

Utilization of Heat from The Reactor's Outlet Stream in Formaldehyde Production to Reduce Energy Usage in The Heat Exchanger

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Abstract

Global production of formaldehyde has consistently risen over the past ten years, highlighting its extensive industrial applications and high demand across various sectors worldwide. However, its production continues to consume a significant amount of energy. The aim of this paper is to investigate and propose strategies for enhancing energy efficiency in formaldehyde production processes. Specifically, the study focuses on harnessing heat from the reactor's outlet stream to minimize the energy consumption associated with heat exchangers. By analyzing and optimizing the utilization of this heat source, the paper aims to contribute to sustainable manufacturing practices by reducing overall energy requirements and operational costs in formaldehyde production facilities. The process modification was simulated using Aspen HYSYS and the comparison of net-energy between the basic and the modified process is calculated using the net-energy formula. The results obtained that the Net-Energy (NE) value for both basic and modified process is 316,286,815.4 kJ/h and 125,757,792.9 kJ/h. This shows that the modified process has better energy efficiency compared to the basic process as the net-energy value zero. Therefore, this modification enhances the energy efficiency of the formaldehyde production process through methanol oxidation.

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1. Introduction

Methanol is one of the most critical chemical intermediates used in the petrochemical sector [1]. It is the raw material for the synthesis of some hydrocarbons and various oxidation products [2]. More than 30% of methanol is used in formaldehyde production every year [3].

Formaldehyde (CH₂O) is an volatile, colorless, flammable, and highly reactive aldehyde [4]. Formaldehyde naturally occurs and is produced from organic matter through photochemical processes in the atmosphere as long as life exists on Earth. It is a significant

metabolic product in plants and animals, including humans. Formaldehyde is also produced when organic material is incompletely burned [5]. Formaldehyde is a crucial raw material extensively utilized in numerous industrial chemical products, including resins, 1,4-butylene glycol (BDO), polyoxymethylene, and pesticides [6]. Additionally, formaldehyde is a crucial industrial chemical, used in the production of numerous industrial products and consumer goods [7]. Inhaling formaldehyde has been linked to sensory irritation of the eyes and respiratory tract, as well as to childhood and adult asthma and changes in lung function, according to clinical studies and epidemiological surveys conducted in workplace and residential settings [8].

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Formaldehyde was first synthesized in 1859 when Butlerov hydrolyzed methylene acetate and noticed the distinctive odor of the resulting solution. In 1867, Hofmann definitively identified formaldehyde by passing methanol vapor and air over a heated platinum spiral. This technique, albeit with different catalysts, remains the primary method of production today. The process for preparing pure formaldehyde was later detailed by Kekulé in 1882 [9].

In recent years, the yearly demand for formaldehyde has surpassed 30 megatons, with production remaining notably consistent. In 2017, approximately 52 million tons of formalin (a 37% by weight aqueous solution of CH_2O) were produced. Global annual consumption of formaldehyde is projected to grow by nearly 5% in the coming years [10]. Formaldehyde is a key ingredient in the production of resins, phenols, urea, and melamine [11]. It is utilized for its weather-resistant properties in adhesives and wood coatings [12]. Additionally, due to its disinfectant properties, it is incorporated into soaps and used for sterilizing surgical instruments in the medical field. Formaldehyde also imparts wrinkle resistance to fabrics [13]. On the other side, formaldehyde can be used in the manufacture of paper, textiles, and paints [14].

Currently, global formaldehyde production is divided almost equally between the silver-catalyzed process, the oxide-based catalyst process. The silver-based method uses a methanol-rich feed, while the oxide-based method (Formox process) utilizes a feed with lower methanol content [15]. The oxide-based catalyst achieves a 93% selectivity, in contrast to the silver-based catalyst's selectivity of less than 90%. Although the silver process involves a lower initial investment cost, selectivity remains crucial because methanol accounts for 94% of the production costs, whereas power and catalyst costs represent 4% and 2% each [16]. In the Formox process, a metal oxide (e.g. iron, molybdenum, or vanadium oxide) is used as a catalyst for the conversion of methanol to formaldehyde. The Formox process has been described as a two-step oxidation reaction in the gaseous state (g) which involves an oxidized (K_{OX}) and a reduced (K_{red}) catalyst [17].

Heat Integration (HI) is employed to enhance energy efficiency, with a focus on optimizing the utilization of heat from reactor output. This procedure is paramount for manufacturing industries striving to minimize their energy consumption. Heat integration involves capturing waste heat from certain stages of a process and reusing it to supply heat to other stages that need it. This practice decreases the total amount of external energy required that

leading to overall environmental impact of the manufacturing processes. These innovations allow for more precise control and optimization of heat flows within the system [18].

In this case, the innovation is proposed by utilizing the heat energy from the reactor's outlet stream. This modification can significantly decrease the requirements for external energy sources by directing the heat generated during the reaction to the heat exchanger. The aim of this research is to reduce energy in the formaldehyde production through methanol oxidation process using Aspen HYSYS.

2. Methods

In order to improve energy efficiency in methanol oxidation process, simulations are conducted using Aspen HYSYS. The process employs a fixed bed reactor where the reaction takes place under non-adiabatic and non-isothermic conditions. During the reaction, significant amount of energy is generated which can be utilized for other processes or utilities [19]. This energy generation is a result of the methanol oxidation process and being classified as an exothermic reaction which leads to a higher reactor output temperature.

In this modification, the methanol is mixed with air and then heated before entering the reactor, where the temperature is 120 °C. The heating process occurs in two stages, first in stream 5 to 65 °C and then in stream 6 to 120 °C. The reactor output has a temperature of 650 °C and needs to be reduced to around 140 °C before entering the absorber. To achieve this, the output is cooled using cooling water (stream 9). After contacting the reactor output, the cooling water heats up and exits through HE-102. This heated water is then recycled (stream 10) to heat HE-101. The output from HE-101 is used to heat HE-100 and the water from HE-100 is recycled again (stream 9) to cool the reactor output. The heat

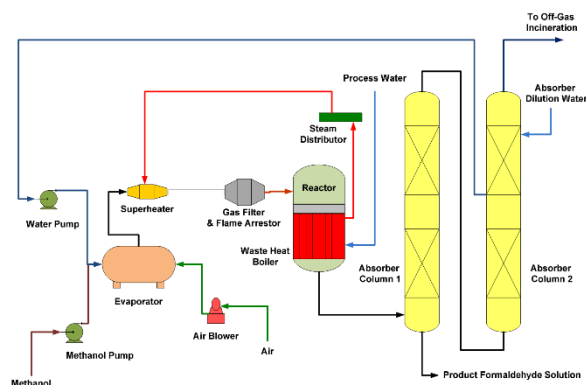


Figure 1. Basic Flowsheet of formaldehyde production through methanol oxidation process from literature [20]

efficiency of basic and modified process can be compared using net-energy calculation as follows:

$$NE = E_P - E_C \quad (1)$$

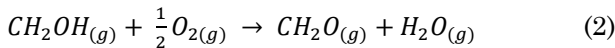
where, NE is net-energy (kJ/h), E_P is energy produced (kJ/h), and E_C is energy consumed (kJ/h) [21]. The effect of this modification process is studied. E_P is the full amount of energy generated by the system, while E_C refers to the total amount of energy consumed throughout the entire lifecycle of the production process.

In the simulation before modification, E_P comes from Q-100, Q-101, and Q-105, while E_C comes from the streams of feed, 9, 11, 13, 16, 10, 12, 14, 17, and 18. In the modified process, E_P comes from Q-100, Q-101, Q-103, Q-104, and Q-105, while E_C comes from Q-102, as well as the streams of feed, 16, 17, and 18.

3. Results and Discussion

3.1. Thermodynamics Review

Determining whether a reaction is exothermic or endothermic and the direction of the reaction reversible or irreversible. It can be obtained by calculating the standard enthalpy of reaction (ΔH°_{298K}) at 1 bar and temperature of 298 K requires using the standard enthalpies of formation of the reactants and products. The value of ΔH°_f and ΔG°_f can be seen in Table 1 [22]. The oxidation process can be expressed as:



Standard heat of reaction at 298 K (ΔH°_{298K}):

$$\begin{aligned} \Delta H^\circ_{298K} &= \sum \Delta H^\circ_f \text{ product} - \sum \Delta H^\circ_f \text{ reactant} \\ &= (\Delta H^\circ_f CH_2O + \Delta H^\circ_f O_2) - (\Delta H^\circ_f CH_2OH - \Delta H^\circ_f H_2O) \\ &= (-201.7 + 0) - (-285.5 + (-241.8)) \\ &= 325.6 \text{ kJ/mol} \end{aligned}$$

Based on the calculations, we get the value $\Delta H^\circ_{298K} = 325.6$ kJ/mol, which is a positive value so that the reaction is endothermic.

Gibbs energy (ΔG°_{298K}):

$$\Delta G^\circ_{298K} = \sum \Delta G^\circ_f \text{ product} - \sum \Delta G^\circ_f \text{ reactant}$$

$$\begin{aligned} &= (\Delta G^\circ_f CH_2OH + \Delta G^\circ_f O_2) - (\Delta G^\circ_f CH_2O - \Delta G^\circ_f H_2O) \\ &= (-162.51 + 0) - (91.2 + (-228.6)) \\ &= -25.11 \text{ kJ/mol} \end{aligned}$$

Equilibrium constant (K_2) in the standart state:

$$\begin{aligned} \Delta G^\circ_{298K} &= -RT \ln K \\ \ln K_2 &= -\frac{\Delta G^\circ_{298K}}{RT} = \frac{25.110 \text{ J/mol}}{8.314 \text{ J/mol} \times 298 \text{ K}} = 10.135 \\ K_2 &= 23.159 \times 10^5 \end{aligned}$$

Equilibrium constant (K_1) at reactor temperature $T = 650$ °C:

$$\ln \frac{K_1}{K_2} = \frac{-\Delta H_R}{R} \left(\frac{1}{T_2} - \frac{1}{T_1} \right)$$

ΔH°_{298K} = Standard heat of reaction at 298 K.

$$\ln \frac{K_1}{23.159 \times 10^5} = \frac{325.600 \text{ J/mol}}{8.314 \text{ J/mol.K}} \left(\frac{1}{923 \text{ K}} - \frac{1}{298 \text{ K}} \right)$$

$$\ln \frac{K_1}{23.159 \times 10^5} = -93,082$$

$$4.533 = \frac{K_1}{23.159 \times 10^5}$$

$$K_1 = 1.05 \times 10^{10}$$

Because the equilibrium constant value is relatively high, the reaction is irreversible and proceeds to the right.

3.2. Comparison between Basic and Modified Processes

The simulation of formaldehyde production by oxidation for both the basic and modified processes using Aspen HYSYS is illustrated in Figures 2–5. Figure 2 shows the basic process simulation, while Figure 3 presents the PFD of the modified process. In the basic process, fewer pieces of equipment are used, and there is no reuse of heat. In contrast, the modified process, depicted in Figure 3, employs more equipment due to the reuse of heat. The main difference between the two is the addition of a heat transfer loop system for heating and cooling in the modified process. In the basic process, there is no heat transfer fluid system, resulting in inefficient energy use for heating and cooling. However, in the modified process, an additional cooler (E-101) and pump (P-101) are included to cool the heat exchanger fluid, enabling its reuse to cool the reactor output in the heat exchanger (E-100). The output of E-100 is then used to heat the reactor feed in HE-101, establishing a continuous looping process.

Table 1. The value of ΔH°_f and ΔG°_f

Compounds	Molecular Formula	ΔH°_f (Kj/mol)	ΔG°_f (kJ/mol)
Methanol	CH ₂ OH	-201.7	-162.51
Air	O ₂	0	0
Formaldehyde	CH ₂ O	-285.5	91.2
Water	H ₂ O	-241.8	-228.6

3.3. Mass Balance and Energy Balance Results

Figure 3 shows that the heat exchanger (E-100) is used to cool the reactor output, which has a flow rate of 6675 kg/h, lowering its temperature from 650 °C to 140 °C. The heat from the reactor output is absorbed by the heat transfer fluid in heat exchanger (E-100), which has an input flow rate of 2330 kg/h at 120 °C, with a duty of 5,164,342.48 kJ/h. In the recycling system, heating occurs in heat exchanger (HE-102), where the reactor feed is heated from 65 °C to 120 °C at a flow rate of 6675 kg/h, requiring 445,611.18 kJ/h. The output from HE-102 is then used to heat the fluid in HE-100 with a flow rate of 6675 kg/h, capturing 2,648,846.80 kJ/h from the heating fluid.

3.3. Heat Transfer Fluid Recycle Effects

The formaldehyde dehydrogenation process requires multiple heat transfer steps to manage both heating and cooling of the compound. Typically carried out in a fixed bed reactor at temperatures ranging from 650-720 °C, this process generates a significant amount of heat due to its exothermic nature, characterized by a negative ΔH [23]. A heat transfer fluid is necessary to facilitate these processes using heat exchangers.

During the process, a stream containing hot fluid from the reactor serves as the heating medium to warm the recycled water in the heat exchanger (E-100). Meanwhile, streams

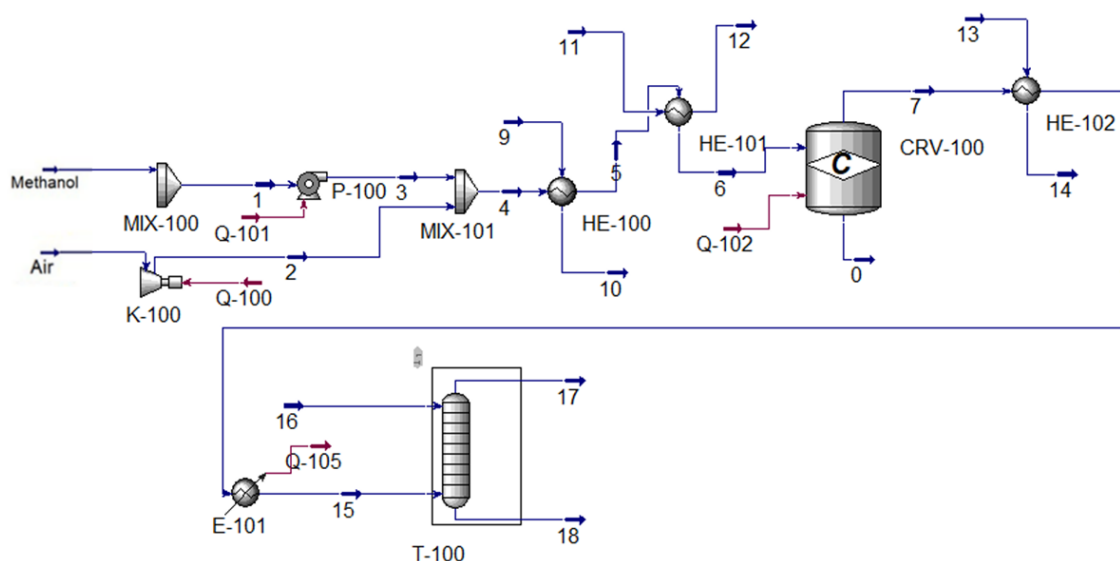


Figure 2. Simulation using Aspen HYSYS of basic (unmodified) process

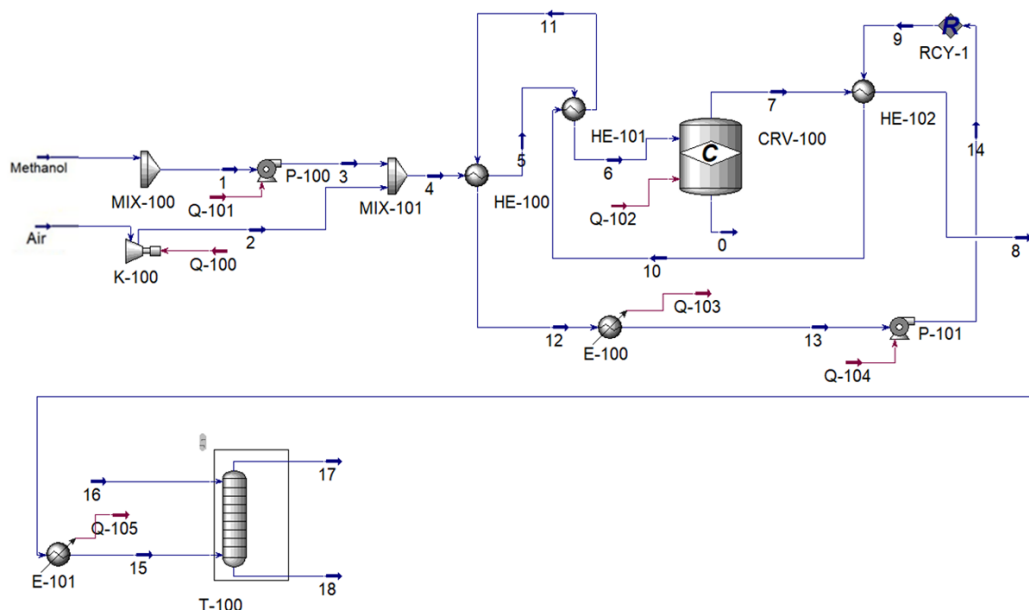


Figure 3. Simulation using Aspen HYSYS of modified process

containing cold fluid enter E-100 to cool the reactor's product [24]. In the basic process, there is no recycling of heat, resulting in wasted energy from the heating processes in the heat exchangers (HE-100, HE-101, and E-100). The absence of a heat recycling system also means that additional heat sources are required to reheat the reactor feed, leading to higher energy consumption.

In contrast, the modified process employs the heat collected by E-100 to heat exchangers HE-101 and HE-100. The cooled heating fluid is then reused to heat the reactor output. This continuous loop ensures that the process's energy is effectively utilized for minimizing waste.

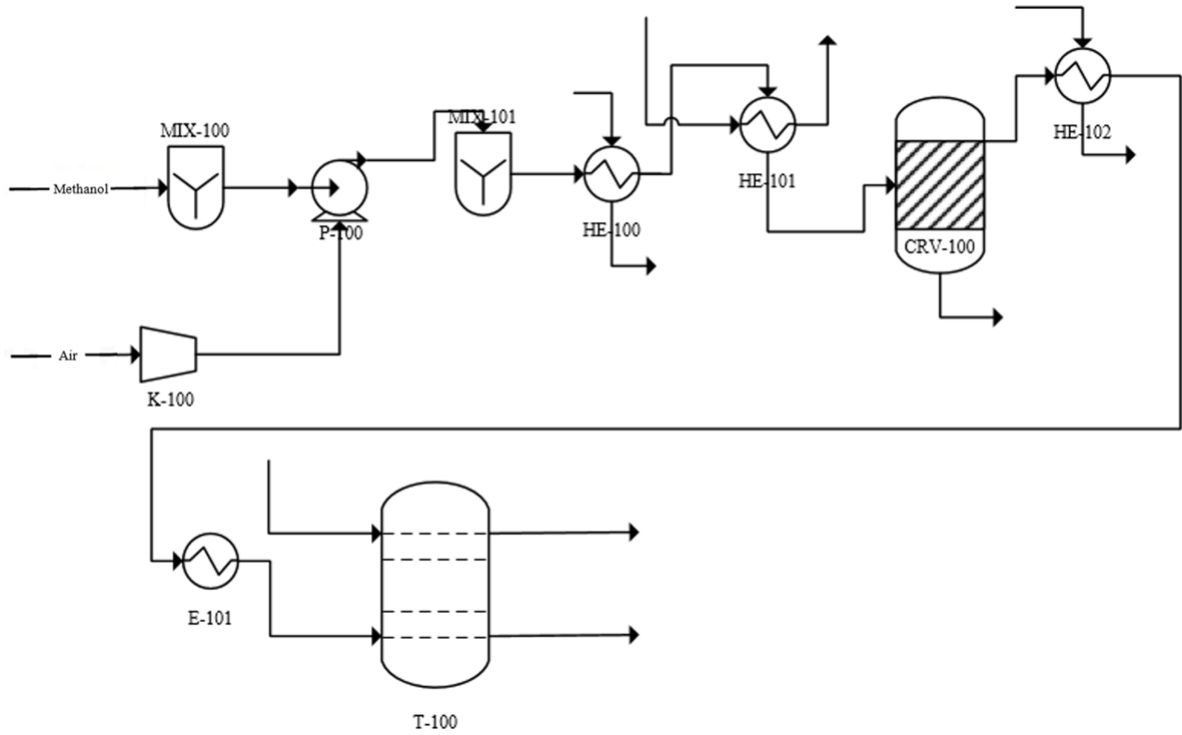


Figure 4. Process flow diagram of basic (unmodified) process

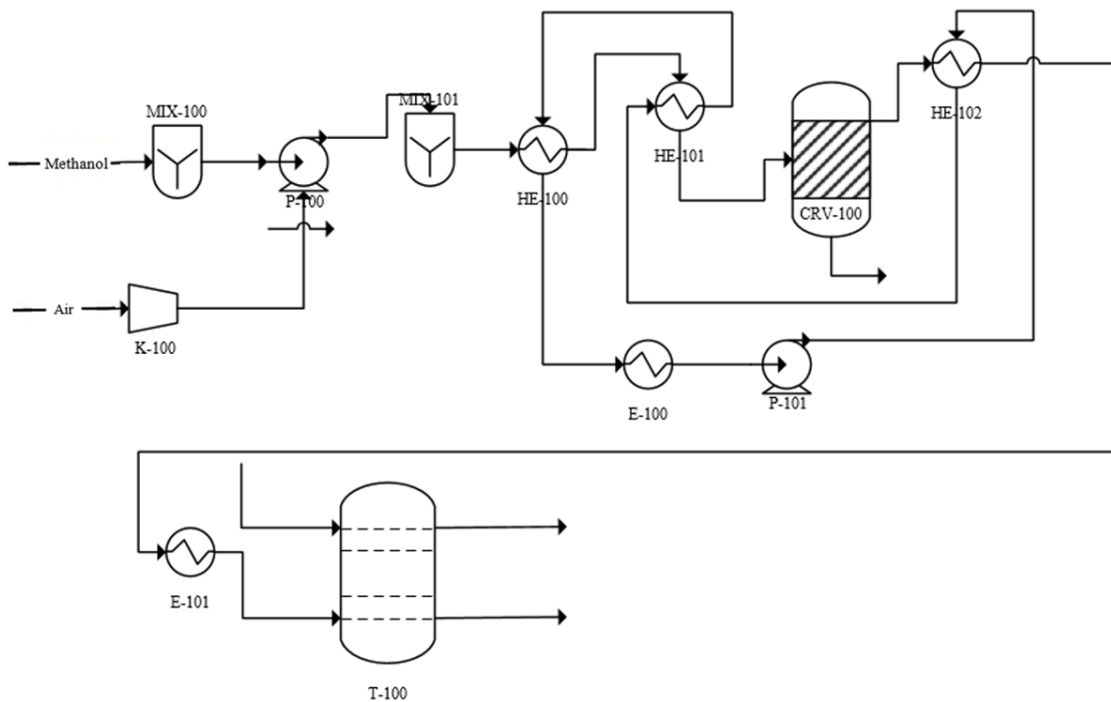


Figure 5. Process flow diagram of modified process

3.5. Energy Source for Heat Exchanger (E-100) Effect

The output from the reactor (CRV-100) serves as the heat source for heat exchanger (E-100), where the reactor output reaches a temperature of 650 °C at a flow rate of 6675 kg/h. Heat exchanger (E-100) generates hot fluid, which is then used to heat the second heat exchanger (HE-101) in the reactor feed exchangers. The cooled fluid from HE-102 continues as the hot fluid supply for the first heat exchanger (HE-100) in the reactor feed exchangers. However, the fluid cooled by HE-100 remains too warm to effectively cool the reactor output liquid in heat exchanger (E-100). Therefore, an additional cooling source is required, which is provided by cooler (E-101). Additionally, pump (P-101) provides the necessary driving force for fluid circulation. The heat absorbed by heat exchangers (E-100, HE-100, and HE-101) facilitates the cooling process for the reactor output, eliminating the need for external energy sources and reducing energy costs. However, this method involves more unit operations, which may increase the overall capital cost of the operation.

3.6. Steam Formation for Heat Recovery Process

The heat from cooling process of the reactor product is wasted without utilizing the heat transfer fluid. Whereas after the modification, this heat can be used for other purposes that require energy. As previously mentioned, some of this heat is used for the recycling process in the heat exchanger (E-100), but additional heat remains for further use. This residual heat generates steam for plant utility purposes. It is common for heat recovery technologies to produce low-pressure steam by vaporizing water at 120.7 °C and 7 bar [25]. When steam is heated above its boiling point at a specific pressure, it becomes "dry steam" also referred to as superheated steam [26].

3.6. Net-Energy Efficiency Improvement Due to Process Modification

Table 2 shows the net-energy values for both the original and altered processes. In the original process, the E_P value is 989,939.8688 kJ/h derived from Q-100, Q-101, and Q-105 with an E_C value is -315,296,875.6 kJ/h. On the other hand, the

modified process shows an E_P value of 3,060,321.915 kJ/h from Q-100, Q-101, Q-103, Q-104, and Q-105 with an E_C value amounts to -122,697,471 kJ/h. It can be concluded that the net-energy (NE) in the original process is 316,286,815.4 kJ/h, whereas in the modified process it increases to 125,757,792.9 kJ/h.

Energy efficiency plays a crucial role in achieving net-zero emissions. Balancing the production and consumption of energy is a key method to achieve this goal of net zero emissions. The energy efficiency is better when the value of net-energy is closer to zero. Energy efficiency improves as the net-energy value approaches zero [27].

4. Conclusion

The formaldehyde production process needs to be modified to avoid excessive energy consumption. Reusing the heat generated from reactor output is one method to contribute to environmental conservation. This is proven by the net-energy value in the modified process is 125,757,792.9 kJ/h. On the other hand, net-energy value in the basic process is 316,286,815.4 kJ/h. It can be seen that in the modified process, the net-energy produced is lower than in the basic process. The heat output from the reactor serves as the heat source for another heat exchanger, enhancing energy efficiency by allowing the energy released and absorbed within the system to be mutually utilized in each processes. The proposed action to be implemented involves engineers finding alternatives to reduce energy usage in benzene production without significantly compromising the quality of the improved process outcome.

CRediT Author Statement

Author Contributions; S.Z. Putri: Conceptualization, Methodology, Investigation, Resources, Data Curation, Writing, Review and Editing, Supervision; S. Chairunnisa: Conceptualization, Methodology, Formal Analysis, Data Curation, Writing Draft Preparation, Visualization, Software, Project Administration; A.S. Nugraha: Validation, Writing, Data Curation. All authors have read and agreed to the published version of the manuscript.

Table 2. Calculation result of net-energy of basic and modified processes

Process	E_P (kJ/h)	E_C (kJ/h)	NE (kJ/h)
Basic	989,939.8688	-315,296,875.6	316,286,815.4
Modified	3,060,321.915	-122,697,471	125,757,792.9

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