



Article

Ammonia—A Fuel of the Future? Economies of Production and Control of NO_x Emissions via Oscillating NH₃ Combustion for Process Heat Generation

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Abstract

This study investigates the viability of using Ammonia as a carbon-free fuel for heat generation in terms of both reactive Nitrogen and Carbon emissions and production cost. As a carbon-free, environmentally friendly energy carrier, Ammonia has the potential to play a significant role in the sustainable, clean energy supply of the future. However, a major drawback of the steady combustion of ammonia for process heat generation is the extremely high levels of NO_x emissions it produces. In this pilot-scale study, the experimental results show that, through the oscillating combustion of NH₃, NO_x emissions can be reduced by as much as 80%. Production costs were compared to evaluate the economic feasibility of Ammonia-based heat; the results reveal the economic challenges associated with using Ammonia compared to natural gas, even when accounting for the development of CO₂ pricing. Only in terms of Carbon Capture and Storage requirements is Ammonia-based heat economically advantageous. This study also scrutinizes the economies of the production of gray and green Ammonia. Considering CO₂ certificate costs, the cost of green ammonia would be competitive in the near future.

Keywords: ammonia; oscillating combustion; NO_x emissions; process heat; production cost



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1. Introduction

Our modern, industrialized society relies heavily on fossil fuels, with such fuels forming the backbone of industries like chemical, steel and non-ferrous metal manufacturing and downstream processing. In addition to electrical power, various industrial processes demand huge amounts of process heat, and its generation consumes a significant portion of the fossil fuels mined today, resulting in massive amounts of anthropogenic CO₂, a well-known greenhouse gas, being released into the atmosphere. Comprising approx. 80% of all greenhouse gas emissions [1], CO₂ is by far the main contributor to the progressive warming of the planet. In order to reduce human-made greenhouse gas emissions, as required by the Kyoto Protocol, adopted in 1997 [2], and keep the Earth's warming, if possible, to 1.5 °C, well below the 2 °C stipulated in the 2015 Paris Agreement [3], humankind must turn to alternative energy sources. Among carbon-free fuels, Hydrogen (H₂) and Ammonia (NH₃) are perhaps the primary candidates of interest [4–9]. Ammonia liquefies readily at −33 °C and has a volumetric energy density of 14 MJ/m³. Hydrogen, in contrast, has a lower volumetric energy of 11 MJ/m³, and its liquefaction takes places at an extremely low temperature of −253 °C [10–13]. These features mean that NH₃ currently

has the advantage over H_2 , particularly in transportation and storage, as NH_3 can be stored at room temperature and a pressure of 0.99 MPa, or at $-33\text{ }^\circ\text{C}$ and atmospheric pressure, alternatively, while the storage of Hydrogen at room temperature requires a pressure of 70 MPa [8,10]. Transportation of Ammonia is generally possible in ships designed for propane, since propane's boiling and condensation temperatures are almost the same as those of NH_3 [10,14]. However, it should be noted that research into alternative methods for the storage and transportation of H_2 —in the form of Ammonia Borane (NH_3BH_3)—is also underway [15–20].

The first attempts to use Ammonia as a fuel date back to the 1940s when, during World War II, a coal gas mixed with NH_3 was used to drive an omnibus in Belgium [14,21]. Later, NASA's X15 airplane, developed in the 1960s, was powered by a fuel comprising mixed liquid Ammonia and Oxygen [22], while the US Army tried, unsuccessfully, to develop an NH_3 -powered gas turbine [23–25].

Efforts to address global warming in the 1990s revived the interest in Ammonia as an energy source, and comprehensive scientific programs have since been launched worldwide, with Japan leading the way. In Japan, the most significant contribution was the launch of the government-supported Strategic Innovation Promotion Program (SIP) for energy carriers in 2014 [26]. In a combined effort within the framework of the program, AIST and FREA succeeded in creating an NH_3 -fueled micro gas turbine [10]. In order to enhance the flame stability and combustion efficiency, it featured an NH_3 vaporizer and a gas compressor, along with a heat regeneration cycle. For NO_x emissions control, an SCR system was installed downstream of the turbine, which resulted in a reduction in the NO_x concentrations in the exhaust gas from approx. 1000 ppm to less than 10 ppm. When fueled by 100% Ammonia, roughly 42 kW of power generation and a combustion efficiency of about 95% were achieved, and the residual NH_3 was used as an additive for the SCR unit. This prototype demonstrated that emissions and combustion efficiency are mostly affected by the combustor inlet temperature [27].

Also in Japan, the Central Research Institute of Electric Power Industry (CRIEPI) developed a furnace co-fired with NH_3 and pulverized coal. It was established that, if Ammonia is injected about 1m downstream of the burner, the NO_x emissions are comparable to those of pulverized coal combustion without co-firing. The added NH_3 made up as much as 20% of the total Lower Heating Value (LHV) of the fuel [10].

A group from Osaka University, in cooperation with Tayo Nippon Sanso, modified a 10 kW furnace to operate with a mixture of ammonia and natural gas using oxygen-enriched air (max. 30% O_2). By altering the location of the secondary air injection port, the NO_x emissions were kept below 150 ppm. The most important outcome was the demonstration of the capability of NH_3 as a fuel for heat generation [10].

The IHI Corporation managed to develop a 2 MW gas turbine in which four parts methane were co-fired with one part NH_3 (Ammonia mixing ratio of 20% LHV). Tailor-made combustor and NH_3 injection nozzles were designed as a way to keep the NO_x emissions below 300 ppm, and NO_x was further reduced to about 6 ppm by means of the SCR unit following the combustion chamber. The project demonstrated the potential of NH_3 co-firing within the field of CO_2 emissions reduction. However, more work, particularly relating to further NO_x reduction, as well as optimization of initial and operating costs, is necessary [13,28].

Outside Japan, a number of other countries, most prominently, the USA [13,29–31], Australia [13,32,33], the UK [13,34–36], the Netherlands [13,37], the UAE [38] and Canada [39,40], also adopted national strategies targeting industry decarbonization based on Ammonia as a fuel or H_2 storage. In addition, different research groups investigated the application of NH_3 as a fuel for SI (spark ignition) and CI (compression ignition)

engines [41–53] and the performance of NH_3 -powered gas turbines [54–56] and NH_3 -fed fuel cells [4,57–66].

However, regardless of its high potential, using Ammonia as an energy carrier has its shortcomings. The heat combustion and the laminar burning velocity of NH_3 are approx. 40% and 20% lower, respectively, compared to those of fossil fuels [10,67–74], and Ammonia also features a higher ignition temperature and low flammability [10,75–79]. The chemistry of Ammonia burning has been extensively studied since 1960s, and, although different chemical mechanisms of NH_3 oxidation have been proposed over the years [51,52,73,80–96], a deeper understanding is needed before NH_3 can be fully exploited as a fuel. Challenges include the control of NH_3 slip due to unreacted ammonia [97–99] and, perhaps most significantly, the extremely high levels of NO_x emissions produced by Ammonia combustion.

NO_x is a common term for the gaseous oxides of Nitrogen— NO , NO_2 , NO_3 , N_2O , N_2O_3 , N_2O_4 , which form during most combustion processes NO_x is of concern for decades, resulting in comprehensive studies of the reaction mechanisms of its formation through the years [80,84,88,100–105]. It is harmful to humans and environment. N_2O is another greenhouse gas with a 265 times higher warming potential than CO_2 [106–108]. In the aspect of air quality, however, only the two most important, NO and NO_2 are considered as NO_x .

Much effort was committed to develop strategies for NO_x reduction. Broadly, the measures for NO_x —emissions control can be divided in two groups—combustion modification or primary measures and post-combustion or secondary measures [109–112]. The secondary measures focus on the NO_x removal from the flue gas. They encompass Selective Catalytic Reduction (SCR), Selective Non-catalytic Reduction (SNCR), wet scrubbing, electron beam, adsorption, electrochemical reduction, as well as non-thermal plasma [85,112–115]. By the means of these processes close to 100% NO_x abatement is achieved, but demand high investment costs and bulky equipment, which makes upgrading of existing plants not always viable [112]. Some are still in the development stage, as in the case of electrochemical reduction [112,116–121].

In contrast, the primary measures focus on avoiding the formation of NO_x during combustion and include fuel and air staging [79,108,120–122], low excess air [108], flue gas recirculation [108,120,122], low- NO_x burners [108] and re-burning [108,123,124], as well as Linde's LoTox [125] and MARTIN's VLN [126] processes. They are low-cost and particularly suitable for the retrofitting of existing facilities. All primary measures should be utilized before secondary measures are considered.

Most relevant to this study is oscillating combustion [127–129], which can be principally seen as air and fuel staging carried out over time, i.e., it is a primary NO_x reduction approach.

The aim of our study was to establish the optimal operating parameters for the low-emission combustion of NH_3 with air relating to NO_x while simultaneously keeping NH_3 slip and N_2O emissions low. The experiments were set up to simulate boiler firing for process heat generation and the experimental results demonstrate how far the application of this primary measure, i.e., an oscillating mode of combustion involving interrupting the NH_3 supply, enables a reduction of NO_x concentration in the exhaust gas.

The theoretical part of our study scrutinized the economies of the production of gray and green Ammonia via the Haber–Bosch process [8,10,13,130]. Gray Ammonia production depends on Hydrogen produced conventionally by the steam reforming of natural gas [8,13,130]. It is estimated that 1.8% to 3% of all global energy is consumed by the fabrication of NH_3 . At present, most of this energy still comes from fossil fuels, meaning NH_3 manufacturing is one of the largest CO_2 emitters [131]. In contrast, the foundation

of green Ammonia production is H_2 obtained via water electrolysis [8,132–136] realized using electrical power from renewable energy sources [137]. Norway has been examined as a green NH_3 fabrication site due to its high abundance of renewable energy, particularly from affordable hydropower, which accounts for 44% of the total energy supply in the country [138–140]. The economic analysis was conducted with and without considering CO_2 pricing.

An assessment of the entire Ammonia process chain was also carried out. The process chain comprises NH_3 production in Norway from regeneratively produced H_2 and N_2 from air separation, transportation to Germany, storage in Germany and, finally, energy recovery through combustion with air and conversion of the enthalpy into steam or process heat.

2. Materials and Methods

All experimental investigations were carried out at the Gas- und Wärme-Institut (GWI) in Essen, Germany.

2.1. Gaseous Fuels

During the experimental studies, the combustion of pure NH_3 , as well as different gas mixtures consisting of natural gas and ammonia, was investigated. NH_3 with a purity of at least 99.98% was supplied in barrels, and natural gas corresponding to gas quality “H” was delivered to the location of the experiments via a pipeline [141]. The quality of the natural gas was additionally continuously monitored with a Type PGC 9303 gas chromatograph from RMG as knowledge of the exact chemical composition is crucial for the proper adjustment of its volume flow and the volume flow of the combustion air. For the furnace tests, the volume flows of the natural gas and the combustion air were adjusted with the help of a sophisticated gas mixing system based on various thermal Mass Flow Controllers (MFCs). The volume flow regulation of NH_3 was realized using a separate system with a similar design.

2.2. Burner

A tailor-made swirl burner developed by an industrial partner exclusively for the combustion of alternative gases was used for the experimental trials. It features a central gas lance which can also be moved axially and offers a rated output of 250 kW. The combustion air, divided into primary air (PA) and secondary air (SA), can be supplied to the combustion chamber via separate annular gaps in a swirling mode. By changing the distribution of the combustion air and adjusting the position of the gas lance, the burner can be adapted to the specific application.

In order to stabilize the Ammonia flame, a flame tube was attached to the inner side of the burner’s port on the furnace wall. The flame tube was of simple design, made of a piece of heat-resistant steel tube welded onto a mounting flange.

To further support the flame stability, the burner was pulled 15 cm back relative to the initial position (Figure 1).

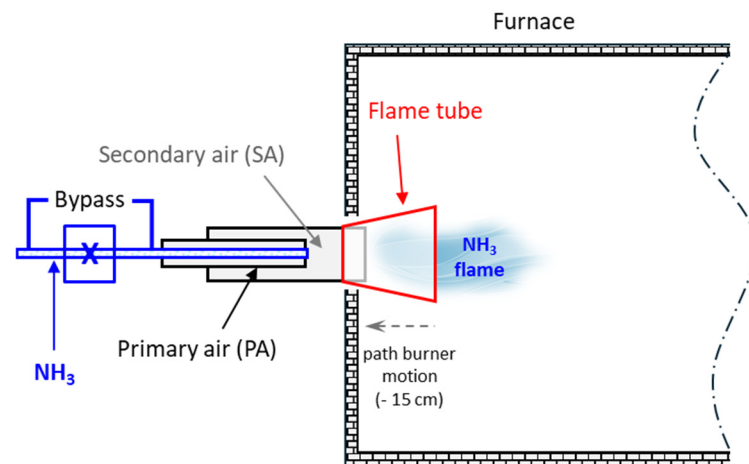


Figure 1. Setup of burner, flame tube and furnace.

2.3. Oscillator

In order to achieve oscillating combustion of the Ammonia fuel, a controllable oscillator was assembled. The core of the oscillator was a gas solenoid valve (Figure 2), enabling a sufficiently high number of switching cycles over a broad range of frequencies. This valve was controlled with the help of a 24 VDC power supply in combination with an adjustable cycle valve. Additionally, a bypass was set up in order to provide a pilot flame with the fuel gas (Figure 2), serving to secure stable re-ignition in the event of an interruption in the gas flow (as in the case of oscillation).



Figure 2. Ammonia fuel supply line equipped with a gas solenoid valve and a bypass, courtesy of the GWI.

2.4. Test Furnace Systems

To resemble a boiler firing system, the ammonia burner was integrated into a **water-cooled combustion chamber** with the following dimensions: length—1.5 m, width—0.8 m, height—1 m (Figure 3a). It simulated the conditions in a boiler firing system and comprised a cylindrical steel tube confined in a liquid-tight housing. The hollow space formed between the steel tube and the housing was filled with water through an inlet connected to a freshwater pipe. The exhaust water was discharged afterwards through an outlet connection in the drainage. The inlet and outlet water temperatures were monitored

continuously by two thermocouples, and the volume flow of the water was supervised by means of a flow meter. This water-cooling system configuration allowed, depending on the burner load, for an exhaust gas temperature between 108 °C and 115 °C. The burner was mounted at the front edge of the chamber and accordingly connected to the supply lines for combustion air and fuel gas (gas mixing device) (Figure 3a). The initial ignition of the burner was supported by a start-up flame lance attached sideways to the combustion chamber, perpendicular to the burner (Figure 3b).

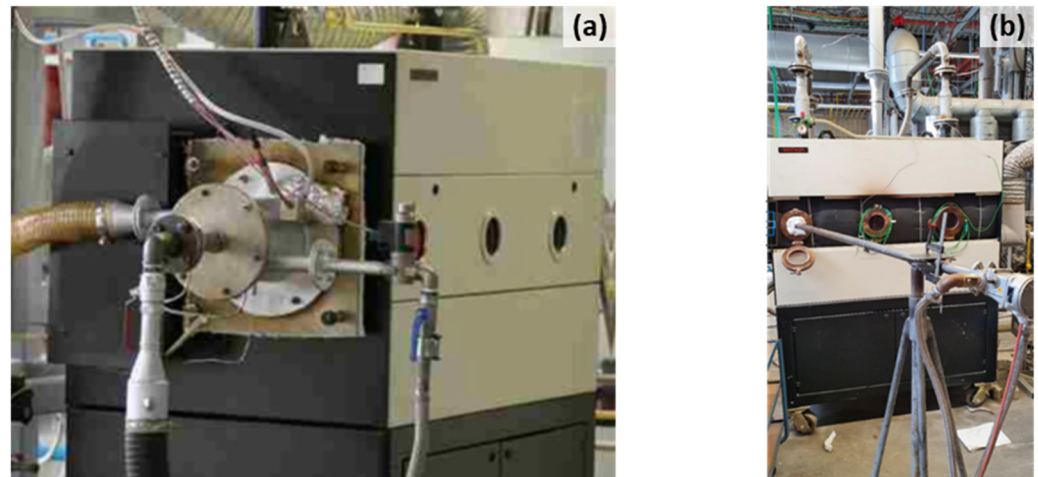


Figure 3. Water-cooled combustion chamber with integrated Ammonia burner (a), start-up flame lance location (b), courtesy of the GWI.

The potential for NO_x reduction via oscillating Ammonia combustion was investigated in another series of experiments executed in an **isothermal combustion chamber**. The basic layout of the system is pictured in Figures 4 and 5. The combustion chamber had a length of approx. 2.5 m, a width of 1.2 m and a height of 1.85 m. The interior was lined with a refractory material (Al₂O₃) and four additional radiant tubes at the bottom of the chamber served to control the temperature inside it. Another four air-operated cooling tubes permitted a load to be mimicked through heat dissipation. Seven closable openings integrated along the side of the furnace (approx. central plane) allowed direct access to the combustion chamber during operation, and a larger viewing window located in the front furnace section could be opened, if necessary, for visualization and measurement of burner flames through optical methods.

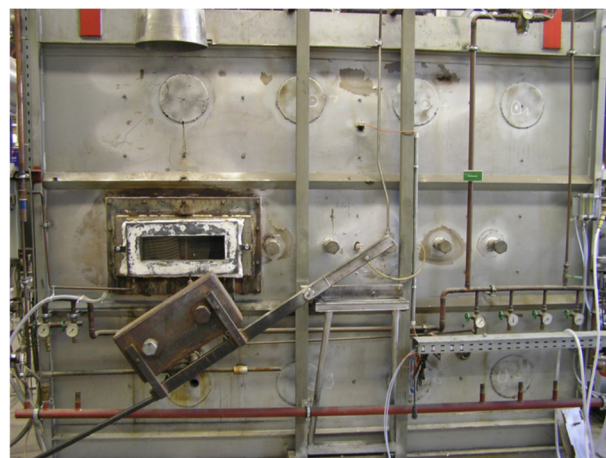


Figure 4. Right-side view of the isothermal combustion chamber, courtesy of the GWI.

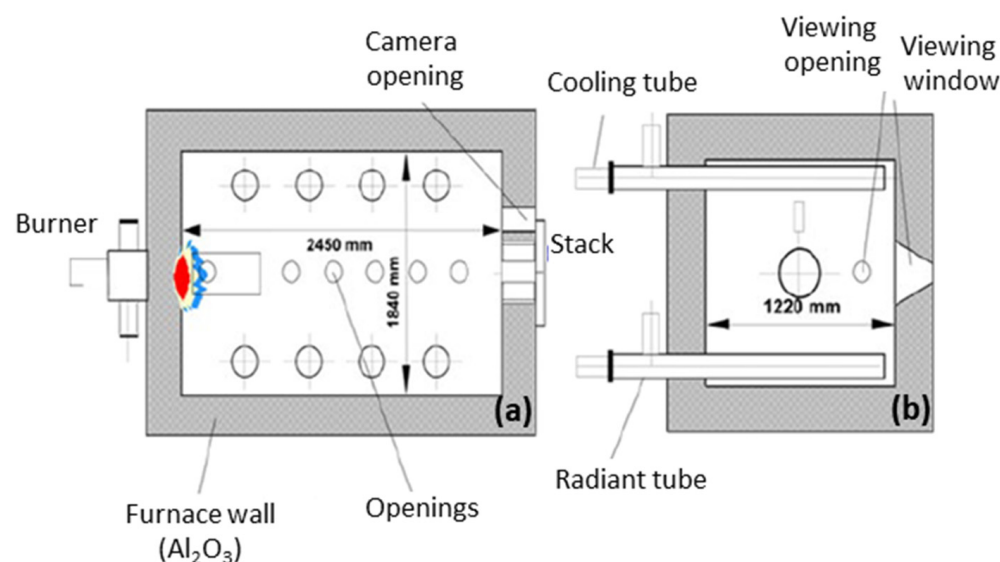


Figure 5. Schematic layout of the isothermal combustion chamber. Right-side view (a), front edge view (b), courtesy of the GWI.

2.5. Flue Gas Analytics

The concentrations of NH_3 (Ammonia slip), N_2O , NO , NO_2 , NO_x , H_2O , CO and CO_2 in the flue gas were measured continuously with a portable Fourier-Transform Infrared (FT-IR) DX 4000 spectrometer from Gaset[®] (Mestartintie 6, FI-01730 Vantaa, Finland) [142]. The spectrometer operates in the wave number range from 900 cm^{-1} to 4200 cm^{-1} and is characterized by a multi-reflection unit and a heated measuring cell with a fixed path length of 5 m, a spectral resolution of 8 cm^{-1} and a scanning frequency of 10 scans/s. After reaching the operating temperature of the measuring cell ($180\text{ }^\circ\text{C}$), prior to each measurement, a new zero (background) spectrum was recorded, to ensure the reliability of the measurement results. The recorded infrared spectra were analyzed by means of the software package Calcm[®] 12.16 [143] utilizing a modified Classical Least-Squares (CLS) method [144]. The unit was calibrated for the flue gas species listed in Table 1.

Table 1. DX 400 flue gas species and concentration ranges.

Component Flue Gas	Concentration Range [mg/m^3]	Concentration Range [Vol. %]	Concentration Range [ppm]
NH_3	0 to 5000		
H_2O		0 to 30	
CO		0 to 2	
CO_2		0 to 20	
N_2O	0 to 180		
NO			0 to 1100
NO_2 dry			0 to 250
NO_x dry			0 to 1300

The parameters of the experimental investigations, which were carried out in two separate series of trial runs, are listed in Table 2.

Table 2. Experimental parameters for NH₃ combustion.

Parameter		First Experimental Series	Second Experimental Series
Volume flow (NH ₃)	[m ³ _N /h]	10.7–19.4	0–32.5
Volume flow (natural gas)	[m ³ _N /h]	11.8–3.1	12–0
Total volume flow fuel (NH ₃ + natural gas)	[m ³ _N /h]	22.5	32.5
NH ₃ fraction	[%]	48–86	0–100
Natural gas fraction	[%]	52–14	100–0
Volume flow (combustion air)	[m ³ _N /h]	110	150
Primary air/secondary air	[%]	15/85	100/0
Air ratio, λ	[-]	1.24–1.06	1.06–1.97
Swirl	[-]	Yes	Yes
Temperature (combustion air)	[°C]	25	25/260
Oscillation frequency	[Hz]	0	0–2
Furnace type		Water-cooled	Isothermal
Furnace interior temperature	[°C]	800	800/1000
Thermal output furnace	[kW]	110	130

2.6. Economic Analysis

The economies of the production of gray and green Ammonia were comprehensively analyzed with the help of the **Palisade @Risk 8** software system, [145], which is particularly suitable for the reliable evaluation of the OpEx [146], CapEx [147] and profitability of green and gray NH₃ production. While CapEx include investment costs for the facility only, OpEx comprise the expenditures for materials, supplies, utilities, capital, maintenance and depreciation costs, as well as taxes and insurance. An amortization period of 20 years was assumed and the cost of capital was calculated with an interest rate of 10%; the annual increase for the maintenance costs was considered to be between 1% and 3% of the investment costs; and tax and insurance costs of 1% to 2% of the investment costs were taken into account [148].

The economic feasibility study evaluated the whole system comprising the process chain for the manufacturing in Norway, the subsequent maritime transportation to Germany, storage in Germany, the combustion of the ammonia in a boiler firing plant and the NO_x reduction in the flue gas achieved via SNCR or SCR. Both the unreduced NO_x concentrations and the values lowered by the oscillating mode of combustion were also considered.

3. Results

3.1. NH₃ Combustion in a Water-Cooled Combustion Chamber

Before investigating the NO_x reduction achieved through the oscillating combustion of Ammonia, the challenge of generating a constant burning/stable NH₃ flame had to be tackled. The burner was mounted on the water-cooled combustion chamber and started with natural gas. While the fuel and the total combustion air volume flows were kept constant throughout, NH₃ was gradually mixed with the natural gas while the natural gas fraction in the fuel mixture was simultaneously reduced by the same proportion. The Ammonia fraction in the fuel mixture, x_{NH_3} , was calculated according to Equation (1):

$$x_{NH_3} = \frac{\dot{V}_{NH_3}}{\dot{V}_{NH_3} + \dot{V}_{CH_4}} \quad (1)$$

The temperature and chemical composition of the exhaust gas, as well as its volume flow, were monitored and recorded continuously.

Figure 6 shows the NO_x and NH_3 concentrations in the flue gas as a function of the NH_3 portion in the fuel mixture achieved by the 110 kW thermal output of the water-cooled combustion chamber. In a swirl mode, $110 \text{ m}_N^3/\text{h}$ combustion air was supplied, 15% as primary air and the rest as secondary air.

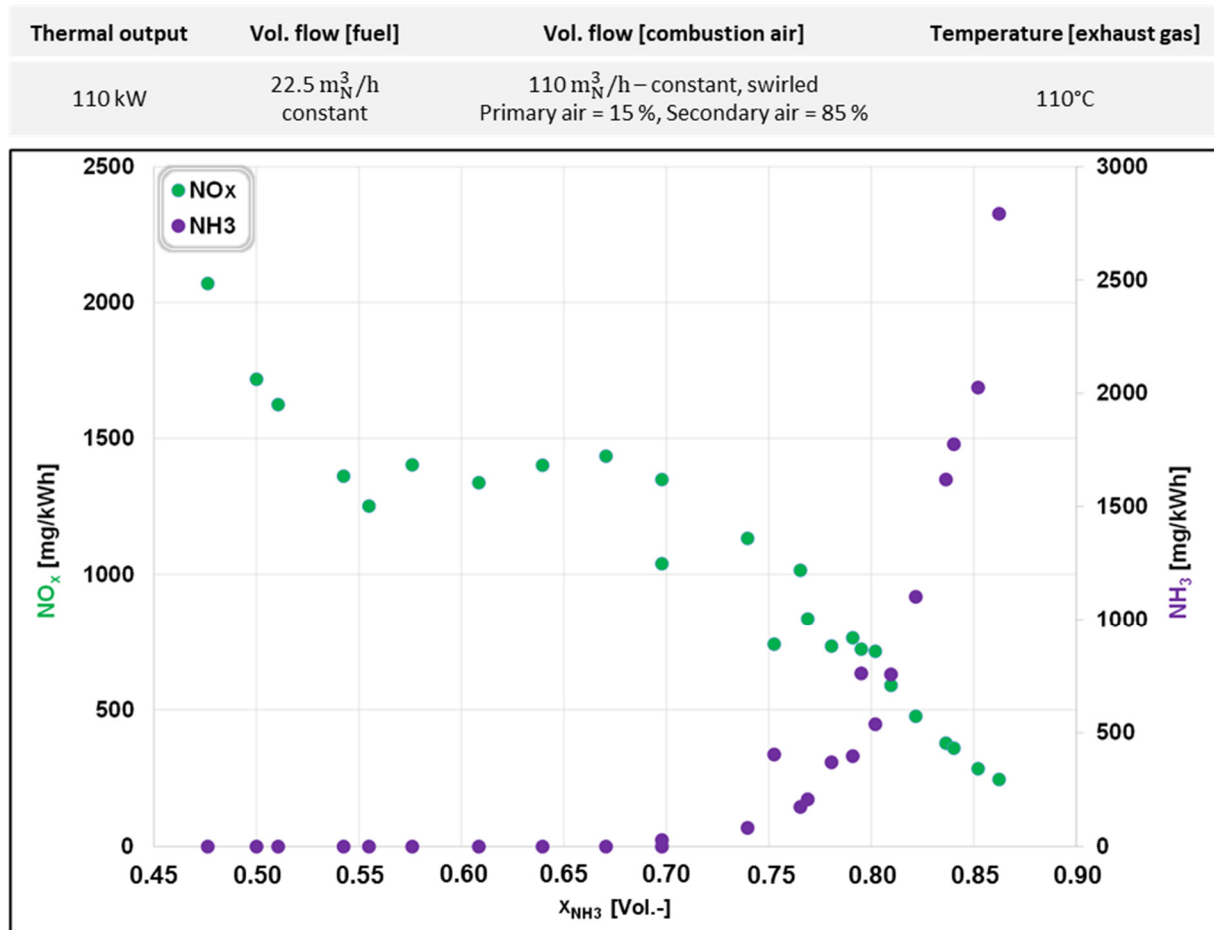


Figure 6. Flue gas NO_x and NH_3 concentrations as a function of NH_3 portion in the fuel mixture in a water-cooled combustion chamber.

To compensate for the difference between the heating values of NH_3 (18.6 MJ/kg) and natural gas ($34\text{--}52 \text{ MJ/m}^3$, depending on location of origin), NO_x and NH_3 concentrations are plotted in energy units, mg/kWh , rather than mg/m_N^3 , as usual. Due to technical issues, the acquisition of the Ammonia volume flow data was interrupted occasionally. In these instances, the NH_3 portion in the fuel mixture was partly calculated from the measured flue gas concentrations of the H_2O vapor, O_2 and CO_2 . The calculated values deviated by 10% to 15% from the experimentally measured ones.

The Ammonia slip in the exhaust gas hit a maximum of 2800 mg/kWh at NH_3 fuel fraction $x_{\text{NH}_3} = 0.86$. The rapid increase in NH_3 concentrations in the flue gas with the increasing NH_3 portion (from $x_{\text{NH}_3} = 0.70$ onwards) in the fuel mixture suggests a significant amount of unburned ammonia. This is highly undesirable due to various possible negative impacts, including the following: (1) In the atmosphere, Ammonia can react with SO_2 and/or NO_x to form fine particles of Ammonium sulfate $((\text{NH}_4)_2\text{SO}_4)$ and/or Ammonium nitrate (NH_4NO_3) . These particles can significantly reduce the air quality or contribute to the formation of haze, which poses a serious risk to human health and the environment [13,42,110,149]. (2) NH_3 is toxic and irritates the eyes and the respiratory system. (3) Ammonia is corrosive and thus can damage equipment, which results in

higher maintenance costs [11,13,149]. (4) Unutilized Ammonia is simply wasted, and will eventually inflate the operational expenditures.

A reaction between the nitrogen oxides in the flue gas and the unburnt Ammonia resulted in a decrease in the NO_x concentration from 2100 mg/kWh to approx. 250 mg/kWh.

Although the combustion air was supplied at maximum swirl all the time, with ammonia fractions in the fuel mixture larger than 0.86, the flame could not be sustained and, consequently, the burner continuously went out. For that reason, further investigations of ignition and flame stability improvements were carried out in the isothermal combustion chamber.

3.2. NH_3 Combustion in an Isothermal Combustion Chamber

Prior to the Ammonia combustion experiments, the isothermal combustion chamber was preheated to 800 °C by means of the radiant tubes and a natural gas flame. All of the combustion air was added as primary air and was not preheated. Similarly to in the trial runs in the water-cooled combustion chamber, a mixture of natural gas and Ammonia with a gradually decreasing natural gas fraction and increasing NH_3 fraction, respectively, was burned, and the flame kept burning until an Ammonia content of 90% was reached. It was not possible to secure operation of the burner with 100% NH_3 under these experimental conditions.

As shown in Figure 7, the NO_x concentration reached 5015 mg/kWh. It increased with the increasing Ammonia fraction in the fuel, x_{NH_3} , until the latter equaled 0.6, after which further increasing the Ammonia content resulted in a decreasing NO_x concentration. For almost the entire duration, the concentration of the NH_3 slip remained low at approximately 3.6 mg/kWh, though a minimal increase was observed at $x_{\text{NH}_3} \approx 0.9$.

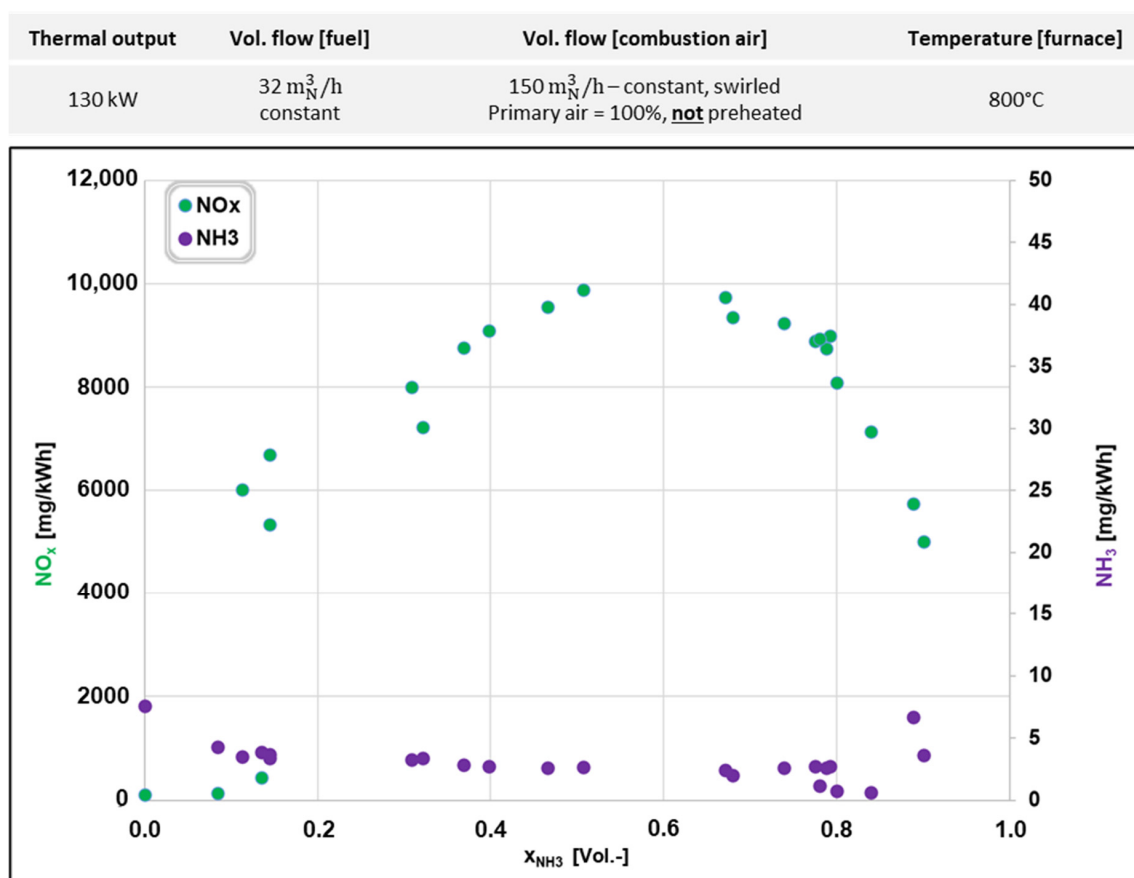


Figure 7. Flue gas NO_x and NH_3 concentrations as a function of NH_3 portion in the fuel mixture in an isothermal combustion chamber with no combustion air preheating.

The CO concentration ranged from 0.15 mg/m³_N (dry) (0.12 ppm) to 0.20 mg/m³_N (dry) (0.16 ppm), indicating very good burnout. The corresponding CO₂ concentration varied between 80 mg/m³_N (dry) (4.07 vol.%) and 213 mg/m³_N (dry) (10.84 vol.%).

In order to secure stable NH₃ ignition, the combustion chamber temperature was increased to 1000 °C with the aid of the radiant tubes, and, to further support the ignition, the combustion air was preheated to 260 °C. The whole volume was supplied again as primary air only in a swirl mode, and the absence of secondary air prevented convective cooling. Consequently, the radiant energy of the red-hot flame tube mounted on the combustion chamber wall (inside, at the burner's entrance port) could be transferred to the premixed flame as a portion of the ignition energy. In this way, by combining an increased furnace temperature, preheated combustion air and the installation of a flame tube, stable ignition/burning of a pure (100%) Ammonia flame was achieved. The resulting nitrogen oxide concentration was as high as 13,900 mg/kWh (approx. 14,900 mg/m³_N) (Figure 8), while the concentration of the Ammonia slip was as low as 0.4 mg/kWh (approx. 0.42 mg/m³_N), which indicates a very good burnout.

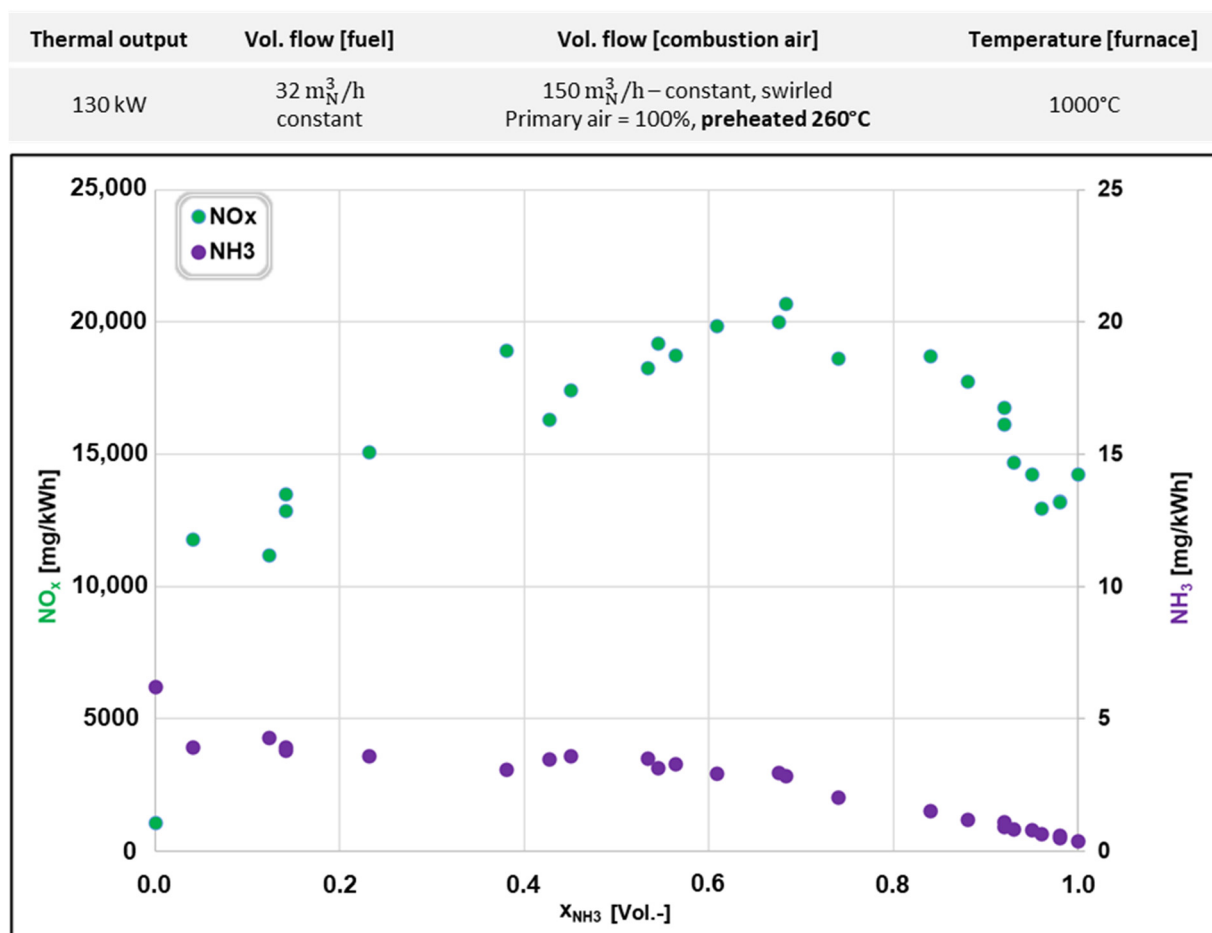


Figure 8. Flue gas NO_x and NH₃ concentrations as a function of NH₃ portion in the fuel mixture in an isothermal combustion chamber with combustion air preheated to 260 °C.

The NO_x concentration reached a maximum of 20,700 mg/kWh (approx. 23,600 mg/m³_N) with an NH₃ fraction in the fuel mixture of 68%.

The NH₃ slip concentration was below the value of 30 mg/m³_N considered the legal limit in Germany [150], but the NO_x concentrations in the exhaust gas very significantly exceeded the legal limit value of 150 mg/m³_N [150].

Before reaching a 100% Ammonia fraction, the CO concentration decreased from approximately 15 mg/m_N^3 (dry) (0.12 ppm) to 0 mg/m_N^3 (dry), indicating a highly efficient combustion process. During the same period, the CO_2 concentration dropped significantly from about 227 mg/m_N^3 (dry) (11.57 vol. %) to just 12 mg/m_N^3 (dry) (0.59 vol. %).

3.3. NO_x Reduction Through Oscillating Combustion of Ammonia in an Isothermal Combustion Chamber

Generally, oscillating combustion is part of air and fuel staging, implemented not on a local scale but in stages over time. In other words, the oxidant or the fuel flow is interrupted in time [127].

In our study, the Ammonia fuel flow rate was periodically interrupted, resulting in alternating periods with different stoichiometric ratios following one after another.

The basic principle of NO_x reduction by oscillation with the amplitude Δ is shown in Figure 9. In the fuel-rich periods (Δt_1), through CH_4 combustion (a), significant amounts of CO, H_2 and possibly residual hydrocarbons (CH_i) are generated, while, in the fuel-poor periods (Δt_2) (with increased air addition), combustion leads to the formation of CO_2 , H_2O and NO_x .

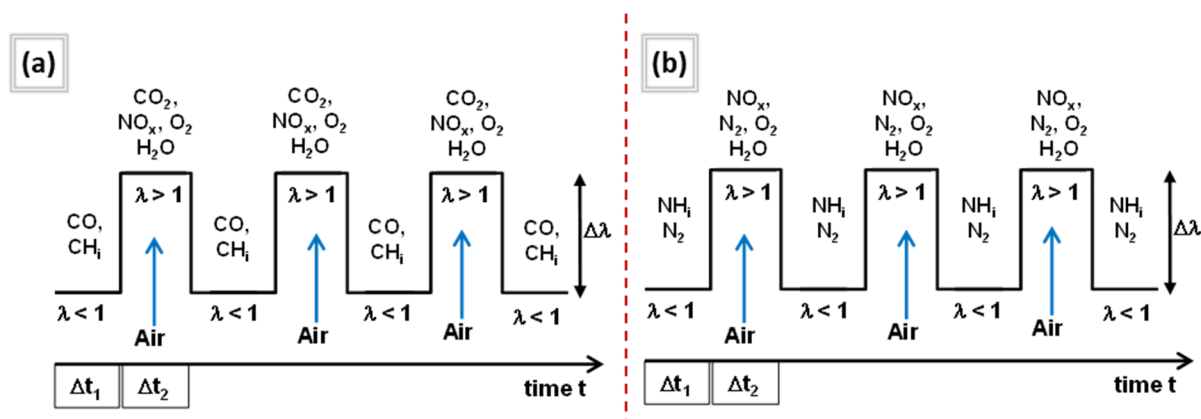


Figure 9. Principle of NO_x reduction by the oscillating combustion of methane (a) and Ammonia (b).

Similarly, in the case of NH_3 combustion (b), during the fuel-rich time periods (Δt_1), the combustion products are predominantly N_2 , H_2 and H_2O . In these periods, the air ratio drops below 1, which leads to a lower flame temperature, and results in reduced NO_x formation [128,151]. Conversely, in the fuel-deficient time intervals (Δt_2), the increased air supply (air ratio > 1) leads to a higher flame temperature (high-temperature reaction zone), which results in the formation of NO_x too [128]. When projected over the time scale, the average NO_x concentration is reduced, and the efficiency of NO_x reduction by the oscillating combustion is influenced by the oscillating frequency and oscillating amplitude [128].

The results from the oscillating combustion of pure Ammonia are shown in Figure 10. Prior to the oscillating mode of operation, the ammonia flame was initiated and run stably for approx. 30 min. In this period, the NO_x emissions remained around 14,200 mg/kWh (corresponding to about $14,500 \text{ mg/m}_N^3$ dry), and the Ammonia slip ran in the range of 0.4 mg/kWh. These values are roughly of the same magnitude as those shown in Figure 8.

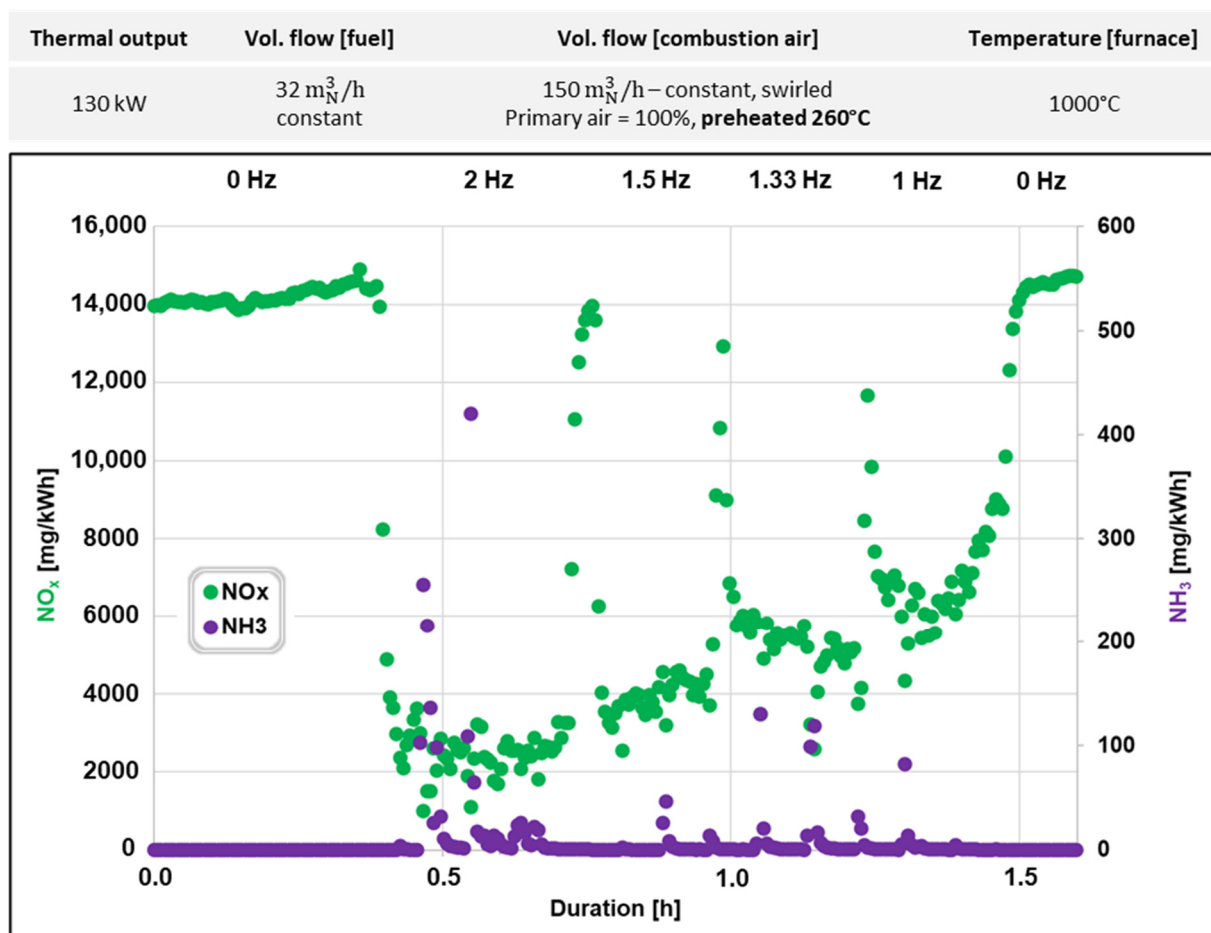


Figure 10. NO_x and NH₃ concentrations as a function of the oscillation frequency in an isothermal combustion chamber.

Afterwards, the NH₃ supply was realized in an oscillating manner. Starting at 2 Hz, the oscillating frequency was gradually lowered to 1 Hz. The highest NO_x reduction of about 80% was recorded at 2 Hz, where its concentration decreased to approx. 2500 mg/kWh ($\sim 2590 \text{ mg/m}_N^3$); however, the Ammonia slip increased to around 33 mg/kWh ($\sim 33.4 \text{ mg/m}_N^3$). At an oscillating frequency of 1 Hz, the nitrogen oxide concentration decreased to 7240 mg/kWh ($\sim 7540 \text{ mg/m}_N^3$), which is not as high the reduction of 48% but is still significant. Due to technical limitations, the effect of oscillating frequencies higher than 2 Hz was not investigated.

3.4. Economic Analysis

3.4.1. Cost Analysis of Ammonia Production in Norway Without CO₂ Pricing

At first, the operating costs (OpEx) of gray and green ammonia production in Norway were calculated without taking into account the price of CO₂ certificates. OpEx include materials, supplies, utilities, capital, maintenance and amortization costs, as well as taxes and insurance. The amortization period was set at 20 years and the capital costs were taken into account at an interest rate of 10%, based on capital expenditures (CapEx).

For gray ammonia production, the literature data for a plant with the best available technology (BAT) are used. Accordingly, the required thermal energy is 7.8 MWth/t_{NH₃} [152], distributed as follows: 5 MWth/t_{NH₃} for the supply of raw materials, 2.18 MWth/t_{NH₃} for fuels, 0.39 MWth/t_{NH₃} for electricity and 0.23 MWth/t_{NH₃} for steam.

The operating costs of green ammonia production are based on an energy balance with an electrical energy power demand of 11.8 MWh_{el}/t_{NH₃}. Since the H₂ for the green

Ammonia synthesis is projected to be supplied by water electrolysis, the costs for water are included in the material costs too. A demand of $9 \text{ l}_{\text{H}_2\text{O}}/\text{kg}_{\text{H}_2}$ is assumed, which corresponds to $1.6 \text{ t}_{\text{H}_2\text{O}}/\text{t}_{\text{NH}_3}$ [153]. Table 3 summarizes the data required for the calculation of the operating costs.

Table 3. Material expenditures considered for the operating costs calculation for green Ammonia production.

Projection Costs	2020	2030	2040	2050	Location	Reference
Natural gas [€/MWh]	33	34	36	37	EU	[154]
Electricity [€/MWh]	42	41	44	49	NOR	[155]
Demineralized water [€/t _{H₂O}]	0.44	0.45	0.45	0.45	NOR	[156,157]

The calculated operating costs for gray and green ammonia production in Norway from 2020 to 2050 without considering CO₂ pricing are shown in Figure 11.

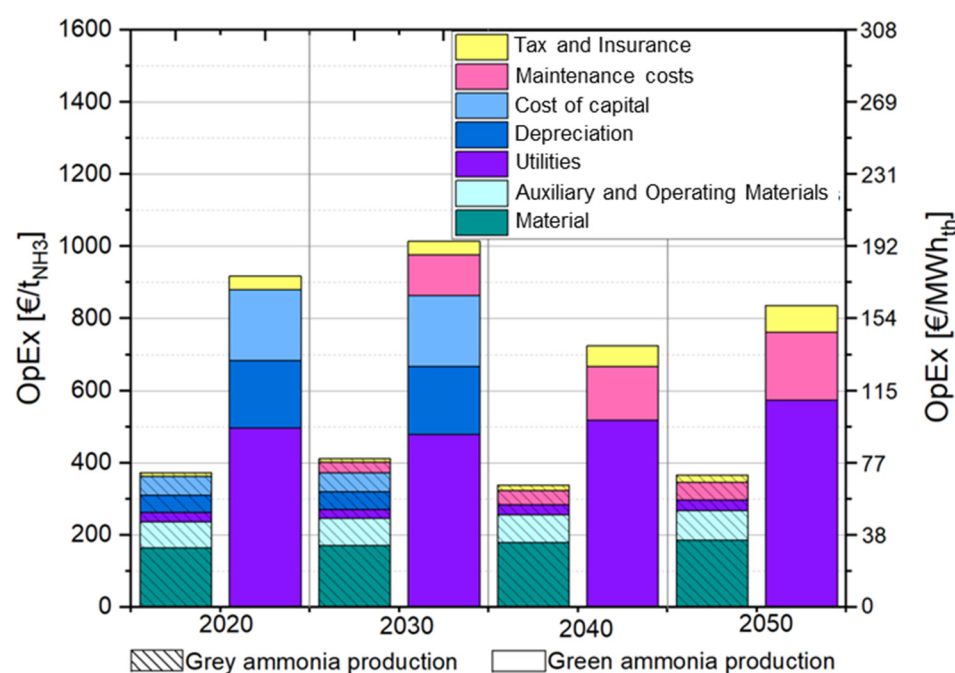


Figure 11. Operating costs of gray and green ammonia production in Norway from 2020 to 2050 without considering CO₂ pricing.

3.4.2. Cost Analysis of Ammonia Production in Norway with CO₂ Pricing

As early as 2003, the European Parliament and Council prepared a directive to establish a trading scheme for certificates for greenhouse gas emissions, hoping to stimulate industry to reduce them [158]. Although Norway is not a member of the EU, it is closely linked to the it through the EEA Agreement and the European Economic Community (EEC); thus, under the EEA Agreement, EU environmental and climate legislation also applies in Norway. Norway has also participated in the EU Emissions Trading System (EU ETS) since 2008. Therefore, the prices for CO₂ emissions must be taken into account. Table 4 shows an estimate of CO₂ certificate costs in the EU from 2020 to 2050.

Table 4. Cost estimate for CO₂ certificates in Europe through 2050.

Cost Estimate	2020	2030	2040	2050	Location	Reference
CO ₂ certificate [€/t _{CO₂}]	25	282	398	515	EU	[159,160]

The CO₂ emissions of gray ammonia synthesis can vary depending on the age of the plant. In this case, a CO₂ emission of 2 t_{CO₂}/t_{NH₃}, that of a state-of-the-art plant, was used [152]. Green ammonia synthesis has a CO₂ emission of 0.17 t_{CO₂}/t_{NH₃}, which is related to the transport of gases within the production plant [152].

The price of CO₂ certificates in 2020 was around 25 €/t_{CO₂} (Table 4) and is estimated to increase to 515 €/t_{CO₂} by 2050 [157,159] (Figure 12).

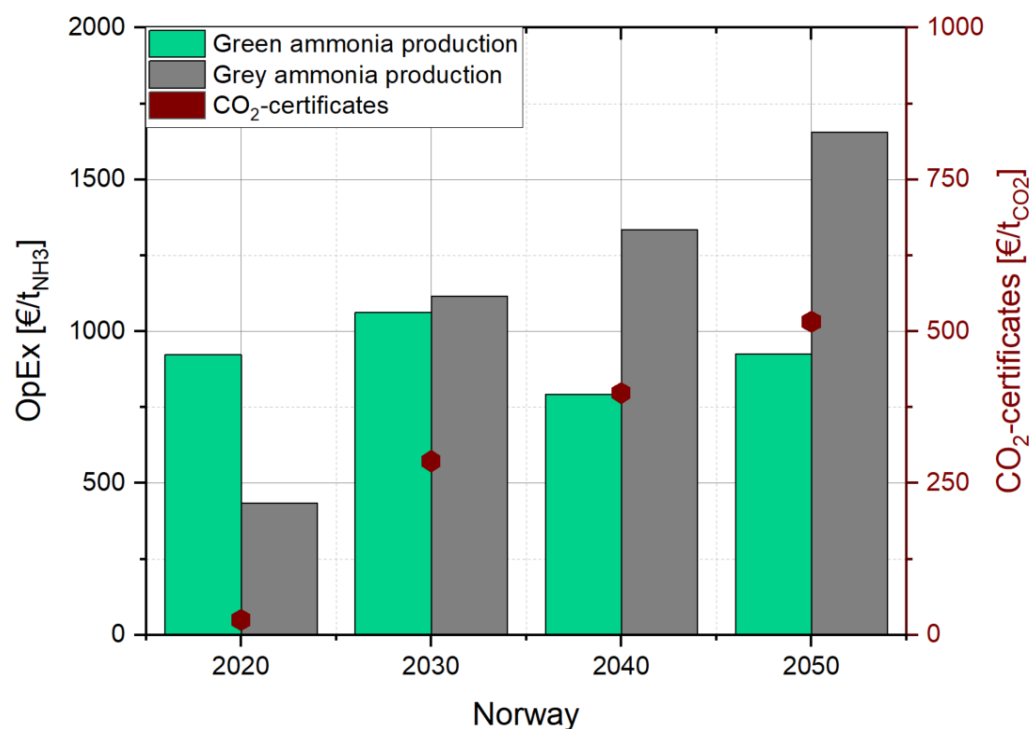


Figure 12. Operating costs of gray and green ammonia production in Norway from 2020 to 2050 considering CO₂ certificate prices.

3.4.3. Cost Analysis of the Entire Ammonia Process Chain

The process chain comprises the production of ammonia in Norway, its subsequent transportation to Germany, storage in Germany, combustion in a boiler combustion plant and, finally, exhaust gas cleaning (Figure 13). The costs are listed in Table 5.

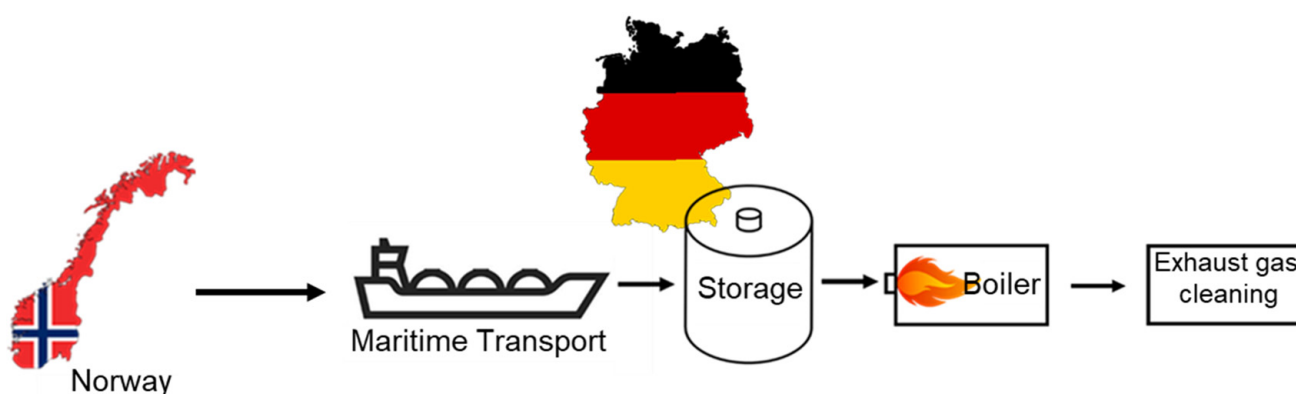


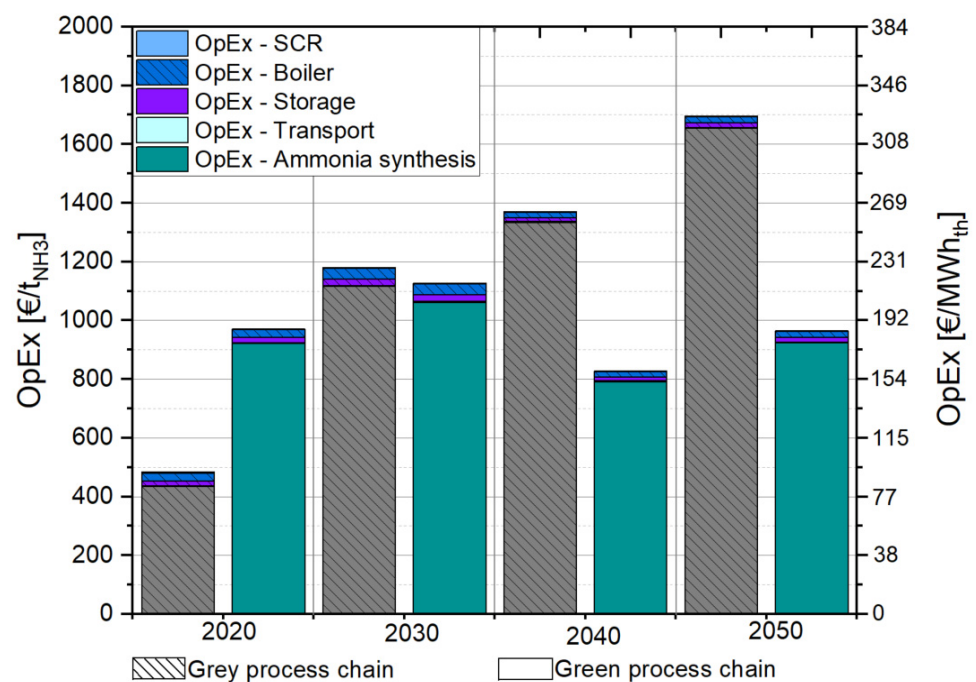
Figure 13. Structure of the Ammonia process chain for the cost analysis.

Table 5. CapEx and OpEx for transportation of Ammonia from Norway to Germany, storage and boiler firing and subsequent flue gas treatment.

	CapEx [Millions of EUR]	OpEx [€/t _{NH₃}]			
		2020	2030	2040	2050
Maritime transport	-	2.6	2.6	2.6	2.6
Storage	16.4	16.6	23	13.3	14.6
Boiler firing	30	26.5	35.6	18.2	20.8
Flue gas treatment	3.5	9.5	10.4	8.5	9.1

A distance of 608 km is assumed for the transportation of ammonia from Norway to Germany. According to [161], sea transportation is more cost-effective than pipeline transportation for distances over 300 km. Therefore, the transportation of Ammonia by ship is considered here, and freight rate, capacity, transport speed and port charges are taken into account. Ammonia storage requires special infrastructure and safety precautions, so construction and operating costs for NH₃ storage facilities are included, as well as maintenance costs. For boiler combustion of ammonia for heat generation, the investment and operating costs are calculated for a 100 MW plant.

The aggregated process chain costs of green and gray NH₃ production are visualized in Figure 14. The largest share of costs for both relates to the production itself; for the green and gray ammonia process chains, respectively, approx. 93% and 89% of the total costs result from Ammonia synthesis. The transport, storage, boiler firing and flue gas cleaning have a smaller impact.

**Figure 14.** Calculated operating costs of the process chains for the production of gray and green ammonia, including further use, from 2020 to 2050.

From the experimental investigations, an NO_x concentration of 2920 mg/kWh in the exhaust gas was determined for oscillating combustion at 2 Hz. Since the NO_x concentration in the exhaust gas is not permitted to exceed 100 mg/m_N³ dry (at 3 vol. % O₂ content) [158], which corresponds to 99.73 mg/kWh, post-combustion treatment of the exhaust gas must take place. For this purpose, a combination of SNCR and SCR was selected.

The total cost estimate is about EUR 3.5 million, without consideration of the expenses for various peripheral systems such as Ammonia storage, dosing and control. Due to the higher exhaust gas concentrations, the combined SNCR and SCR process requires more raw materials, considered in the OpEx as material and auxiliary expenditures. The material costs amount to about 6 €/t_{NH₃} and thus account for the largest share of the total costs (Figure 15).

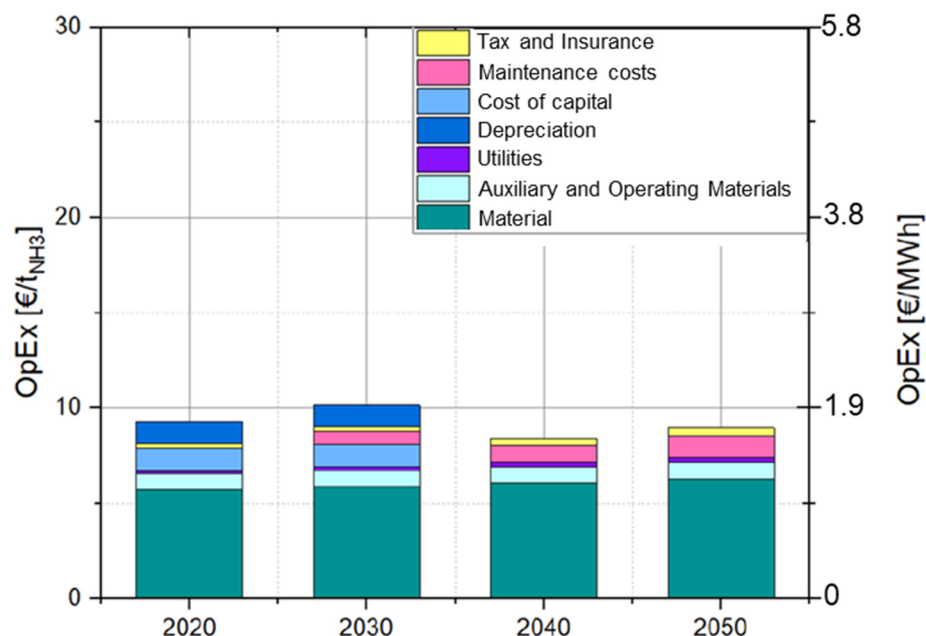


Figure 15. Calculated cost for the operation of a combined SCR and SNCR exhaust gas treatment facility from 2020 to 2050.

Based on the experimentally obtained NO_x emissions data from the combustion of NH₃, Table 6 shows the investment and operating costs for operation with and without oscillation.

Table 6. Investment and operating costs for the reduction of NO_x concentrations.

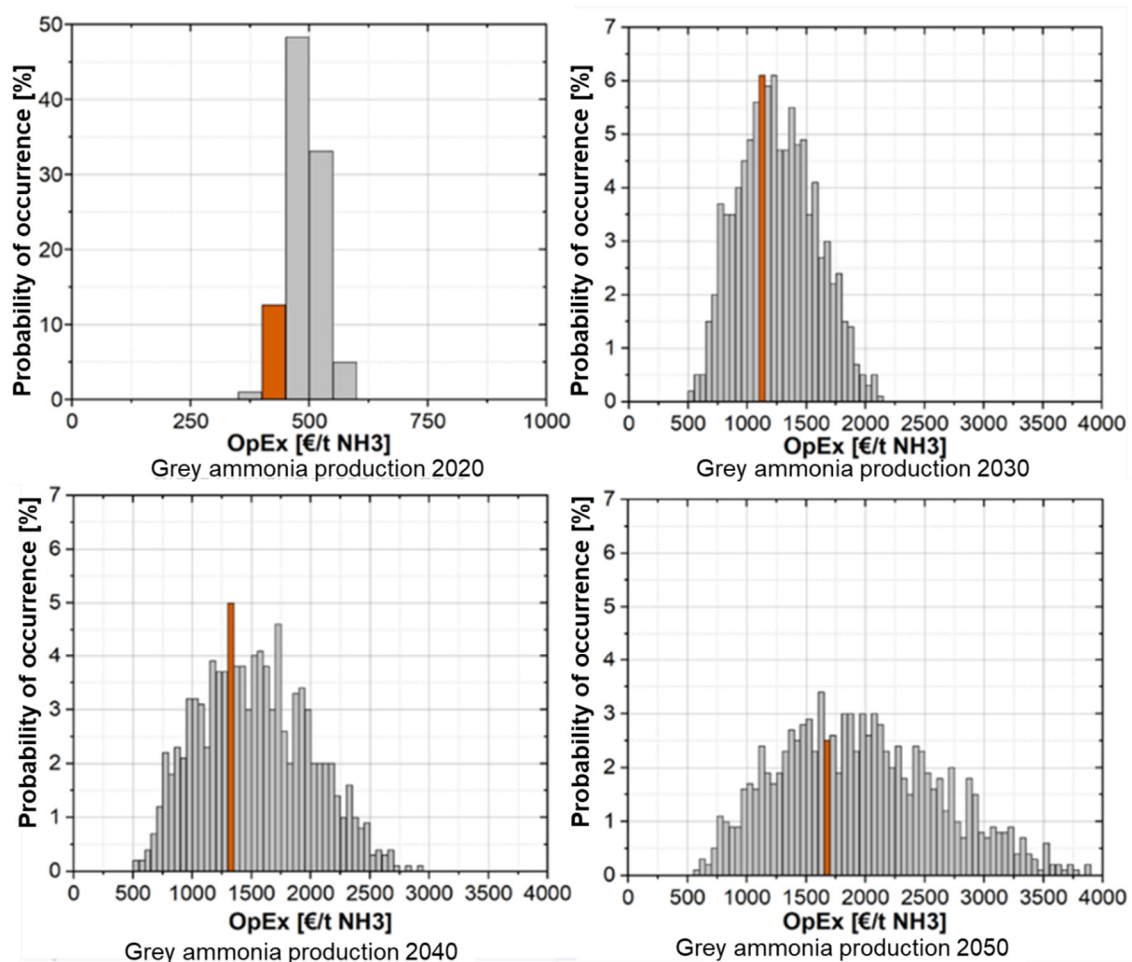
Operation Mode	Investment SNCR and SCR	Reduction Rate [%]		2020	2030
		SNCR	SCR	[€/t _{NH₃}]	
Without oscillation (14,000 mg/kWh)	EUR 3.5 million	86	95	21.8	23.2
With oscillation (3000 mg/kWh)		70	90	9.5	10.4

The reduction rates are adapted to the input concentrations. A simplified assumption is made of a comparably high investment volume for both scenarios, as a catalyst is always needed for the secondary NO_x reduction (SCR or SNCR). Due to the differently distributed reduction rates, the consumption of operating resources is not reduced linearly with the NO_x concentrations, but is halved under the assumptions made here. The corresponding operating costs for exhaust gas cleaning are listed in Table 7.

To evaluate the influence of the Ammonia synthesis on the total costs, a sensitivity analysis was carried out. The results showed that the total operating costs of gray Ammonia production in 2020 are slightly higher than those calculated in the profitability analysis (Figure 16).

Table 7. Price forecasts for operating resources in flue gas cleaning.

	2020	2030	2040	2050	Reference
Ammonia water [€/t]	103	106	109	113	[157]
Compressed air [€/m ³ _N]	0.03	0.03	0.03	0.03	[158]
Electricity [€/MWh]	45	76	72	59	[160]

**Figure 16.** Sensitivity analysis of the total cost of ownership of gray ammonia production (the red bars represent the calculated value from the profitability calculation).

Costs between 350 €/t_{NH₃} and 600 €/t_{NH₃} were calculated, meaning the value from the profitability calculation (434 €/t_{NH₃}) is in the lower calculation quantile. The cost estimate for the year 2030 is in the range between 500 €/t_{NH₃} and 2150 €/t_{NH₃}, but the value of 1115 €/t_{NH₃} determined from the profitability calculation has a high probability of occurrence. Likewise, for 2040, operating costs of up to 2750 €/t_{NH₃} are possible, while the calculated value of 1334 €/t_{NH₃} has a high probability of occurrence. The results for 2050 show that costs vary between 550 €/t_{NH₃} and 3900 €/t_{NH₃}, with the calculated value of 1655 €/t_{NH₃} in the medium range of the probability of occurrence.

As shown in Figure 17, the total costs of green ammonia production from 2020 to 2050 are likely to be lower than those calculated in the economic analysis, mainly due to the price ranges shown, particularly in the supply sector.

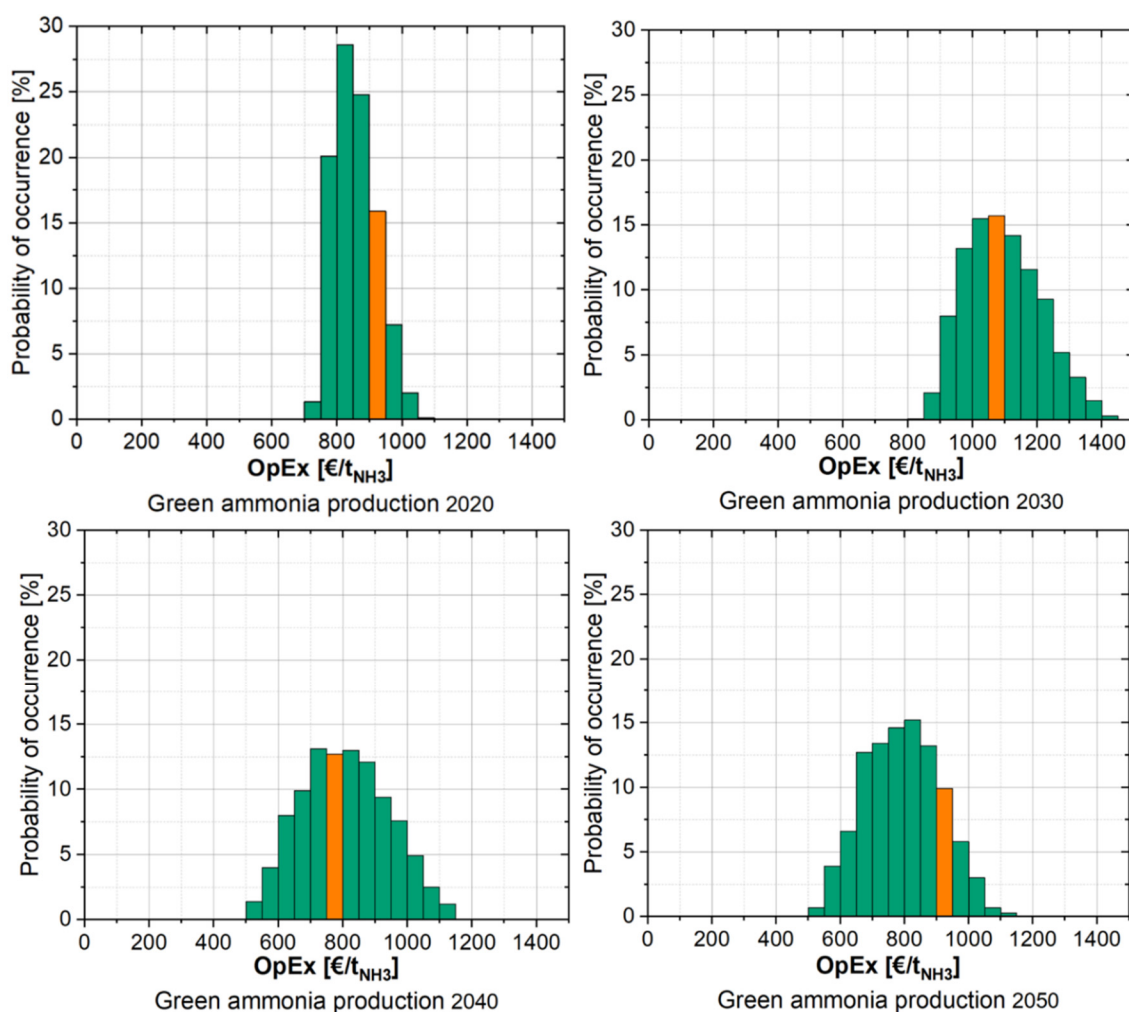


Figure 17. Sensitivity analysis of the total operating costs of green ammonia production (the red bars represent the calculated value from the profitability calculation).

Costs between 750 €/t_{NH₃} and 1100 €/t_{NH₃} were calculated for 2020 and costs in the range of 550 €/t_{NH₃} to 1150 €/t_{NH₃} for 2050. The values from the profitability calculation were 902 €/t_{NH₃} for 2020 and 924 €/t_{NH₃} for 2050, respectively. The OpEx determined from the profitability calculation, 1062 €/t_{NH₃} for 2030 and 792 €/t_{NH₃} for 2040, are correspondingly close to the OpEx determined with the highest probability of occurrence. The sensitivity analysis confirmed the assumptions made in the economic analysis and thus the credibility of the results.

3.4.4. Cost Analysis of Heat Generation by Boiler Firing with Natural Gas and Ammonia

The combustion of ammonia for heat generation has not yet been established. Therefore, the combustion of Ammonia for heat generation in a boiler firing system was compared with that of natural gas as a reference fuel. Because of the well-developed natural gas pipeline network in Germany, transportation and storage were omitted from the analysis. For a direct comparison of the combustion of natural gas and ammonia, the investment costs for the boiler firing system for both fuels were assumed to be the same, and thus, accordingly, the capital costs for the combustion of ammonia and methane were equally high. Flue gas cleaning costs were not regarded in this approach. The stoichiometric combustion of one ton of methane results in 2.73 t_{CO₂}, and so this value is considered in the economic analysis in the pricing of CO₂ certificates. To generate 100 MW of thermal output, natural gas is burned in the boiler firing system at a rate of 7.2 t_{CH₄}/h, and the corresponding

carbon dioxide emissions equal $19.8 \text{ t}_{\text{CO}_2}/\text{h}$. The operating costs for the procurement of natural gas (material costs), auxiliary and operating materials (demineralized water) and operating resources (electricity for the blower and feeding water pump), as well as the CO_2 costs, are represented graphically in Figure 18.

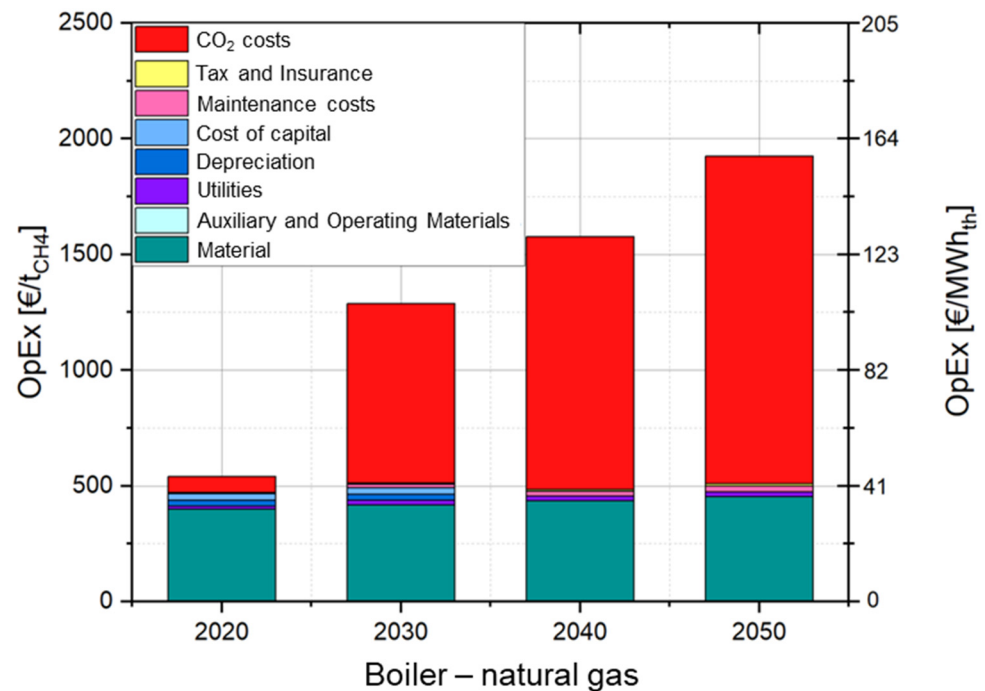


Figure 18. Operating cost of boiler firing with natural gas from 2020 to 2050.

Figure 19 shows the direct cost comparison of the combustion of green ammonia and the combustion of natural gas.

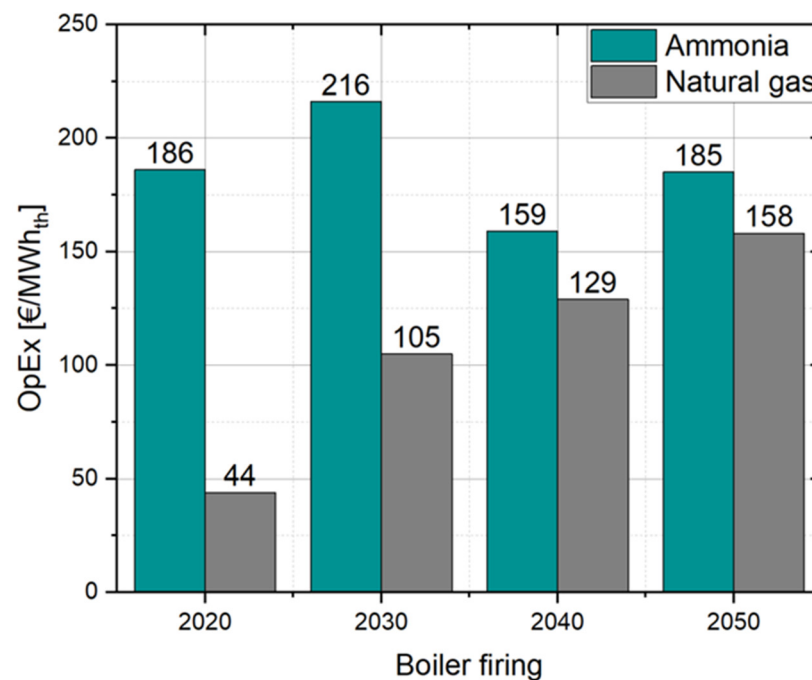


Figure 19. Operating cost of boiler firing with natural gas and ammonia for heat production from 2020 to 2050.

Considering the estimated CO₂ costs and natural gas prices, the combustion of natural gas is more cost-effective than the combustion of ammonia between 2020 and 2050. Due to CO₂ pricing and the costs for the procurement of natural gas, the overall costs for boiler firing with natural gas are projected to increase by a factor of three and a half over the period under consideration.

4. Discussion

4.1. NH₃ Flame Stabilization and Combustion Under Oscillating Conditions

Although promising, the utilization of NH₃ as a fuel has so far been limited mainly to research applications and demonstration plants. One of the most challenging tasks is the stabilization of Ammonia flames due to NH₃'s combustion properties [162]. As our results also show, even with the application of a burner particularly designed for NH₃ combustion, achieving a pure (100%) Ammonia flame without specific measures targeting flame stabilization is very challenging. The change from the water-cooled to the isothermal combustion chamber contributed minimally to the flame stability. It proved too, that reducing the methane amount and simultaneously increasing the NH₃ fraction in the fuel is not enough (Figures 6 and 7). A pure NH₃ flame was not realized before installing the flame tube, increasing the combustion chamber temperature from 800 °C to 1000 °C and preheating the combustion air to 260 °C (Figure 8).

Due to the air preheating and the associated stronger swirl, higher nitrogen oxide concentrations were observed compared to those obtained without combustion air preheating (Figure 7). In both cases, the nitrogen oxide concentrations behaved similarly (max. NO_x concentration at an NH₃ portion of 68%); a further increase in the ammonia content in the gas fuel mixture was accompanied by decreasing NO_x concentrations, while NH₃ slip remained continuously low. This NO_x concentration reduction was due to in situ reduction with excess NH₃ from the gas fuel within the flame, as described elsewhere [163].

The results from the oscillating combustion of Ammonia indicate that the NO_x concentration increased with decreasing oscillating frequency (Figure 10), presumably due to deteriorating mixing conditions in the combustion chamber. Unfortunately, technical limitations prohibited an oscillation frequency of more than 2 Hz; therefore, at present, it is not possible to fully confirm this dependence.

4.2. Production Cost of Ammonia and Heat

The economic analysis showed that, even without taking into account costs for CO₂ certificates, the production of green ammonia in Norway is two and a half times more expensive than the production of gray ammonia in 2020. By 2050, it will still be twice as expensive as gray NH₃ due to higher capital costs and amortization. The synthesis of green ammonia requires considerable amounts of electricity, which accounts for around 54% of the total costs, and electrical power costs are estimated to rise from EUR 40/MWh in 2020 to EUR 48/MWh in 2050. In the case of gray ammonia, the biggest cost driver is the expenses for natural gas, which accounts for around 50% of all material costs.

To understand the importance of the CO₂ expenditures for the production of gray and green NH₃, it is important to look at the CO₂ amounts emitted by the two processes. The production of gray ammonia releases approx. $2 \text{ t}_{\text{CO}_2} / \text{t}_{\text{NH}_3}$ into the atmosphere [154], and, correspondingly, the production costs should increase from 434 €/t_{NH₃} in 2020 to about 1655 €/t_{NH₃} by 2050. In contrast, only $0.17 \text{ t}_{\text{CO}_2} / \text{t}_{\text{NH}_3}$ is emitted by the production of green ammonia. Thus, the prices for CO₂ certificates have only a minor influence on the operating costs of green ammonia synthesis, and green ammonia production could be more profitable than gray ammonia production as early as 2029. Considering the production of green ammonia in Germany, it is obvious that production costs in the future will be roughly

twice as high as production costs in Norway. This significant difference can be attributed to various factors, in particular energy costs and raw material prices.

The examination of the entire Ammonia process chain (Table 5) shows the structure of the OpEx in more detail. As precise cost forecasting for trade by sea is extremely complex and volatile, it is assumed that transportation costs stay constant at 2.6 €/t_{NH₃}, without including the cost of CO₂ emissions. Currently, research is focused on the development of ship engines using Ammonia as a fuel. Generally, there is potential to eliminate the CO₂ emissions resulting from maritime transport and to reduce the cost of transporting Ammonia, since the transported raw material can also be used as fuel for the ship's propulsion system. Starting in 2040, the costs decrease due to the elimination of depreciation costs for the facilities after 20 years. The operating costs for Ammonia storage result from the need for electrical power for liquid storage in tanks, for which an energy expense of 2% energy loss was determined.

The operating costs for boiler firing contain the electricity required for the supply of combustion air, the induced draught and the feeding water pump. The pump circulates a water volume of 113 t/h, which is then used for the boiler. The operating costs of the utilities are thus 4.7 €/t_{NH₃} in 2020 and 7.2 €/t_{NH₃} in 2050. Costs for ammonia as a fuel are not included in the material costs. The considerable costs of the flue gas cleaning processes SCR and SNCR are mainly due to depreciation and capital costs of about 2 €/t_{NH₃}. Maintenance costs amount to about 1 €/t_{NH₃}. The consumption of ammonia water and utilities is comparatively low and accounts for a share of about 1 €/t_{NH₃}.

We assumed that, for heat generation by the combustion of natural gas and Ammonia, the investment costs for the boiler firing plant would be the same. Consequently, the capital, depreciation, maintenance and insurance costs, as well as the taxes incurred, add up to the same value for the combustion of NH₃ and methane. By far, the main cost driver for methane combustion is the price of CO₂ emissions (Figure 18). Due to their high levels from 2030 onwards, the costs increase significantly, and the outcome will be a three-and-a-half-fold OpEx increase for natural gas boiler combustion in the period from 2020 to 2050. In contrast, the firing of the carbon-free fuel NH₃ does not generate any CO₂, so there are no CO₂ costs. At present, operation with natural gas remains economically more attractive than operation with ammonia. However, the cost calculation does not consider any expenses for Carbon Capture and Storage (CCS) technology, which would add further financial load to natural gas combustion. The inclusion of these costs could maybe turn the tide towards boiler firing with Ammonia between 2040 and 2050.

5. Conclusions

Continuous, stable combustion of pure Ammonia, without adding hydrogen or natural gas, was demonstrated in a pilot-scale isothermal furnace. This was achieved with a combustion chamber temperature of 1000 °C and the utilization of a supercritical swirl and combustion air preheated to 260 °C. Under these conditions, NO_x concentrations amounted to around 14,500 mg/m_N³ at the stack, and, with fuel (NH₃) oscillation of 2 Hz, they could be reduced by around 80% to approx. 2650 mg/m_N³. The NH₃ slip reached an average of 34 mg/m_N³ (*all concentrations normalized on 3 vol.% O₂ in the exhaust gas*). Considering increasing CO₂ certificate costs, the production of green ammonia would be more economical than the production of gray ammonia as early as 2029. A sensitivity analysis supports this result.

Based on the experimental findings on NO_x reduction by the primary measure, the costs of heat production using oscillating ammonia combustion were compared to those of natural gas-based heat generation, considering the use of both SCR and SNCR to maintain NO_x emission limit values. Assuming similar capital investment for all options, heat

generation from natural gas combustion will remain cheaper than that from Ammonia firing up to 2050. However, by making conservative economical calculation assumptions and considering expenses for Carbon Capture and Storage, the switch to Ammonia as a fuel could actually pay off by the early 2050s.

Outlook

This study demonstrated that NO_x reduction through oscillating combustion of pure ammonia is generally possible. However, due to technical limitations, the effects of oscillation frequencies higher than 2 Hz could not be investigated. Future research should explore the influence of higher frequencies and identify additional factors, beyond oscillation frequency, that contribute to NO_x reduction. In the present work, flame stabilization was achieved using empirical/qualitative methods only. Subsequent studies should focus on quantitative flame diagnostics, including measurements of ignition delay, flame temperature and chemiluminescence imaging, to gain a deeper understanding of the combustion behavior. The initial economic modeling of green ammonia production primarily considered electricity prices, as they represent the main expenses. Nevertheless, further, more detailed sensitivity analysis is needed to develop the comprehensive economics of ammonia as a fuel for heat supply.

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