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Research Article

Enhancing Nitric Acid Production Efficiency Using Tail Gas Recycle

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Abstract

This study simulates and compares the conventional Ostwald process, and a modified full tail-gas recycle configuration to evaluate the enhancement in nitric acid production efficiency. Using simulation software with the Peng–Robinson model, the conventional Ostwald process and a modified recycle configuration were simulated and compared. In the standard process, unabsorbed NO_2 leaves with the tail gas, limiting nitric acid formation. Recycling this tail gas back to the absorber increases NOx contact time and promotes further conversion. Process efficiency, evaluated through production intensity (PI), improved from 0.4702 to 1.0320 kg HNO_3 per kg NH_3, a 119% increase. These results show that tail-gas recycling is an effective and straightforward method to boost nitric acid yield and reduce emissions without significant changes to the existing flowsheet.

Keywords: Nitric Acid; Ostwald Process; Tail-Gas Recycle; Aspen HYSYS; Process Intensification; NOx Recovery; Production Efficiency

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1. Introduction

Nitric acid is a strong acid that is naturally found only in the form of nitrate salts [1]. Nitric acid ranks second after sulfuric acid in terms of production volume [2]. It is widely used in various industrial and chemical processes. Its applications are not limited to fertilizer production but also extend to the food processing and dairy industries [3]. Modern methods of nitric acid production generally employ catalytic oxidation of ammonia. This process is highly exothermic and is carried out in high-pressure plants. The heat from exhaust gases is often recovered through turbines to support gas compression needs [4].

The production process of nitric acid consists of several stages. Ammonia is fed together with air into a reactor to undergo oxidation, producing nitric oxide. This compound is then further oxidized into nitrogen dioxide, which reacts with water to form nitric acid. In the final stage, the nitric acid solution passes through purification and separation units to achieve the required level of purity and concentration [5]. The reactions involved are as follows [1]:

$$4NH_3 + 5O_2 \rightarrow 4NO + 6H_2O \quad \Delta H^o_{298K} = -904 \text{ kJ/mol} \tag{1}$$

$$2NO + O_2 \rightarrow 2NO_2 \quad \Delta H^o_{298K} = -1127 \text{ kJ/mol} \tag{2}$$

$$3NO_2 + H_2O \rightarrow 2HNO_3 + NO \quad \Delta H^o_{298K} = -72.8 \text{ kJ/mol} \tag{3}$$

The occurrence of NO_2 slip in the absorber section of the Ostwald process has been identified as a critical operational challenge. The primary cause lies in the bottleneck during the oxidation stage, where the conversion of NO to NO_2 is

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inherently incomplete before the gas stream enters the absorber [6]. This limitation is attributed to the relatively short residence time and restricted oxygen availability in the process gas, resulting in a fraction of NO remaining in its less reactive form and therefore resisting absorption [7]. Operating conditions within the absorber further exacerbate this issue. Elevated temperatures reduce the tendency of NO_2 to dimerize into the more soluble N_2O_4, while low pressures diminish the driving force for absorption. Recent studies on modern nitric acid plants confirm that the combination of high temperature and low pressure directly increases the fraction of NO_2 escaping from the column [8].

In addition to these kinetic and thermodynamic constraints, hydrodynamic and mass transfer factors significantly contribute to NO_2 slip. An inadequate liquid-to-gas ratio, uneven liquid distribution, and limited gas–liquid contact area collectively reduce the absorption rate, preventing it from keeping pace with the supply of NO_2 [9]. Investigations into NOx capture have demonstrated that mass transfer limitations in conventional absorbers are a dominant factor leading to tail gas enriched with NO_2 [10]. Furthermore, even advanced adsorption technologies remain highly sensitive to fluctuations in liquid-to-gas ratio and residence time, underscoring the systemic nature of the problem. Taken together, these findings highlight that NO_2 slip results from a complex interplay of kinetic, thermodynamic, and hydrodynamic bottlenecks. Addressing this issue requires a holistic process intensification strategy that not only reduces emissions but also enables recovery of NO_2, thereby improving both environmental performance and economic efficiency of nitric acid production [11].

Despite significant progress in improving nitric acid plant performance, most recent process intensification studies have primarily focused on enhancing reaction kinetics through catalytic oxidation of nitric oxide rather than employing process-level recycle strategies to recover unconverted NOx. Catalytic oxidation has been shown to effectively alleviate kinetic limitations in the NO to NO_2 conversion step; however, such approaches generally require additional reactors and noble metal catalysts, increasing process complexity and operational cost [12]. In parallel, industrial-scale investigations have demonstrated that absorber operating conditions, particularly temperature and pressure, play a dominant role in determining NO_2 absorption efficiency, confirming that a substantial fraction of NOx can still escape with the tail gas under typical operating conditions [8]. These observations indicate that tail-gas NOx constitutes a recoverable loss and highlight the potential of process configuration modification, rather than fundamental reaction redesign, as a practical route for improving overall plant efficiency.

The objective of this study is to quantitatively evaluate the effectiveness of implementing a full tail-gas recycle strategy to enhance nitric acid production efficiency and NOx utilization in the Ostwald process. A steady-state process simulation is conducted to compare a conventional once-through configuration with a modified flowsheet incorporating tail-gas recycle using Aspen HYSYS. Process performance is evaluated using production intensity (PI), defined as the mass of nitric acid produced per unit mass of fresh ammonia feed, as this metric provides a more representative measure of overall process improvement achieved through recycle-based intensification than single-pass conversion alone [13].

2. Methods

This study applies a steady-state process simulation approach to evaluate nitric acid yield enhancement through a full tail-gas recycle. All simulations were developed and executed in Aspen HYSYS V15 using the Peng–Robinson (PR) property package. PR was selected because it is a robust equation-of-state model for gas-dominated, industrial-pressure systems, providing stable prediction of non-ideal multicomponent gas behaviour (N_2, O_2, NO, NO_2/N_2O_4, and H_2O vapor). In addition, PR offers consistent estimation of key thermophysical properties for vapor–liquid equilibrium, density, and enthalpy under medium-to-high pressure conditions typical of nitric acid plants. The modelled process consists of an ammonia oxidation reactor, where NH_3 reacts with excess air to form NO and H_2O in a highly exothermic reaction; reaction-gas cooling/condensation to reduce the temperature of the reactor effluent to levels suitable for downstream absorption; a nitric acid absorber column, where NOx is absorbed and reacts with water to form liquid HNO_3 and an absorber overhead tail-gas stream containing N_2, O_2, unabsorbed NOx, and traces of H_2O/HNO_3 vapor. The chemical components included in the model were NH_3, O_2, N_2, NO, NO_2/N_2O_4 (as NOx species), H_2O, and HNO_3.

To enhance nitric acid yield, the absorber overhead tail gas is fully recycled back to the absorption section rather than being discharged. This recycle is proposed because, in the basic process, a portion of NOx leaves the absorber unreacted, representing a direct loss of potential HNO_3 formation and a contributor to NOx emissions. By returning the entire tail-gas stream to the absorber inlet, the unabsorbed NOx is given additional contact opportunities with the liquid phase, thereby increasing its absorption and subsequent conversion to nitric acid. The performance of the full-recycle configuration is then compared with the basic non-recycle process to evaluate its effect on nitric acid yield and NOx recovery.

The parameter used to evaluate the efficiency improvement provided by tail-gas recycle will be the production intensity (PI). PI is defined as the amount of nitric acid produced per kilogram of fresh ammonia feed.

$$PI = \frac{HNO_3 \text{ produced} \left(\frac{kg}{h}\right)}{\text{Ammonia fed} \left(\frac{kg}{h}\right)} \quad (4)$$

The reason for this parameter being introduced in this study is because it directly measures how effectively the plant converts the primary reactant into product. Conventional parameters such as single-pass conversion or stochiometric yield do not fully capture the improvement introduced by tail-gas recycle because they describe only the performance of individual reaction steps rather than the overall plant output [13].

3. Results and Discussion

3.1. Basic Process Flowsheet and Simulation

The basic method for nitric acid production is carried out through the Ostwald process, which consists of three main stages, such as, ammonia oxidation, cooling of the reaction gases, and absorption of nitrogen dioxide in water as depicted in Figure 1. In this process design, liquid ammonia is first pumped and vaporized using a dedicated vaporizer unit, producing ammonia in the gaseous phase. The gaseous ammonia is then mixed with compressed air in a mixer unit, forming a mixture with the appropriate stoichiometric ratio required for efficient oxidation. This mixture is subsequently directed into a catalytic oxidation reactor operating at a pressure of approximately 8 bar and a temperature range of 850 – 950 °C. The reactor employs a platinum (Pt) – rhodium (Rh) gauze catalyst to facilitate the reaction. The principal reaction occurring in this stage is the catalytic oxidation of ammonia to nitric oxide, which serves as the foundation for subsequent process steps in nitric acid production.

$$4NH_3 + 2O_2 \rightarrow 4NO + 6H_2O \quad (\Delta H < 0) \quad (5)$$

The reaction gas stream, which is rich in nitric oxide (NO), is directed to the waste heat boiler (WHB) where its temperature is reduced while simultaneously generating high-pressure steam as a valuable energy by-product. Following this stage, the cooled gas exits the WHB and passes through a condenser unit designed to remove a portion of the water vapor present in the stream. At this point in the process, secondary air is introduced to enhance the conversion of NO into nitrogen dioxide (NO_2), in accordance with the established oxidation reaction. This step is critical for maximizing NO_2 yield prior to the absorption stage, thereby improving overall process efficiency and ensuring optimal conditions for subsequent nitric acid formation.

$$2NO + O_2 \rightarrow 2NO_2 \quad (6)$$

The secondary oxidation stage is essential to ensure sufficient availability of nitrogen dioxide (NO_2) prior to the absorption process. In this step, the gas stream enriched with NO_2 is directed into the absorber column, where intimate contact with water takes place. The absorption mechanism facilitates the conversion of NO_2 into nitric acid (HNO_3) according to the established reaction pathway. This stage represents the final and most critical step in the Ostwald process, as it directly determines the yield and quality of nitric acid produced. Proper control of operating conditions within the absorber, such as liquid distribution, temperature, and pressure is therefore vital to maximize absorption efficiency and minimize NO_2 slip to the tail gas.

$$3NO_2 + H_2O \rightarrow 2HNO_3 + NO \quad (7)$$

The liquid product obtained from the absorber consists of an aqueous nitric acid solution with a concentration range of 55 – 68% by weight. This product is collected at the bottom section of the absorber column and represents the primary output of the Ostwald process. Meanwhile, the residual gas stream (tail gas), which still contains trace amounts of NOx, is directed to the tail-gas treatment unit for further purification to meet environmental standards. With this basic configuration, the process can produce nitric acid of industrial - grade purity while simultaneously utilizing the thermal energy released from the exothermic reactions for the generation of high-pressure process steam. This integration of product recovery and energy utilization ensures both economic efficiency and compliance with industrial specifications.

3.2. Modified Process Flowsheet and Simulation

The Ostwald process has a clear inefficiency issue since the tail gas out of the absorber still contains a fair amount of NO_2. To address this inefficiency, the process in this study was modified by incorporating a tail gas recycle loop, a strategy previously that was used to increase the yield of hydrogen production up to 8.7% [14]. In this modification, the entirety of the tail gas coming out of the absorber is recycled back into the inlet. This allows an increase in the overall NO_2 utilization efficiency and nitric acid yield. The key operating parameters of this modification is the recycle ratio (R) defined as the fraction of the absorber tail gas that is directed toward the recycle loop [15]. Typical industrial processes use 0.9 or higher values of recycle ratios with 5 % of the total recycle stream used as the purge fraction [16]. In this simulation, this standard is utilized.

Overall, the addition of the recycle and purge stream modifies the flowsheet only minimally. The entire addition only includes two additional splitters (tee) and an expander to adjust the pressure of the inlet. Even so. It all yields a substantial improvement of NO_2 utilization. By increasing the effective number of absorption cycles experienced by the NO_2, the modification enhances nitric acid production efficiency while simultaneously reducing emissions.

3.3. Process Simulation using Aspen HYSYS

The simulation was conducted using Aspen HSYS V15 using the Peng-Robinson fluid package. To evaluate the effect of tail-gas recycling on the overall plant output, two cases, one incorporating a recycle stream and one operating without it. The basic unmodified case (without recycle) was simulated first, as shown in Figure 2. The simulation shows that the end stream (stream 13) contains 343.7256 kg/h of HNO_3. Given that the fresh feed (stream 1) contains 731 kg/h of ammonia. Thus, the base-case simulation yields a production intensity of 0.4702 kg of HNO_3 per kg of NH_3 fed. Next, we simulate the recycle configuration to evaluate the impact of tail-gas return on overall plant performance. This flowsheet is shown in Figure 3.

In this configuration, the tail gas stream (stream 10) is split into two fractions: a 10% bleed stream (stream 23) and an intermediate stream (stream 24). The intermediate stream is then divided further into a purge stream (stream 25) and a recycle stream (stream 26). Stream 26 has

Table 1. Process stream data summary.

Line no. Stream Component	1 Ammonia Feed	1A Ammonia vapour	2 Filtered air	2A Oxidiser air	3 Oxidiser feed
NH_3 (kg/h)	731.0	731.0	-	-	731.0
O_2 (kg/h)	-	-	3036.9	2628.2	2628.2
N_2 (kg/h)	-	-	9990.8	8644.7	8644.7
NO (kg/h)	-	-	-	-	-
NO_2 (kg/h)		-	-	-	-
HNO_3 (kg/h)		-	-	-	-
H_2O (kg/h)		-	-	-	-
Total		-	-	Trace	-
Press (bar)	8	8	1	8	8
Temp. (°C)	15	20	15	230	204

Line no. Stream Component	4 Oxidiser outlet	5 W.H.B. outlet	6 Condenser gas	7 Condenser acid	8 Secondary air
NH_3 (kg/h)	Nil	-	-	-	-
O_2 (kg/h)	935.7	(935.7)$^{(1)}$	275.2	Trace	408.7
N_2 (kg/h)	8668.8	8668.8	8668.8	Trace	1346.1
NO (kg/h)	1238.4	(1238.4)$^{(1)}$	202.5	-	-
NO_2 (kg/h)	-	Trace	(?)$^{(1)}$	967.2	-
HNO_3 (kg/h)	-	Nil	Nil	-	850.6
H_2O (kg/h)	-	1161.0	1161.0	29.4	1010.1
Total	12,003.9	12,003.9	10,143.1	1860.7	1754.8
Press bar	8	8	8	1	8
Temp. °C	907	234	40	40	40

Line no. Stream Component	9 Absorber feed	10 Tail (2) gas	11 Water feed	12 Absorber acid	13 Product acid
NH_3 (kg/h)	-	-	-	-	-
O_2 (kg/h)	683.9	371.5	-	Trace	Trace
N_2 (kg/h)	10,014.7	10,014.7	-	Trace	Trace
NO (kg/h)	202.5	21.9	-	Trace	Trace
NO_2 (kg/h)	-	967.2	(Trace)$^{(1)}$	-	Trace
HNO_3 (kg/h)	-	-	-	-	1704.0
H_2O (kg/h)	-	29.4	26.3	1376.9	1136.0
Total	11,897.7	10,434.4	1376.9	2840.0	4700.6
Press bar	8	1	8	1	1
Temp. °C	40	25	25	40	43

a higher pressure than the working pressure of the absorber hence it needs to be put through an expander first. Looking at the compositions of stream 13, we find that the amount of nitric acid produced has increased to 754.3905 kg/h. Again, given that the fresh feed (stream 1) contains 731 kg/h of ammonia. Thus, the recycle-case simulation yields a production intensity of 1.0320 kg HNO_3 per kg of NH_3 fed.

The introduction of tail-gas recycling resulted in a production intensity rise from 0.4702 to 1.0320 kg HNO_3 per kg of NH_3. This corresponds to an improvement of 119%. This increase occurs because the recycle stream returns unreacted NO_2 back to the absorber, allowing them to undergo further conversion to nitric acid rather than being discharged. As a result, a greater fraction of the fresh ammonia feed can contribute to product formation, leading to a much higher overall plant output.

Table 1. Comparison of Production Intensity (PI).

Parameter	Value
Production intensity before modification	0.4702 kg HNO_3/kg NH_3
Production intensity after recycling	1.0320 kg HNO_3/kg NH_3
Increase in production	+119% NO_2 Recovery

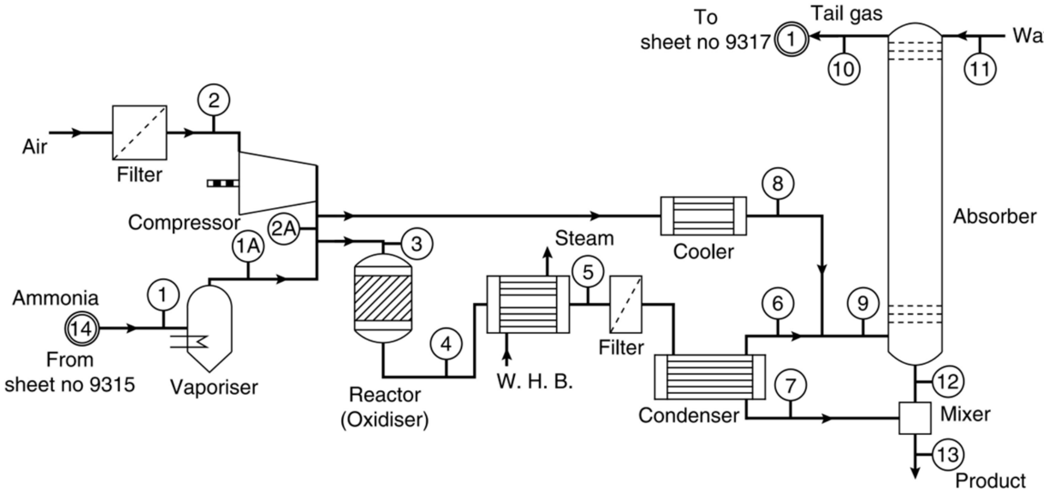

Figure 1. Basic process flow diagram (PFD) of nitric acid production.

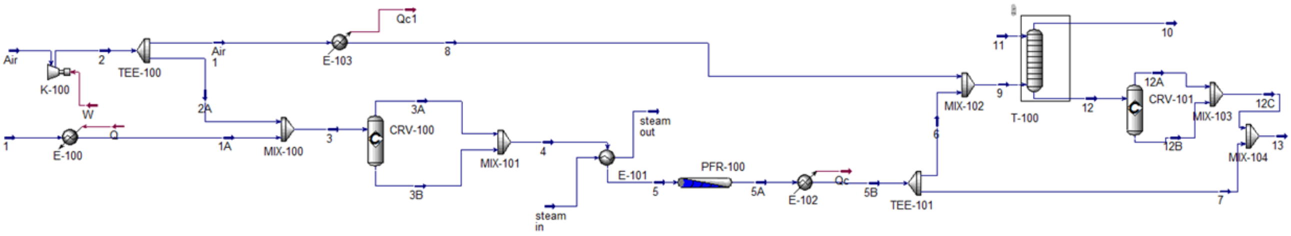

Figure 2. Process simulation of the conventional nitric acid production process.

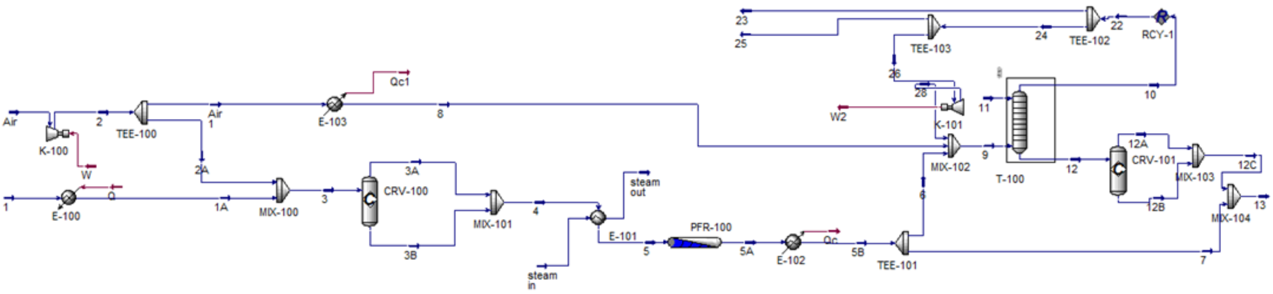

Figure 3. The improved nitric acid production process using a tail gas recycling system.

4. Conclusions

The enhancement of tail gas recycling on nitric acid production was analyzed by simulating the conventional nitric acid production process with (the conventional way) and without a recycle stream, focusing on the overall production efficiency using the production intensity (PI) as the key metric. In the base case without tail-gas recycle, unreacted NO_2 in the tail stream are discharged, limiting the overall conversion of ammonia and resulting in a lower PI value. On the other hand, the recycle configuration shows a significant increase in PI. This happens because unreacted NO_2 is allowed to return to the absorber, increasing their opportunity for further reaction and allowing a larger fraction of the fresh ammonia feed to be converted into nitric acid. This modification substantially enhances process performance, increasing the PI of 0.4702 to 1.0320 kg HNO_3 per kg of NH_3. This corresponds to an improvement of 119%. Based on these results, the incorporation of a tail-gas recycle system is an effective strategy for boosting nitric acid output without major process redesigns.

CRedit Author Statement

Author Contributions: S.O.H Simangunsong: Conceptualization, Methodology, Resources, Data Curation, Writing, Software, Formal analysis; D.R Ruslan: Conceptualization, Methodology, Formal Analysis, Data Curation, Writing, Draft Preparation, Software; M. Silviana: Validation, Writing, Review and Editing, Conceptualization, Visualization, Data Curation, Project Administration; K. Theni: Investigation, Resources, Writing, Review and Editing, Visualization, Validation, Conceptualization; H.N. Lerrick: Investigation, Resources, Writing, Review and Editing, Validation, Resources, Conceptualization. All authors have read and agreed to the published version of the manuscript.

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