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Journal of Chemical Engineering Research Progress, 1 (2) 2024, 97-107

Research Article

Minimizing Energy Usage in the Production of Benzene through Hydrodealkylation Toluene Process by Optimizing Heat Transfer Unit in Reactor System

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Received: 15th June 2024; Revised: 19th June 2024; Accepted: 19th June 2024
Available online: 25th June 2024; Published regularly: December 2024

Abstract

Benzene is a chemical raw material that is widely used without alternatives in the production of high energy solid liquid fuels and polymers. As a result, the global demand of benzene reaches 51 million per year. The process simulator has been utilized to simulate the reactor system of benzene production through the hydrodealkylation of toluene using Peng-Robinson equation-of-state property package. This system is designed to reach 200,000 tons of benzene per year with an optimized heat flow mechanism. By using a heat recovery strategy that utilizes the heat stream outlet from the waste heat boiler (WHB-01) and the partial condenser (PC-01), the net-energy in the simulation has been effectively minimized by saving a total of -23,915,490.40 kJ/h by directing the heat streams to heaters H-01 and H-02, respectively. Considering this strategy, the modified process within the reactor system is conclusively more optimized than the basic process system.

Keywords: Benzene; toluene; hydrodealkylation; simulation; net energy optimization

How to Cite: E. I. Maulana, A. Tarikh, R. T. Widaranti, (2024). Minimizing Energy Usage in the Production of Benzene through Hydrodealkylation Toluene Process by Optimizing Heat Transfer Unit in Reactor System. *Journal of Chemical Engineering Research Progress*, 1 (2), 97-107 (doi: 10.9767/jcerp.20167)

Permalink/DOI: https://doi.org/10.9767/jcerp.20167

1. Introduction

Benzene is a petrochemical product that has a single hexagonal ring structure and is an aromatic compound with the molecular formula C_6H_6 [1]. Benzene is a chemical raw material that is widely used without alternatives in the production of high energy solid liquid fuels and polymers. Produced from fossil carbon, benzene consumption contributed to increased CO_2 levels in the atmosphere at the end of life [2]. Benzene is a colorless substance that is volatile, flammable, toxic, and only slightly soluble in water. Benzene has many applications in industry, including as a solvent in extraction and distillation processes. Additionally, benzene is widely used as an intermediate in the production of various other chemicals, with about two-thirds of all chemicals containing benzene, making its presence in the environment very common [1]. Benzene is used as a light component in fuels to improve knocking characteristics and increase octane rating [2]. At present, almost all benzene is produced via crude oil refining, unlocking fossil stored carbon and contributing to global warming [3]. The estimation of global benzene demand reaches 51 million tons per year [2].

Hydrodealkylation of toluene is a suitable reaction to be coupled with naphtha reforming process [4]. Hydrodealkylation of toluene to benzene is an important petrochemical process which is the largest since benzene is more applicable than toluene [5]. Most of the produced toluene is converted to benzene via hydrodealkylation or disproportionation reactions [6]. Hydrodealkylation of toluene is carried out

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both catalytically and homogeneously. The catalytic reaction is performed on hydrogenation dehydrogenation catalyst such as nickel or clinoptilolite. However, higher temperatures can be used instead of a catalyst [3].

The hydrodealkylation (HDA) process is a higher aromatic change process to benzene. This process produces benzene using raw materials hydrogen and toluene with a high purity of 99.8%. This process takes place at high temperature and pressure. The hydrogen removes alkyl groups in aromatic compounds and produces benzene and light paraffin gas. The hydrodealkylation process uses a chromia-alumina catalyst with operating conditions of temperature 575-650 °C, pressure 25-60 atm and conversion 85% [7]. Product quality is controlled through the product composition in the distillation column and the control variables chosen are product purity in the benzene column and reactor inlet temperature. Use of the HDA process to demonstrate flowsheet design condition procedures and presents an integrated heuristic hierarchical approach to process and control system design and uses the entire HDA process using the HDA factory model where steam generated from waste-feed heat exchanger waste (FEHE) via series of steam coolers [8].

From a material balance point of view, the control variable chosen are fresh toluene feed flow rate (control production rate) and recycled gas flow rate. Quality of the product is controlled through the product composition in the distillation column and variables the selected control is the purity of the product on the benzene column and reactor inlet temperature. Utilizing the heat generated from a cooler to reheat another heat exchanger is to improve heat efficiency in the benzene production process [8]. The aim of this research is to enhance heat efficiency in benzene production by utilizing a fluid loop heat transfer system for heating and cooling processes in the reactor system using Aspen HYSYS

2. Methods

2.1. Process Simulator

Many chemical engineering programs available such as Aspen Plus, Aspen HYSYS, UniSim, CHEMCAD, PRO/II, and DWSIM. These programs enable the efficient resolution of intricate process engineering challenges, providing chemical engineers with the means to perform rapid, sophisticated calculations and the accuracy of the computations make it practicable to design better processes [9-10]. Aspen HYSYS has been selected for simulating the processes because of its high popularity among researchers [11].

Aspen HYSYS stands as a versatile process simulation platform serving a wide range of processing industries. It is very popular in the oil, gas, and refining fields, offering a powerful thermodynamic framework for precise calculations of physical and transport properties, as well as phase behavior [12]. This user-friendly and extensible software comes with many add-ons, thereby increasing its usability in various industrial sectors. Users can create detailed steady-state and dynamic models for simulate factory processes and designs. Additionally, Aspen HYSYS facilitates monitoring, troubleshooting, operational improvement, business strategy formulation, and asset management. Its fully interactive interface simplifies the manipulation of process variables and configuration of unit operations, applicable in both steady-state and dynamic operations [13].

2.2. Basic Process Flow Diagram

Figure 1 shows the simplified process flow diagram for HDA Toluene process [14]. Benzene is produced by the HDA of toluene. Liquid toluene, hydrogen gas and a smaller amount of methane gas are entering furnace to reach the HDA temperature requirement inside the reactor. Then, the reactor outflow is fed into the phase split system. The liquid phase mostly contains benzene and diphenyl, while the gas phase mostly contains hydrogen and methane gas. The liquid phase flow is directed to liquid separation system to separate benzene and diphenyl. With a certain ratio, the separation output stream is recycled into the reactor system to increase the process yield. The same thing is done in the vapor recovery system. For this research, the modification process focuses only on the reactor system.

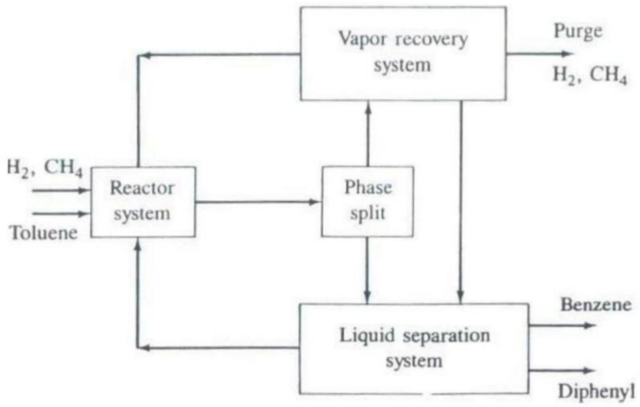

Figure 1. Simplified process flow diagram of HDA toluene to produce benzene [9].

2.3.1 Reaction mechanism

The process of making benzene using the thermal hydrodealkylation method is carried out in conversion reactor with the main reaction:

$$C_7H_8\,_{(g)} + H_2\,_{(g)} \rightarrow C_6H_6\,_{(g)} + CH_4\,_{(g)} \qquad \Delta H^o_{298} = -41952 \text{ kJ/kmol}$$

and side reactions:

$$2C_6H_6\,_{(g)} \leftrightarrow C_{12}H_{10}\,_{(g)} + H_2\,_{(g)} \qquad \Delta H^o_{298} = 0 \text{ kJ/kmol}$$

Because Aspen HYSYS V10 can not use conversion and equilibrium reaction in one reaction set within the conversion reactor (CRV-100), the side reactions then divided into two conversion reaction; one for the forward reaction and two for the reverse reaction:

$$2C_6H_6\,_{(g)} \rightarrow C_{12}H_{10}\,_{(g)} + H_2\,_{(g)} \quad \Delta H^o_{298} = 8118 \text{ kJ/kmol}$$

$$C_{12}H_{10}\,_{(g)} + H_2\,_{(g)} \rightarrow 2C_6H_6\,_{(g)} \qquad \Delta H^o_{298} = -16236 \text{ kJ/kmol}$$

The conversion of the side reactions can be calculated using dummy equilibrium reactor (ERV-100) to replicate the CRV-100 system as shown in Figure 2. The reaction set for the dummy equilibrium reactor was set only for the pre-divided side reactions, so that the product result will not be mixed with the product from main reaction. The main reactor simulation was run with the conversion of side reactions each assumed to be 1%, then the molar composition from the CRV-100 inlet — which are '*Tol. Mixed Heated*' and '*H2 FH-01*'— was replicated to '*Toluene (Dummy)*' and '*Hydrogen (Dummy)*', respectively. The heat (Q-ERV100) was added into the dummy equilibrium reactor to match the outlet product temperature from the main simulation. Result of this simulation can be seen in Table 1. By using the conversion formula for Benzene, the actual conversion for the side reaction was calculated to be 1.31%.

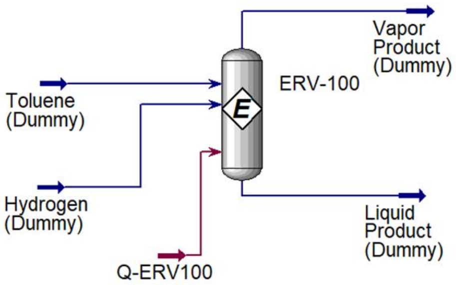

Figure 2. Simulation of dummy equilibrium reactor.

2.3.2 Operating Conditions

The selection of the reaction temperature within the reactor requires careful consideration of the reaction phases and a thorough thermodynamic analysis. For this reason, the reaction temperature is maintained at an optimum temperature of 621-694 °C. Exceeding the specified temperature range induces hydrocracking, leading to a lower conversion. Conversely, temperatures below this range result in a slower reaction progression [15]. Under these operating conditions, a conversion of 75% and selectivity of 97% is achieved.

The reaction pressure in the reactor is determined at 34 atm (range 14-68 atm) with a review of the condition of the reactants in the reactor is the gas phase. In this hydrodealkylation reaction, a mole ratio is used hydrogen and toluene feed is 5:1. The mole ratio of these reactants is related to their presence. The side reaction involves diphenyl formation, which contributes to coke production. The formation of this coke can be limited by keeping the hydrogen concentration high. In other words, the mole ratio of hydrogen and toluene feed 5:1 is used to obtain

Table 1. Simulation result of dummy equilibrium reactor

	Unit	**Toluene (Dummy)**	**Hydrogen (Dummy)**	**Q-ERV100**	**Vapor Product (Dummy)**	**Liquid Product (Dummy)**
Vapour Fraction		1.00	1.00	-	1.00	0.00
Temperature	*C*	570.60	641.80	-	637.30	637.30
Pressure	*atm*	34.00	34.00	-	34.00	34.00
Molar Flow	*kgmole/h*	4,003.95	2,860.00	-	6,863.95	0.00
Mass Flow	*kg/h*	110,932.98	5,816.62	-	116,749.62	0.00
Heat Flow	*kJ/h*	192,197,599.31	51,519,328.27	24,587,898.95	268,304,826.53	0.00
Master Comp Molar Flow (Toluene)	*kgmole/h*	642.18	0.00	-	642.18	0.00
Master Comp Molar Flow (Hydrogen)	*kgmole/h*	2,374.79	2,856.37	-	5,234.44	0.00
Master Comp Molar Flow (Benzene)	*kgmole/h*	501.72	0.00	-	495.18	0.00
Master Comp Molar Flow (Methane)	*kgmole/h*	485.26	3.63	-	488.89	0.00
Master Comp Molar Flow (BiPhenyl)	*kgmole/h*	0.00	0.00	-	3.27	0.00

the speed of coke formation for the small one [15]. Hydrogen was chosen as the excess reactant because for the same number of moles, weight of hydrogen needed is smaller than toluene. Reaction to form benzene by thermal hydrodealkylation toluene process is run under non-isothermal adiabatic conditions where the reaction is maintained at an optimum temperature of 621-694 ^{o}C with a pressure of 34 atm.

2.3.3 Raw Materials

The raw material used in this process is toluene (C_7H_8) and hydrogen gas (H_2). According to [11], the most optimal mole ratio of H_2 to C_7H_8 is 5:1. Fresh feed hydrogen with a purity of 99.99% by weight flowed using a pipe at pressure 20 atm and temperature 35 ^{o}C. The pressure of fresh feed hydrogen was increased using a compressor (K-01) to 34.1 atm. After increasing the pressure, the temperature of hydrogen was increased to 192 ^{o}C by flowing it to the heat exchanger (HE-01) utilizes the heat produced by the reactor. Furthermore, hydrogen flows to the furnace (FH-01) together with toluene to be heated to a temperature of 621 ^{o}C before being reacted into the reactor.

Toluene with a purity of 98.5% by weight is stored in liquid phase at a temperature of 30 ^{o}C and pressure 1 atm. Toluene from the storage tank is then pumped. Toluene mixture of mixing pipe which is at a temperature of 61 ^{o}C with a pressure of 1 atm then pumped and flowed to the operating furnace (FH-01) at 34 atm conditions to increase the temperature to 621 ^{o}C. The toluene phase transformed from liquid to gas in the furnace because the reaction in the reactor occurs in the gas phase. The temperature of the reactor output product is 694 ^{o}C. Reactor products flowed to the heat exchanger (HE-01) to use the heat so the temperature drops to 642 ^{o}C. Then, the heat from reactor product is reused to produce steam in the waste heat boiler (WHB-01) and further cooled in partial condensor (PC-01) so that the temperature decreases to 190 ^{o}C before carrying out to the product purification stage.

2.3. Modification Strategy for Heat Transfer Unit

The strategy modification is to change the components used. In the references found, the factory design utilities used heaters and coolers separately. Modifications are made by reusing the heat from waste heat boiler and partial condensor into the heaters. This aims to improve energy efficiency. In a typical chemical process, a heat exchanger network can be designed to facilitate the transfer of heat from streams that need to be cooled to materials that need to be heated [17]. This can be achieved by using the output of a cooler as a heating medium in a heater for another part of the process [18]. This method can be used to reduce utility consumption in chemical processes which can lead to reduced utility costs and energy consumption [19]. In utility operations, conserving energy through its effective utilization correlates with a decrease in energy use and/or a lesser dependence on traditional energy sources, thereby impacting the cost of production favorably. Thus, the implementation of heat exchanger networks by reusing cooling water from the coolers into the heaters is expected to result in an optimal compromise between equipment and operation, potentially establishing the most energy-efficient configuration for chemical processing plants [20]. The net-energy of basic and modified process can be compared using net-energy formula as follows:

$$NE = EP - EC \tag{1}$$

where, NE is net-energy, EP is energy produced, and EC is energy consumed with all units in kJ/h [21]. In other word, EP is the energy output of a process and EC is the energy input of a process. The effect of process modification is studied with this calculation.

3. Results and Discussion

3.1. Basic Process Flow Diagram and Modification

The design capacity of the industrial benzene production device from a factory is 200,000 metric tons of benzene per year. The Peng-Robinson equation-of-state (PR EOS) is used to determine the thermodynamic properties in this study. Oil, gas and petrochemical applications uses the PR EOS because of its accuracy for variety of systems over a wide range of conditions. The system efficiently and reliably computes solutions for a majority of single-phase, two-phase, and three-phase systems with rigorous precision [22]. The PR equation-of-state may be more accurate, as it specifically meant for real gases [23].

The basic process flow diagram was taken from [24]. Hydrodealkylation reaction is carried out in a conversion reactor. Conversion reactor simulations are carried out by determining the conversion value of a reaction, i.e. 75%. Because the reaction proceeds in gas phase, the mass flow in liquid product stream is zero. The vapor products were then divided into two streams, directed to a heat exchanger (HE-01) and the latter recycled. Then heat the reactor product is reused to produce steam in the waste heat boiler (WHB-01), prior of cooling in partial condensor (PC-01) to decrease the crude product stream temperature to 190 ^{o}C.

Figures 3-4 shows that the heat from waste heat boiler and partial condensor is not used. Modifications were made by directing those unused heat streams into the process that needs a heat source. The heat from waste heat boiler (WHB-01) was directed to heat the toluene feed in H-01. Meanwhile, the heat from partial condensor (PC-01) was directed to heat the toluene after they were mixed with the pure toluene stream and the recycled stream in H-02 as depicted in Figures 5-6. A cooler (C-01) then added for the modified process so that the temperature of Crude Product stream match the temperature of Crude Product stream in the basic process flow diagram, i.e. 190 °C. The mass and energy balances is reported in Table S1 as Supporting Information (SI). This modifications is expected to have a more energy-efficient process.

3.2. Reduced Energy Consumption by Process Modification

In this study, the heat transfer unit of the benzene production process was modified. The heat transfer unit H-01 was modified by using heat recovery from PC-01, and the unit H-02 used heat recovery from WHB-01. A cooler C-01 was added to the heat transfer unit modification to achieve the same temperature as the crude product stream before modification. The net-energy produced decreased significantly from 52,238,925.441 kJ/h to 28,352,155.071 kJ/h, saving a total of 23,886,770.37 kJ/h worth of energy (Table 2).

In terms of energy efficiency, achieving zero net-energy is the ideal target. It represents a perfect balance between the amount of energy

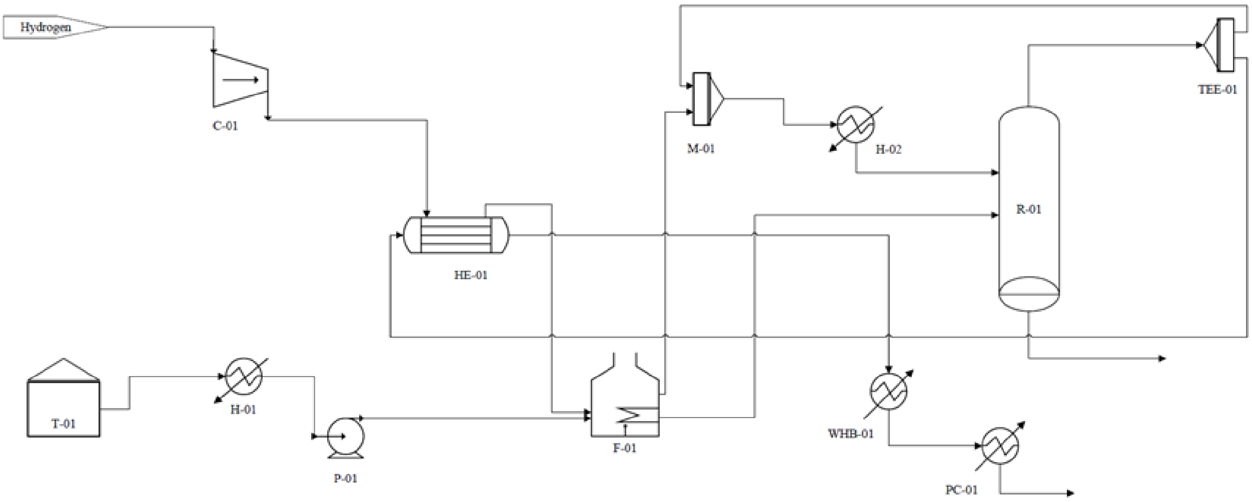

Figure 3. Basic Process Flow Diagram (PFD) before process modifications

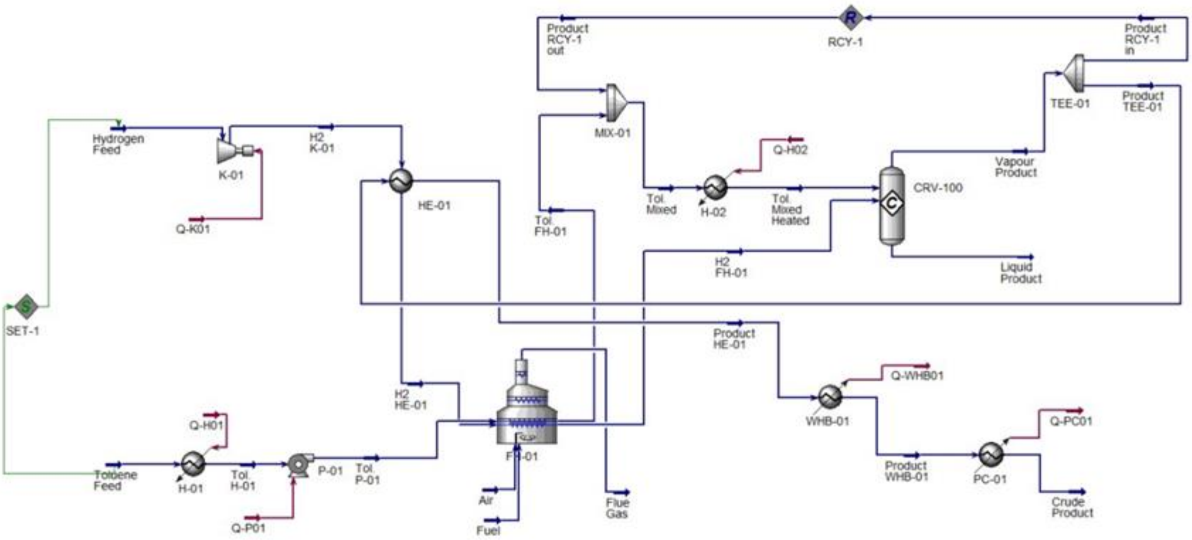

Figure 4. Aspen HYSYS simulation of the basic PFD before process modifications

Table 2. Modification effect in heat transfer unit

Energy stream (kJ/h)	Before modification	After modification
	Input	
Q-K01	5,024,216.057	5,024,216.057
Q-H01	2,685,924.398	21,156,060.949
Q-H02	17,480,718.172	2,685,924.398
Q-P01	282,157.902	282,157.902
	Output	
Q-C01	-	57,496,522.836
Q-WHB01	66,254,474.755	-
Q-PC01	11,457,467.214	-
Net-Energy	52,238,925.441	28,352,155.071

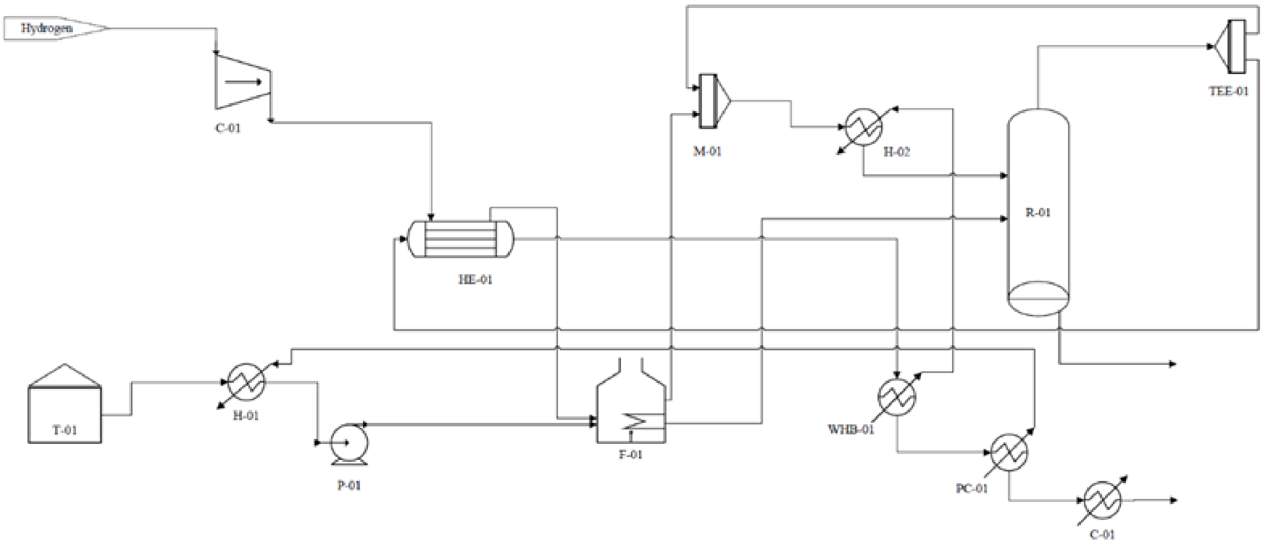

Figure 5. Revised PFD after process modifications

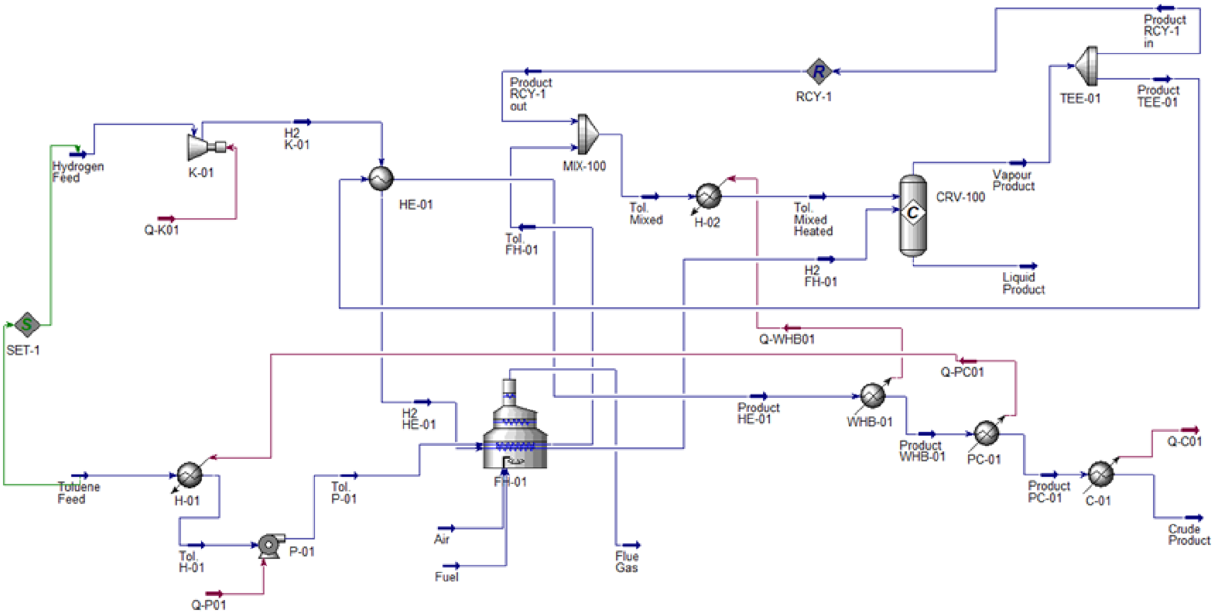

Figure 6. Revised simulated PFD after process modifications using Aspen HYSYS

produced and the amount consumed. Value of net-energy closer to zero suggests that the energy efficiency is better [25]. Thus, the modified process is demonstrably proven to be more energy-efficient than the basic process.

4. Conclusion

Simulation of benzene producton through hydrodealkylation toluene process in reactor system with optimized heat flow system capable of producing benzene with 200,000 tons per year is done using Aspen HYSYS V10 process simulator. PR EOS is chosen as the property package in the simulation. Energy consumption of simulation is reduced from 52,238,925.441 kJ/h to 28,352,155.071 kJ/h by reusing heat output from waste heat boiler (WHB-01) and partial condensor (PC-01) to H-01 and H-02. Based on the findings, it was concluded that the production of benzene from hydrodealkylation toluene with lower net-energy consumption in reactor system itself is technically feasible. Further research into energy integration to the vapour recovery system and liquid separation system is necessary to optimize the entire hydrodealkylation process.

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