REDUCING THE N_2O FOOTPRINT OF YOUR NITRIC ACID PLANT

STAMICARBON

NEXTCHEM
MAIRE Sustainable Technology Solutions

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SUMMARY

ABSTRACT

This paper evaluates various technical options proposed in the market for emission reduction in nitric acid plants, which are necessary for lowering their overall environmental footprint. It shows why the tertiary abatement system is the most effective and technically feasible solution for reducing NO_x and N_2O emissions. The paper also presents the development of the tertiary abatement system by Stamicarbon, the nitrogen technology licensor of NEXTCHEM (MAIRE Group), which can incorporate both extruded catalyst and monolith catalyst beds. These solutions are tailored to different situations, with examples illustrating primary challenges posed by various scenarios.

1 INTRODUCTION

Nowadays, most countries strictly regulate emission levels with an Emissions Trading System (ETS). The ETS is a market-based approach used to control pollution by providing economic incentives for achieving reduction in the emissions of pollutants. The most well-known ETS is the European Union Emissions Trading System (EU ETS), which was launched in 2005. It works by setting a cap on the total amount of certain greenhouse gases that can be emitted by installations covered by the system. Typically, companies receive or buy emission allowances, which they can trade with one another as needed. The cap is reduced over time, ensuring that total emissions fall.

For emerging countries where the ETS is not yet in place, there are possibilities to receive financing to implement an abatement system. The most well-known is probably the one sponsored by the Nitric Acid Climate Action Group (NACAG).

NACAG is an initiative launched by the German government during the 21st Conference of the Parties in Paris (COP 21) in 2015. Its goal is to drive a global shift toward climate-friendly nitric acid production by promoting the installation of effective nitrous oxide (N_2O) abatement technology in nitric acid and caprolactam plants worldwide. NACAG works by providing guidance, information, and financial support for the procurement and installation of N_2O abatement technologies. Partner countries commit to permanently reducing emissions, ensuring the sustainability of the initiative.

In addition to these initiatives, there is an emerging trend to promote cleaner production beyond European countries through mechanisms like the Carbon Border Adjustment Mechanism. Currently, regulations are being developed to impose a carbon levy on imported goods to the EU, such as fertilizers and hydrogen.

These systems are crucial for driving global efforts to reduce greenhouse gas emissions and combat climate change.

2 N_2O ABATEMENT SYSTEMS

In the nitric acid plant, there is a single emission point: the stack where tail gas is released. This tail gas primarily consists of nitrogen (around 98% vol), along with some oxygen and water, and contains pollutants such as NO_x and N_2O. While most existing plants have had NO_x abatement systems since the 1990s, many operate without N_2O abatement or with outdated systems.

N_2O, formed as a byproduct in the ammonia burner during nitric acid production and consequently emitted during this process, is a greenhouse gas with a global warming potential 300 times greater than carbon dioxide (CO_2). Due to its stability in the atmosphere, eliminating N_2O has become a significant concern.

There are several locations in the plant where N_2O can be removed, known as primary, secondary or tertiary abatement. Primary abatement targets the reduction of N_2O formation directly within the platinum gauze pack, where ammonia oxidation occurs. However, in practice, gauze selection is typically driven by plant-specific operational goals - most notably, optimizing selectivity toward nitric oxide (NO) rather than minimizing N_2O formation. As a result, the effectiveness of primary abatement is inherently limited, since N_2O is an unavoidable byproduct of ammonia combustion, regardless of the gauze configuration.

The second alternative is installing a secondary abatement system downstream the primary gauzes. In this case, the catalyst - typically composed of metal oxides - is placed in a basket and may be shaped either as a honeycomb or as extrudates, depending on the supplier. While this approach is sometimes perceived as a straightforward solution for N_2O abatement, particularly in plants equipped with a $deNO_x$ system, it often involves more complexity than initially assumed. In existing facilities, inspections often reveal significant mechanical challenges associated with modifying older equipment, including issues with increased weight compromising mechanical stability of the ammonia burner/reactor. In the case of installing secondary abatement, the catalyst supplier needs to provide guarantees on N_2O conversion. Additionally, the ammonia burner supplier must ensure the mechanical stability of the basket and burner. Furthermore, it is important to consider the additional load due to the new catalyst and the resulting additional pressure drops, along with the fact that the burner may already be operating beyond its original guaranteed lifetime.

The tertiary abatement system is a solution for removing N_2O upstream of the tail gas expander, enabling in most cases simultaneous elimination of both NO_x and N_2O in a single vessel. This represents the standard solution applied in Stamicarbon grassroot plants, as the process conditions of the tail gas are ideal for its implementation. Additionally, there is no need to add natural gas as a reducing agent for N_2O.

Techno-economic comparisons between secondary and tertiary abatement systems have revealed major benefits of the tertiary system. Integrating secondary catalysts into existing equipment is usually not feasible due to space constraints that consequently cause lower achievable conversion rates compared to international standards. Therefore, when planning to install secondary catalysts in long-standing existing plants, a new burner is frequently needed.

When implementing a secondary abatement system in conjunction with a new burner, it is advisable to assess the potential for simultaneously increasing plant capacity. By integrating these upgrades, the return on investment associated with new equipment can be optimized, offsetting the higher capital costs. However, to proceed in that direction, it is essential to consider additional risks first. Integrating a secondary abatement installation with the plant capacity increase necessitates careful planning and coordination.

A comprehensive inspection of the entire facility is essential, ensuring that all systems and components are operational and ready to support increased production levels. Specialized equipment, particularly rotating machinery, will require input and possibly intervention from external suppliers. Coordination of supplier lead times with the schedule for the installation of new burners is crucial. Furthermore, detailed evaluations may identify additional constraints or bottlenecks within the system that were not previously apparent. These expanded requirements often lead to increased financial investment, a broader scope of work, and potentially a reassessment of project goals. Increased complexity often leads to higher financial expenditure, an escalation in the scope of required activities, and a re-evaluation of the project's objectives. As such, meticulous project management and strategic oversight are essential to mitigate potential risks and ensure successful completion.

Even if guarantees are managed, the tertiary abatement system offers superior benefits, given that the burner remains unchanged, and it can be added in a dedicated structure without affecting the core of the plant.

Furthermore, when considering environmental (and consequently economical) advantages, it is necessary to mention that with tertiary abatement, it is possible to reduce the N_2O emissions up to 99%, while for secondary abatement, in most cases, it is not common to go above 90% N_2O reduction. Considering the ETS, this difference has an impact on the yearly savings due to CO_2 credits.

A ton of N_2O is equivalent to 273 tons of CO_2[1] and, considering that the average price of emitting a ton of CO_2 is EUR 70, this means that sending one ton of N_2O to the atmosphere costs EUR 19110. In the case of an 800 MTPD nitric acid plant, implementing secondary abatement in a plant without an existing N_2O reduction system leads to EUR 26M savings per year. However, if a tertiary abatement system is implemented, then this amount can increase up to EUR 29M savings per year.

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[1] *IPCC sixth assessment report, working group 1, chapter 7, section 7.6.1.1.*

3 INTEGRATION OF TERTIARY ABATEMENT IN STAMICARBON GRASSROOT PLANTS

Stamicarbon offers mono- and dual-pressure nitric acid production processes for a wide range of plant capacities up to 600 MTPD (mono-pressure) or up to 2000 MTPD and above (dual-pressure).

The technology uses the Ostwald process, where ammonia is converted to nitric acid in two main steps.

1. **Oxidation of ammonia**:
    - In this step, ammonia (NH_3) is oxidized to form nitric oxide (NO). This reaction takes place in the presence of a platinum-rhodium catalyst at high temperatures. The chemical equation for this reaction is:

      $4NH_3+5O_2 \rightarrow 4NO+6H_2O$

      The nitric oxide produced is then further oxidized to nitrogen dioxide (NO_2) in the presence of oxygen. The chemical equation for this reaction is:

      $2NO+O_2 \rightarrow 2NO_2$
2. **Absorption of nitrogen dioxide**:
    - In this step, nitrogen dioxide is absorbed in water to form nitric acid (HNO_3). This reaction occurs in an absorption tower where nitrogen dioxide is dissolved in water, producing nitric acid. The chemical equation for this reaction is:

      $3NO_2+H_2O \rightarrow 2HNO_3+NO$

      The nitric oxide produced in this reaction is re-oxidized to nitrogen dioxide, ensuring efficient use of the reactants.

The main characteristic of Stamicarbon’s nitric acid technology is an effective heat exchanger network downstream of ammonia oxidation. The heat exchanging network has specific process conditions selected to prevent corrosion and ensure no proprietary materials are required for equipment manufacturing. The placement of the third tail gas heater in the ammonia burner vessel, very close to the gauzes, allows for achieving a tail gas temperature of 480°C. This high temperature of tail gas has the following advantages:

- **Nitrous oxide decomposition**: High tail gas temperature facilitates the decomposition of nitrous oxide (N_2O) without the need for an external agent like natural gas or ammonia.
- **Energy recovery**: The process configuration significantly enhances energy efficiency through multiple integrated strategies. It generates higher power output in the expansion turbine, which decreases the steam consumption of the compressor train and enables increased steam export for other uses. In addition, the system incorporates an extra heat recovery stage just before the tail gas is released into the atmosphere, improving the overall energy balance.
- **Possibility of combining NO_x and N_2O removal in one bed:** The selected temperature by Stamicarbon process design makes it feasible to remove NO_x in the same vessel and even in the same bed. The selected tail gas temperature is appropriate for NO_x catalytic reduction by ammonia.
- **Operational safety**: Accurate control of the process minimizes ammonia slippage, preventing unsafe situations due to potential reaction of ammonia and NO_x.

4 INTEGRATION OF TERTIARY ABATEMENT IN EXISTING PLANTS

Stamicarbon offers emission reduction solutions for all nitric acid plants. The different solutions regarding abatement systems depend on the tail gas temperature in the process. There are three differentiated categories:

- **High tail gas temperature (> 550 °C):** The standard solution for N_2O reduction can be implemented, since this temperature is optimal for reducing N_2O without requiring a reducing agent. Stamicarbon can offer an axial or radial reactor depending on the plant characteristics. For example, Stamicarbon designed a tertiary abatement reactor for Soluciones Químicas nitric acid plant in Mexico.
- **Medium tail gas temperature (340ºC-550 °C):** For this temperature range, Stamicarbon can offer its standard solution as implemented in grassroot plants. This solution consists of two catalyst beds where N_2O is eliminated without any reducing agent required in the first bed. At this temperature the NO_x present acts as co-catalyst for N_2O removal. These NO_x gases will be removed in the presence of NH_3 as well in another catalyst bed downstream, depending on the amount of NO_x of each specific plant. Both can be abated using a single bed as well depending on the client's wishes. This process is described in detail in Section 5.
    - As an exception, in the range between 340-450°C, most likely, a reducing agent, such as natural gas, will be required to reduce N_2O. Each case will be evaluated individually.
- **Low tail gas temperature (<350°C):** When the tail gas temperature is below 350-400°C, direct implementation of a tertiary abatement system is not feasible. This scenario is common in older nitric acid plants, which typically operate with lower tail gas temperatures. Figure 1 shows a potential solution.
    - To enable proper functioning of the tertiary abatement system while minimizing impact on energy balance and existing equipment, the temperature upstream of the tail gas expander must be adjusted:
        - Increase the temperature of the process gas to reach the required operating temperature for the tertiary abatement catalyst.
        - Cool the gas back down before entering the expander to remain within its design temperature limits.
    - A dedicated heating and cooling system will be engineered to meet these needs, with customization based on the client preferences and plant-specific conditions.

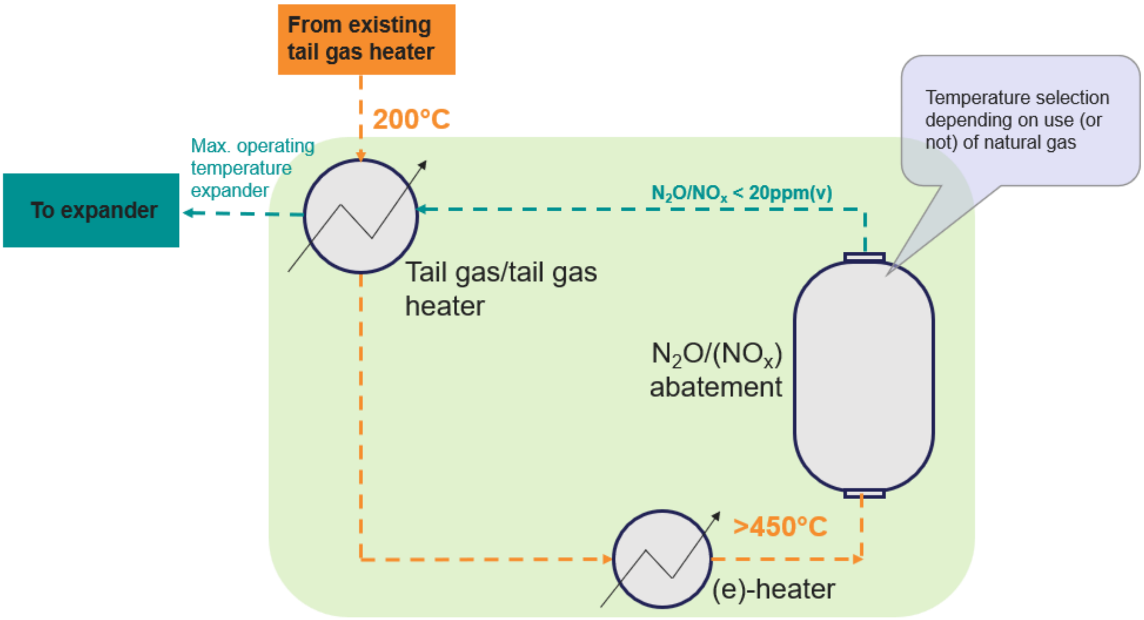

Figure 1. A potential solution for N_2O (and NO_x) removal for low tail gas temperature plants.

5 STAMICARBON DESIGN

Stamicarbon offers three solutions for the tertiary abatement reactor, tailored to both grassroot plants and revamp projects:

1. **Standard one-bed reactor:** As seen in Figure 2, ammonia vapor is introduced as a reducing agent for NO_x and mixed with the tail gas upstream of the tertiary reactor. The resulting mixture enters the reactor from the bottom center, flowing into a radial catalyst bed. The treated tail gas exits from the top of the reactor and is then directed to the tail gas expander.

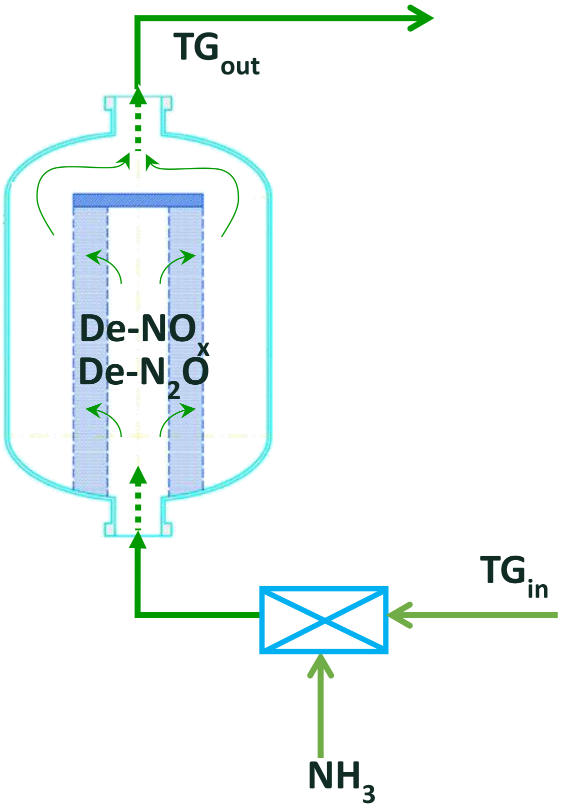

Figure 2. Standard one-bed solution for N_2O and NO_x removal.

2. **Two-bed reactor (especially high NO_x scenarios):** For cases where NO_x concentrations in the tail gas are significantly elevated, Stamicarbon has developed a two-bed reactor configuration, see Figure 3. This design includes several distinctive features that enhance both process performance and operational flexibility. It is engineered to efficiently handle higher pollutant loads while maintaining compatibility with plant constraints.

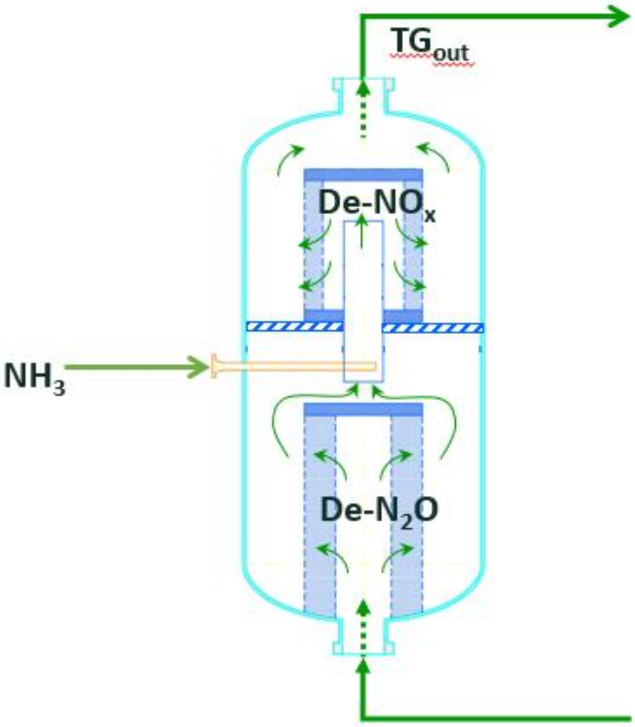

Figure 3. Two-bed solution for N_2O and NO_x removal.

3. **Axial reactor:** When space constraint is a critical factor, the axial reactor presents an efficient and compact solution. This design enables simultaneous removal of both N_2O and NO_x on a single monolithic catalyst bed, see Figure 4. Its minimal footprint and reduced pressure drop make it particularly advantageous for plant revamping projects, where optimizing existing space is key.

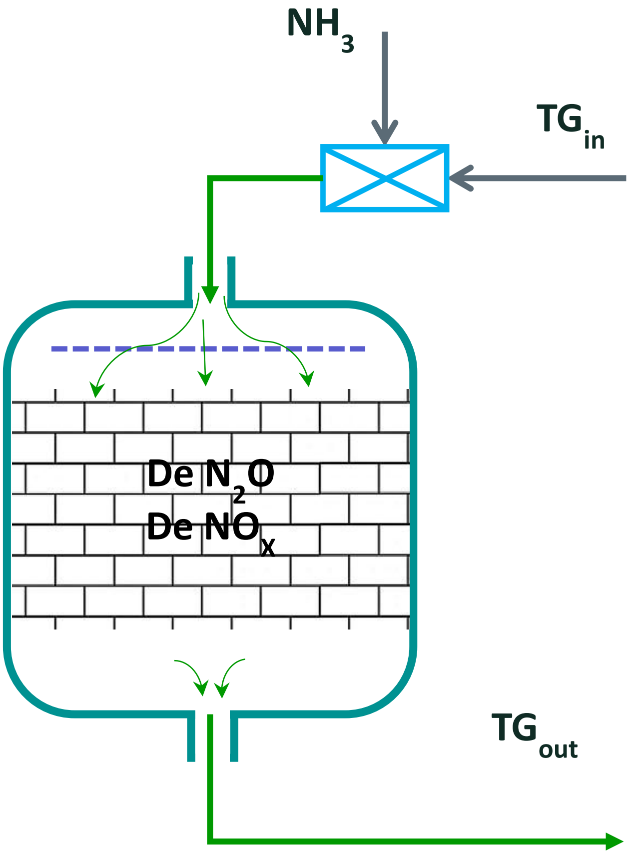

Figure 4. Axial bed solution for N_2O and NO_x removal.

6 DIFFERENT CATALYST TYPES

Extruded catalysts are formed by forcing a mixture of catalyst materials, binders, and pore-forming agents through a specific mold to create a desired shape. This process is known as extrusion molding. Extruded catalysts are often used in radial bed reactors to optimize the flow and reduce pressure drop.

Monolith catalysts are characterized by their large open frontal area, which allows for high space velocities and minimal resistance to flow through the channels. Monolith catalysts are often used in axial bed reactors and are known for their lower pressure drop and ease of maintenance. As a relatively newer technology compared to pellet catalysts, monolith catalysts can be especially advantageous in revamp projects due to their smaller volume and overall reduced reactor size. Both types of catalysts have their advantages and are selected based on specific process conditions and requirements.

For grassroot plants, the standard solution recommended by Stamicarbon is to use extrudate-type catalysts, as multiple existing references demonstrate high performance with this material. While monolithic catalysts are also expected to deliver high efficiency, their coating on cordierite substrates may result in a shorter operational lifetime compared to extrudates. However, in revamp scenarios, in which space is limited and there might be a need to minimize pressure drop to maintain the energy balance in the plant, monolithic catalysts may be a more suitable choice.

7 CONCLUSIONS

Stamicarbon provides emission reduction solutions not only for new grassroot plants, where the process is designed to achieve optimal values for tertiary abatement to reduce N_2O and NO_x in a single reactor, but also for plants with a wide range of tail gas temperatures. This includes potential evaluations of secondary and tertiary abatement, with feasibility studies encompassing various suppliers for catalysts and critical equipment. Each case undergoes detailed evaluation, and a tailor-made solution is developed, prioritizing simple schemes while considering factors such as plant location, required emission reductions, and client preferences (e.g., reducing agents, electricity availability, available area, etc.).

Stamicarbon collaborates with catalyst suppliers for both secondary and tertiary abatement considering monolith and extrudate catalysts to offer the most optimized design for each case, adopting a flexible business approach and considering the most attractive business model for each scenario.

Recently, Stamicarbon was awarded a NACAG project for a tertiary abatement system to be installed at a non-Stamicarbon plant, Soluciones Químicas site in Mexico. Stamicarbon is also highly active in revamp projects for emissions reduction, offering the best available technology tailored to each unique case.

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