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Research Article

Improving Heat Efficiency of Aniline Production Process by Modifying Heat Transfer Fluid Looping System in Heating and Cooling Process

Muhammad Ahdan *[a], **Alfan Rizky Saputra** [b], **Rifqy Ivan** [c], **Yohanes Mangarohontua Panjaitan** [d]

Department of Chemical Engineering, Faculty of Engineering, Universitas Diponegoro, Indonesia.

[a] ahdan02@gmail.com; [b] aaxyrs1723@gmail.com; [c] rifqyivan23@gmail.com; [d] yohanes1506@gmail.com

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Abstract

The nitrobenzene hydrogenation process is a highly exothermic reaction which causes the product temperature will increase and then needs to be cooled down with a heat exchanger. The modification process is needed to improve the heat efficiency of the nitrobenzene hydrogenation process by utilizing the heat from reactor product. Heat transfer fluid is used as a medium of heat transfer for both heating and cooling, so that will create a looping heat transfer process. The looping system of heat transfer fluid receives heat from reactor product and then reuses the heat to reheat the recycled hydrogen and for other utilities, so additional energy for these processes is not required. The process modification was simulated using Aspen HYSYS and the comparison of heat efficiency 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 96,714,359.832 kJ/h and 9.869 kJ/h. This shows that the modified process has better energy efficiency compared to the basic process as the net-energy value is closer to zero. Therefore, this modification increases the heat efficiency of the aniline production through nitrobenzene hydrogenation process.

Keywords: Aniline; Nitrobenzene; Hydrogenation; Process modification; Heat efficiency; Net-energy

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1. Introduction

Aniline ($C_6H_5NH_2$) is an organic aromatic amine compound that has a phenyl ring (C_6H_5) and it is bonded to an amino group ($-NH_2$). Aniline has the characteristic of a colorless oily liquid which is an important organicc synthetic product that widely used in the production of aniline-formaldehyde resins, rubber additives and dyes. Aniline is classified as a volatile organic compound that can evaporated at room temperature and normal pressure. Aniline was evaluated using the traditional USEPA 2005 method which is identified as having an associated risk to human health [1-5]. However, despite all of that, at the end of 2022, aniline production in the world was about 9.4 million tons. In 2027, the aniline production is expected to reach about 12.47 million tons [6,7]. Commonly used aniline production methods in industry are by nitrobenzene hydrogenation and phenol

* Corresponding Author.
Email: ahdan02@gmail.com (M. Ahdan)

amination. However, more yields are obtained when using the nitrobenzene hydrogenation process, so it is used as a commercial route and supplying about 85% of the global aniline production [8]. Aniline production can be carried out either in gaseous or in liquid phase. For the reactions in the vapor phase, fluidized bed and fixed bed reactors are usually used at a temperatures of 200-400 °C and a pressures of 1-10 bar with nitrobenzene conversion almost 99.9% [9,10]. Catalysts that were often used in the nitrobenzene hydrogenation process is in the form of metal, such as nickel (Ni) and copper (Cu). The aniline in the isolated species, while having stability in the inverted orthonated form, has a closer energy balance. A commercial process of highly exothermic catalytic hydrogenation of nitrobenzene (ΔH_r^o = -544 kJ/mol, at 200 °C) in the vapor and liquid phases was carried out [11-13].

The process of heat management is an important policy in reaction engineering design. Heat management not only aims to improve energy efficiency, but also to extend life of system and provide operational stability [15,16]. Heat transfer process is the main procedure for energy efficiency, so it is very important to improve the heat transfer process in the system to achieve more energy efficiency [17]. In recent years, there has been an increasing intention to minimize energy costs and to improve overall operational efficiency of such facilities. One route to achieve this goal is to ensure efficient heat recovery in highly exothermic reactions. It is because there is a significant amount of heat that usually lost in the process or there is a certain amount of heat that must be transferred out of the reactor, either as a heat or work to keep the temperature below the set point, so that the product safety and quality will be maintained, or else the system temperature will increase [18-20]. Based on the explanation, it is shown that the high energy output that produced by operating reactors can be utilized to produce superheated steam for further chemical processes or to generate electricity [21]. One commonly used way is to use a multifunctional reactor where an exothermic reaction is integrated with an endothermic reaction. This can be beneficial to lower the amount of energy used and reduced operational costs by using the heat source from the exothermic reaction to drive the endothermic reaction rather than using external heat to improve efficiency [22-24]. For example, combining the nitrobenzene hydrogenation to produce aniline with the ethylbenzene dehydrogenation to produce styrene in a catalytic fixed bed membrane reactor has a potential to increase significance of both processes [25]. In the process, a certain amount of heat released from the hydrogenation process is used to break the endothermality in the dehydrogenation process where the heat is transferred through the membrane layer that heats the reacting mixture on the dehydrogenation side [26,27]. In addition, a certain amount of hydrogen generated from the dehydrogenation process permeates through the hydrogen-selective membrane, so as to improve the equilibrium conversion of ethylbenzene and react with nitrobenzene on the permeate side to produce aniline [26]. Although the energy used is more efficient, by combining the endothermic reactions and the exothermic reactions in one unit, it is possible to require additional separation and recycling units [28].

In this case, the innovation is proposed by using the heat generated from reaction in the reactor to reheat the excess hydrogen which will be reused for further process. This modification improves heat efficiency of the nitrobenzene hydrogenation process [29]. The aim of this research is to improve heat efficiency in the aniline production through nitrobenzene hydrogenation process by utilizing heat transfer fluid looping system in heating and cooling process using Aspen HYSYS.

2. Methods

In order to improve heat efficiency in nitrobenzene hydrogenation process, it is simulated using Aspen HYSYS. Fixed bed reactor is used in the nitrobenzene hydrogenation process and the reaction occurs non-adiabatically and non-isothermically. During the reaction, a large amount of heat is generated which can be used for other processes or for other utilities [30]. It is because the nitrobenzene hydrogenation process is classified as an exothermic reaction which leads to a higher reactor output temperature. In this

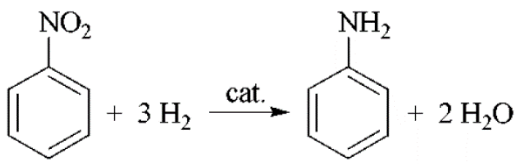

Figure 1. Hydrogenation of nitrobenzene [14]

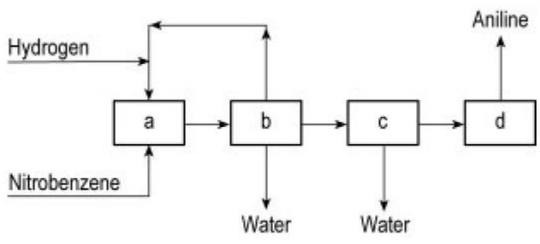

Figure 2. Simplified basic process flowsheet of hydrogenation of nitrobenzene [14]

modification, the heat received by a heat transfer fluid in the reactor product cooling process is used to reheat the recycled hydrogen. The excess hydrogen is separated from the product and then recycled [31]. The reactor output temperature is 310 °C and cooled to 38.7 °C using a mixture of ethylene glycol and water with a mass ratio of 1:1 as the heat transfer fluid. The heat received by the ethylene glycol-water mixture is used to reheat the recycled hydrogen from the knock-out drum and to generate low-pressure steam for other utility usage. The heat efficiency of basic and modified process can be compared using net-energy calculation as follows:

$$NE = E_P - E_C \tag{1}$$

where, NE is net-energy (kJ/h), E_P is energy produced (kJ/h), and E_C is energy consumed (kJ/h) [32]. The effect of this modification process is studied.

3. Results and Discussion

3.1. Comparison Between Basic and Modified Processes

The simulation of aniline production through nitrobenzene hydrogenation process for basic and modified process using Aspen HYSYS is depicted in Figures 3-6. The basic process simulation is shown in Figure 3, while the PFD of the basic process is shown in Figure 5. In the basic process, less equipment is used without any reusage of the heat. Whereas the modified process simulation is shown in Figure 4, while the PFD is shown inFigure 6. In the modified process, more equipment is used due to reusage of heat. The difference between those two is in the addition of a heat transfer fluid looping system for heating and cooling which can be seen in the modified process. For the basic process, there is no heat transfer fluid looping system so the energy that obtained and needed in heating and cooling process did not run efficiently. But for the modified process, there is an additional heat exchanger (HE-04) which is used to cool the heat exchanger fluid so it can be reused to cool the reactor output in the heat exchanger (HE-03), so the looping process of the heat transfer fluid system can be established and take place.

3.2. Mass Balance and Energy Balance Results

Based on the Table S1 (Supporting Information), heat and material balance for modified process is reported. In Figure 4, it is shown that the heat exchanger (HE-02) is used to cool the reactor output with a flow rate of

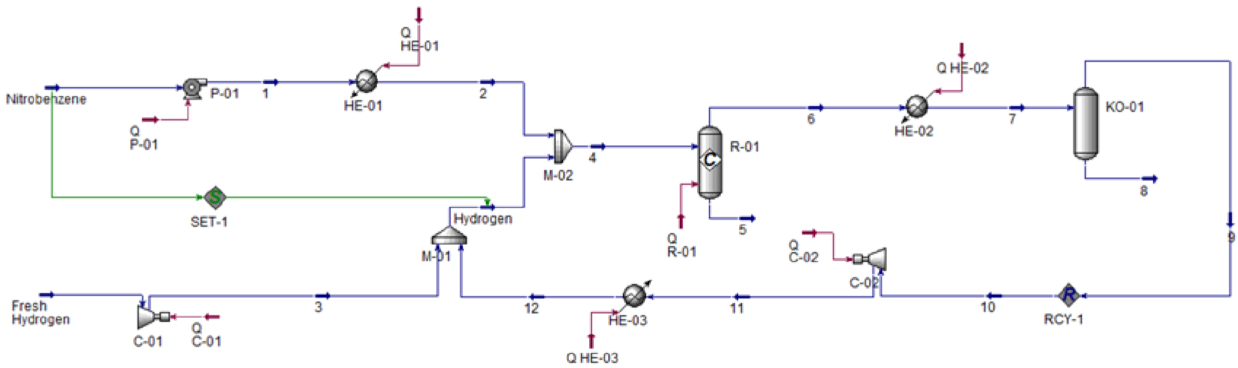

Figure 3. Simulation using Aspen HYSYS of basic (unmodified) process

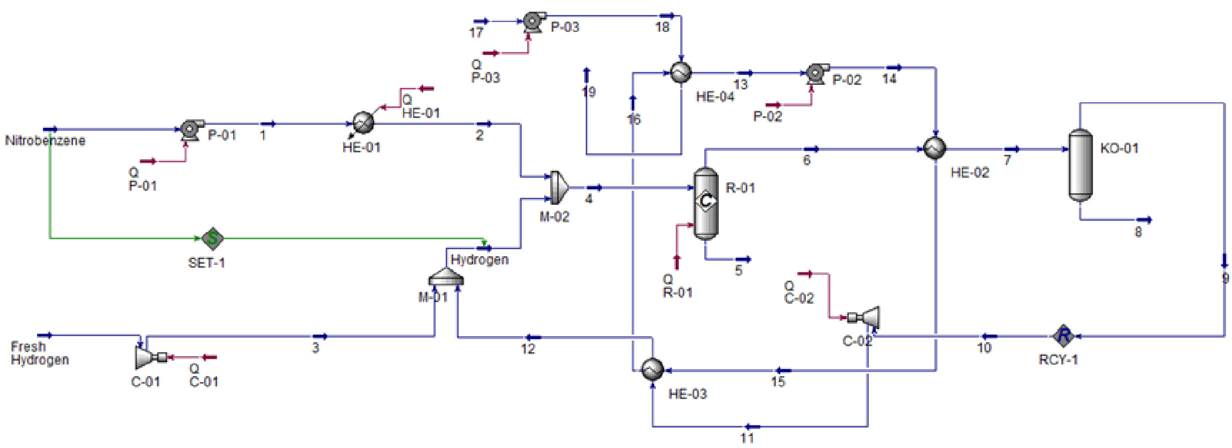

Figure 4. Simulation using Aspen HYSYS of modified process

41,271.456 kg/h from a temperature of 310 °C to 38.7 °C. The heat from reactor output will be received by the heat transfer fluid inside the heat exchanger (HE-02) with an input flow rate of 42,440.965 kg/h at 30 °C and a total duty of 96,714,359.832 kJ/h. The heating process in the recycle system occurs in the heat exchanger (HE-03) which is used to reheat the recycled hydrogen with a flow rate of 28,348.492 kg/h from a temperature of 73 °C to 223.8 °C with a required heat of 42,638,014.109 kJ/h. Meanwhile, the heat exchanger (HE-04) is used to recool the heat transfer fluid with a flow rate of 42,440.965 kg/h from a temperature of 122.1 °C to 30 °C using a water with a flow rate of 20,857.610 at 27 °C as a coolant and captures a heat of 54,079,258.071 kJ/h from the heat transfer fluid.

3.3. Heat Transfer Fluid Recycle Effect

Several heat transfer processes are required in the nitrobenzene hydrogenation process, both to heat and cool the compound. The nitrobenzene hydrogenation process usually manufactured in fixed bed reactor with the temperature between 300-475 °C, so the reaction generates a large amount of heat because it is classified as an exothermic reaction with the negative value of ΔH [33]. A heat transfer fluid is needed during the heat transfer process using the heat exchangers. To illustrate the process, the stream that contains hot fluid enters as a heating fluid to heat the the recycled hydrogen in the heat exchanger (HE-03) and the stream that containing cold fluid will enter as a cooling fluid to cool the product from reactor in the heat exchanger (HE-02) [34]. In the

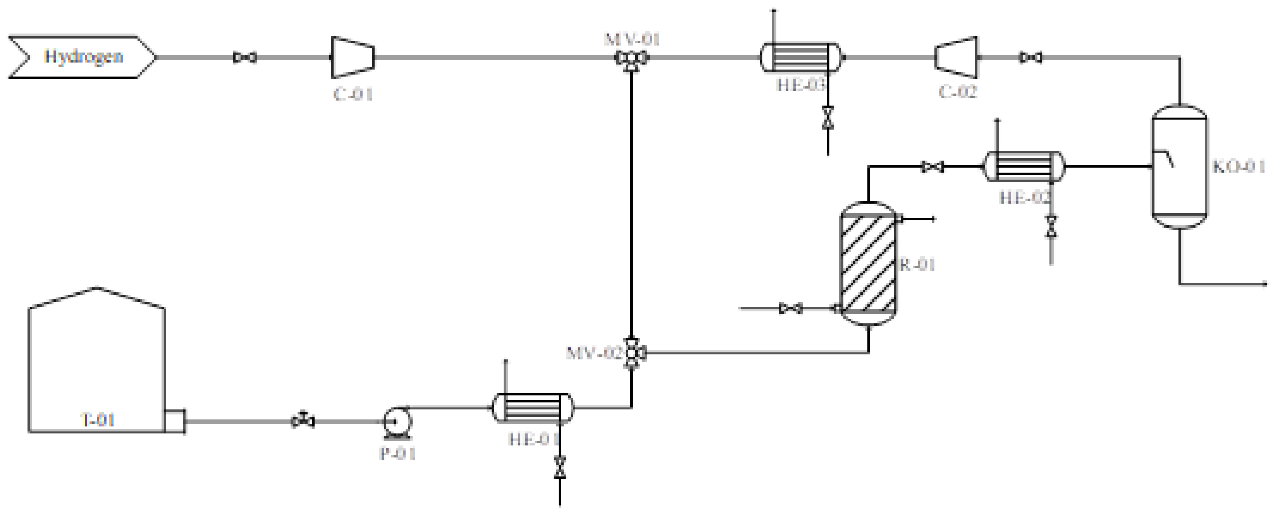

Figure 5. Process flow diagram of basic (unmodified) process

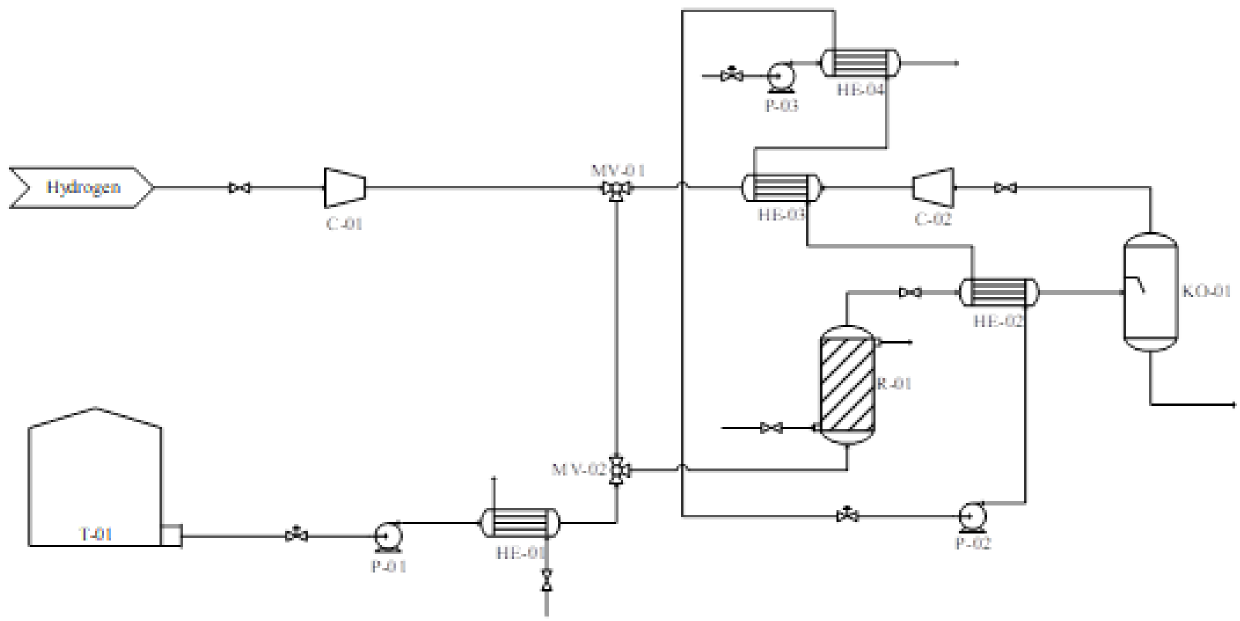

Figure 6. Process flow diagram of modified process

basic process, there is no such heat recycle process applied, so the heat obtained in the cooling process by the heat exchanger (HE-02) is wasted. The absence of heat recycle process also causes the reheating process of recycled hydrogen needs an additional heater and obviously consumes more energy. If the modified process is applied, the heat received by the heat transfer fluid in the heat exchanger (HE-02) can be used to reheat the recycled hydrogen and the residual heat can be used to generate steam for other utilities. The cooled heat transfer fluid then reused to cool the reactor output. This cycle runs continuously as a looping process, so that the energy from the process is not expected to be wasted.

3.4. Energy Source for Heat Exchanger (HE-03) Effect

The excess hydrogen as the upper product of the knock-out drum (KO-01) is reused to be fed into the reactor through the recycling process. The knock-out drum (KO-01) output temperature is 38.7 °C with a pressure of 1.398 atm. The excess hydrogen temperature needs to be increased using a heater or heat exchanger. The selection of this tool is based on the presence or absence of heat transfer fluid. If there is no heat transfer fluid, then the heater is used by using energy supply from outside. If there is a heat transfer fluid, an additional heat exchanger (HE-03) can be used to increase the excess hydrogen temperature. Heat received by the heat transfer fluid is from the cooling process of the reactor output by the heat exchanger (HE-02). The use of the heat exchanger (HE-03) compared to the heater is more efficient because the energy obtained in the cooling process is not wasted and does not require additional energy supply for the heater.

3.5. Steam Formation for Heat Recovery Process

Without utilizing the heat transfer fluid, the heat from cooling process of the reactor product is wasted. Whereas after the modification, this heat can be used for other purposes that require energy. Previously, it has been explained that some of the heat is used for the recycled hydrogen reheating process in the heat exchanger (HE-03), but there is still some heat that can be utilized further. This residual heat is used to generate steam for plant utility purposes. It is usual for heat recovery technologies to produce low-pressure steam, which is accomplished by vaporizing water at 120.2 °C and 2 bar [35]. As steam is heated over its boiling point for a given pressure, "dry steam" often referred as superheated steam is formed [36].

3.6. Net-Energy Efficiency Improvement Due to Process Modification

The net-energy for both basic and modified process is shown by Table 1. In the basic process, E_P value is 96,714,359.832 kJ/h which is obtained from reactor product cooling, but E_C value is equal to zero as there is no reusage of heat produced. On the other hand, in the modified process, the E_P value is 96,717,291.918 kJ/h which is obtained from pump and reactor product cooling. The E_C value is 96,717,282.049 kJ/h due to reusage of heat produced to reheat the recycled hydrogen and generate low-pressure steam with values of 42,638,014.109 kJ/h and 54,079,267.94 kJ/h, respectively. It can be concluded that the net-energy (NE) value in the basic process is 96,714,359.832 kJ/h and in the modified process is 9.869 kJ/h.

In the context of energy efficiency, zero net-energy is considered the ultimate goal. It refers to the balance between the amount of energy produced and the amount of energy consumed. A net-energy value that is closer to zero indicates that the energy efficiency is better [37]. Therefore, the modified process has better energy efficiency than the basic process.

4. Conclusions

The heat transfer fluid looping system for heating and cooling process improves the heat efficiency of the aniline production through nitrobenzene hydrogenation process. This is proven by the net-energy value in the modified process is 9.869 kJ/h. On the other hand, net-energy value in the basic process is 96,714,359.832 kJ/h. It can be seen that the net-energy value of modified process is much lower and closer to zero than the basic process. The looping process in the heat exchange fluid allows for better energy efficiency due to the process of releasing and receiving energy in the system which can be mutually utilized in each processes.

Table 1. Calculation result of net-energy of basic and modified processes

Process	E_P (kJ/h)	E_C (kJ/h)	NE (kJ/h)
Basic	96,714,359.832	0	96,714,359.832
Modified	96,717,291.918	96,717,282.049	9.869

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