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Research Article

Improving Energy Efficiency in Sulfuric Acid Production from Sulfur and Air by Adding Heat Exchanger and Reducing Cooler

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Abstract

Sulfuric acid is a strong mineral acid and is useful for processing mineral ores, chemical synthesis, waste water processing, and others. The increasing need for sulfuric acid is not balanced with the amount of sulfuric acid production due to the use of quite a lot of energy in the production process. Therefore, innovation is being carried out, namely energy efficiency in sulfuric acid production from sulfur and air, which is expected to help increase production of sulfuric acid so that it can meet market needs. The sulfuric acid production process uses a contact process which is carried out with 3 reaction stages where the raw materials in the form of sulfur and free air are reacted in the reactor and SO2 gas is obtained, then SO2 reacts with oxygen to form SO3 gas in the converter bed, and finally the formation of H2SO4 from the reaction between SO3 and water in the absorber column. Modifications were made using Aspen HYSYS to improve energy efficiency in the sulfuric acid production.

Keywords: Sulfuric Acid Production; Sulfur; Air Oxidation; Energy Consumption; Energy Efficient

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1. Introduction

Sulfuric acid (H_2SO_4) is one of the basic raw materials for the chemical industry. It is used in numerous processes as a reagent, as a catalyst and as a drying agent. Examples of the wide-ranging applications of this acid include the treatment of raw phosphates in the fertilizer industry, the treatment of titanium ores for the production of titanium dioxide, the manufacture of phosphoric and hydrofluoric acid and the wide field of organic chemical synthesis, for example, in sulfonation and nitration [1]. Sulfuric acid is a colorless liquid, highly reactive, and capable of dissolving various metal [2]. Since more than 3 decades, reduced organic sulfur compounds have been recognized as substantial biogenic emissions contributing to Earth´s sulfur cycle [3]. Hydrogen sulfide (H_2S) can be produced in various industrial processes, such as anaerobic digestion, landfills, petroleum refining, paper milling and food processing. For example, the concentration of H_2S in Anaerobic digestion for biogas production can range from 1000 to 20000 ppmv [4]. H_2S is not only corrosive to gas storage tanks and pipelines but also harmful to the human body, especially the nervous system [5]. These highly versatile capabilities can be attributed to the complicated nature of this acid. Sulfuric acid is a corrosive with a continuously changing character. It changes from the reducing nature of dilute acid to the oxidizing nature of concentrated acid [6]. Sulfuric acid (H_2SO_4) annual production is the highest among chemicals worldwide. It is an intermediate compound in most industries such as fertilizers, water treatment, oil refining, and chemical synthesis [7]. The sulfuric acid production with contact process is the most popular [8].

The Contact Process is the most common industrial method for the production of H_2SO_4.

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The process involves the oxidation of sulfur dioxide (SO_2) to sulfur trioxide (SO_3), which is then dissolved in water to produce sulfuric acid. The sulfuric acid production process is very exothermic [9]. The sulfuric acid production process using the contact process is carried out in 3 reaction stages where first the raw materials in the form of sulfate and free air are reacted in the reactor and SO_2 gas is obtained, then SO_2 is reacted with oxygen to form SO_3 gas in the reactor. converter layer, and finally the formation of H_2SO_4 from the reaction between SO_3 and water in the absorber column [10].

To enhance sulfuric acid (H_2SO_4) production from both economic and production perspectives, this research will investigate the design of a sulfuric acid plant using simulations that consider aspects of energy efficiency. Research studies indicate that addressing this issue can be accomplished by implementing sustainable design procedures at various stages of the process. Optimizing energy usage in the industry is crucial, aligning with key targets in the industrial sector.

As for previous research on energy efficiency in the contact process, studies have focused on the integration of advanced heat recovery systems and modifications in process design, such as optimizing heat exchanger networks and reducing the number of coolers required during SO_2 oxidation. The objective of this research work is to improve net energy efficiency in sulfuric acid production by adding heat exchangers and reducing the use of coolers, and to review the net energy efficiency calculations in the contact process. The basic PFD from the literature sulfuric acid production is depicted in Figure 1 [11].

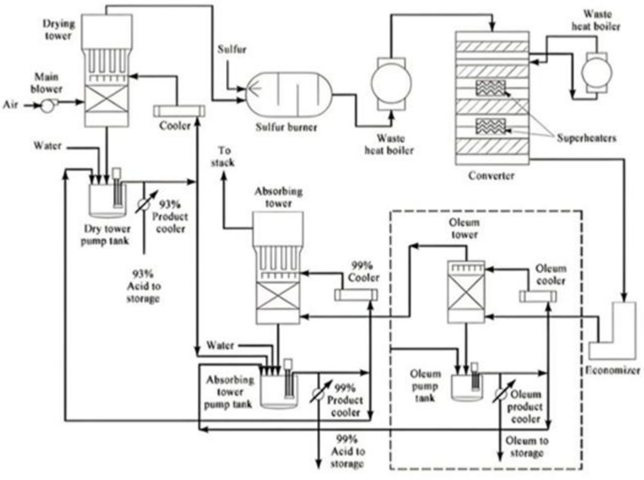

Figure 1. PFD sulfuric acid production from literature

2. Method

2.1 Contact Process

During contact process, manufactured sulfuric acid underwent four stages process starting from melting the elemental sulfur:

$$S + O_2 \rightarrow SO_2 \quad \Delta H_{298} = -296.80 \text{ kJ/mol} \tag{1}$$

$$SO_2 + \tfrac{1}{2}O_2 \leftrightarrow SO_3 \quad \Delta H_{298} = -98.9 \text{kJ/mol} \tag{2}$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad \Delta H_{298} = -115.63 \text{ kJ/mol} \tag{3}$$

The initial step, melting of elemental sulfur, is the predominant step of sulfuric acid manufacture, which gives the S element needed in its formula [12].

2.2. Thermodynamics Consideration

The thermodynamic aspect can be seen from the value ΔH and ΔG from the reaction process. The value of ΔHf can be seen in Table 1.

First reaction:

$$\begin{aligned}\Delta H_{298} &= \Delta \text{Hf product} - \Delta \text{Hf reactant}\\ &= [(\Delta H_{r,SO2}) - (\Delta H_{r,S} + \Delta H_{r,O2})]\\ &= -296.80 - (0+0)\\ &= -296.80 \text{ kJ/mol}\end{aligned}$$

Second reaction:

$$\begin{aligned}\Delta H_{298} &= \Delta \text{Hf product} - \Delta \text{Hf reactant}\\ &= [(\Delta H_{r,SO3}) - (\Delta H_{r,SO2} + 0.5 \times \Delta H_{r,O2})]\\ &= -395.70-(-296.80+0)\\ &= -98.9 \text{ kJ/mol}\end{aligned}$$

Third reaction:

$$\begin{aligned}\Delta H_{298} &= \Delta \text{Hf product} - \Delta \text{Hf reactant}\\ &= [(\Delta H_{r,H2SO4}) - (\Delta H_{r,SO3} + \Delta H_{r,H2O})]\\ &= -735.13-(-395.70+241.80)\\ &= -115.63 \text{ kJ/mol}\end{aligned}$$

From the data above, it is known that the reaction takes place exothermic so that the more the temperature increases, the reaction speed will decrease [14]. The value of ΔGf can be seen in Table 2.

First reaction:

$$\begin{aligned}\Delta G_{298} &= \Delta \text{Gf product} - \Delta \text{Gf reactant}\\ &= [(\Delta G_{f,SO2}) - (\Delta G_{f,S} + \Delta G_{f,O2})]\\ &= -300.10 - (0+0)\\ &= -300.10 \text{ kJ/mol}\end{aligned}$$

$$\Delta G_{298} = -RT \text{ In } K$$

$$\text{In } K_{298} = \frac{\Delta G_{298}}{-RT}$$

$$\text{In } K_{298} = \frac{-300.10 \times 10^3}{-(8.314 \frac{J}{mol°K} \times 298°K}$$

$$\text{In } K_{298} = 121.12$$

$$K_{298} = 4.024 \times 10^{52}$$

It is known that the reaction takes place at an operating temperature of 1230 ^oC. Then, the value of K at the operating temperature is 1230 ^oC (1503 K) can be calculated.

$$\text{In}\, K_{298} = -\frac{\Delta \text{H}}{\text{R}}\left[\frac{1}{298} - \frac{1}{\text{T}}\right]$$
$$\text{In}\frac{K_{298}}{K_{1503}} = \frac{-296.80}{8.314\frac{\text{J}}{\text{mol°K}}}\left[\frac{1}{298} - \frac{1}{1503}\right]$$
$$\text{In}\frac{4.024\times10^{52}}{K_{1503}} = -96.043$$
$$\frac{2.013\times10^{35}}{K_{1503}} = 1.9456\times10^{-42}$$
$$K_{1503} = 1.955\times10^{77}$$

It is known that the value of the reaction equilibrium constant is very large, so the reaction process is irreversible.

Second reaction:

$$\Delta \text{G}_{298} = \Delta\text{Gf product} - \Delta\text{Gf reactant}$$
$$= [(\Delta \text{G}_{\text{f,SO3}}) - (\Delta \text{G}_{\text{f,SO2}} + \Delta \text{G}_{\text{f,O2}})]$$
$$= -300.10 - (0+0)$$
$$= -300.10 \text{ kJ/mol}$$
$$\Delta \text{Gf}_{298} = -\text{RT In } K$$
$$\text{In}\, K_{298} = \frac{\Delta \text{G}_{298}}{-\text{RT}}$$
$$\text{In}\, K_{298} = \frac{-71\times10^{3}\ J/mol}{-(8.314\frac{\text{J}}{\text{mol°K}}\times 298\text{°K})}$$
$$\text{In}\, K_{298} = 28.65$$
$$K_{298} = 2.790\times10^{12}$$

It is known that the reaction takes place at an operating temperature of 610 °C. Then, the value of *K* at the operating temperature is 610 °C (883 K) can be calculated.

$$\text{In}\, K_{298} = -\frac{\Delta \text{H}}{\text{R}}\left[\frac{1}{298} - \frac{1}{\text{T}}\right]$$
$$\text{In}\frac{K_{298}}{K_{883}} = \frac{-98.9\times10^{3}}{8.314\frac{\text{J}}{\text{mol°K}}}\left[\frac{1}{298} - \frac{1}{883}\right]$$
$$\text{In}\frac{2.790\times10^{12}}{\text{K}_{883}} = -26.446$$
$$\frac{2.790\times10^{12}}{K_{883}} = 3.269\times10^{-12}$$
$$K_{883} = 3.535\times10^{-12}$$

It is known that the value of the reaction equilibrium constant is very large, so the reaction process is irreversible.

Third reaction:

$$\Delta \text{G}_{298} = \Delta\text{Gf product} - \Delta\text{Gf reactant}$$
$$= [(\Delta \text{G}_{\text{f,H2SO4}}) - (\Delta \text{G}_{\text{f,SO3}} + \Delta \text{G}_{\text{f,H2O}})]$$
$$= -653.47 - (371.10-228.60)$$
$$= -53.77 \text{ kJ/mol}$$
$$\Delta \text{Gf}_{298} = -\text{RT In } K$$
$$\text{In}\, K_{298} = \frac{\Delta \text{G}_{298}}{-\text{RT}}$$
$$\text{In}\, K_{298} = \frac{-53.77\times10^{3}\ J/mol}{-(8.314\frac{\text{J}}{\text{mol°K}}\times 298\text{°K})}$$
$$\text{In}\, K_{298} = 21.70$$
$$K_{298} = 2.656\times10^{9}$$

It is known that the reaction takes place at an operating temperature of 172 °C. Then, the value of *K* at the operating temperature is 172 °C (445 K) can be calculated.

$$\text{In}\, K_{298} = -\frac{\Delta \text{H}}{\text{R}}\left[\frac{1}{298} - \frac{1}{\text{T}}\right]$$
$$\text{In}\frac{K_{298}}{K_{445}} = \frac{-115.63\times10^{3}}{8.314\frac{\text{J}}{\text{mol°K}}}\left[\frac{1}{298} - \frac{1}{445}\right]$$
$$\text{In}\frac{2.656\times10^{9}}{K_{445}} = -15.417$$
$$\frac{2.656\times10^{9}}{K_{445}} = 2.016\times10^{-7}$$
$$K_{445} = 1.317\times10^{16}$$

From the data, it is known that the value of the reaction equilibrium constant is very large, so the reaction process is irreversible.

2.3. Method to Improve Net Energy Efficiency of Sulfuric Acid Production Process from Sulfur and Air

Method to increase energy efficiency by using additional tools in the Aspen HYSYS simulator [15]. Aspen HYSYS is a market-leading process modeling tool for conceptual design, optimization, business planning, asset management, and performance monitoring for oil and gas production, gas processing, petroleum refining, and air separation industries [16]. The simulation was performed in Aspen HYSYS which contains 30 different thermodynamic models. Property package in this process is Antoine. This model is used because it is suitable for ideal mix systems at low pressure. The ideal mixture includes heavier hydrocarbon systems and its liquid phase behavior is close to ideal [17].

In modifying the process system using Aspen HYSYS, the temperature of the raw material is increased using an additional E-108 heat exchanger with the output from the CRV-101

Table 1. The value of ΔHf for each component [13]

Component	ΔHf_{298} (kJ/mol)
S	0
O_2	0
SO_2	-296.80
SO_3	-395.70
H_2SO_4	-735.13
H_2O	-241.80

Table 2. The value of ΔGf for each component [13].

Component	ΔGf_{298} (kJ/mol)
S	0
O_2	0
SO_2	-300.10
SO_3	-371.10
H_2SO_4	-653.47
H_2O	-228.60

reactor then, enter the E-101 heater. The addition of a heat exchanger can save energy from the heater. This because the temperature of the raw material is not much different from the desired temperature of the heater output so it can save energy from the heater. A reduction in the E-103 cooler was also carried out to increase energy efficiency in this process.

3. Result and Discussion

3.1 Simulation Process from Sulfuric Acid Production

Figures 2 and 4 show that the basic process flow diagram was taken from. In terms of energy efficiency, achieving zero net-energy is the ideal target. It represents a perfect balance between amount of energy produced and the amount consumed. Value of net energy closer to zero suggest that the energy efficiency is better [18]. Thus, the modified process is demonstrably proven to be more energy efficient than the basic process as show on Figures 3 and 5.

The temperature of sulfur at 660 ℃ and 1500 kPa and free air at 50 ℃ and 1500 kPa is increased using a heat exchanger first with the temperature of the reactor outlet so that the temperature can be close to the output of the heater. The next is heated using a heater according to the reactor specifications, namely 1230 ℃. The output is then flowed to the reactor [19].

$$S + O_2 \rightarrow SO_2 \quad \Delta H_{298} = -296.80 \text{ kJ/mol} \qquad (4)$$

The reactor operates at a temperature of 1230 ℃ and a pressure of 1500 kPa. The reactor output is unreacted SO_2 and oxygen. Then cooled using a cooler so that the temperature drops to 422 ℃. The process gas then enters first converter bed where SO_2 gas will be converted into SO_3 [9]. The following reaction:

$$SO_2 + \tfrac{1}{2} O_2 \leftrightarrow SO_3 \quad \Delta H_{298} = -98.9 \text{ kJ/mol} \qquad (5)$$

In the first bed, the reaction conversion is 65.89%. Gas containing SO_3 coming out of first bed, with a temperature of 610.7 ℃, will be put into the cooler so that the gas temperature drops to 450 ℃. The gas will enter second bed where, a

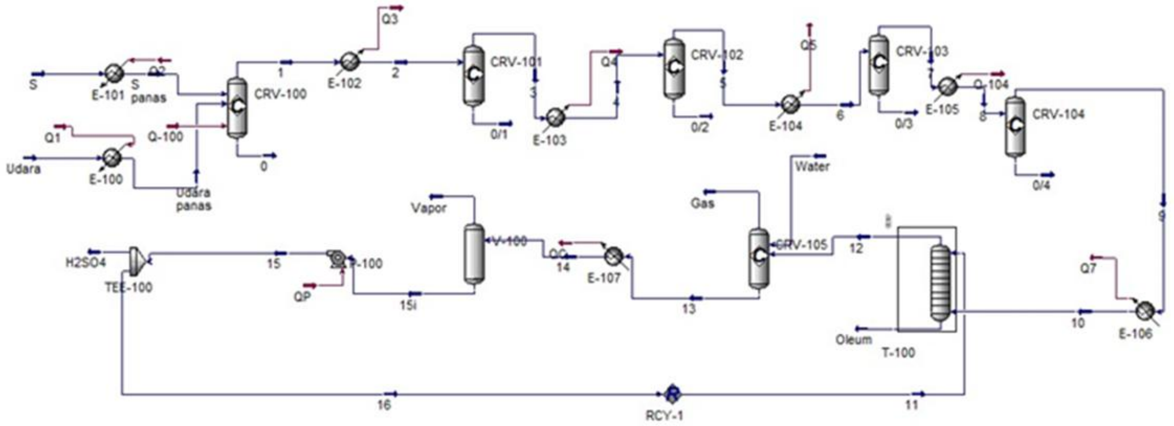

Figure 2. Process simulation of sulfuric acid production without modification using HYSYS

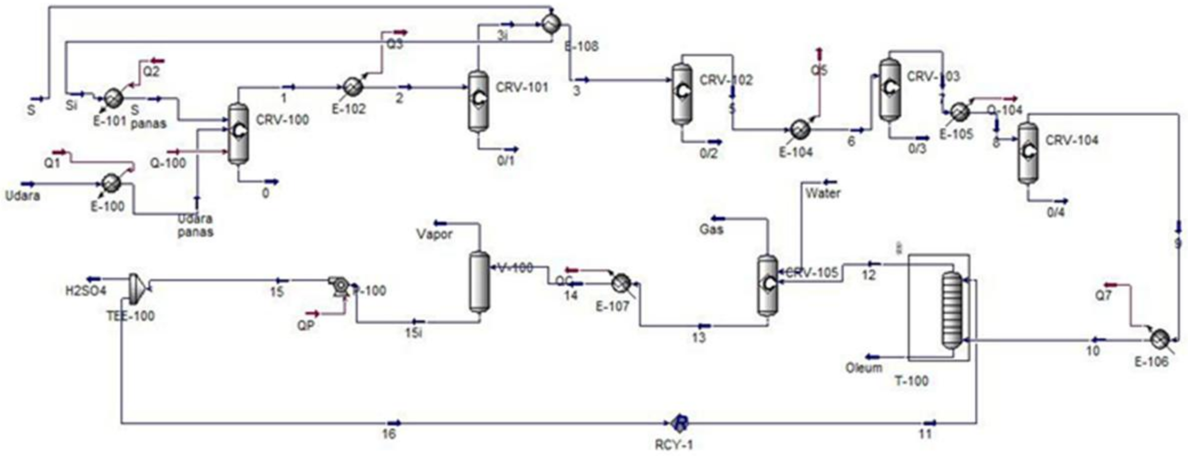

Figure 3. Process flow diagram sulfuric acid production with modification in HYSYS

conversion of 89.99% occurs. Second bed outlet gas, with a temperature of 535.5 ℃, will be entered into the cooler and the temperature will drop to 420 ℃. The next gas will enter third bed where, a conversion of 97.06% occurs. Third bed outlet gas with a temperature of 429 ℃ is then flowed to the cooler so that the gas temperature drops to 420 ℃. The gas then enters the fourth bed and a conversion of 10% is obtained. The bed output gas is then cooled from a temperature of 420.4 ℃ to 205 ℃ with a final pressure of 350 kPa. Next, gas a lot of SO_3 output from fourth bed will be absorbed by H_2SO_4 in the first absorber column. The use of circulating water supply in the energy sector is due to the conservation of water resources because, based on water treatment technologies, it is possible to minimize water consumption [20]. The results obtained are oleum (containing 99.17% H_2SO_4) and SO_3 gas is flowed to the second absorber column and a reaction to form sulfuric acid occurs following the following reaction:

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad \Delta H_{298} = -115.63 \text{ kJ/mol} \qquad (6)$$

The conversion in this reaction reached 89.18%. This gas, contains a lot of H_2SO_4, then cooled to a temperature of 30 ℃, separated, and purified from other gases such as oxygen, nitrogen and water. The gas is discharged into the wild, while the H_2SO_4 gas flows to the pump to increase the pressure from 100 kPa to 250 kPa, then the results are collected into the final product and part of it is returned to the first absorber column. The final product is H_2SO_4 with a purity level of 98.61% at a temperature of 30 ℃ and a pressure of 250 kPa.

3.2 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

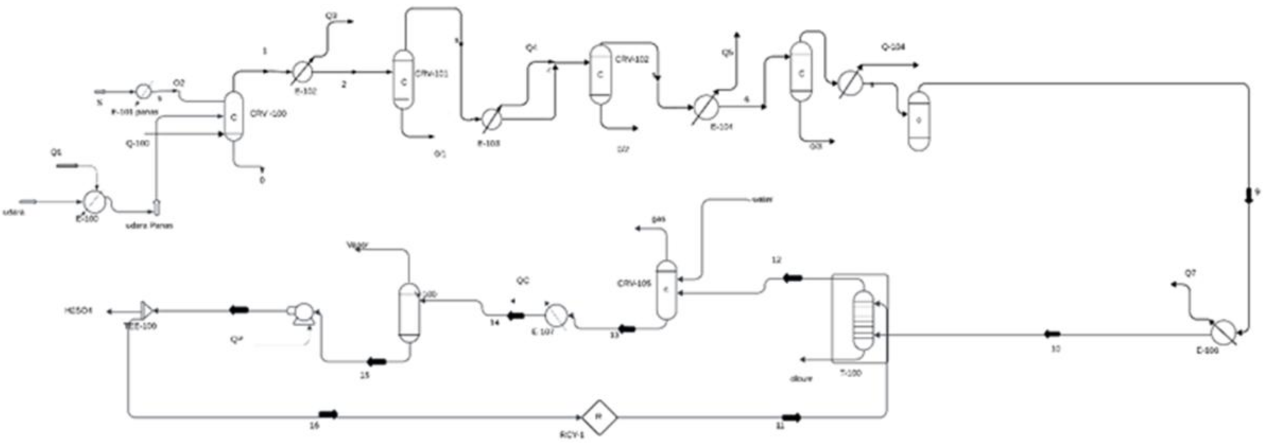

Figure 4. Process flow diagram sulfuric acid production without modification

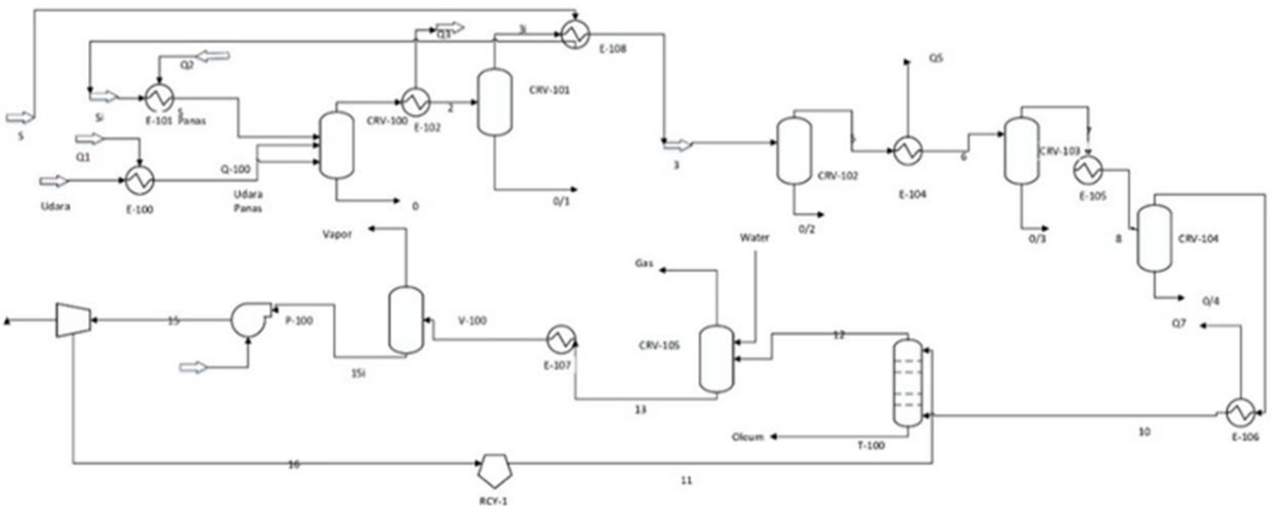

Figure 5. Process flow diagram of modified sulfuric acid production process

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 3 and Table 4.

$$\Delta H_{\text{Unmodified}} = H_{Q-1} + H_{Q-2} + H_{Q-100} + H_{Q-3} + H_{Q-4} + H_{Q-5} + H_{Q-104} + H_{Q-7} + H_{Q-C} + H_{Q-P}$$

$$\Delta H_{\text{Unmodified}} = 430803802 \text{ kJ/h}$$

$$\Delta H_{\text{Modified}} = H_{Q-1} + H_{Q-2} + H_{Q-3} + H_{Q-5} + H_{Q-104} + H_{Q-7} + H_{Q-C} + H_{Q-P}$$

$$\Delta H_{\text{Modified}} = 31253320 \text{ kJ/h}$$

$$\%\text{Energy Efficiency} = \frac{430803802 \text{ kJ/h} - 31253320 \text{ kJ/h}}{430803802 \text{ kJ/h}} \times 100\%$$

$$\%\text{Energy Efficiency} = 92.75\%$$

4. Conclusions

Based on this research, net energy efficiency can be increased by adding a heat exchanger and reducing a cooler will result in an increase in net energy efficiency of 92.75%. Combining heat recovery through the use of a heat exchanger and reducing the energy consumption of cooling systems is recommended to significantly improve overall energy efficiency. This approach minimizes energy required for both heating and cooling, allowing more of the energy generated within the system to be used for driving reactions, such as sulfur burning and oxidation, instead of being lost as waste heat or used in inefficient cooling.

CRediT Author Statement

Author Contributions: D.P. Andini: Conceptualization, Methodology, Investigation, Resources, Data Curation, Review and Editing, Supervision; L.F. Santoso: Conceptualization, Methodology, Formal Analysis, Data Curation, Writting Draft Preparation, Visualization; S.P. Bersa: Investigation, Software, Resources, Writing, Review and Editing, Validation, Supervision. All authors have read and agreed to the published version of the manuscript.

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Table 3. Energy analysis of the unmodified processes

	Unit	Q-1	Q-2	Q-100	Q-3	Q-4
Heat Flow	kJ/h	167800	12340000	167800	15530000	1630000
	Unit	Q-5	Q-104	Q-7	Q-C	Q-P
Heat Flow	kJ/h	10911071	88022900	182507650	75051560	44475021

Table 4. Energy analysis of the modified processes

	Unit	Q-1	Q-2	Q-3	Q-5	Q-104
Heat Flow	kJ/h	167800	10710000	15530000	1091000	88020
	Unit	Q-7	Q-C	Q-P		
Heat Flow	kJ/h	1825000	750500	1091000		

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