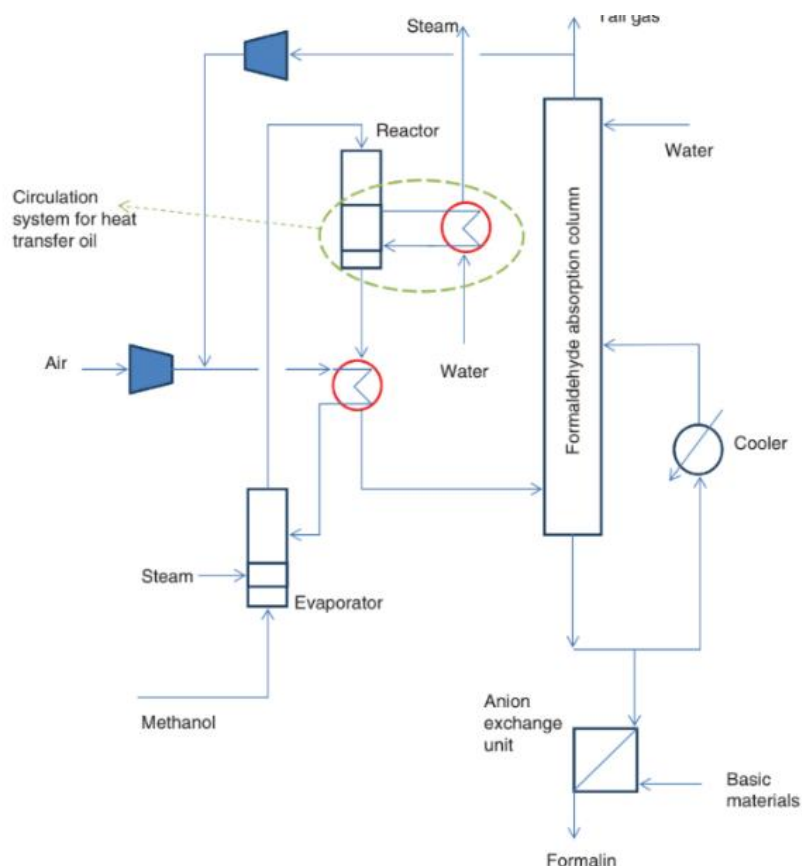


Figure 2. Formox Method Process

This paper focuses on the modification of the pretreatment of the reactor feed and its subsequent product. The feed of the reactor is to be heated as a pretreatment process to increase the overall yield of the process. Through several exothermic chemical processes that is going to happen within the reactor, the feed is then transformed into a hot mixture of formaldehyde and other by-products which need to be cooled down to not damage other components and equipment within the production line, this process can be seen in Figure 3. As can be seen from the figure, the unmodified process uses a series of pumps, heaters, and coolers to process water so that it can be used in the pre-treatment process as a heater or as a coolant for the reactor products. Arrangements such as these are inefficient, redundant, and expensive to run. The modified process (Figure 4) seeks to remedy this issue by combining the pre-treatment and product cooling process into a single refrigerant-heater cycle as can be seen on Figure 4. A refrigerant-heater cycle such as these can eliminate the need of multiple heaters and coolers by using the residual heat from the product which turns the cooling water into a heater that can be used in the pre-treatment process. Only a single cooler and pump is necessary to process the water back into a suitable cooler, thus completing the cycle. With this cycle, the water required to effectively run the system is also reduced, further reducing the operational cost of the system.

One of the most important steps in the production of formaldehyde is the absorption process that needs to be done to separate formaldehyde products from its impurity. On a basic level, absorption is a technique that separates gas or liquid mixture using mass transfer of one or several components from the feed into a solvent liquid to purify it from unwanted by-products or impurities. Most formaldehyde production uses this technique because the components of the reactor products have similar boiling points, rendering other methods of separation like distillation ineffective. Several solvents can be used in this process depending on the nature of its feed. Water in the form of distilled or deionized water is used to aid the absorption process of Formaldehyde to reduce further contamination.

Mass transfer within the absorption column takes place when the elements are specifically absorbed by the liquid phase, resulting in the gas or liquid leaving the column being separated and purified. Following this, the absorbent, which now holds the absorbed component, undergoes a regeneration process in a distinct unit to ready it for reuse in the absorption column. Designing absorption columns requires meticulous attention to factors like the selection of the absorbent, determination of column height, choice of packing material, and optimization of operating conditions, all aimed at enhancing the effectiveness of the separation process. One of

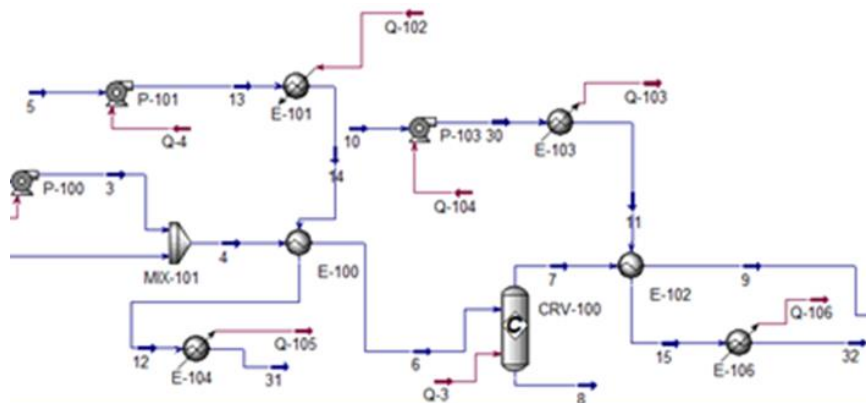


Figure 3. Unmodified pre-treatment and product cooling process

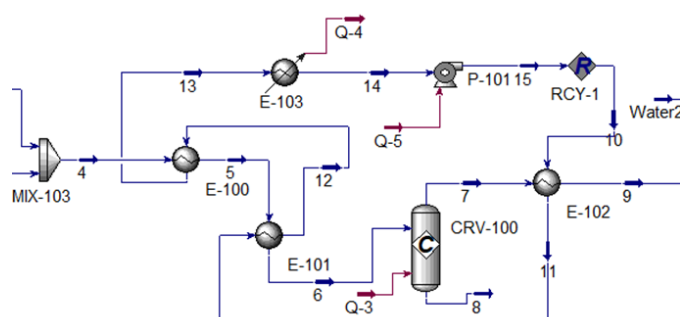


Figure 4. Modified pre-treatment and product cooling process

them is the adjustment of a column absorber's tray count.

The number of trays in an absorber, such as an absorption column used in the synthesis of formaldehyde, significantly influences the separation and refinement of components in gas or liquid mixtures. Augmenting the tray count improves mass transfer by increasing the contact surface between gas and liquid phases, thereby enhancing the efficiency of component separation. This heightened efficiency arises from multiple equilibrium stages, where each tray contributes to achieving partial equilibrium. Nevertheless, the advantages are accompanied by considerations, as a greater tray count may necessitate a taller absorption column. This, in turn, can impact energy consumption and introduce intricacy and cost to the design. Engineers must adeptly balance these factors to optimize the effectiveness and cost-efficiency of absorbers across a range of industrial applications.

3. Results and Discussion

3.1. Simulation of Basic Chemical Process and Modified Chemical Process.

To simulate and demonstrate the production and optimization of the formaldehyde synthesis process, Aspen HYSYS software was used. The HYSYS software can be used to simulate, optimize, and also demonstrate a chemical reaction process either as a one-off unit or as a system as a whole. This makes the process of optimization and analyses of the system far more flexible and thorough, as we can see the effects of individual unit's specifications and its effect on the production process. But there are still some limitations with HYSYS that must be mitigated in some way. The HYSYS itself does not have a few processes and conditions that must be specified in the production of formaldehyde, and thus some creative liberties have been taken to be able to implement the optimization that we want to do within the scope of this literature. The basic process diagram to be simulated is depicted in Figure 5.

As can be seen on the diagram above, fresh feed of air and methanol is to be compressed and pumped into the system before getting mixed, heated and reacted into the reactor. The result of which is to be cooled before being absorbed with water in the absorption column and the reboiled absorber column. The product of the absorption process is to be mixed with a fresh feed of deionized water to set the concentration of the formaldehyde products to 37%. The top products of the absorber are to be released as off gas or to be separated and recycled as new feed for the reactor or returned to the absorption tower to extract the remaining formaldehyde.

The process pre-treatment of the feed and the cooling of the reactor products require a substance to transfer the heat to or from the product/feed as a heater or a cooler. Water is usually used as an easy to obtain and generally non-corrosive medium to heat and cool the feed and products. The problem presented by the diagram above is the repetitive heating and cooling of the water so that it can be used in the system and so that it can be safely discharged or contained. This leads to unnecessary energy consumption that raises the emission caused by the use of more energy and unwanted cost associated with it. One of the optimizations that can be done is to utilize a loop system which allow the residual heat from the reactor's product to be used to heat the feed as its pre-treatment process. The new, optimized and modified process diagram can be to be simulated is depicted in Figure 6.

As can be seen from the Figure 6 that the water from each heat exchanger is connected in a serial manner, which allows the water to be reused as a cooler or heater in each process. This can be done as the residual heat from the cooler of the reactor's product is carried by the water into the heat exchanger before the reactor to pre-treat the feed before entering the reactor. Two exchangers are used to avoid temperature shock that may be causing the excess heat the water carrier way; only one unit of cooler and pump is needed to run the system as the water is cooled

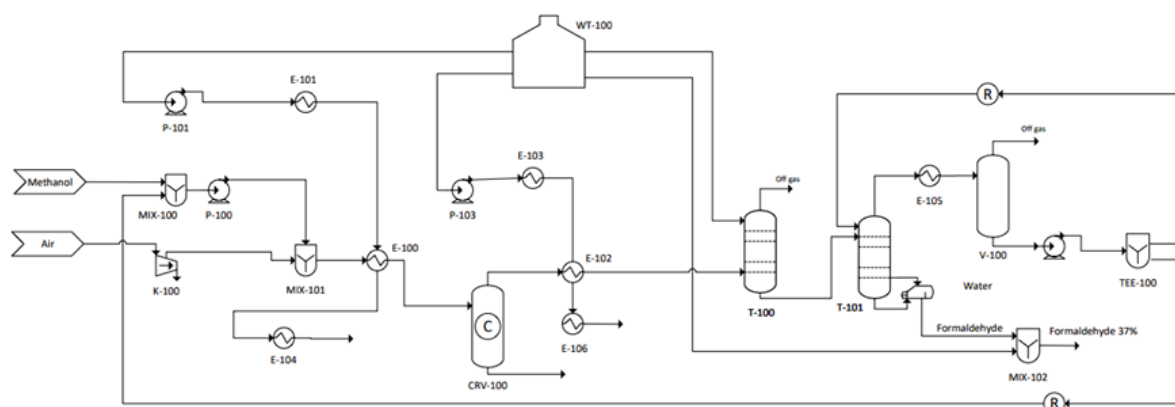


Figure 5. Unmodified process flow diagram to be simulated

and recycled into the heat exchanger after the reactor is reused again. Not only does it conserve the energy used by the process, but it also reduces the need for water, as the loss of water caused by the loop is minimal compared to the continuous flow needed before. Both diagrams (Figures 5 and 6) were simulated using Aspen HYSYS to estimate the energy that can be saved from this optimization process. Energy saving by analyzing the heat flow between each system as an analog of the system's energy consumption. Both systems are to use the exact specification and feed basis to eliminate unwanted variables from the analysis process.

3.2. Modification of Reboiled Absorption Process in Formaldehyde Production

In the HYSYS simulation, the synthesis process of formaldehyde must be set up with its reactions in mind. This means that we have to define every reaction and side reactions within the process. Besides that, the simulation process is straight forward. As the basis of production, a feed of 6,000 kg/h of air containing 79% Nitrogen and 21% oxygen and 4,000 kg/h of pure methanol. This excess addition of air is to promote the production of formaldehyde rather than other side reactions. The HYSYS simulation of process flow diagram of both unmodified and modified processes and their differences can be seen in Figures 7 and 8, while mass and energy balances are presented in Table S1 (Supporting Information).

As stated before, in the simulation process, a conversion reactor is used. Unlike other reactor such as batch reactor or plug flow reactor,

conversion reactor uses a basis or known conversion rate as a dummy for the reaction process itself. It is estimated that the rate of conversion within the system is 49% for its main reaction. The reactions within the production of formaldehyde follows Equations (3-5).

The diagram shows that the synthesis process begins with the compression and pumping of methanol and air feed before mixing them alongside the recycle stream and going to the pretreatment process. Here, the temperature of the feed mixture is to be raised to 120 °C. This is because the formation of formaldehyde happens more effectively in higher temperatures. After that, the reactor products will be cooled with a heat exchanger system before going through the two absorption columns. Here, the stages are set to the optimal step of 30. The bottom product of the absorption process is a mixture of formaldehyde and water, which is watered down to formaldehyde 37%. The top product of the absorption column is to be separated with a flash drum before being recycled as the feed of the second absorption column to extract its formaldehyde contents further or as reactor feed to convert the leftover methanol within the feed.

It can be seen in the modified process that a system loop of three heat exchangers (E-102, E-101, E-100) is connected to cooler E-103 and a pump to function as both a refrigerant and heater cycle. The water within this loop will leech residual heat from the reactor's product within E-102 for pre-treatment reactor feed. This reduces

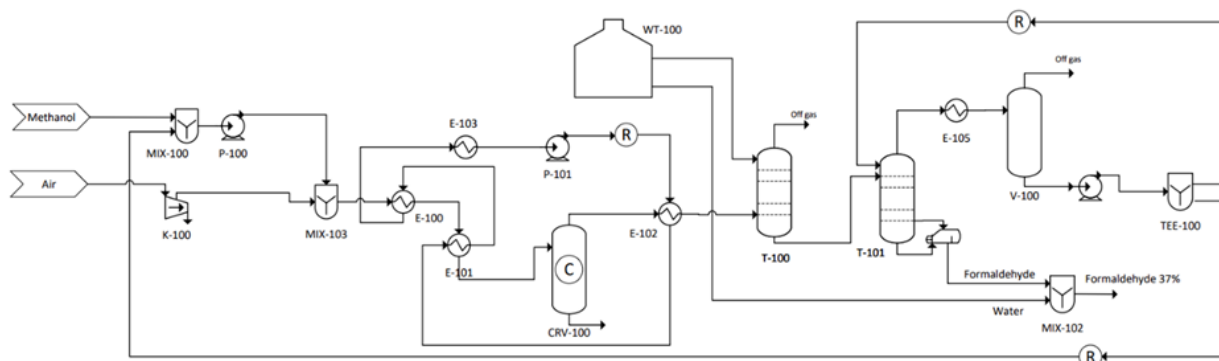


Figure 6. Modified process flow diagram to be simulated

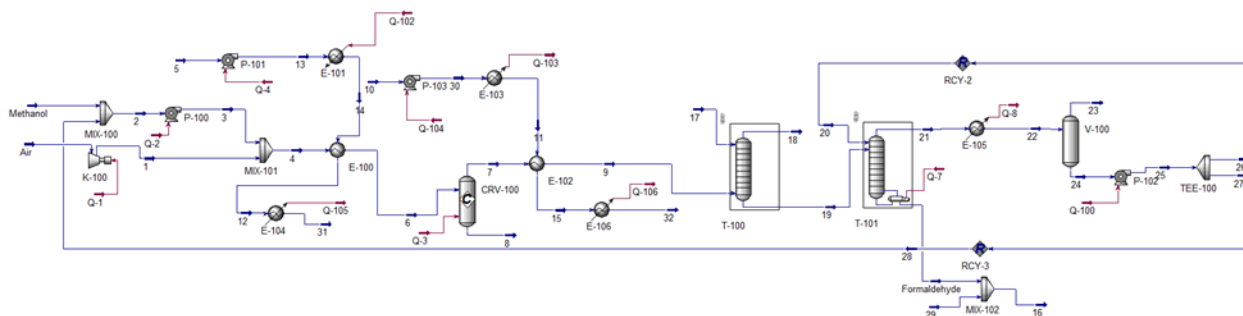


Figure 7. Unmodified/basic process flow diagram from HYSYS simulation

the need for additional heaters or more excellent components and allows the system to reduce the energy required to run and reduce utility costs [18].

3.4. Improving Energy Efficiency by reducing Net Energy Required

From the simulation, it can be obtained data of the total energy that's needed by the system. This energy is measured by heat flow (kJ/h) of the system itself. This method serves as an analogue for how much energy can be conserved compared to the unmodified system. From there we can measure the energy saved by the modified process mathematically and measure its efficiency. The heat stream data obtained from the simulation is presented in Table 1.

$$\Delta H_{Unmodified} = H_{Q-1} + H_{Q-2} + H_{Q-3} + H_{Q-4} + H_{Q-7} + H_{Q-8} + H_{Q-100} + H_{Q-102} + H_{Q-103} + H_{Q-104} + H_{Q-105} + H_{Q-106}$$

$$\Delta H_{Unmodified} = 153,735,360.4 \text{ kJ/h}$$

$$\Delta H_{Modified} = H_{Q-1} + H_{Q-2} + H_{Q-3} + H_{Q-4} + H_{Q-7} + H_{Q-8} + H_{Q-100}$$

$$\Delta H_{Modified} = 9,256,646.618 \text{ kJ/h}$$

$$\text{Energy reduction} = 153,735,360.4 \frac{\text{kJ}}{\text{h}} - 9,256,646.618 \text{ kJ/h}$$

$$\text{Energy reduction} = 144,478,713.8 \text{ kJ/h}$$

$$\% \text{Energy Efficiency} = \frac{\Delta H_{Unmodified} - \Delta H_{Modified}}{\Delta H_{Unmodified}} \times 100\%$$

$$\% \text{Energy Efficiency} = \frac{153,735,360.4 \text{ kJ/h} - 9,256,646.618 \text{ kJ/h}}{153,735,360.4 \text{ kJ/h}} \times 100\%$$

$$\text{Therefore, \%Energy Efficiency} = 93.978\%$$

3.5. Absorption Column Optimization

One of the most important steps in the synthesis of formaldehyde is the absorption process. The goal of the absorption process is to separate the formaldehyde products of the reactor from its by-products [12]. This is to ensure that the formaldehyde products obtained in the end of the process is pure and free of unwanted components and can be utilized in various industries such as the production of fertilizer, paper and urea-formaldehyde resin [13]. Several

Table 1. Energy analysis of the unmodified and modified processes

| Unmodified Process | | Modified Process | |
|--------------------|------------------|-------------------|------------------|
| Heat stream items | Heat flow (kJ/h) | Heat stream items | Heat flow (kJ/h) |
| Q-1 | 885,831 | Q-1 | 885,831 |
| Q-2 | 1,968.06 | Q-2 | 1,968.06 |
| Q-3 | 225,272 | Q-3 | 225,083 |
| Q-4 | 6,693.86 | Q-4 | 1,606,062 |
| Q-7 | 3,321,074 | Q-5 | 527.856 |
| Q-8 | 3,219,086 | Q-7 | 3,319,596 |
| Q-100 | 35.6754 | Q-8 | 3,217,543 |
| Q-102 | 74,280,962 | Q-100 | 35.658714 |
| Q-103 | 1,688,889 | | |
| Q-104 | 720.942 | | |
| Q-105 | 62,943,811 | | |
| Q-106 | 8,681,016 | | |

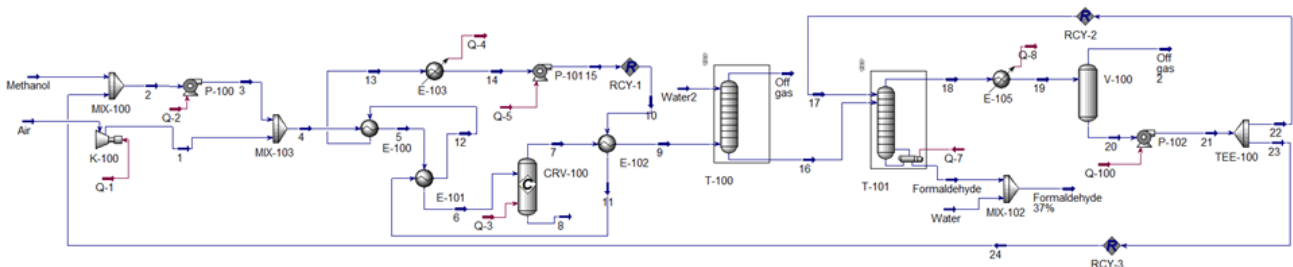


Figure 8. Modified process flow diagram from HYSYS simulation

variables are crucial for the operation of an absorption tower, one of them is how many stages does a column have. A stage typically refers to the individual trays or plates within the column where mass transfer occurs [14]. Each stage represents a specific horizontal level at which the gas or vapor phase and the liquid absorbent come into contact, facilitating the absorption or desorption of components [17].

If the absorption column does not have enough stages in its column, mass transfer becomes inefficient which may lead to several issues. One of them is the reduction of pure products that can be produced in a given time. Another is that leftover formaldehyde that has not been leached will be discarded as off gas alongside other unwanted components [15]. Even so, too much stages can lead to excessive operational cost without much gain in yield. This is why optimization of absorption stages need to be done [16].

In this process, the water that's used for the absorption process is set at 2,612 kg/h whilst the inlet and outlet pressure is kept at 140 kpa. The adjustment of the stages is done in the first absorption process while the reboiled absorber specification is constant. The dependent variable of this process is how much final product (formaldehyde 37%) is obtained in certain number of stages. The results can be presented in Table 3 and Figure 7.

From the Table 3 and Figure 7, the yield of formaldehyde rises alongside the addition of additional stages of the absorption process. As the number of stages in the column increases, the area in which absorption can happen alongside mass transfer also increases. Because of this increase, more of the formaldehyde within the feed can be leached out, causing an increase in the yield of the system. It can be seen from the graph that with ten stages in the absorption column, the final yield

of the process is 2,420.445 kg/h. Increasing the tray count to 14 increases the yield to 2,660.313 kg/h. This trend continues until the last data point of 38 trays, where the yield of the process increases to 3,123.251779 kg/h. Although the more stages added to the column, the yield that can be gained reduces and eventually stagnates. This means that a further increase of stages does not increase the yield but increase the operating cost of the column. That is why it is identified that 30 stages are the optimal stage count for the system, where the yield is 3,064.792 kg/h of formaldehyde 37%.

4. Conclusion

From the data that can be observed from the figures above, it can be seen that the addition of a refrigerant-heater cycle within the pre-treatment and product cooling of the formaldehyde production process substantially reduces the energy required to run the production process. Measured as an overall heat flow of the process, the modified process reduces the energy required to run the process by 144,478,713.8 kJ/h which increases energy efficiency by 93.8%. It can also be seen from the results that the stages within the absorption tower does indeed correlate with the product yield in a linear manner where an increase of the absorption stages increases the yield produced yet with an ever-increasing number of stages, the addition of yield becomes overshadowed by the cost associated with the addition of the stages. We have concluded that 30 stages are the optimal number of stages for the production process with the formaldehyde yield of 3,064.792 kg/h. Through the research done within this paper, it can be seen that the addition of a refrigerant-heater cycle is incredibly beneficial for the production process by reducing the power

Table 3. Correlation between tray count of absorption column and yield

| Number of absorption column tray | Mass flow rate of formaldehyde product (kg/h) |
|----------------------------------|---|
| 10 | 2420.445 |
| 14 | 2660.313 |
| 18 | 2799.285 |
| 22 | 2892.324 |
| 26 | 2958.330 |
| 30 | 3064.792 |
| 34 | 3098.944 |
| 38 | 3123.252 |

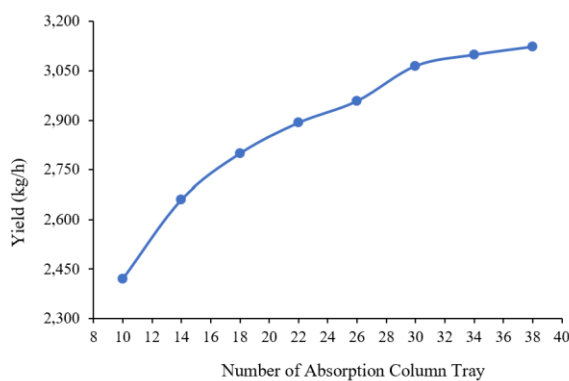


Figure 7. Correlation between absorption column tray number and formaldehyde product mass flow rate

and water required to run the production process. We hope this may be a stepping stone so that further study of how to further integrate such system into the industry can be done.

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