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Ammonia as Fuel for Gas Turbines – The Impact of Heat Integrated Partial Decomposition

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ABSTRACT

Ammonia has received in recent years significant attention as potential carbon free fuel. However, its combustion properties limit its direct application for both providing heat and in power generation through gas turbines. Ammonia cracking is one potential solution to circumvent the problem by producing hydrogen. When using the ammonia in gas turbines, it is possible to heat integrate the endothermic decomposition reaction with the exhaust gas from the gas turbine. Thermodynamic and kinetic limitations have however a major impact on the achievable ammonia conversion. Based on the consideration of these limitations, this paper presents a detailed investigation of key design parameters affecting the overall process efficiency utilizing both an equilibrium reactor model and a reactor model based on detailed kinetics and heat transfer. Ammonia decomposition should occur at sufficiently high pressure to avoid a) the com-pression energy demand for achieving the pressure of the combustion chamber and b) to reduce equipment size although the increased pressure results in a reduced conversion when considering an equilibrium reactor. It is crucial for the energy efficient integration of ammonia decomposition with gas turbines to avoid a partial combustion of the decomposed ammonia even as an increased temperature of the reactor results in an increased ammonia conversion. Furthermore, it is beneficial to operate the compression stages without intercooling to reduce the required fuel flow to the gas turbine.

Keywords: Alternative Fuels, Process Design, Modelling and Simulations, Surrogate Model

INTRODUCTION

The application of ammonia in the energy sector has received significant attention in recent years [1]. Ammonia can be considered as both a hydrogen carrier and a fuel in itself. The utilization of Ammonia as fuel results in no direct CO_2 emissions and has the potential to drastically reduce the emissions associated with energy use when produced from decarbonized hydrogen, *i.e.*, hydrogen produced from natural gas reforming with CO_2 capture or electrolysis [2]. In addition, it has in its liquid state the advantage of a relatively high volumetric energy density and established logistics chains. Hence, it can solve the problem of transporting hydrogen from regions with high renewable potential to regions with high energy demand [3]. Ammonia is also seen as direct fuel and not only as hydrogen carrier. It is considered as one alternative for dispatchable power generation [4] with a benefit at low capacity utilizations compared to other fuels when utilized in gas turbines [5]. Its potential application in gas turbines for electricity generation is however hindered by its poor combustion properties [6]. Consequently, decomposition of ammonia into hydrogen and nitrogen is required to maintain a stable flame [5]. Ammonia decomposition, that is the reverse of its synthesis, is an endothermic reaction given by

$$2NH_3 \rightleftharpoons N_2 + 3H_2 \quad (1)$$

High conversions can either be achieved at high temperatures or at a low pressure. Furthermore, the activity of ammonia decomposition catalysts at the low temperatures of gas turbine flue gas is low [7]. As a consequence, analyses normally focus on providing the heat for the decomposition reaction by burning a part of the (partially) decomposed) ammonia as fuel [5]. The decomposition of the ammonia leads however to an efficiency penalty, resulting in calls for the industry to develop novel

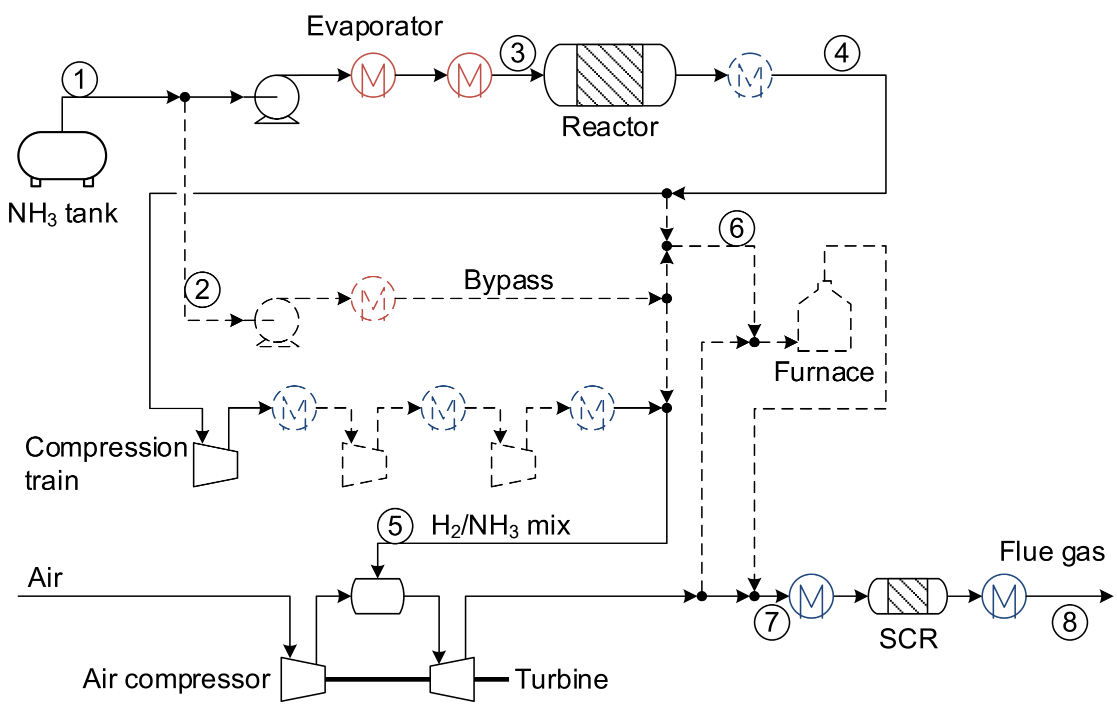

Figure 1. Process flow diagram of the proposed process. Blue heat exchangers correspond to coolers while red heat exchangers to heaters or boilers.

gas turbine burner types to be able to utilize pure ammonia. An alternative route for increasing the efficiency is to focus on partial decomposition of ammonia [8]. The partial decomposition has the advantage of a) having a reduced energy demand for the reaction, b) smaller reactor sizes as full conversion is not required, and c) the potential for heat integration with the flue gas of the gas turbine as less energy is required at lower temperatures.

The application of ammonia in gas turbines has however still open questions. While Cesaro et al. investigated as well the efficiency of partially cracked ammonia [5], their investigation did not consider the thermal integration of the gas turbine flue gas with the decomposition reactor. Instead, the decomposed ammonia (in this case a mixture of hydrogen and nitrogen) is used for providing the energy to the endothermic reaction while the flue gas is only utilized for preheating. Similarly, Paschchenko focused extensively on the operation of the gas turbine, but assumed an equilibrium reactor for the conversion of ammonia [8]. The investigation of the partial decomposition of ammonia has, however, still open questions with respect to the optimal process configuration due to the large number of design parameters.

Hence, we present in this study a thorough analysis of a novel integrated process for partial ammonia decomposition coupled with a gas turbine for exhaust heat recuperation. A focus of the work is to understand how the different design choices may affect the process energy efficiency and potential complexity. A detailed model of the decomposition reactor at low temperatures is included in the process flowsheet as surrogate model to account for the catalytic performance and potential limitations in the heat transfer. This allows the analyses of heat transfer limitations, and hence, both direct integration for heat recuperation and indirect through a heating fluid with a high heat capacity are investigated.

PROCESS DESCRIPTION

Modelling approach

The overall process is modelled in Aspen HYSYS and illustrated in Figure 1. All dashed components are optional depending on the chosen process configuration. Ammonia is stored (*NH_3 tank*) at refrigerated conditions (-33 °C and 1.1 bar pressure) to avoid explosive spillage in the case of a leak [9]. The liquid ammonia (*stream 1*) is first pumped before being evaporated (*Evaporator*) to avoid excessive compression requirements. The gaseous ammonia is then heated to the inlet temperature of the *Reactor*. In this study, two different reactor models are considered; initial analyses focusing on investigating the reactor pressure utilize an equilibrium reactor with an approach to equilibrium of 250 K while in-depth analyses are conducted utilizing a conversion reactor in which the conversion and outlet temperature of the different streams are calculated using a table lookup method given the inlet temperature to the reactor of ammonia and the heating medium (exhaust gas or a heating liquid). The detailed reactor model for creating the table is implemented in MATLAB and models a shell and tube reactor with experimentally calculated kinetics for low temperature ammonia decomposition using a Co-Ce catalyst. These kinetic values are seen as optimistic as the used weight-hourly space velocity is below the experimental range, but potential trends in the analyses are considered to be independent of the exact kinetic values. After the decomposition reactor, the partially cracked ammonia is cooled (to *stream 4*) and compressed to the combustion chamber pressure at 30 bar (*Compression train*). Both the cooling after the reactor and interstage cooling between the compression stages can be deactivated to analyze their impact. The number of compressor stages is automatically calculated based on a maximum allowed compression ratio of 2.6. The process allows furthermore the inclusion of a bypass of the decomposition reactor (*stream 2*). The bypass corresponds to pumping the liquid ammonia directly to the combustion chamber pressure before evaporating it. The bypass can be seen as an additional degree of freedom for controlling the composition to the gas turbine.

Subsequently, the *Bypass* and the stream from the *Compression train* are mixed and fed to the combustion chamber of the gas turbine (*stream 5*). The mass flow of the *Air* to the combustion chamber is used for adjusting the temperature in the combustion chamber to 1160 °C. Electricity is subsequently generated using an expander model corresponding to a turbine. The exhaust gas contains significant amounts of NO_x which is removed by first adding ammonia (not shown in the Figure but modelled) to the exhaust gas and subsequent selective catalytic reduction (*SCR*) modelled as an equilibrium reactor. Heat recuperation from the exhaust gas is achieved both before and after the *SCR* reactor to allow for the required temperature in the *SCR* reactor.

As the temperature of the exhaust gas of the turbine may be too low to provide heat to the decomposition reaction, a potential *Furnace* is included. The fuel to the furnace (*stream 6*) is partially cracked ammonia at the same composition as the fuel to the combustion chamber while the air is a fraction of the exhaust gas. However, as the furnace is operating at a lower pressure, it utilizes the partially cracked ammonia before the compression to the pressure of the combustion chamber to avoid unnecessary energy demands. The flow to the furnace is controlled by pinch analysis and a specified minimum temperature approach ΔT_{min} of 25 °C. In the case of the detailed reactor model, we specify the required temperature of *stream 7* as input parameter while the furnace flow is calculated through a piecewise linear surrogate model. If the outlet temperature of the gas turbine exceeds the specified value, we use instead the outlet temperature directly. The hot liquid requires furthermore a ΔT_{min} of 12.5 °C for the heat transfer between the exhaust gas and the hot liquid. This assures the potential for designing an optimal heat exchanger network. Different approaches are chosen as the minimum temperature approach can occur at the reactor inlet in the case of the equilibrium reactor, while this is already included in the detailed reactor model.

Design parameters

The complete process has several important design parameters. These design parameters are explained in detail in the following sections.

Reactor pressure and temperatures

The reactor pressure has an impact on both the potential parasitic electricity requirements for the compression of the partially cracked ammonia and the equilibrium conversion of ammonia. Higher conversions can be achieved at low reactor pressures, but additional electricity for compression is required in this case. In addition, lower pressures result in larger equipment sizes. Similarly, higher reaction temperatures result in higher conversions, while they may require more combustion of partially cracked ammonia to achieve the temperatures as the temperature of the exhaust gas may be too low for providing the heat to the decomposition reaction.

The equilibrium reactor approach utilizes a constant temperature change in the reactor to approximate a counter current reactor while the table look-up method includes the temperature and relative flow of the heating medium as input parameters. The latter is implemented through the temperature of *stream 7* in Figure 1.

Cooling of the partially cracked ammonia and interstage cooling

The outlet stream from the ammonia decomposition reactor can be cooled to a desired temperature in *stream 4*. In addition, inter-stage cooling in the *compression train* can be applied or not. If the outlet stream is not cooled and no interstage cooling is applied, the efficiency of the compressors is reduced due to the increased temperature. The benefit of this approach is a simplified compressor design (albeit with potentially more expensive materials due to elevated temperatures) and a reduction in required fuel flow to the combustion chamber due to the increased temperature of the fuel.

Bypass of the decomposition reactor

The bypass of the decomposition reactor (*stream 2*) is an important factor for both the overall cracking ratio and the required compression energy demand. Introducing a bypass reduces the compression demand after the decomposition reactor and increases the conversion in the decomposition reactor due to a longer residence time for a given reactor size. In addition, it reduces the equipment size for compressors and heat exchangers However, care must be taken to avoid a significant reduction in overall cracking ratio to maintain a stable flame in the combustion chamber. The bypass does not affect the heat required for the cracking ratio for a constant overall cracking ratio.

Key performance indicators

The overall cracking ratio X_P provides information regarding the conversion of ammonia. It has an impact on the flame in the combustion chamber as well as the potential NO_x formation of the process. It is calculated through the conversion in the decomposition reactor X_R and the bypass ratio, $BR = \dot{m}_2/\dot{m}_1$. $\dot{m}_1$ corresponds here to the complete ammonia feed, including the bypass and the required ammonia for the SCR reaction, while $\dot{m}_2$ represents the flow through the *Bypass*. The stream numbers are also included in Figure 1. It is given by

$$X_P = BR \times X_R \tag{2}$$

The overall lower heating value efficiency η_{LHV} can be calculated as

$$\eta_{LHV} = \frac{P_{turb} - P_{para}}{\dot{m}_1 \times LHV_1} \tag{3}$$

where P_{turb} corresponds to the electricity generation in the *Turbine* and P_{para} to parasitic electricity demand for pumping and compression. It is closely related to the energy efficiency of the gas turbine η_{GT}, calculated as

$$\eta_{GT} = \frac{P_{turb} - P_{comp}}{\dot{m}_5 \times LHV_5} \tag{4}$$

with P_{comp} representing the electricity demand for the *Air compressor* while the denominator is based on the inlet flow to the combustion chamber (*stream 5*). The gas turbine efficiency is higher than the overall efficiency of the process. It can provide an overview of the losses incurred by the partial ammonia decomposition.

The ratio of feed stream to the furnace stream, $FR = \dot{m}_6/\dot{m}_1$, is another important key performance indicator. While it is indirectly included in the overall efficiency η_{LHV}, it is still important to understand what fraction of ammonia must be combusted to achieve the desired decomposition reactor temperature.

ANALYSIS

The analysis is split into two distinctive sections, an analysis of the impact of the reactor pressure on the performance of the process through the application of the equilibrium reactor and a subsequent in-depth analysis utilizing the detailed reactor model and a table lookup method for additional parameters. In both cases, one-dimensional sensitivity analyses are conducted. The reference parameters are provided in Table 1**.** Note that the heating fluid temperature is only relevant for the table lookup method.

Table 1: Reference parameters for the analysis.

Parameter	**Value**	**Unit**
Reactor inlet temperature T_3	400	°C
Reactor inlet pressure p_3	10	bar
Cooling temperature T_4	23	°C
Bypass ratio BP	0	
Heating fluid temperature T_7	455	°C

Equilibrium reactor

The equilibrium reactor model is first utilized for analyzing the impact of the reactor pressure as the impact of the reactor pressure is significantly affected by the chosen reactor size in the detailed reactor model. Interstage cooling is utilized to avoid high temperatures after the compression train affecting the results significantly. Instead, it is assumed that the compressed gas after the compression section is cooled to 100 °C.

Figure 2 shows the impact of the reactor inlet pressure on the performance of the integrated process. Due to the chemical equilibrium of the decomposition reaction, low pressures are beneficial for achieving a high ammonia conversion; the conversion is reduced from 79.5 % at a reactor pressure of 2 bar to 32.9 % at a reactor pressure of 15 bar. However, due to the increased conversion at lower pressures, it is necessary to have a higher furnace bypass ratio to provide sufficient high temperature heat for the decomposition reaction resulting in a similar behavior of both KPIs as shown in Figure 2 a). In the case of a high conversion, the minimum temperature approach is at the reactor inlet corresponding to an increased ΔT at the reactor outlet. While the turbine efficiency is largely unaffected by the reactor inlet pressure, we see large increases in the process efficiency from 36.7 % to 43.0 % when increasing the reactor inlet pressure from 2 bar to 15 bar.

The increase is especially pronounced in the range of 2 bar to 10 bar where the latter already results in 42.6 %. The efficiency increase of 17 % can be explained by 1) the reduction in the required furnace bypass ratio due to the reduced head demand in the decomposition reaction and 2) the reduced compression energy demand. The latter is especially important as the compression energy requirement at a reactor pressure of 2 bar corresponds to 7 % of the lower heating value of the partially cracked ammonia. This electricity demand is reduced to 1.9 % at a pressure of 10 bar and 1.2 % at a pressure of 15 bar.

Given these initial analyses utilizing the simplified equilibrium reactor model, detailed analyses are conducted utilizing a reactor pressure of 10 bar. This pressure can be seen as a beneficial compromise between the required conversion of ammonia for the gas turbine and the process efficiency.

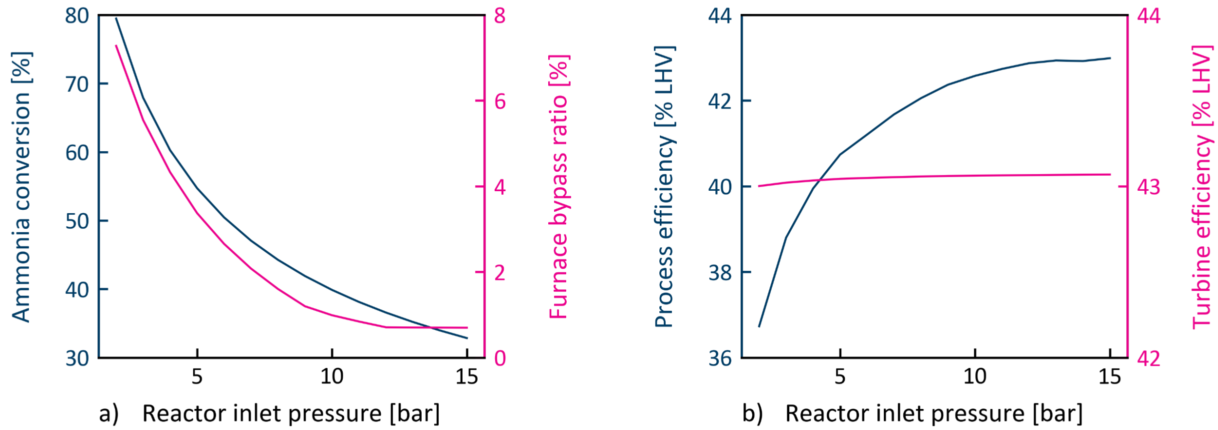

Figure 2. Results for the eqilibrium reactor model when varying the reactor inlet pressure. The results show a) (blue) the ammonia conversion, a) (pink) the furnace bypass ratio, b) (blue) the process efficiency, and b) (pink) the gas turbine efficiency.

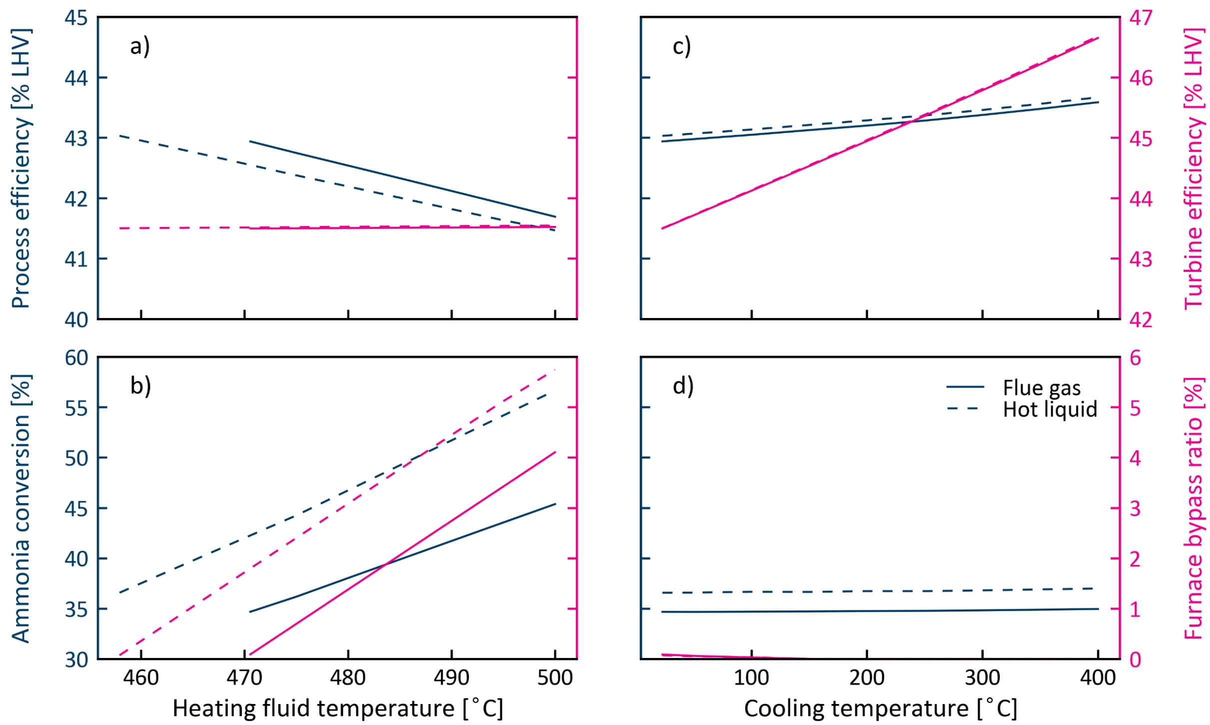

Figure 3. Results for the detailed reactor model when varying in a) and b) the heating fluid temperature or in c) and d) the cooling temperature when utilizing the exhaust gas (solid) or a hot liquid (dashed) as heat transfer medium. The results show a) and c) (blue) the process efficiency, a) and c) (pink) the gas turbine efficiency, b) and d) (blue) the ammonia conversion, and b) and d) (pink) the furnace bypass ratio.

Detailed reactor model

Figure 3 illustrates the results for varying the heating fluid temperature, a) and b), and the cooling temperature, c) and d) for both using the flue gas and the hot liquid for providing heat to the decomposition reactor. When investigating the impact of the heating fluid temperature, shown in Figure 3 a) and b), we see directly the difference of 12.5 °C for the minimum value used for the heating fluid temperature, representing the chosen ΔT_{min} when using the hot liquid. This difference leads to a shift of the complete analysis. Increasing the heating fluid temperature results in a linear decrease in the process efficiency while the turbine efficiency increases slightly. The linear decrease of the process efficiency is nearly independent of the chose heating fluid when considering

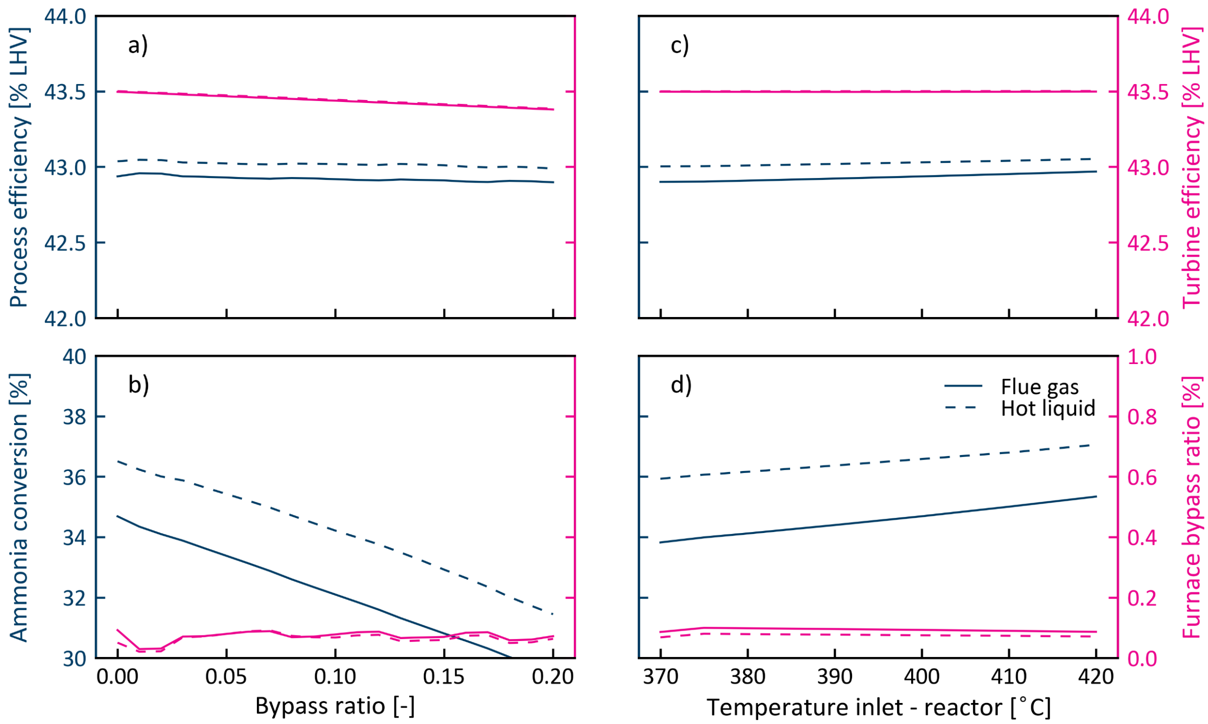

Figure 4. Results for the detailed reactor model when varying in a) and b) the heating bypass ratio or in c) and d) the reactor inlet temperature of the NH_3 feed when utilizing the exhaust gas (solid) or a hot liquid (dashed) as heat transfer medium. The results show a) and c) (blue) the process efficiency, a) and c) (pink) the gas turbine efficiency, b) and d) (blue) the ammonia conversion, and b) and d) (pink) the furnace bypass ratio.
Note the change in y-axis ticks compared to Figure 3.

the 12.5 °C temperature difference. When increasing the heating fluid temperature, the temperature of the exhaust gas from the gas turbine is not sufficient for providing the heat to the decomposition reactor. It is hence necessary to combust part of the partially cracked ammonia to reach the required temperature level. The observed increase in the cracking ratio is furthermore different between utilizing the exhaust gas directly or utilizing a hot liquid due to the differing heat transfer properties with a steeper slope when using the hot liquid.

The impact of the cooling temperature on the efficiencies is more pronounced as shown in Figure 3 c). Increasing the cooling temperature, *i.e.*, cooling less after the reactor, has a profound effect on both the turbine efficiency and the process efficiency, although the effect on the latter is smaller. The increase in the gas turbine efficiency can be explained by the chosen boundary conditions. As the temperature of the feed increases with increasing cooling temperature, less fuel must be provided to the combustion chamber for achieving the desired temperature level after the combustion chamber. The benefit for the process efficiency is, however, reduced as the increased temperature in the compressor results in an increased electricity demand of the compressor by 120 % when increasing the temperature before the compression from 23 °C to 400 °C. There is however still a significant net benefit of not cooling the partially cracked ammonia. On the other hand, the cooling temperature has next to no effect on both the ammonia conversion and the furnace bypass ratio as the reactor conditions remain unchanged, and hence, the conversion and required additional heat through the furnace bypass.

Figure 4 shows the results when varying the bypass ratio, a) and b) and the inlet temperature to the reactor, c) and d). The reactor bypass ratio has only a minor direct impact on both the gas turbine and the process efficiency as shown in Figure 4 a). Increasing the bypass ratio reduces both efficiencies, but the reduction is only 0.04 percentage points for the process efficiency and 0.1 percentage for the gas turbine efficiency. Both numbers are considered to be within the margin of error in the table lookup method. The slight change in the efficiency is on the one hand explainable by the reduced ammonia conversion as shown in Figure 4 b). Due to the bypass, we reduce the overall ammonia conversion. However, the reduction from 34.7% to 29.5 % in the case of using the exhaust gas directly and from 36.5 % to 31.4 % in the case of using an intermediate hot liquid does not reflect the actual loss as the conversion within the reactor is increased. If the conversion in the reactor remains the same, one would expect a conversion of 27.7 and 29.2 %, respectively. Hence, the conversion in the reactor is increased if the mass flow of the heating medium is increased compared to the mass flow of ammonia. As a consequence, using a bypass would allow for a smaller reactor size at the cost of a slightly reduced process efficiency. The furnace bypass ratio is in practice independent of the chosen bypass of the decomposition reactor. The observed non-linearities can be attributed to steps within the table lookup method resulting in less accurate simulations. The observed value of 0.1 % is also rather small.

Similar to the reactor bypass ratio, we do not observe a significant impact of the reactor inlet temperature on the efficiencies as shown in Figure 4 c). While the process efficiency experiences a small increase with a higher reactor inlet temperature, the turbine efficiency remains unaffected by this change despite the increased ammonia conversion as shown in Figure 4 d). The furnace bypass ratio is again small and unaffected by the changes, similar to the results shown in Figure 4 b) for the cooling temperature. The result can be explained by the chosen size for the reactor model. The reactor outlet temperature of the partially cracked ammonia is for the chosen size nearly independent of the reactor inlet temperature within the given range

DISCUSSIONS AND LIMITATIONS

Impact of the reactor model approach

On the one hand, the chosen reactor model and its representation has a significant impact on the identification of key operating parameters in the analysis. Specifically, utilizing an equilibrium reactor requires assumptions regarding the approach to equilibrium and the heat transfer within the reactor to approximate the temperature change ΔT in the reactor. Neither parameters can be directly deduced from kinetic studies. Especially the assumed ΔT in combination with the inlet temperature has a major effect as the equilibrium is calculated at the reactor outlet conditions. In addition, when analyzing the reactor bypass, we cannot observe the increased conversion in the reactor when using an equilibrium reactor.

On the other hand, it is not possible to utilize a kinetic reactor when designing a process due to the requirement of specifying a size for the reactor. As can be seen in the results for varying the inlet temperature to the reactor, the assumption of a constant size provides a suboptimal configuration in some of the values for the inlet temperature.

One improvement would hence be to operate iteratively between the detailed reactor model and the process model to adjust the size of the reactor model based on the analyses from the process model or alternatively use a complete surrogate model of the detail reactor model.

Cooling before compression

As shown in the analyses, with a required combustion chamber pressure of 30 bar, cooling before the compression chamber is disadvantageous. Although compression at high temperatures results in an increased electricity demand, it is compensated by the reduced feed flow rate for a constant electricity generation in the turbine. However, the feed gas to the combustion chamber has without cooling a temperature of 675 °C resulting in potential problems when compressing a mixture consisting of ammonia and hydrogen. When assuming cooling of the partial cracked ammonia post the reactor to 23 °C, the temperature is reduced to 175 °C without interstage cooling. While temperatures of 600 °C are observed for air compression, the corrosive nature of ammonia and potential hydrogen embrittlement can lead to problems. Consequently, it is important to consider the practical limits for the compression section.

Inclusion of a steam cycle for combined cycle operation

The current model does not include a steam cycle utilizing the still available exhaust heat for increased electricity generation. Due to the utilization of parts of the heat for the decomposition reaction, less heat is available for the steam cycle. Specifically, we observe a reduction in the temperature of the exhaust gas stream available for steam generation. Consequently, electricity generation in a potential steam cycle will be reduced compared to the standard process without ammonia decomposition. When not cooling the reactor before the compression steps, ammonia is heated using the exhaust flue gas from the reactor. This is especially a problem when choosing high reactor inlet temperatures for ammonia as the temperature exceeds in this case the exhaust of the heating medium from the reactor. This implies that the optimal process parameters may differ when including a steam cycle for an increased efficiency.

Dynamic flexibility of the process and operation at part-load

The overall process requires a constant heat supply to the decomposition reactor for maintaining a constant cracking ratio to the combustion chamber. As this is not necessarily feasible, introducing the bypass will allow a tight control of the composition to the combustion chamber. In this situation, operational flexibility can be achieved through sacrificing efficiency.

Part-load operation of gas turbines is in general achieved through a reduced pressure in the combustion chamber. As a consequence, the temperature of the exhaust gas is reduced. Hence, more partially cracked ammonia must be combusted to compensate for the reduced temperature of the exhaust gas. This implies that the efficiency penalty of the gas turbine at reduced load is furthermore exacerbated by the need for additional combustion of ammonia for achieving the required temperature in the decomposition reactor.

CONCLUSION AND FUTURE WORK

In this work, we provide an in-depth analysis of a coupled process for electricity generation from ammonia. The process, in which the ammonia decomposition reactor is heat-integrated with the gas turbine exhaust gas, allows for a high efficiency in the operation of the gas turbine. Lowering the required ammonia conversion results in improved overall performance due to a reduction in the required high temperature heat, and hence, the removal of combusting a fraction of the partially cracked ammonia for providing the heat to the decomposition reactor. Similarly, higher operating pressures for the reaction are beneficial to avoid the compression of the partially decomposed ammonia.

Future work will focus on part-load operation and analyzing the dynamic behavior of the complete process to propose potential control strategies given the increasing share of variable renewable power generation and the demand for adjustable dispatchable generators.

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