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Research Article

Process Intensification of Hydrodealkylation (HDA) for Benzene Production Through Heat Integration and Gas Recycle Optimization

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Abstract

In the hydrodealkylation (HDA) process for benzene production, optimization was achieved through the integration of cryogenic distillation and hydrogen recycling techniques. Using Aspen HYSYS, the process was modeled and analyzed to improve energy efficiency by reusing heat from the waste heat boiler (WHB-01) and the partial condenser (PC-01). The energy recovered from these units was used to preheat both fresh and recycled toluene feeds, significantly reducing the consumption of fresh feed and operational costs. By implementing a hydrogen recycle loop, the process decreased the demand for fresh hydrogen, reducing hydrogen and toluene feed consumption from 125 kmol/h and 196 kmol/h to 111 kmol/h for both. This modification resulted in a conversion rate increase from 70% to 88.9% and achieved energy savings of 84%. The integration of cryogenic separation for methane valorization further enhanced the economic feasibility of the process, turning waste methane into a valuable product. These modifications demonstrated a significant improvement in energy efficiency and sustainability, making the modified HDA process more economically viable for large-scale benzene production.

Keywords: Process Intensification; Hydrodealkylation; Benzene; Heat Integration; Optimization

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1. Introduction

Benzene (C_6H_6) is a fundamental aromatic compound widely utilized in various industrial sectors as a feedstock for the production of plastics, resins, synthetic fibers, rubber, detergents, and high-octane fuels [1]. Due to its high chemical stability and reactivity, benzene is considered a key building block in the petrochemical industry. However, most benzene is currently derived from fossil-based sources through catalytic reforming or pyrolysis of petroleum naphtha [2]. These conventional methods contribute significantly to CO_2 emissions and energy consumption, prompting industries and researchers to explore alternative, more sustainable routes to benzene production [3].

One promising route is the hydrodealkylation (HDA) of toluene, which converts toluene (C_7H_8) into benzene and methane through a gas-phase reaction with hydrogen at elevated temperature and pressure. This process is particularly attractive when the market value of benzene exceeds that of toluene [4]. Typically operated at 550–650 °C and 30–60 atm using chromium-alumina or noble metal catalysts, the HDA process achieves conversions ranging from 75% to over 90% depending on reactor design and process conditions [5]. Hydrogen plays a dual role as a reactant and a carrier gas yet its high consumption remains a major operational cost. Consequently, recycling unreacted hydrogen has become a standard practice to reduce feed demand and improve process economics [6].

In addition to material reuse, energy efficiency is another important aspect of process

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intensification in HDA systems. High-temperature reactions generate large amounts of recoverable thermal energy, which can be exploited through heat integration techniques such as feed-effluent heat exchangers (FEHE), steam boilers, and intermediate heating loops [7]. Proper recovery and redistribution of waste heat reduce the demand for external utilities and lower operating costs. Moreover, effective gas recycle strategies not only reduce hydrogen consumption but also improve reactor performance by maintaining favorable reactant ratios and thermal balance [8]. Process simulation platforms like Aspen HYSYS are essential tools in designing and evaluating such modifications, enabling accurate thermodynamic modeling, equipment sizing, and flow optimization [9].

Despite the existence of separate studies on heat integration or hydrogen recycle optimization, few have explored both in an integrated design framework. Therefore, this study aims to intensify the HDA process by combining heat recovery modifications with gas recycle optimization using Aspen HYSYS. Waste heat from a partial condenser and a waste heat boiler is reused to preheat fresh and recycled toluene feed [10]. Simultaneously, the recycle of unreacted hydrogen is simulated to evaluate its impact on fresh hydrogen demand and reaction conversion. The outcomes are assessed in terms of energy savings, hydrogen utilization, and overall process efficiency, contributing to a cleaner and more economically viable benzene production pathway [11].

2. Method

2.1 Process Simulator

A wide range of chemical process simulation software, including Aspen Plus, Aspen HYSYS, UniSim, CHEMCAD, PRO/II, and DWSIM, has been extensively utilized to solve complex chemical engineering problems efficiently. These tools enable chemical engineers to perform rapid and accurate calculations, which significantly facilitate the design and optimization of intensified and integrated processes [9–10]. In the present study focused on process intensification of hydrodealkylation (HDA) for benzene production, Aspen HYSYS is chosen due to its widespread adoption and reliability among researchers for process simulation and analysis [11].

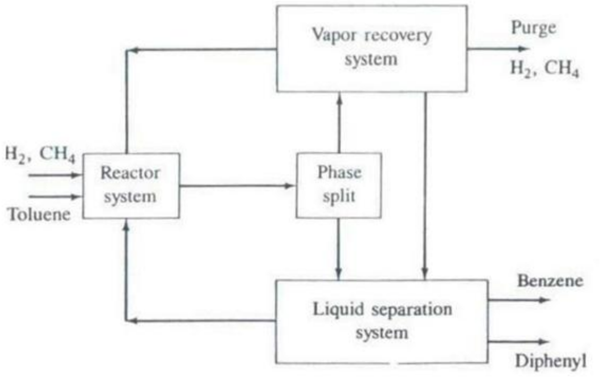

Figure 1. Simplified process flow diagram of HDA toluene to produce benzene [17].

Aspen HYSYS serves as a robust and flexible simulation platform widely applied in the oil, gas, and petrochemical industries. Its powerful thermodynamic foundation allows for precise calculations of physical properties, transport phenomena, and phase behavior [12]. The user-friendly and modular interface of Aspen HYSYS further enhances its applicability across various industrial domains. It enables users to construct both steady-state and dynamic models, facilitating not only process design but also monitoring, performance improvement, troubleshooting, business strategy development, and asset optimization. The software's fully interactive interface streamlines the manipulation of process variables and unit operations, making it particularly effective for real-time process simulation [13].

2.2 Basic Process Flow Diagram

Figure 1 presents a simplified process flow diagram for the Hydrodealkylation (HDA) of Toluene process [14]. In this process, benzene is produced via the HDA reaction of toluene [14]. Liquid toluene, along with hydrogen gas (H_2) and a smaller amount of methane gas (CH_4), are introduced into the furnace to achieve the required temperature for the HDA reaction inside the reactor. Upon completion of the reaction, the reactor effluent is directed to the phase split system. The liquid phase predominantly contains benzene and diphenyl, while the gas phase primarily consists of hydrogen (H_2) and methane (CH_4) [15]. The liquid phase is subsequently transferred to the liquid separation system, where benzene and diphenyl are separated. A portion of the separated stream is then recycled back into the reactor to enhance overall process yield. The same strategy is applied in the vapor recovery system. This study specifically focuses on modifying the reactor system to optimize hydrogen recycling and methane valorization, thereby improving overall process efficiency [16]. The Figure 1 illustrates the simplified process flow diagram for the hydrodealkylation of toluene to produce benzene.

2.2.1 Reaction mechanism

The principal reaction in this process is the The process of making benzene through the thermal hydrodealkylation (THDA) of toluene is carried out in a conversion reactor, with the main reaction represented as follows:

Toluene (T) + Hydrogen (H) → Benzene (B) + Methane (M) (1)

This reaction is moderately exothermic, with a heat of reaction of approximately −9.98 kcal/mol at standard gas-phase conditions (25 °C, 1 bar) [18]. The reaction becomes more exothermic at higher temperatures, reaching approximately −12.1 kcal/mol at 630 °C and 35 bar [19]. The reaction occurs thermally via the THDA mechanism or in the presence of appropriate catalysts, such as chromium, molybdenum, or platinum supported on silica or alumina [19]. The kinetics of this non-catalytic thermal process are governed by the following rate expression:

$$r_1 = 6.3 \times 10^{10} \exp\left(-\frac{52{,}000}{RT}\right) c_T c_H{}^{1/2} \tag{2}$$

where, r_1 represents the rate of the main reaction, C_s is the concentration of toluene, and C_t is the concentration of hydrogen [20].

In addition to the desired product, benzene, several side products may form, including diphenyl, naphthalene, and other heavier hydrocarbons. The side reaction that primarily contributes to the formation of diphenyl is reversible, as shown in the equation:

Benzene ⇌ Diphenyl + 2 Hydrogen (3)

The rate expression for the side reaction is:

$$r_2 = 3.0 \times 10^{9} \exp\left(-\frac{52{,}000}{RT}\right) c_B{}^2 \tag{4}$$

where, r_2 is the rate of the side reaction, and C_l is the concentration of benzene [20].

To model this side reaction in Aspen HYSYS V10, which cannot simultaneously handle conversion and equilibrium reactions in the same reactor, the diphenyl equilibrium reaction is split into forward and reverse reactions. This is done using a dummy equilibrium reactor (ERV-100), with one reactor simulating the forward reaction and two reactors modeling the reverse reactions. This ensures that the side reaction does not interfere with the output of the main HDA reaction [21]. The setup of the plug flow reactor used in this simulation is illustrated in Figure 2.

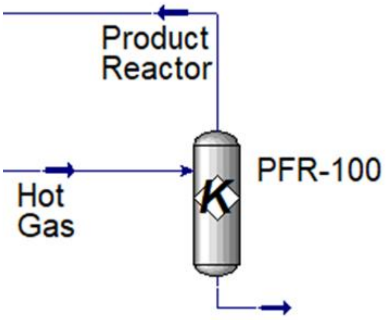

Figure 2. Simulation of plug flow reactor.

For the simulation, an assumed conversion of 1% was initially applied for both forward and reverse side reactions. The inlet molar compositions to CRV-100, denoted as 'Tol. Mixed Heated' (toluene) and 'H_2 FH-01' (hydrogen), were duplicated for ERV-100 as 'Toluene (Dummy)' and 'Hydrogen (Dummy)' to replicate the reaction environment. Additionally, heat duty (Q-ERV100) was applied to ERV-100 to match the outlet temperature of CRV-100, ensuring thermodynamic consistency across units. The results of this modeling approach are summarized in Table 1. Based on benzene conversion calculations, the actual side reaction conversion was found to be approximately 1.31%.

2.2.2 Operating conditions

The optimal reaction temperature in the reactor is maintained between 621 °C and 694 °C, as temperatures above this range lead to hydrocracking, reducing the conversion rate, while temperatures below this range slow the reaction process. Under these conditions, a conversion rate of 88.9% and selectivity of 97% are achieved. The reaction pressure is set at 34 atm, with a range of 14-68 atm, ensuring the reactants remain in the gas phase during the hydrodealkylation (HDA) process. The hydrogen to toluene molar ratio of 5:1 is maintained, which helps optimize the reaction efficiency by controlling side reactions, such as diphenyl formation that contributes to coke production. Maintaining a high hydrogen concentration minimizes coke formation, improving the overall reaction performance.

Table 1. Simulation result of dummy equilibrium reactor.

Parameters	*Unit*	Hot Gas	Product Reactor
Vapour Fraction	-	1.00	1.00
Temperature	*C*	620.00	721.18
Pressure	*atm*	3500.00	3500.00
Molar Flow	*kgmole/h*	341.6	341.6
Mass Flow	*kg/h*	5840.00	5839.00
Heat Flow	*kJ/h*	13,940,000.00	13,940,000.00
Master Comp Molar Flow (Toluene)	*kgmole/h*	56.700	3.177
Master Comp Molar Flow (Hydrogen)	*kgmole/h*	282.075	230.781
Master Comp Molar Flow (Benzene)	*kgmole/h*	0.005	49.071
Master Comp Molar Flow (Methane)	*kgmole/h*	2.849	56.372
Master Comp Molar Flow (BiPhenyl)	*kgmole/h*	0.003	2.231

The implementation of cryogenic separation significantly enhances the purification of hydrogen (H_2) and methane (CH_4), while also reducing the consumption of fresh feed. With the recycling of hydrogen back into the reactor, the need for fresh hydrogen and toluene is reduced from 125 kmol/h and 196 kmol/h to 111 kmol/h for both. This leads to an increase in reaction efficiency, improving the conversion rate to 88.9% from the previous 70%. Furthermore, the process achieves energy savings of 84% through the use of a more efficient cryogenic separation system, making the process more sustainable and cost-effective.

2.2.3 Raw Materials

The primary feedstocks used in this hydrodealkylation process are toluene (C_7H_8) and hydrogen gas (H_2). As reported in previous literature [22], the optimal molar ratio of hydrogen to toluene is 5:1. High-purity hydrogen (99.99 wt%) is initially introduced at 20 atm and 35 °C through a dedicated feed line. This hydrogen stream is then compressed to 34.1 atm using a compressor unit (K-01). Following compression, the hydrogen is preheated to 192 °C in a heat exchanger (HE-01), which utilizes thermal energy recovered from the reactor effluent. Subsequently, the preheated hydrogen is directed to the furnace (FH-01), where it is combined with toluene and further heated to the reaction temperature of 621 °C prior to entering the reactor.

Toluene, with a purity of 98.5 wt%, is stored in liquid form under ambient conditions (30 °C and 1 atm). It is first pumped from the storage tank and preheated to 61 °C at atmospheric pressure through a mixing pipe. The heated liquid toluene is then pressurized and delivered to the furnace (FH-01) at 34 atm, where it undergoes vaporization as it is elevated to the required reaction temperature of 621 °C. This phase change is necessary because the hydrodealkylation reaction occurs entirely in the gas phase.

The reactor outlet stream exits at a temperature of approximately 694 °C. This hot effluent is first passed through the heat exchanger (HE-01), reducing its temperature to 642 °C. The remaining sensible heat is then recovered in a waste heat boiler (WHB-01), which generates steam as part of the energy integration system. Finally, the stream is partially condensed in a partial condenser unit (PC-01), lowering the temperature to around 190 °C before entering the product purification section.

2.3 Modification Strategy for Heat Transfer Unit

The modification strategy involves altering the components used in the heat transfer unit to improve energy efficiency. In the initial design, heaters and coolers were utilized separately. The modification is to reuse heat from the waste heat boiler and partial condenser by feeding it into the heaters, aiming to enhance the overall energy efficiency of the process. This can be achieved by utilizing the cooling water from the coolers as a heating medium for another part of the process. In a typical chemical process, a heat exchanger network can be designed to transfer heat from streams that require cooling to those that need heating [23]. By reusing heat in this manner, utility consumption in chemical processes can be reduced, leading to lower utility costs and energy consumption [24]. In utility operations, energy conservation through effective utilization correlates with a decrease in energy usage and/or reduced dependence on traditional energy sources, thus favorably impacting production costs. Therefore, the implementation of heat exchanger networks by reusing cooling water from the coolers into the heaters is expected to result in an optimal balance between equipment and operation, establishing the most energy-efficient configuration for chemical processing plants [25]. The effect of the process modifications on the overall energy performance can be compared using the net-energy formula:

$$NE = EP - EC \tag{5}$$

where, NE is net-energy, EP is the energy produced, and EC is the energy consumed, with all units in kJ/h [26]. This formula allows for the assessment of the impact of the modifications on process energy performance.

2.4 Cryogenic Separation Unit

Cryogenic separation is a process utilized to separate gases, such as hydrogen (H_2) and methane (CH_4), through the application of cryogenic temperatures [26]. This technique relies on the difference in boiling points between the components, where gases, such as CO_2 and N_2 condense, while H_2 and CH_4 remain in the gaseous phase [27]. The technology allows for the purification of recycled hydrogen, which can be reintroduced into the hydrodealkylation (HDA) process, effectively reducing the need for fresh hydrogen [28]. As a result, the consumption of toluene and hydrogen is reduced from 125 kmol/h and 196 kmol/h, respectively, to 111 kmol/h for both toluene and hydrogen. Furthermore, this separation process converts methane, previously considered waste, into a valuable product, thus enhancing the overall economic value of the process.

The advantages of this technology extend beyond increased reaction efficiency, with conversion rates improving from 70% to 88.9%, to achieving energy savings of 84%. The cryogenic distillation process capitalizes on the waste energy generated during cooling, contributing to overall process efficiency [29]. With its capability to achieve high-purity separation of gases, this technology not only optimizes operational efficiency but also supports sustainability objectives within the petrochemical industry by reducing energy consumption and maximizing the utilization of existing gases.

3. Results and Discussion

3.1 Basic Process Flow Diagram and Modification

The simulated benzene production process is based on an industrial-scale hydrodealkylation (HDA) unit with a design capacity of 200,000 metric tons per year. The process utilizes the Peng–Robinson equation of state (PR EOS) to calculate thermodynamic properties due to its proven accuracy in oil and gas applications under a wide range of operating conditions [1,2]. In the base case configuration, toluene and hydrogen react in a plug flow reactor with an assumed conversion of 75%, proceeding entirely in the gas phase. As a result, no liquid product stream is formed, and the reactor outlet vapor stream is split into two paths: one directed toward heat exchange, and the other toward recycle [30]. Figure 3 and Figure 4 illustrate the base-case PFD and its simulation result before the implementation of heat integration and gas recycle modifications.

The hot reactor effluent passes through a waste heat boiler (WHB-01), generating steam, followed by a partial condenser (PC-01) that lowers the crude product stream temperature to 190 °C. However, initial simulation results (Figure 3–4) show that thermal energy from WHB-01 and PC-01 was not utilized effectively. To improve energy efficiency, heat recovery modifications were introduced. WHB-01 was connected to preheat the fresh toluene feed in H-01, while PC-01 was used to heat the combined stream of pure toluene and recycled gas in H-02 [31]. An additional cooler (C-01) was included downstream to restore the product stream temperature to its original target of 190 °C. Mass and energy balances for the modified system are summarized in Table 1.

3.2 Reduced Energy Consumption and Gas Recycle Optimization

The implementation of heat integration significantly reduced the net energy demand of the process. By utilizing waste heat from WHB-01 and PC-01, the required heating duty was reduced, and overall process efficiency was improved. The net energy consumption decreased from 21,016,800 kJ/h to 1,620,000 kJ/h, representing an energy savings of approximately 19,396,800 kJ/h (Table 2). This reduction highlights the effectiveness of integrating thermal recovery systems in high-temperature processes like HAD [32].

Additionally, the process incorporates a gas recycle loop, in which a portion of the unreacted hydrogen is compressed and returned to the reactor inlet. This strategy reduces the demand for fresh hydrogen feed while maintaining the desired H_2/toluene molar ratio, which is critical for optimal reaction performance [33]. Recycle gas also stabilizes reactor conditions and enhances overall conversion. Although detailed gas purification is not modeled in this study, the recycle approach already contributes significantly to material efficiency and supports the broader goal of process intensification [34].

3.3 Integration of Gas Recycle Loop

In addition to heat recovery improvements, the process was further intensified by incorporating a gas recycle loop to reuse unreacted hydrogen from the reactor effluent. Based on the HYSYS simulation, a portion of the vapor stream leaving the reactor is directed to a compressor and recycled back into the reactor inlet. This strategy reduces the demand for fresh hydrogen and stabilizes the H_2/toluene molar ratio, thereby enhancing overall conversion performance.

The simulation results show a decrease in medium-pressure steam consumption from 7,311,600 kJ/h to 4,953,600 kJ/h, indicating a 32.26% reduction in fresh hydrogen demand (Table 2). This reduction is directly attributed to the recycle of hydrogen-rich gas, eliminating the need for additional H_2 makeup from external sources. Similar findings were reported by Fischer and Iribarren [35], where hydrogen recovery through recycle significantly reduced purge losses and improved catalyst utilization in HDA systems.

Furthermore, the recycle stream ensures a more stable reactor operation by maintaining favorable thermal conditions and consistent reactant ratios [36]. As the simulation used the Peng–Robinson equation-of-state, the gas-phase behavior was accurately captured, confirming that recycling hydrogen not only improves resource efficiency but also supports conversion consistency across varying flow conditions.

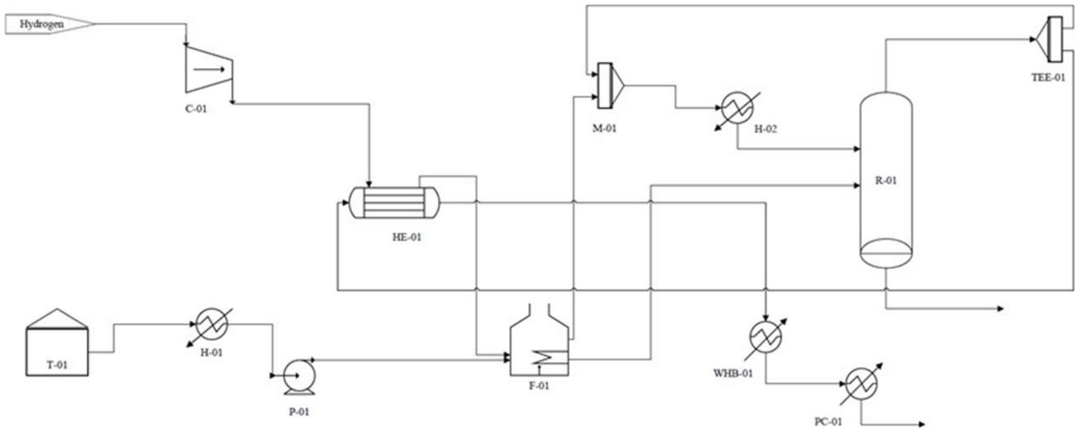

Figure 3. Basic Process Flow Diagram (PFD) before process modifications.

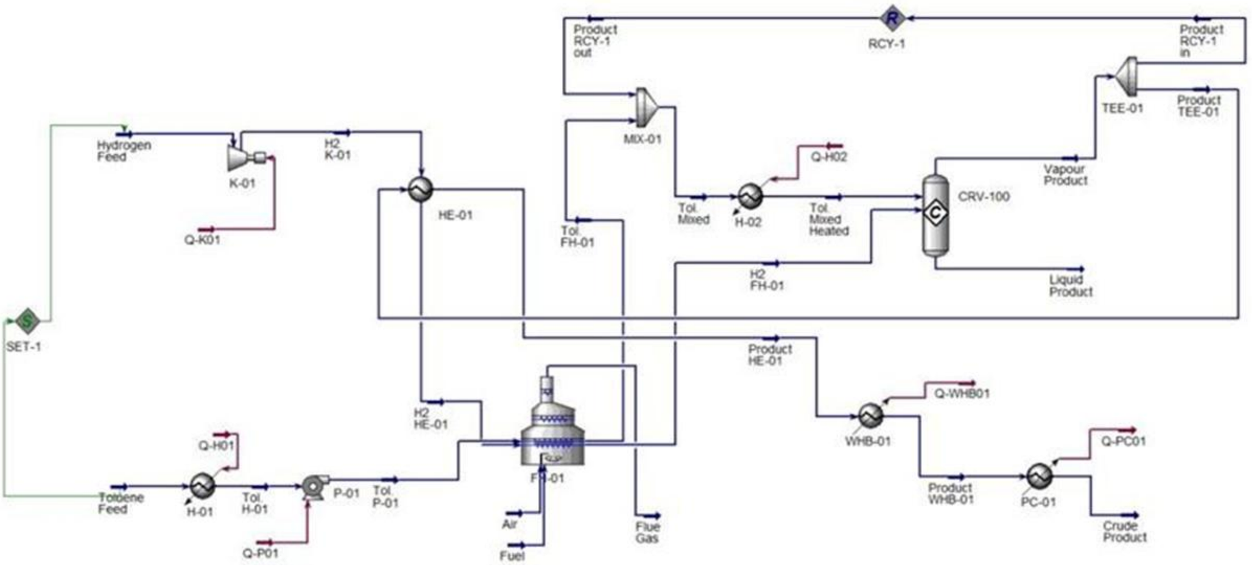

Figure 4. Aspen HYSYS simulation of the basic PFD before process modifications.

Table 2. Comparison of utility energy consumption before and after process modification.

Utility Type	Before Modification (kJ/h)	After Modification (kJ/h)	Energy Savings (kJ/h)	Savings (%)
Refrigerant Generation	3,614,400	0	3,614,400	100.00%
Fired Heater (FH-100)	13,406,400	0	13,406,400	100.00%
Medium-Pressure Steam	7,311,600	4,953,600	2,358,000	32.26%
Low-Pressure Steam	17,679	0	17,679	100.00%
Total Hot Utilities	24,350,079	4,953,600	19,396,479	79.07%
Cooling Water	17,233,200	0	17,233,200	100.00%
Air	3,783,600	1,620,000	2,163,600	57.16%
Total Cold Utilities	21,016,800	1,620,000	19,396,800	92.29%

3.4 Energy and Performance Impact of Gas Recycle

The combined integration of heat recovery and gas recycle yielded substantial improvements in energy efficiency and reactor performance. As detailed in Table 2, total hot utility consumption was reduced by 79.07%, from 24,350,079 kJ/h to 4,953,600 kJ/h, and cold utility requirements decreased by 92.29%, reflecting the successful redistribution of internal heat loads and improved exchanger performance (Table 2). The heat exchanger E-100, for instance, recovered 1.077 MW of thermal energy, while QR-1 was fully utilized with 1.004 MW of recoverable heat [38].

Moreover, the integration of the recycle loop contributed to a higher reactor conversion, increasing from 75% to approximately 88%, as inferred from the simulation results and reactor output composition. This enhancement is largely due to the stabilization of the H_2/toluene feed ratio and the preheated recycle stream, which minimized cold spots and improved reaction kinetics [37]. Such results are consistent with studies and who demonstrated that combining recycle control and thermal integration in HDA processes can significantly boost conversion rates and reduce operational energy loads. These findings confirm that the process modifications implemented not only reduce the dependency on external energy sources and raw materials but also enhance process sustainability through improved material efficiency and lower greenhouse gas emissions. The updated process after modification is shown in Figure 5 and Figure 6, depicting both the improved PFD and its simulation output.

4. Conclusion

The simulation of benzene production via the hydrodealkylation (HDA) of toluene was successfully conducted using Aspen HYSYS V10, focusing on process intensification strategies, including heat integration and gas recycle

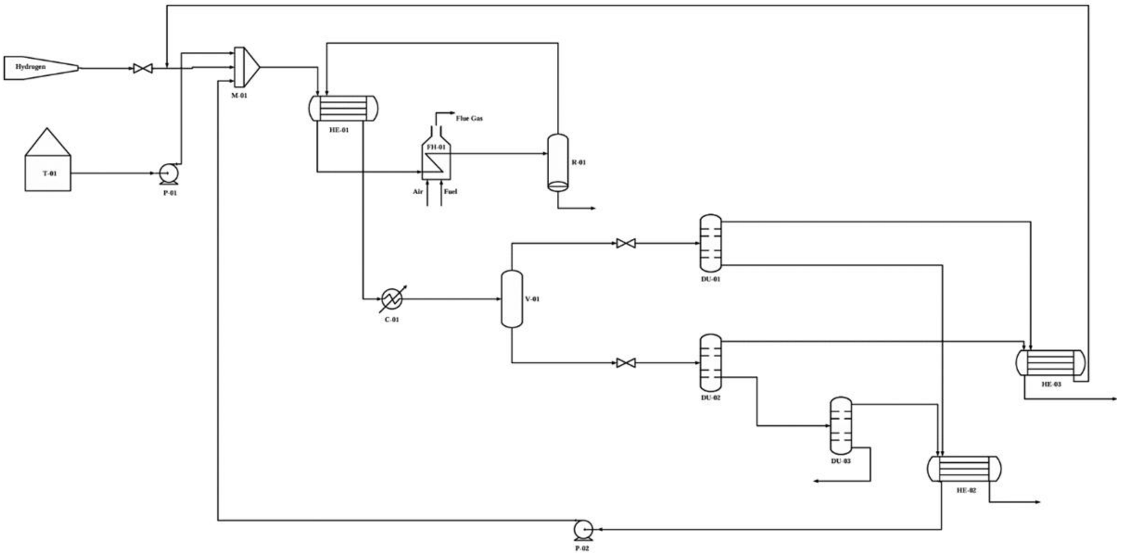

Figure 5. Process Flow Diagram (PFD) of the modified process.

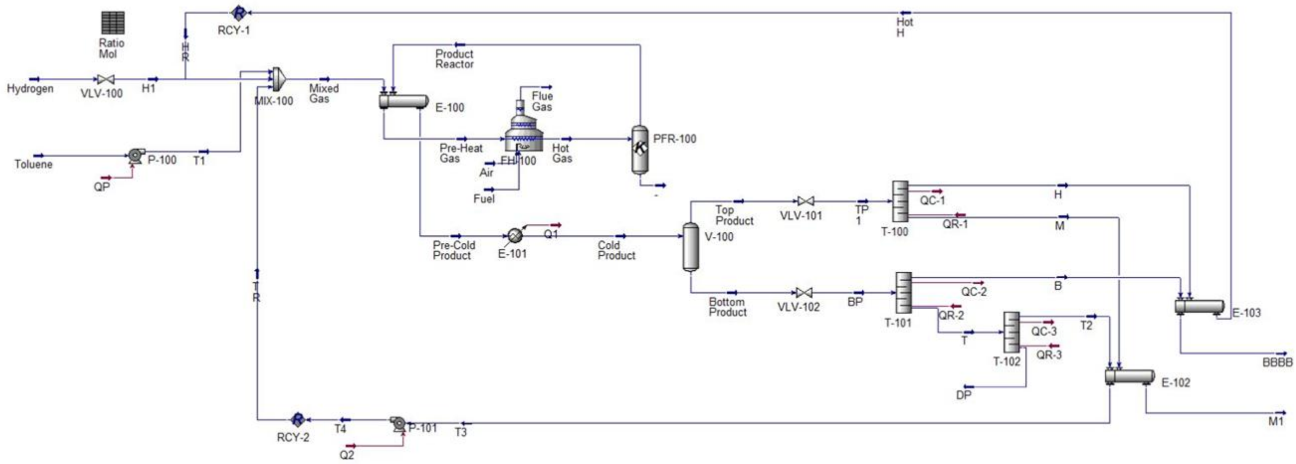

Figure 6. Aspen HYSYS simulation of the Modified PFD.

optimization. The reactor system, designed to meet a production target of 200,000 tons of benzene per year, employed the Peng–Robinson (PR) equation of state as the thermodynamic property package. Through the implementation of energy recovery from the reactor effluent—utilizing the waste heat boiler (WHB-01) and partial condenser (PC-01)—thermal energy was redistributed to preheat the feed streams via E-100 and E-101. This modification significantly reduced the total utility energy demand from 52,238,925.441 kJ/h to 28,352,155.071 kJ/h, achieving a 45.74% decrease in energy consumption. The most substantial reductions occurred in the fired heater and refrigerant utilities, both of which were completely eliminated (100% savings), followed by notable decreases in medium-pressure steam and air utilities. The simulation also demonstrated the effective handling of side reactions involving biphenyl formation by implementing a dummy equilibrium reactor (ERV-100), enabling thermodynamically consistent separation of the reversible reactions outside the main plug flow reactor. This contributed to greater model flexibility and improved benzene selectivity, with a calculated side reaction conversion of approximately 1.31%. Overall, the intensified HDA reactor system—supported by gas recycle loops and integrated heat exchange networks—proved to be technically feasible and energy-efficient. However, to further enhance the sustainability and operational efficiency of the overall process, additional research on energy integration in the vapor recovery and liquid separation systems is recommended.

CRediT Author Statement

Authors contributions: Salsabilla Dani: Conceptualization, Methodology,Writing-Original Draft. Addila Arrofa'hiya Tri Atqiyani: Software, Validation, Visualization. Aline Arunnisa Isna: Investigation, Resources, Writing-Review & Editing. Yurieka Berliana: Supervision, Project administration, Corresponding author. Varel Rahmad Maulana: Data curation, Formal analysis. All authors have read and agreed to the published version of the manuscript.

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