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# Industrial Process Design for the Production of Aniline by Direct Amination

The objective is to design a plant from raw material to product for the production of aniline by direct amination of benzene. The process design is started on a conceptual level and ended on a basic engineering level as well as a techno-economical evaluation. The amination of benzene by hydroxylamine was used as basis. For the production of hydroxylamine four routes are proposed. The most promising route is the chemical reduction of nitric oxide with hydrogen. The process evaluation shows that 27 % of the atomic nitrogen is lost. The atomic carbon efficiency is close to unity. Furthermore, a significant amount of steam can be produced. From an economical perspective, there is still room for improvement because the return of investment is quite low and the payback period is quite high.

**Keywords:** Aniline, Conceptual process design, Direct amination, Hydroxylamine, Techno-economical evaluation

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## 1 Introduction

Aniline is a frequently used bulk chemical. At the moment, it is used to produce methylenedianiline (MDA), which is a precursor for generating methylene diphenyl di-isocyanate (MDI). MDI accounts for 75 % (volume-based) of the global aniline market. The demand for MDI in the coatings, adhesives, sealants, and elastomers (CASE) industry and the polyurethane industry boosted the aniline market. It is expected that this trend will continue until 2020. The use of aniline as a corrosion inhibitor in the microelectronics industry offers new opportunities to market growth [1].

In industry, there are two main routes to produce aniline from benzene. The first route is the catalytic hydrogenation of nitrobenzene and the second one is the amination of phenol with ammonia. Both routes employ a two-step process: firstly, either nitrobenzene or phenol has to be produced from benzene, and secondly, aniline has to be formed. The drawbacks of these routes are the low atomic efficiencies and the expensive reactants. The intention of this study is to design a one-step process, i.e., via direct amination of benzene to aniline, to reduce the number of reaction steps and to achieve a higher atomic efficiency. It is expected that this design will reduce the number of unit operations and with this the capital expenditure (CAPEX), lower the cost on reactants, and decrease the energy consumption. Therefore, it is supposed to be more economically profitable [2].

A process is designed and evaluated for an aniline plant of 250 kton per year aniline by direct amination. This plant will produce aniline as a feedstock to a MDI production plant, which uses aniline in its protonated form. Therefore, this study focuses on the production of aniline with a concentration of 75 wt % which is set by the MDI process that follows.

## 2 Design

### 2.1 Methodology

The methodology for this project has its base in Douglas' theory for chemical process design [3]. In this theory, the creativity of the designer plays an important role. Douglas states that the best way to come to a conceptual design is to open the mind and think creatively. This way of thinking can be structured with a hierarchy of design decisions. In this way, a large problem can be solved by creatively solving several smaller problems. In Douglas' theory, all design alternatives are kept until there are sufficient arguments to reject a particular design alternative. In this way, it is prevented that important design options are neglected in an early stage. By using rules of thumb, a lot of different alternatives can be evaluated in a limited amount of time.

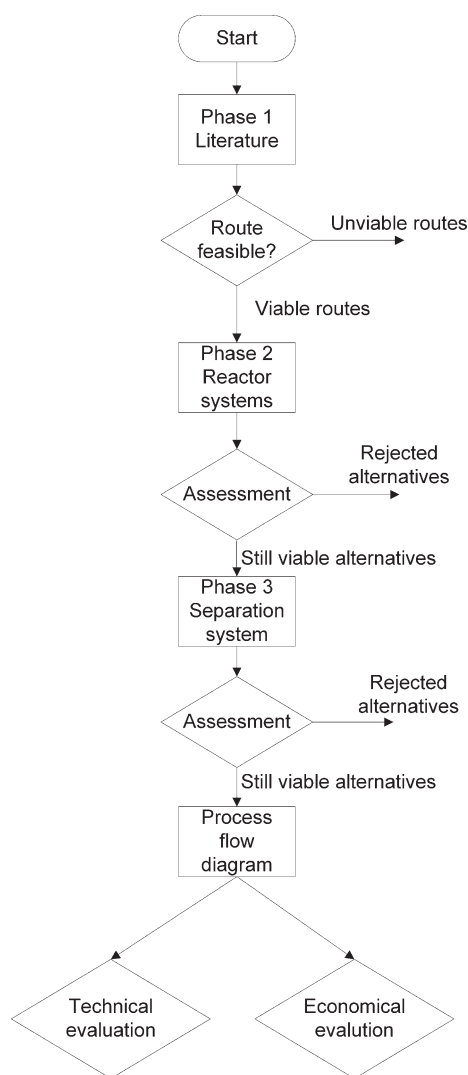
The conceptual design (Fig. 1) starts with the identification of possible raw materials and corresponding chemical routes. Based on simple black box mass balances for raw materials and (side) products, it was assessed if a particular route could be profitable. At the end of this phase the most promising chemical route was investigated. This route acts as starting point for the next level of the process design.

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**Figure 1.** Schematic of the applied methodology.

The second phase of the conceptual design is the identification of the required reactor systems and recycle flows which resulted in a number of conceptual design alternatives. The separation is considered to be one process function. The third phase of the process design is the design of the separation system, again resulting in a number of conceptual designs. Note that sequentially laws of conservation for mass, heat, and momentum are considered. Eventually, based on (a) the magnitude of the calculated recycle flows, (b) the number of unit operations, and (c) the early removal of inerts and corrosive components, the most promising conceptual design is selected and developed up to a basic engineering level. This includes a process flow diagram and corresponding mass and energy balances calculated in UniSim Design.

## 2.2 Chemical Routes

A number of routes for the direct amination of aniline have been investigated by a literature study. Tab. 1 shows the benzene conversion and selectivity to aniline as reported. From this table, it can be concluded that Route B and D have very low benzene conversions. Therefore, these routes are considered to be non-viable. When comparing Route C with Route A, it is obvious that both benzene conversion and selectivity are reasonable and both routes can still be competitive. Route C has already been investigated by means of a process design and evaluation in a previous study. So, the focus of this study is the amination of benzene by hydroxylamine [4].

**Table 1.** Routes for the direct amination of benzene.

Route	Process	Conversion [%]	Selectivity [%]	Ref.
A	Amination of benzene by hydroxylamine	42.5	> 99.99	[5]
B	Amination of benzene by ammonia with oxygen	1.5	87	[6]
C	Oxyamination of benzene with ammonia and hydrogen peroxide	18	96	[7]
D	Amination of benzene by ammonia with carbon monoxide	3	50	[6]

Now that the choice for hydroxylamine has been made, one has to think of ways to obtain hydroxylamine. Of course, one can buy it on the market, but hydroxylamine is not a bulk chemical and, thus, it is most likely cheaper to produce it autonomously. Therefore, the production of hydroxylamine will be in the scope of this process design. Hydroxylamine can be produced in six ways which are depicted in Tab. 2.

**Table 2.** Routes for the production of hydroxylamine.

Route	Process	Conversion [%]	Selectivity [%]	Yield [%]	Ref.
1	Raschig process	–	–	50–80	[8]
2	Direct conversion of ammonia with hydrogen and oxygen	–	–	11	[9]
3	Electrochemical reduction of nitric oxide	–	–	80–90	[10, 11]
4	Chemical reduction of nitric oxide	–	–	48–77	[12, 13]
5	Electrochemical reduction of nitric acid	–	–	$0.57 \times 10^{-3}$ – $1.9 \times 10^{-3}$	[14]
6	Chemical reduction of nitrates	60–70	80–85		[15]

Route 1 is rejected because it introduces the element sulfur, which is not consumed or produced in the reaction. Besides data on yields, conversions, and selectivity, the amount of information available for a proper process design is also an important factor in this stage of process design. Therefore, Route 2 is excluded because only one patent is available and there is no evidence that this process is applied in industry. The reduction of nitric acid in an electrochemical cell (Route 5) is also disregarded because it has a low yield, high residence time, and no evidence for industrial application exists. This means that only Route 3, 4, and 6 are left. From these routes it was concluded that Route 4, the chemical reduction of nitric oxide, was the most promising process because it is used in industry and has a high yield. Note that according to Douglas' theory Route 3 and 6 cannot be rejected because there are not enough arguments. Nevertheless, the most promising route was selected.

Nitric oxide will be used as feedstock for the production of hydroxylamine. Nitric oxide is, however, not commercially available on a bulk scale but it can be produced relatively easy by oxidation of ammonia. This is an established technology in the production of nitric acid, also known as the Ostwald process. A challenge in this technology is to control the explosion limits of ammonia in an oxygen environment.

## 2.3 Conceptual Design

To conclude, the proposed process for the production of aniline by direct amination consists of three challenges: the production of nitric oxide, the production of hydroxylamine, and the production of aniline. This will be the boundary of the process design.

### 2.3.1 Reactor Systems

At this level all possible configurations of reactor systems were listed. An important consideration was the combination of the production of hydroxylamine and the amination of benzene in one reactor. The rules of thumb of Douglas' theory that were used to reject a conceptual reactor system were: (a) separated streams should not be mixed again, (b) no circulation of plentiful by-products, and (c) limit the amount of purges, i.e., prevent material loss. Eventually, eleven conceptual reactor systems were listed and seven were rejected based on these rules of thumb.

### 2.3.2 Separation System

Of each conceptual reactor system that is left, the conceptual separation system was investigated according to Douglas' theory. At this level it is important to not think of separation technologies, like distillation, that could fulfill a particular separation function. Again all possible configurations of the separation system were listed. Rules of thumb for rejecting a particular separation system include, but are not limited to: (a) remove corrosive components as soon as possible, (b) remove reactive components as soon as possible, (c) most plentiful separations first, (d) lightest separations first, and (e) difficult separations last. Eventually, ten conceptual separation systems were listed and eight were rejected.

### 2.3.3 Preliminary Conclusion

At this stage, three conceptual designs are left. Of these three, one conceptual design stood out because it produced protonated aniline. The remaining two produced aniline in its neutral phase, which is undesirable for the consecutive MDI process. The most promising conceptual design is given in Fig. 2.

## 2.4 Choice of Unit Operations

The conceptual design is translated in a detailed process (Fig. 3). For each reaction or separation function a proper unit operation was chosen. For R1, a reactor type similar to the reactor type of the Ostwald process (production of nitric acid) was chosen: the gas phase is contacted to catalyst gauzes. The reactor type for R2 is discussed in more detail in Sect. 2.6.1. R3 is considered to be a black box since too little information, e.g., kinetics, is available to make a sound choice.

The separation functions S1 and S2 are conventional flash drums where the gas and liquid phase are separated. A pressure swing adsorption (PSA) was chosen for S3. Please note that PSA is not necessarily the most appropriate unit operation for this separation. Alternatives include membrane units for gas-phase separation and cryogenic distillation. A conventional three-phase separator was chosen for S4 to separate the gas, aqueous, and organic phase.

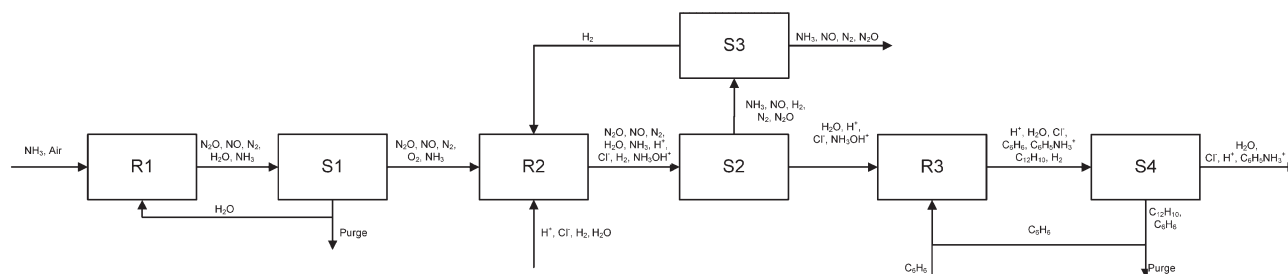
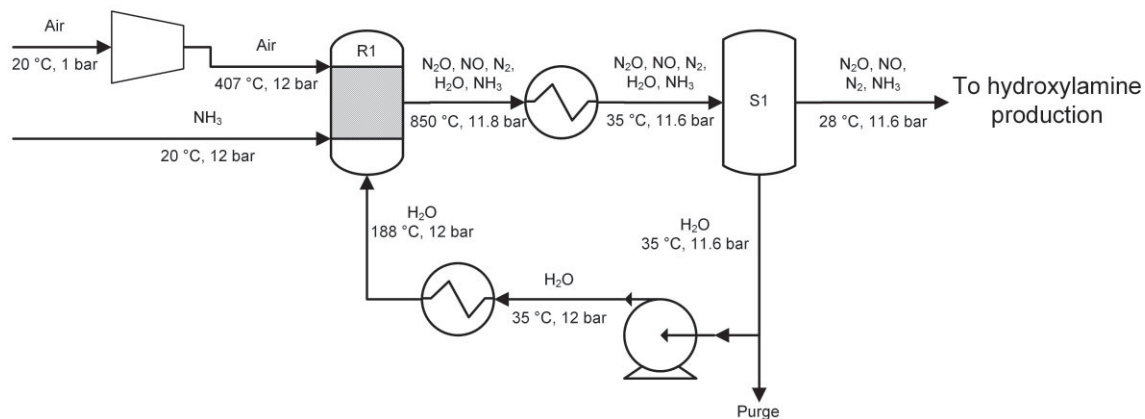
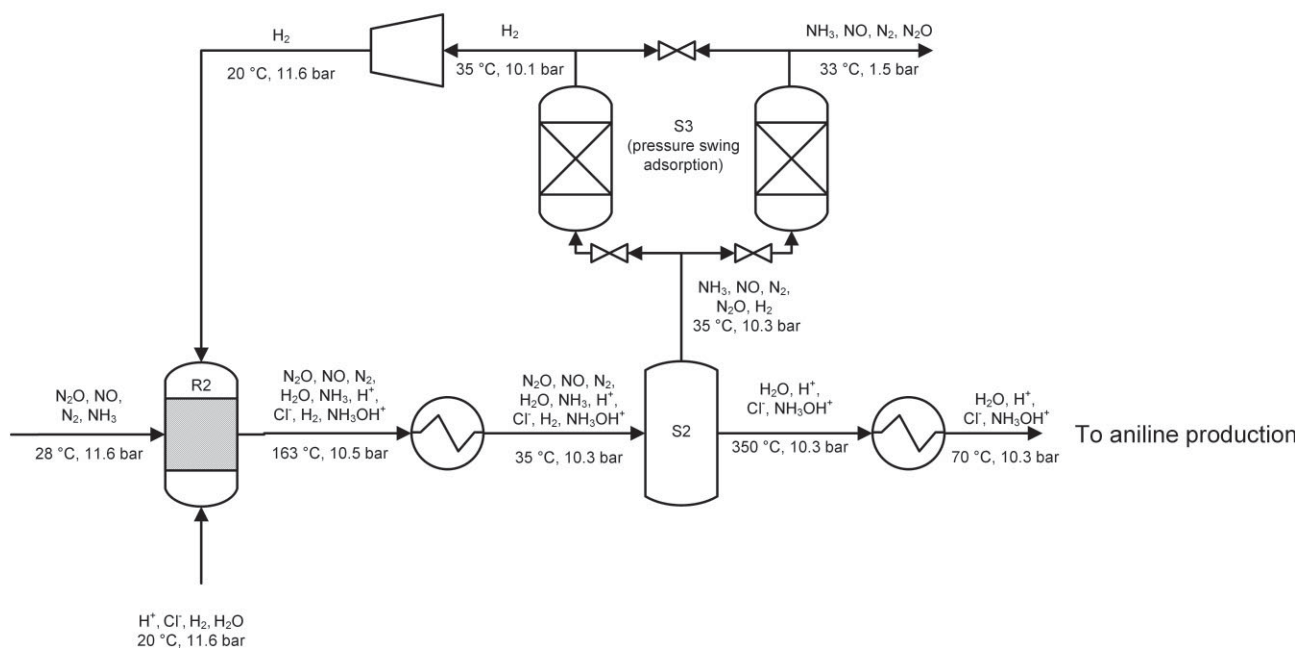


Figure 2. The most promising conceptual design.

a) Nitric oxide production



b) Hydroxylamine production



c) Aniline production

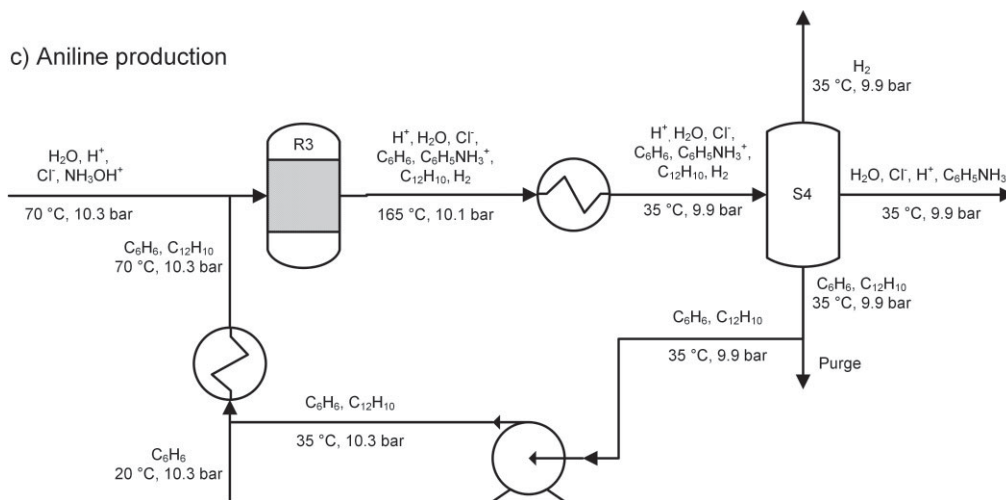


Figure 3. Process flow diagram (PFD) of the proposed process. The PFD is divided in three subsequent sections.

## 2.5 Detailed Process

In the first stage of design it was assumed that the process could operate under atmospheric pressure. It was readily observed that this leads to large volume flows of gas and this would require large equipment. Therefore, it was decided to increase the operating pressure to 10–12 bar as indicated in Fig. 3.

The first reactor (R1) is fed with air and ammonia in which ammonia is oxidized to produce nitric oxide. A platinum/rhodium catalyst applied as fine mesh gauzes are used for obtaining the highest yield of nitric oxide [16].



Via a flash drum (S1), water is removed from the gas phase and recycled to the first reactor for controlling the explosion limits of ammonia (15.5 mol %, lower explosion limit, [16]). The gas phase is fed to the second reactor in which hydroxylamine is produced. Fresh hydrogen gas and hydrochloric acid are added. The following main reaction takes place using a carbon-supported platinum catalyst [12]:



An excess of hydrogen is used (5 mol H<sub>2</sub>/mol NO) because this would favor the reaction kinetics [12]. Hydroxylamine is kept in its protonated form throughout the whole process to enhance the chemical stability of this compound [17]. The products are separated in a gas phase and a liquid phase. The gas phase is purified via PSA and hydrogen gas is recycled to the second reactor. The other gases are purged after the PSA. For the PSA, various sorbents including activated carbon, molecular sieve 13X, and zeolite 5A were considered and the PSA size for each sorbent was estimated [18–20]. Eventually, molecular sieve 13X showed the smallest PSA and hence, this sorbent was selected.

The liquid phase, containing protonated hydroxylamine, is fed to the third reactor in which benzene is converted into aniline using a Mn-MCM-41(20) catalyst [21]. Fresh benzene is added to the recycle stream containing benzene. The following main reaction takes place:



Via a three-phase separator, the gas phase, organic phase, and aqueous phase are separated. Aniline remains in the aqueous phase because it is protonated due to the acidic environment. The organic phase containing a significant amount of benzene is recycled to the third reactor.

This process has been modeled to obtain mass and energy balances with UniSim R410. The non-random two-liquid model (NRTL) is applied as thermodynamic model, together with a Peng-Robinson equation of state for the gas phase.

## 2.6 Equipment Design

To gain more information on the CAPEX, all equipment sizes were estimated.

### 2.6.1 Reactors

The ammonia oxidation reactor (R1) was sized based on the approach given by Temkin et al. [22]. The design of this reactor is less challenging than R2 and R3 because this reactor is commonly used in industry.

The hydroxylamine reactor (R2) was selected based on the approach published by Krishna and Sie [23]. The first level of selection is the catalyst design. Using the reaction kinetics of Polizzi et al., it was concluded that the reaction is very fast and, therefore, very small catalyst particles ( $d < 10 \mu\text{m}$ ) are used [12]. The second level of selection is the reactant and the energy injection strategies. The adiabatic temperature rise of this reaction is 140 °C because of the slightly exothermic nature of the reaction. Therefore, external cooling is required. The third level of selection is the hydrodynamic flow regimes and the reactor type selection.

For this gas-liquid-solid system, basically there are two options: a packed-bed reactor or a bubble slurry column. Because of the very small catalyst particles, the pressure drop in a packed-bed reactor would be too high, so a bubble slurry column is chosen as reactor type. This reactor has been modeled based on the model published by Van der Laan et al. [24].

The size of the amination reactor (R3) has been estimated by a simple approach because the reaction kinetics is unknown. Based on volume flow and required residence time, the reactor has been sized.

### 2.6.2 Flash Drums and Three Phase Separators

The flash drums and three phase separators have been sized based on the method published by Svrcek and Monnery [25, 26].

### 2.6.3 Pressure Swing Adsorber

The size of one adsorption column is estimated based on a typical minimum and maximum sorbent loading, sorbent bulk density, and a cycle time of 2.5 min. As in industry practice, it is assumed that four adsorption columns are needed for a sound operation [27].

### 2.6.4 Heat Exchangers

Fig. 3 does not show any heat exchange between process streams because heat integration was separately investigated by pinch analysis. All heat exchangers are assumed to be shell- and tube-type heat exchangers. The size of the heat exchanger was estimated using the approach of Sinnott and Towler [28].



### 2.6.5 Pumps and Compressors

The size and type of all pumps and compressors was based on the approach given by Towler and Sinnott [28].

## 3 Technical Evaluation

The process was evaluated based on a heat management study and atomic balances for nitrogen, carbon, and hydrogen, which are presented as Sankey diagrams.

### 3.1 Heat Management

Heat integration was separately investigated by pinch analysis. The total heating and cooling duty without heat integration is 13 and 102 MW, respectively. Assuming a temperature approach of 10 °C in the heat exchanger, it was estimated that 13 MW can be saved by applying heat integration.

Because of the exothermic nature of all reactions, there is still a huge amount (88 MW) of energy left. Especially R1 is very exothermic. A part of this excess energy (48 MW) can be used to produce high-quality steam that can be taken for other unit operations at the same chemical manufacturing complex. The total amount of steam that can be produced is calculated in Tab. 3. From the results it can be concluded that this process can deliver a significant amount of steam.

### 3.2 Nitrogen Balance

The Sankey diagram for the nitrogen atomic efficiency is given in Fig. 4. Inert molecular nitrogen (N<sub>2</sub>) is excluded from the efficiency calculations, because it is not involved in the reactions. The results indicate that 72 % of the nitrogenous components, which are present in the system, end up in the product stream. Furthermore, obviously 27 % of the nitrogenous com-

**Table 3.** Estimated steam production.

	$\Delta H_{\text{vap}}$ [MJ kg <sup>-1</sup> ]	Q [MW]	F [kg s <sup>-1</sup> ]
High pressure (40 bar, 250 °C)	1.72	40.4	25.5
Medium pressure (10 bar, 180 °C)	2.02	6.3	3.1
Low pressure (3.5 bar, 140 °C)	2.2	0.8	0.4
Total		47.5	29.0

ponents, mainly nitric oxide, is lost via the purge after the PSA section (S3).

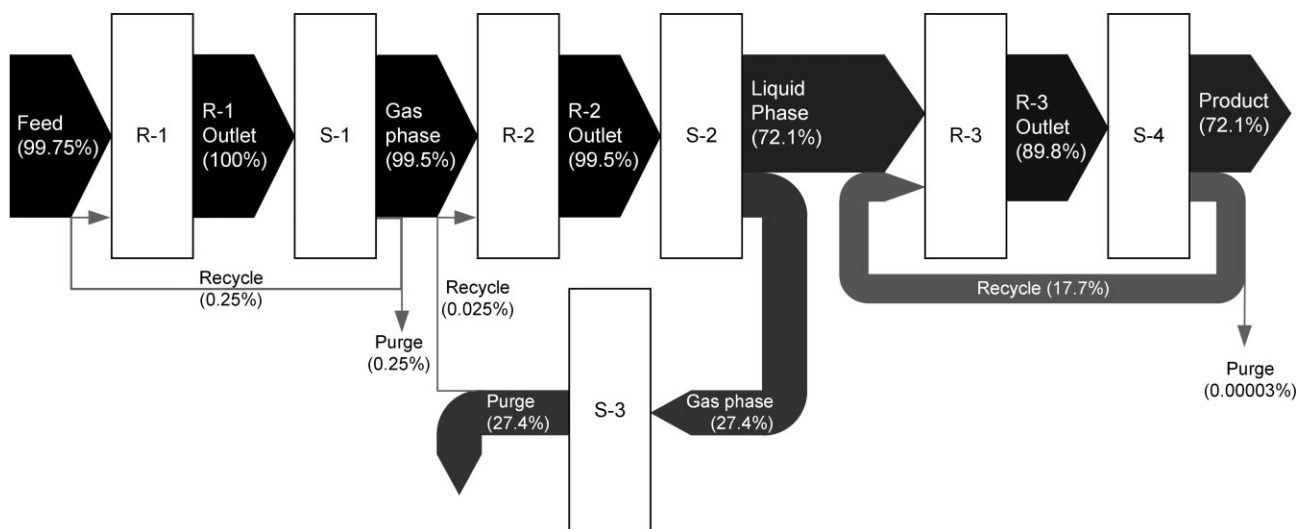
There are three options to prevent or lower the loss of nitric oxide. The first option is to selectively separate hydrogen and nitric oxide from the rest of the separator (S2) gas outlet. The second possibility is to separate nitric oxide from the purge. The last one is to improve the conversion in the hydroxylamine reactor (R2), e.g., by choosing a better catalyst. In this way, the process can be even improved further.

### 3.3 Carbon Balance

In Fig. 5, the Sankey diagram for the carbon atomic efficiency is presented. It shows that the carbon, which does not end up in the product stream, is negligible. Due to the large recycle of carbon, benzene can be almost completely converted in aniline. No improvements are required for the carbon system.

### 3.4 Hydrogen Balance

The hydrogen atomic efficiency is illustrated in Fig. 6. It shows that 56 % of the hydrogenous components end up in the product stream. In addition, 13 % of the hydrogenous components, i.e., water for controlling the ammonia explosion limit in R1,



**Figure 4.** Sankey diagram of atomic nitrogen.

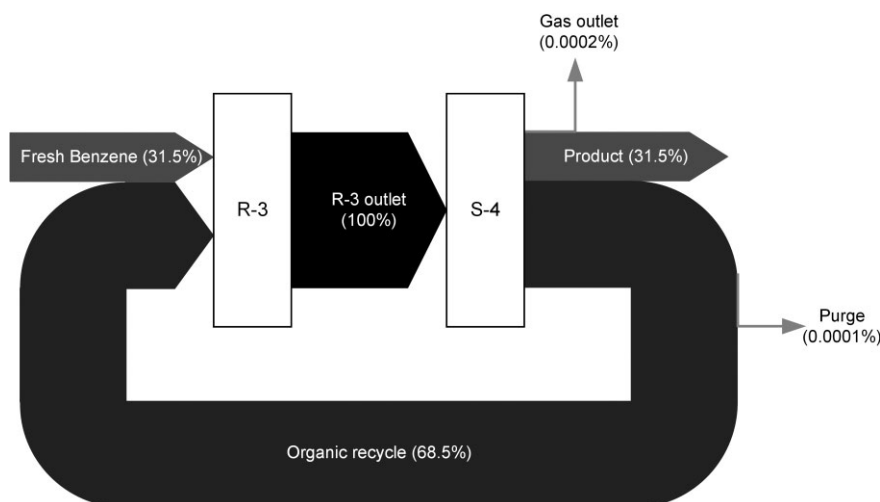


Figure 5. Sankey diagram of atomic carbon.

are lost after the first separation step. This stream consists mainly of water and a small amount of ammonia. Furthermore, a small amount of hydrogenous components (0.9%) is lost via the purge after the PSA section (S3). This purge stream also consists of water and ammonia. Considering the overall reaction of the process, i.e., by summing Eqs. (1), (2), and (3), one can calculate that the maximum theoretical hydrogen atomic efficiency is 62%. The obtained hydrogen atomic efficiency of 56% is very close to the maximum theoretical hydrogen atomic efficiency of 62%: the difference is only 10%. All in all, at this stage no enhancement is needed for the hydrogen system because other improvements such as recovering nitric oxide from the PSA purge would have stronger impact.

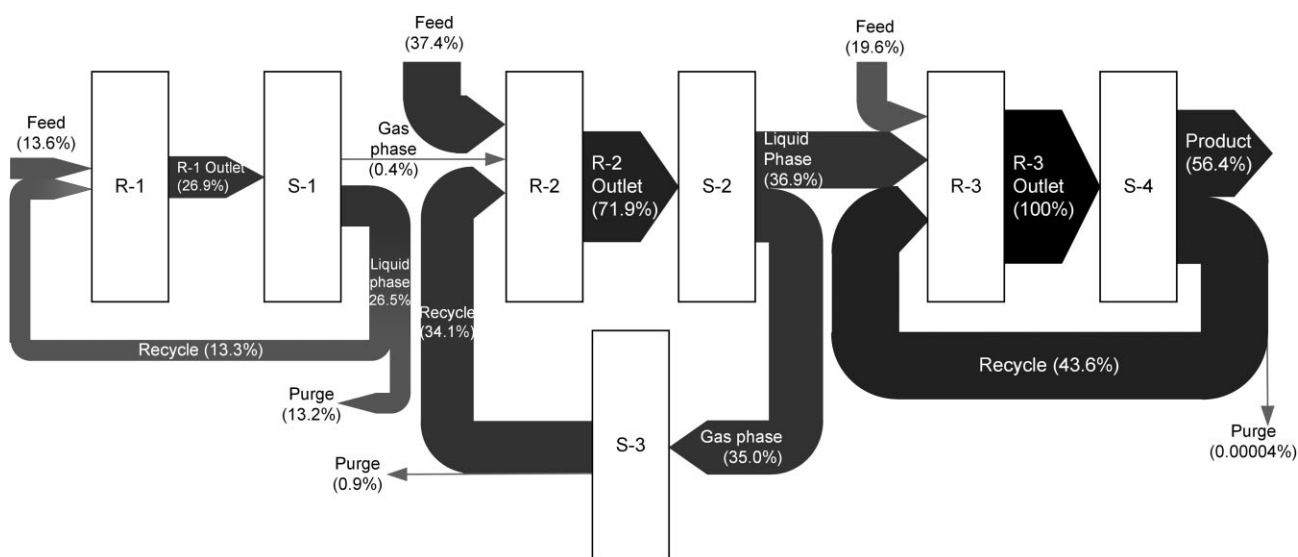


Figure 6. Sankey diagram of hydrogen.

## 4 Economical Evaluation

The process was evaluated on an economic basis by estimating the capital expenditure (CAPEX) and operational expenditure (OPEX).

### 4.1 Capital Expenditure (CAPEX)

The bare module cost of each unit is calculated from cost correlations and multiplied with material cost factors, pressure factors etc. Each equipment cost is multiplied with the Hand factor to account for the installation cost of equipment, inflation factor (Chemical Engineering Plant Cost index), and location cost factor (Rotterdam, The Netherlands). Furthermore, costs are added for other investment costs including but not limited to storage facilities, service facilities, allocated costs for utility plants (OSBL), initial catalyst charges, contingencies, and plant start-up [29]. The total capital expenditure is estimated to be 460 M\$. Note that the error interval of this estimate is  $\pm 30$ –50%.

### 4.2 Operational Expenditure (OPEX)

The OPEX consists of the variable production costs (raw material costs and utilities), fixed production costs (operating labor and maintenance), taxes, depreciation, and general expenses. The method of Seider et al. is applied to estimate all contributions to the operational expenditures [29]. These contributions are depicted in Fig. 7 and Tab. 4. The main driver for these costs is the benzene price; 77% of the variable production cost

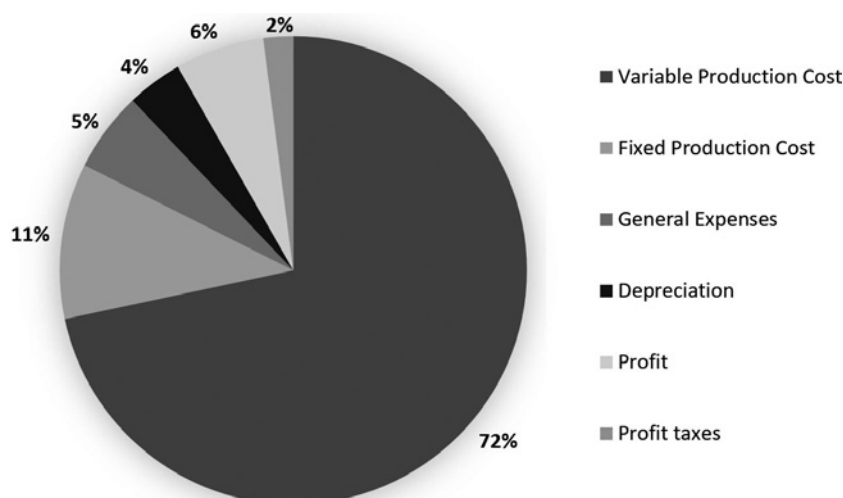


Figure 7. Allocation of the estimated operational expenditure.

Table 4. Cost allocation.

	Costs [M\$ a <sup>-1</sup> ]
Variable production costs	300
Fixed production costs	45
General expenses	23
Depreciation	16
Profit taxes	9
Sales	418
Profit	26

is due to benzene cost. The profit margin of this process is 8 % of the total sales, this includes both aniline and steam sales.

### 4.3 Cost Evaluation

From CAPEX and OPEX, the return on investment and the payback period can be calculated. The return on investment (ROI) is found to be 6 %, which is rather low compared to industry standards. The payback period (PBP) is eight years.

The process can be made more economically attractive via a few recommended options. The first recommendation is to investigate if enriched air can be used instead of air. This would reduce equipment sizes (i.e., CAPEX) because the transportation of inert nitrogen is avoided. The second recommendation is to recover aniline from the organic purge by extraction, adsorption, or membrane technology. And the third recommendation, as mentioned in Sect. 3.2, is to lower the purge of nitric oxide. The latter two would reduce the consumption of feedstock. Most of these recommendations can be handled by available technologies, so this would increase the potential of this process.

## 5 Conclusions

A systematic approach was used to develop a process to produce aniline from benzene by direct amination. In this process design, the route using hydroxylamine was investigated in more detail. The proposed process contains three major sections: nitric oxide production, hydroxylamine production, and benzene amination. The ROI of 6 % is still rather low, but the relatively short payback period is promising. Furthermore, it is shown that the loss of atomic carbon is nihil in contrast there is a loss of atomic nitrogen. However, there are opportunities to prevent this loss. The process can deliver a significant amount of high-quality steam. To conclude, the proposed process is economically and technically feasible and there are even options to improve this process.

The authors have declared no conflict of interest.

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