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Dynamic Simulation of a Novel Small-scale Power to Ammonia Concept

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ABSTRACT

Ammonia is a promising energy vector and storage means for hydrogen. Power to ammonia (P2A) processes employ renewable energy to split water to provide the hydrogen for the Haber-Bosch ammonia synthesis. The fluctuating nature of the renewables requires a good dynamic behavior of these cycles.

Employing the software Aspen Plus Dynamics®, this paper investigates the dynamic behavior of a novel containerized P2A solution, which is going to be tested at the University of Genova in 2023.

The simulation results of the start-up, various load changes and the shutdown of the process suggest that the control architecture can handle all cases in a satisfactory way.

However, there seems to be room for improvement regarding the parameters of some controls.

Keywords: Power to ammonia, hydrogen fixation, Aspen Plus Dynamics® simulations

NOMENCLATURE

Abbreviations	
FC	Flow control
FLEXnCONFU	Flexibilize combined cycle power plant through power-to-X solutions using non-conventional fuels
H_2	Hydrogen
HYSPR	Aspen HYSYS® Peng Robinson
N_2	Nitrogen
NH_3	Ammonia
P2A	Power to ammonia
PC	Pressure control
PID	Proportional integration differential controller
TC	Temperature control
Symbols	
$\Delta\overline{h}^{\circ}$	Molar standard reaction enthalpy (1 bar, 298 K) (kJ/mol)
p	Partial pressure (bar)
r	Rate of reaction (mol/($g_{Catalyst}$·s))
T	Temperature (K)

1. INTRODUCTION

The renewable energy sources are characterized by their variation in space and time [1]. Power storage systems are required to smooth out these fluctuations [2][3]. Ammonia is a promising long-time chemical storage medium and has the potential to become the carbon-free substitute for conventional fuels for multiple purposes: thermal engines, gas turbines and fuel cells [4][5][6][7][8][9].

This paper presents the simulated dynamic behavior of a novel P2A solution introduced in [10], which produces a maximum of 35 kg/d of ammonia at mild reaction conditions below 420 °C and 80 barg and with a close to zero carbon footprint, if electricity from renewable sources is being employed.

The containerized concept is designed for both a fast start-up behavior and low investment costs, thus aiming at harvesting the fluctuating renewables even in remote locations and at low investment costs, not least to lower the market entry barrier of ammonia as the dominant future fuel.

On contrast, traditional large-scale ammonia plants mostly employ methane as the feedstock for hydrogen and run continuously at higher conditions of 100 – 350 bar and 400 – 550 °C, with ammonia capacities of up to 3 000 t/d, emitting on average 1,33 t of carbon dioxide per ton of ammonia [11][12].

In recent years, several small-scale P2A solutions have been suggested [13] and tested [14][15][16]. However, mostly the dynamic behavior of large-scale traditional ammonia plants has been discussed and simulated in literature [17][18][19] and seldom that of small P2A plants [20].

This work aims to fill this gap by investigating the dynamic behavior of a novel small-scale P2A concept, employing the software Aspen Plus Dynamics®. The containerized solution of the concept, which was developed in the EU FLEXnCONFU project, is currently under construction and will be tested at the Savona Campus of the University of Genova, Italy, starting from 2023.

Specifically, this work investigates the start-up, load-changes and shutdown behavior of the cycle, trying to determine, first, if the control architecture is suitable, second, if it should and how it could be improved and third, which of the simulated scenarios, if any, could be a challenge for the real plant.

2. SYSTEM DESCRIPTION

The process diagram including controls is depicted in Fig. 1. The streams are numbered in capital roman letters from I to XII plus the hydrogen (H_2) and nitrogen (N_2) inlet streams. The H_2 is produced in the water electrolyzer and the N_2 is supplied by bottles. The N_2 is flow controlled (FC) to match the flow of H_2 from the electrolyzer. The H_2 and N_2 are mixed with the recycle stream XII to give stream I at 8 barg. The compressor increases the pressure to 80 barg of stream II. The compressor controls the pressure of stream II (PC 1).

Stream II is preheated to the required reactor inlet temperature. The electric preheater controls the temperature of stream III (TC 1). Stream III enters the first of the three reactor sections, where the gaseous exothermic ammonia (NH_3) equilibrium reaction, given in Eq. (1), takes place [12].

$$1{,}5\ H_2 + 0{,}5\ N_2 \rightleftharpoons NH_3\ \ ;\ \ \Delta\bar{h}^{\circ} = -46{,}22\frac{kJ}{mol_{NH_3}} \qquad (1)$$

Each reactor section is temperature-controlled by negative or positive heat streams, which are the result of a combination of electrical heating (for start-up), insulation, and air-cooling (TC 2, TC 3, and TC 4). The reactor is a fixed bed tube-in-tube reactor and is divided into three sections to ensure a good temperature control. The inner tube has an inner diameter of 10 cm and a length of 180 cm (60 cm each section) and it is filled with an iron-based catalyst [10]. Wrapped around the outside of the inner tube of each section are separate electric heating coils. Three different cooling air streams can flow through the outer tube of each section. Each section of the outer tube is insulated on the outside.

Stream VI with the produced NH_3 and the not fully converted H_2 and N_2 exits the third reactor section and enters the condenser, in which the mixture is cooled down to 15 °C and separated into a liquid stream VII of mainly NH_3 and a gaseous stream VIII of mainly H_2 and N_2. The condenser temperature (TC 5) and level (LC) are controlled by a negative heat stream provided by air-cooling and via stream VII. The liquid stream VII flows to the NH_3 storage vessel, not depicted in Fig. 1.

The gaseous stream VIII is split into stream IX entering the purge valve and stream XI entering the recycle valve. The purge valve controls the pressure of the condenser via the purge stream X (PC 2). The recycle valve controls the pressure in the recycle via the recycle stream XI (PC 3). In total, there are ten controls: One FC, one LC, three PC and five TC.

The innovativeness of the design primarily lies

1. in the anticipated fast and efficient electrical preheater and reactor temperature control and in the non-use of an internal gas-gas heat exchanger
2. and in the pressure control of the recycle via the recycle valve, which is normally ensured by a recycle compressor.

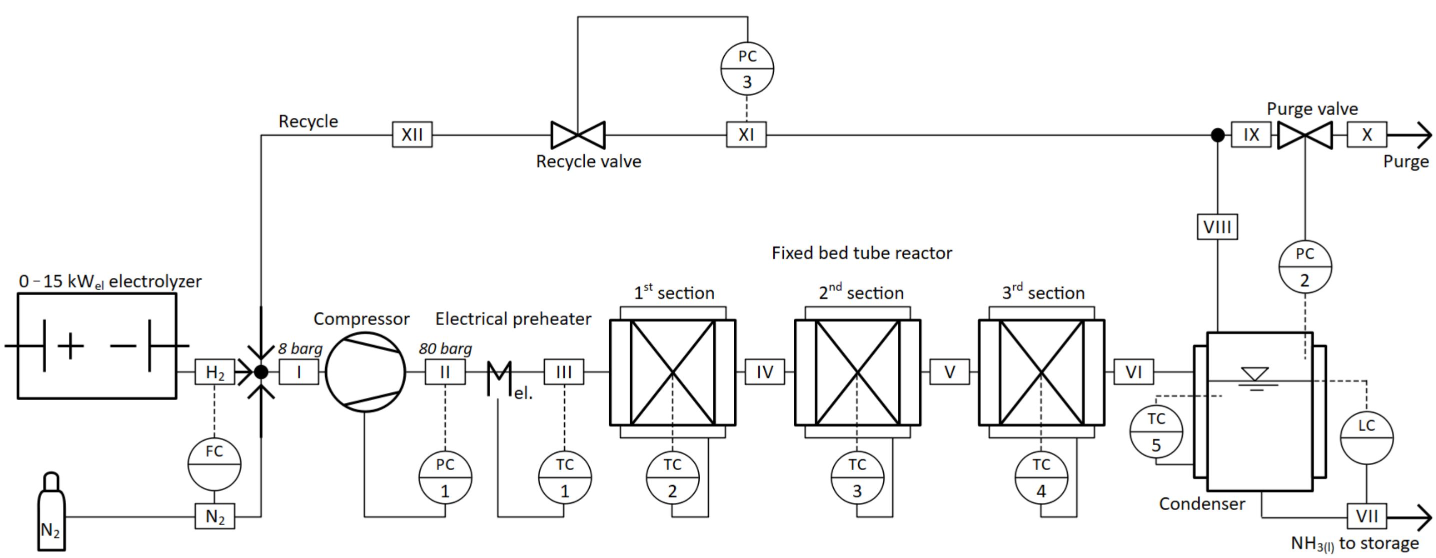

Fig. 1 Process and control diagram

Regarding 1., the fast temperature control tenders to the dynamic nature of the renewables. Most NH_3 production cycles rely on an internal heat exchanger, that transfers heat from the reactor outlet to the reactor inlet [14][17][18][19][20][21]. However, for a fast start-up, the internal heat exchanger brings no noticeable advantages, as first, the system is not heated up, yet, and as second, the internal heat exchanger may even cause instabilities [22].

Regarding 2., most NH_3 production cycles also rely on a recycle compressor [13][14][17][18][19][20][21] to maintain the high-pressure in the whole cycle. However, the recycle valve, just as the expansion valve used in household fridges instead of an expansion machine, reduces the investment cost of the system by saving the recycle compressor. This lowers the market entry barrier of the containerized system. Furthermore, the cycle is divided into a low- and high-pressure part. This is beneficial, as the low pressure of 8 barg in the first half of the cycle is in the typical outlet pressure range of electrolyzers and pressure swing adsorption applications [13], for H_2 and N_2 production respectively.

3. SIMULATION SET-UP

The cycle dynamics and control behavior is simulated with the Aspen Plus Dynamics® software. The chosen property method for the three components is HYSPR with modified parameters. The electrolyzer is modeled as a black box linear model with 0 – 15 kW_{el} input and 0 – 3 Nm^3H_2/h output with a simplified minimum ramp-up and downtime of 1 min/kW_{el}. The isentropic efficiency of the compressor is assumed to be 80 %.

The reactor is modeled as a plug flow reactor with a decreasing optimum temperature profile of 380, 380, 350, and 340 °C for TC 1 to TC 4 [10]. The reaction kinetics, valid for the range of 75 – 125 barg and 300 – 450 °C, is given in Eq. (2) and was fitted to test data provided by the catalyst supplier [10].

$$r = \frac{0{,}016\left(\exp\left(1{,}604 - \frac{10488\ \mathrm{K}}{T}\right) p_{H_2}^{1{,}5} p_{N_2} - \exp\left(29{,}194 - \frac{23235\ \mathrm{K}}{T}\right) p_{H_2}^{-1{,}5} p_{NH_3}^{2}\right)}{1 + 2p_{NH_3}} \tag{2}$$

For simplicity, the pressure drop in each unit operation, excluding the compressor, the recycle and purge valve, was set to 1 mbar. The simulation is set-up in Aspen Plus® and then converted to an Aspen Plus Dynamic® pressure driven simulation. The steady state solution of Aspen Plus® is taken as the starting point of the dynamic simulation. The controls are of type proportional integration differential (PID), are implemented successively and tuned via the integrated Ziegler-Nichols method. As the integration method for each time step for the coupled mass, momentum and energy balances, the implicit Euler method is chosen. The mixed Newton method solves the resulting non-linear set of equations for each time step. The suggested solver settings by Aspen Plus Dynamics® are kept, e.g. an absolute and relative tolerance of 0,0005 for the implicit Euler method.

For the start of every scenario, the purge stream X is set to a value of 0,5 mol% of stream VIII. In practice, purging will take place intermittently, to counter accumulating impurities, e.g. argon. However, this discontinuity cannot easily be simulated in Aspen Plus Dynamics®. Additionally, impurities are neglected in the simulation. At the start, the condenser is set to be half-full of liquid ammonia, i.e. a level of 0,15 m. The following scenarios, all starting from a steady state point of operation after 5 min, were simulated:

a) A start-up from 5 to 50 % electrolyzer capacity shall emulate the scenario of leaving a phase of dark doldrums. The realistic case of 0 % capacity at the start is not a valid steady state point that can be simulated in Aspen Plus®. The chosen 5 % capacity yields a stable simulation and is assumed to be close enough to 0 %. Alternatively, the P2A system is constantly kept at a minimum of 5 %, as is done in a similar way for some fuel cells that require a constant high-temperature. The P2A system would be kept running requiring the minimum amount of electricity whilst being even more ready for a fast start-up.
b) A gradual linear change from 50 up to 90 and back to 50 %, remaining there for 7,5 min and then down to 10 and finally up again to 50 % capacity shall emulate a combination of smooth changes in the renewables, starting from and returning to the point of a relatively moderate baseload.
c) A rapid change from 50 up to 60, followed by another rapid change down to 40, followed by a third rapid change back to 50 % capacity at 15 min, shall emulate rather sudden and big changes in the renewables, again, starting from and returning to a relatively moderate baseload.
d) Small but frequent fluctuations in the renewables, again assuming a relatively moderate occurrence of renewables, shall be emulated by a pseudo-random binary signal with a change every 30 s around 50 % between the upper and lower amplitude of 55 and 45 % capacity for the time period of 5 min and then returning to the 50 % capacity at 10 min.
e) A downward change from 50 to 5 % electrolyzer capacity shall emulate the shutdown of the cycle, using the same arguments raised as in scenario a).

These scenarios go beyond simulations in literature, with small disturbances e.g. a 5 % compressor power increase

[21], a feed increase of 5 Ma% [19] or a ramp-up from 50 to 100 % capacity [20].

4. RESULTS AND DISCUSSION

A selection of the resulting mass flows of the scenarios a) to e) are displayed in Fig. 2 to Fig. 6.

In Fig. 2, the H_2 and N_2 inlet mass flows, the NH_3 mass flows at the reactor compartment in- and outlets (III to VI) as well as the NH_3 mass flow leaving the cycle (VII) for scenario a) are depicted. It can be seen, that the NH_3 flows follow the inlet flows of H_2 and N_2. In addition, it can be seen, that in each compartment the amount of NH_3 increases (streams III to VI). This means, that the optimum decreasing reactor temperature profile for the steady sate from [10] also seems to be suitable for the dynamic start-up of the NH_3 production. However, streams III to VII display a sort of undulating behavior and do not possess such a smooth character as streams H_2 and N_2. Furthermore, streams III to VII display an overshooting and oscillating behavior after 12 min, the end of the start-up. However, the oscillating behavior ends at approximately 25 min and the new steady state is assumed to be reached, judging from the flat lines of the streams from 25 up to 60 min, as shown in Fig. 2. The undulating behavior as well as the over-and undershoots and the time it takes to reach a new steady state lead to believe, that the controls, e.g. the LC for stream VII, can be optimized to minimize these phenomena.

In Fig. 3, the same mass flows are depicted as in Fig. 2. Again, the NH_3 streams follow the inlet streams. Nevertheless, undulating behavior (e.g. 31 to 37 min), over- (37 min) and undershoots (17 min) and time lacks for reaching the new steady states (17,5 to 25 min) are visible in this scenario, too. Comparing the ramp-ups of 5 to 11 min and 31,5 to 37,5 min, there seems to be an undulating behavior only for the latter, i.e. for the ramp up from 10 to 50 % capacity. The same undulating behavior was also visible in Fig. 2, also for the ramp-up from a low capacity. During ramp-up, the mass in the system increases as well as the mass flow that is being recycled. The higher the recycle stream the more open the recycle valve. At low capacities, the recycle valve is almost closed, whereas at high capacities, the recycle valve is almost 100 % open. It seems, that the control parameters for the recycle valve PC 3 fit better, when the recycle valve is more open than closed. In addition and generally speaking, simulations tend to be more unstable, as they reach their boundaries, in this case the limit of a zero recycle stream. Furthermore, judging from the rather sudden up and down behavior of stream VII between 10 and 13 min, the control parameters of the LC should be optimized. Nevertheless, Fig. 3 shows, that the P2A cycle can handle great, but rather smooth, upward and downward inlet changes.

Compared to Fig. 3, in Fig. 4, the inlet changes of H_2 and N_2 are more rapid in terms of change in magnitude per time but less in terms of change in magnitude. Again, the NH_3 streams follow the inlet streams and the amount of NH_3 continues to increases along the reactor. However, the same undulating behavior, over- and undershoots as well as time lacks are visible in this scenario, too. Interestingly, between 12 and 15 min, the streams III to VII rise in an undulating manner, even though the inlet streams are constant for that time. This can only be explained by the aftermath of the sudden decrease in the inlet streams at 10 min.

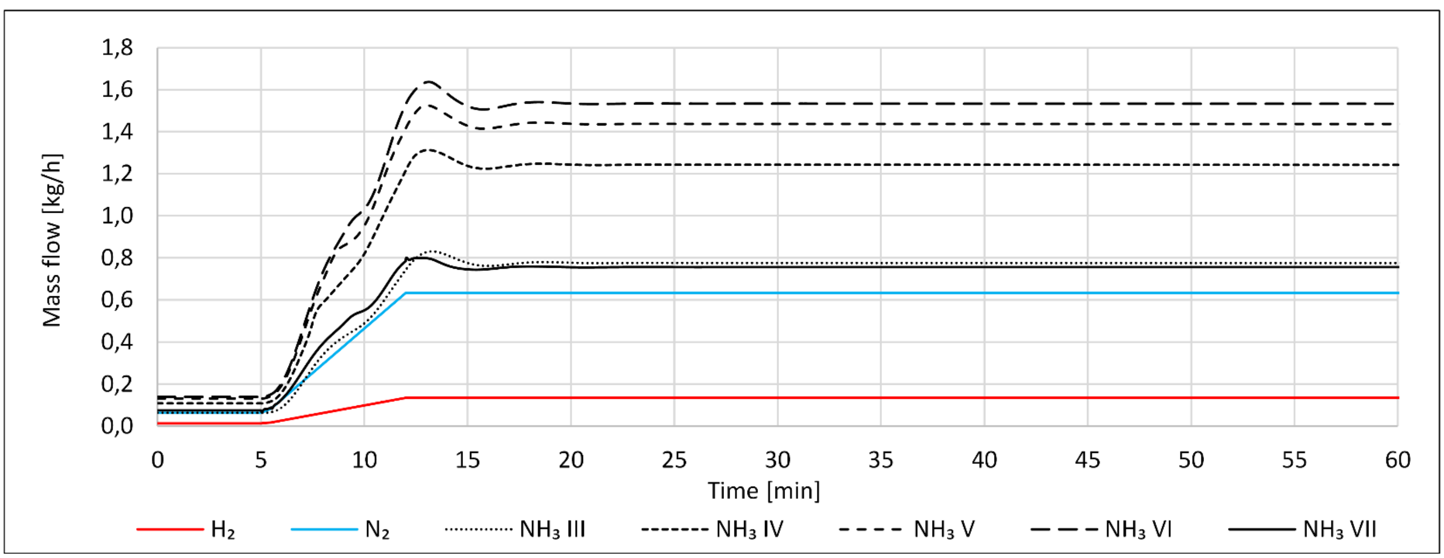

Fig. 2 Selected mass flows of scenario a)

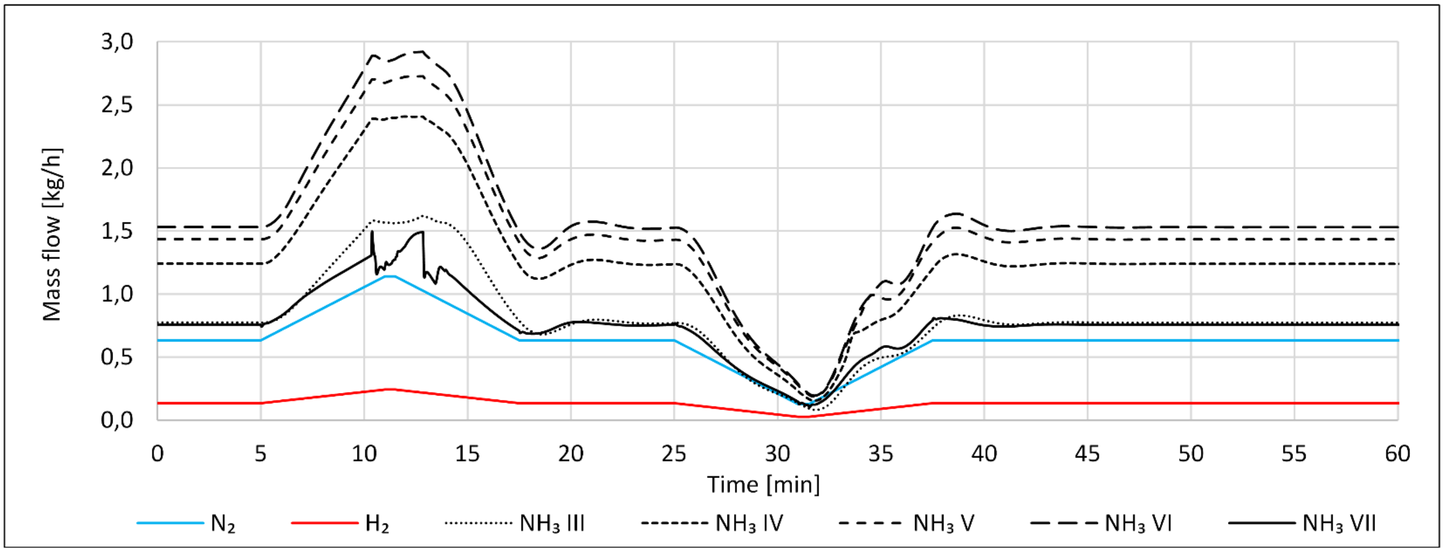

Fig. 3 Selected mass flows of scenario b)

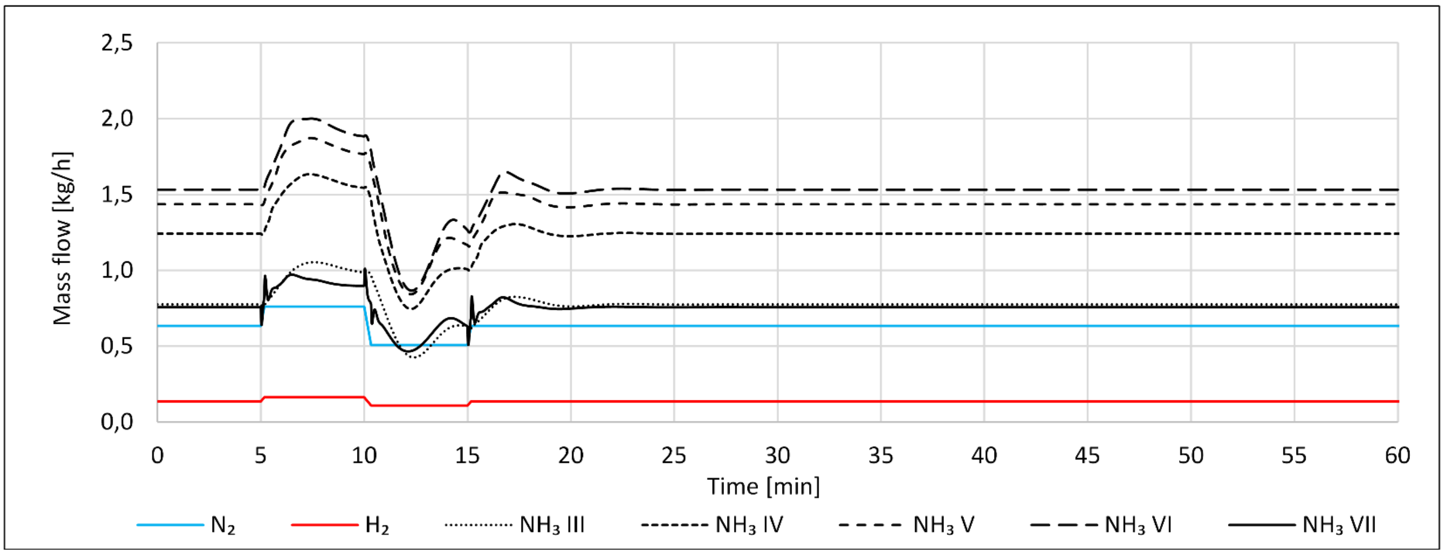

Fig. 4 Selected mass flows of scenario c)

The reaction time of the controls, especially the recycle valve PC 3 and the LC, seems to be too low, which is why an undershoot at 12 min seems to occur, but which is being revised between 12 and 15 min, resulting in the undulating rise. Furthermore, stream VII displays the greatest peaks, i.e. sudden and steep over- and undershoots, always at the changing points of the inlet streams, i.e. at 5, 10 and 15 min. This again points to the fact, that the LC control parameters are not optimal, yet. Nevertheless, Fig. 4 shows, that the P2A cycle can also handle more drastic upward and downward inlet changes.

Second to last, Fig. 5 shows, that the cycle can adequately manage small and frequent inlet changes, too, and, as in the previous cases, end in a stable steady state. However, again, stream VII displays the greatest peaks and lows whereas the other streams seem to be fluctuating more smoothly.

Finally, Fig. 6 displays the simulated shutdown of the P2A system. As in the prior scenarios, the NH_3 streams follow the decrease in the inlet streams. However, as the streams III to VII go down, a small undulating behavior can be seen. Furthermore, at 12 min, when the inlet streams have reached their minimum of 5 %, streams III to VII seem to undergo an undershoot, followed by an oscillating behavior, which only comes to an end after 35 min, when the new steady state is reached. In the shutdown of the real plant, the flows of stream III to VII would reach a value of zero and no oscillations would be visible. However, the oscillating behavior again suggests, that the parameters of at least the LC and PC 3 controls are not optimal with regards to a fast and efficient reaction time.

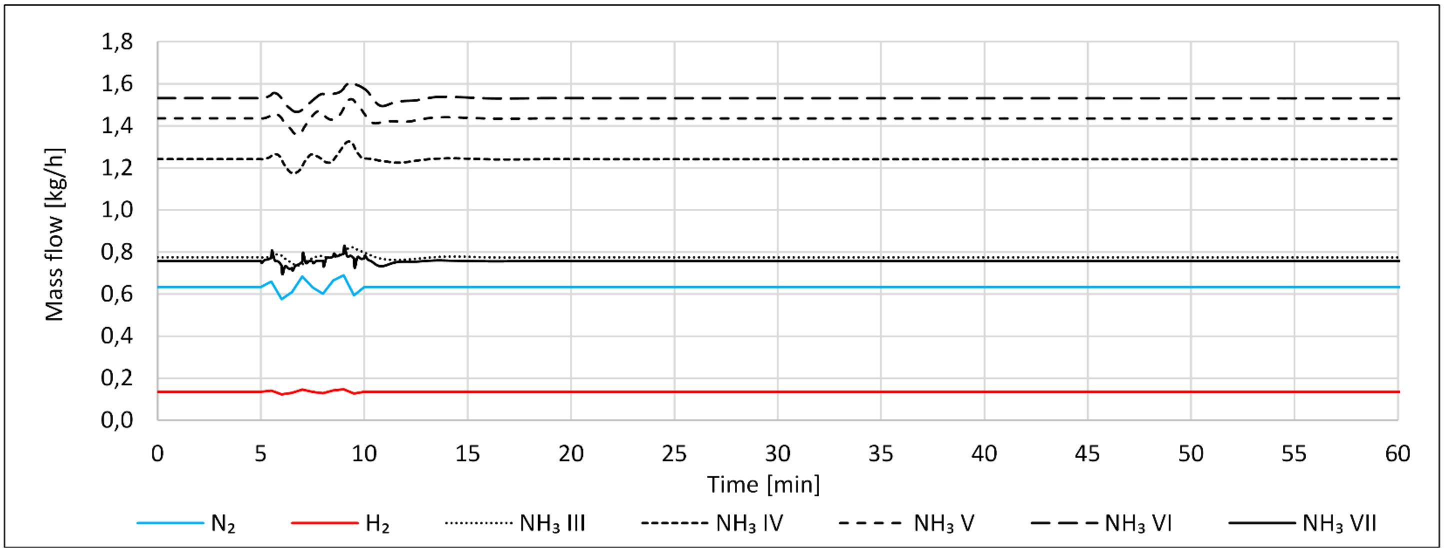

Fig. 5 Selected mass flows of scenario d)

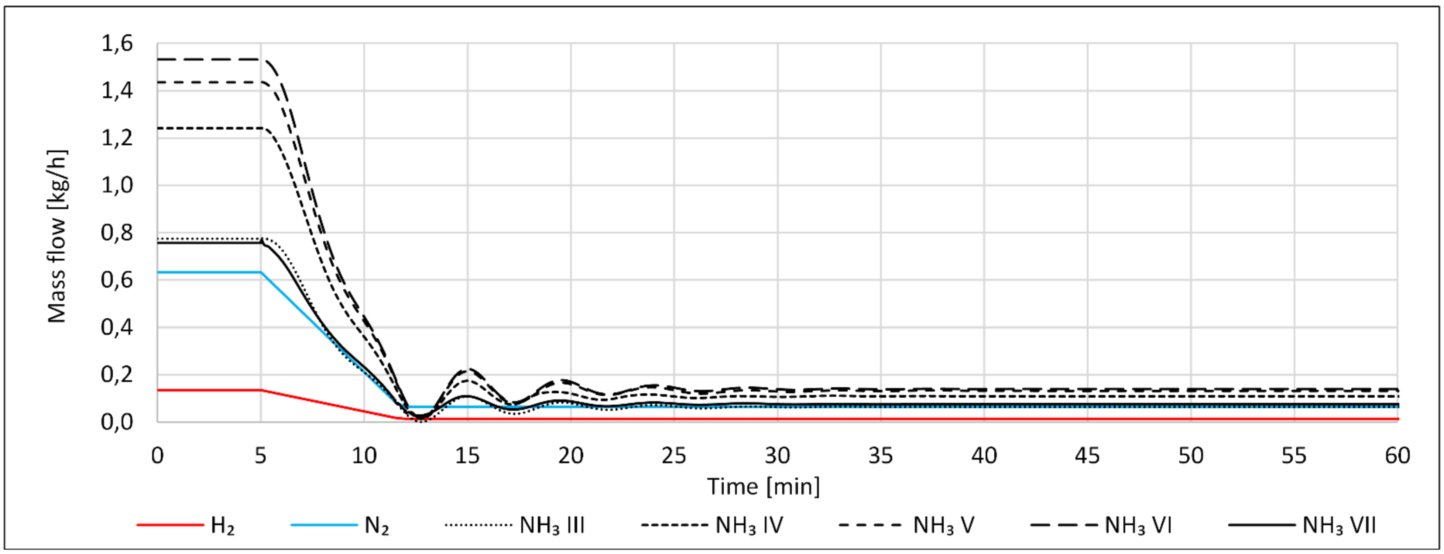

Fig. 6 Selected mass flows of scenario e)

Nevertheless, the overall trend of Fig. 6 is plausible, suggesting, that the control narrative of the P2A cycle is also suited for a shutdown of the system.

In all scenarios, the controls are able to keep the specified set points for pressure, temperature, etc. within reasonable ranges. The optimal inlet H_2/N_2 ratio of 2,96 from [10] is ensured by the FC. The electrical preheater and reactor temperature controls TC 1, TC 2, TC 3, and TC 4 ensure the optimum steady state temperature profile of 380, 380, 350, and 340 °C from [10]. In addition, the desired cycle pressures as well as the condenser temperature and liquid level are ensured by PC 1 to PC 3, TC 5, and LC.

Exemplary for scenario b), selected pressure and temperature values are depicted in Fig. 7 and Fig. 8 as well as the liquid level in the condenser in Fig. 9. As can be seen in Fig. 8 and Fig. 9, the temperature and level values remain relatively constant, pointing to the fact, that the TC controls and the LC control seem to work in a satisfying way. However, as can be seen in Fig. 7, some of the pressure values, i.e. for streams VIII and XI, PC 2 and PC 3, fluctuate to a greater extend. However, it can be seen in Fig. 7, that PC 2 and PC 3 seem to work quite well in unison, as the pressure values of streams VIII and XI are more or less equal, i.e. the dotted line VIII is inside line XI. On contrast, PC 1, line II, does not display fluctuations to such an extend and seems to work better.

Moreover, the fact that the pressure values of streams VIII and XI are above the pressure value of stream II in some short instances, e.g. at 13, 33 and 37 min, seems physically impossible. It can be explained, though, by an insufficient cooling in the condenser, leading to an evaporation in the condenser, which increases the pressure in stream VIII and XI for a short while. The resulting temperature fluctuations in the condenser are not visible in Fig. 8, as the scale of the vertical axis in Fig. 8 is not suited for such small changes. For this reason, the temperature value of stream VII, being the temperature of the condenser, is displayed again in Fig. 10, but in a close up, which makes the temperature fluctuations more visible. Especially at 13 but also at 33 and 37 min, the temperature peaks are high, explaining the pressure peaks for streams VIII and XI discussed for Fig. 7.

As a subsidiary result of comparing the temperature values of stream VII in Fig. 8 and Fig. 10, it can be argued, that the control parameters of TC 5 can be improved as well. Going further, one may presume, that also the other temperature controls can be improved. Nevertheless, the overall trends in Fig. 7 to Fig. 9 suggest, that the control architecture of the cycle can maintain the specified set points for pressure, temperature, etc. within acceptable margins.

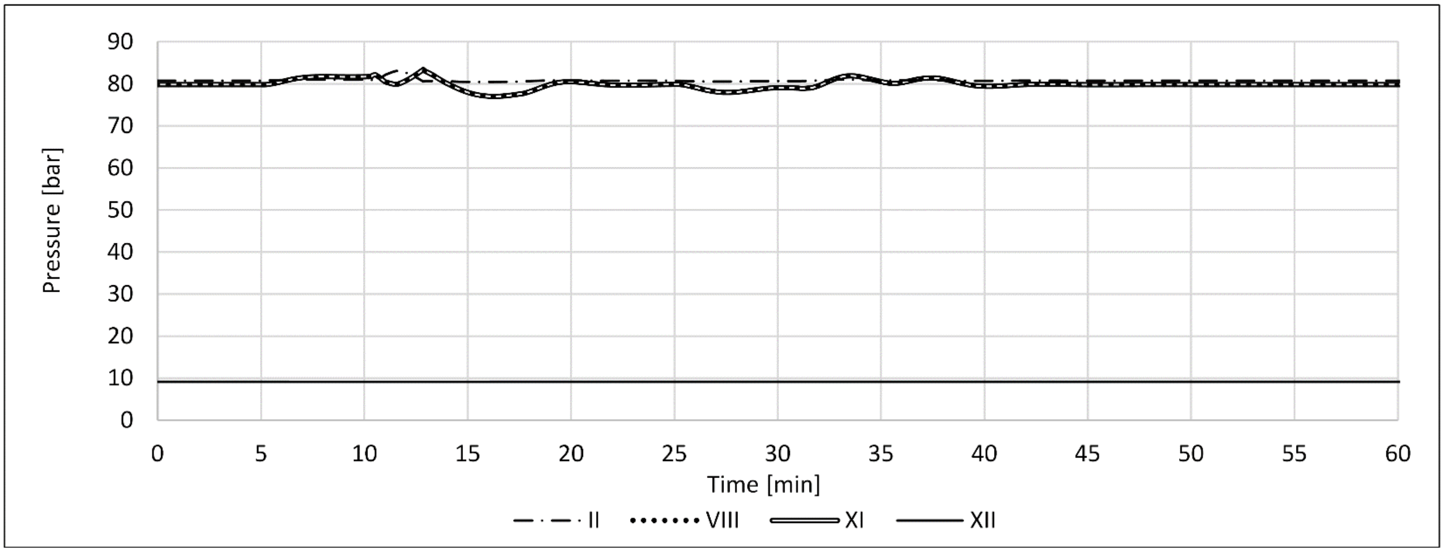

Fig. 7 Selected pressures of scenario b)

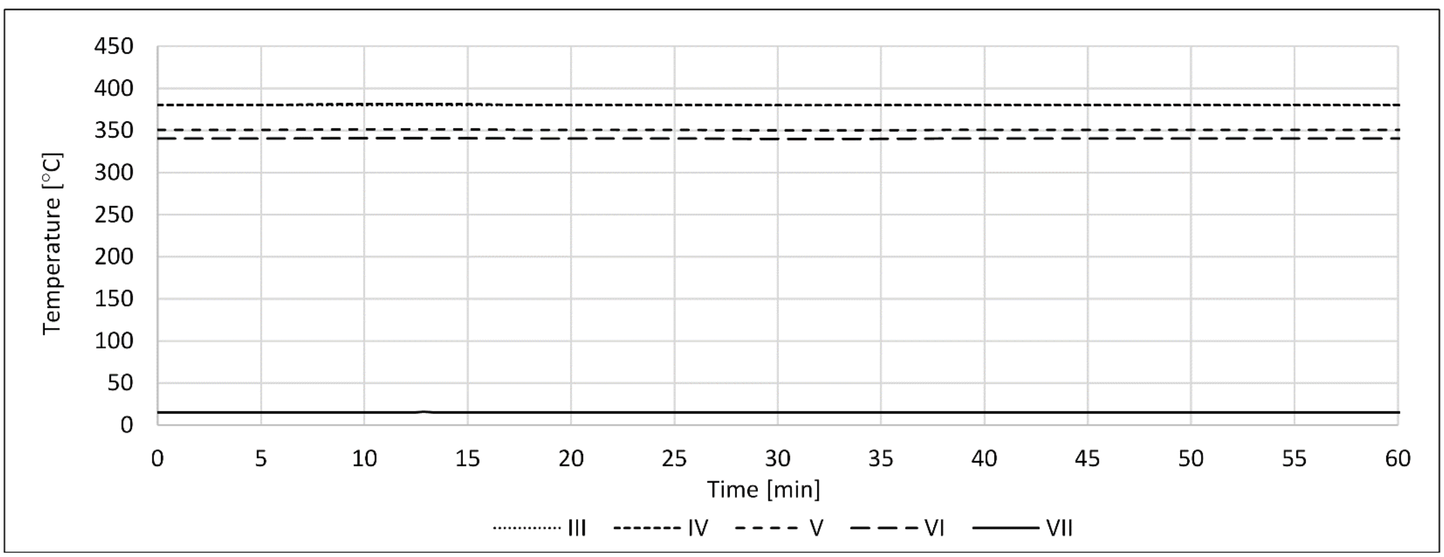

Fig. 8 Selected temperatures of scenario b)

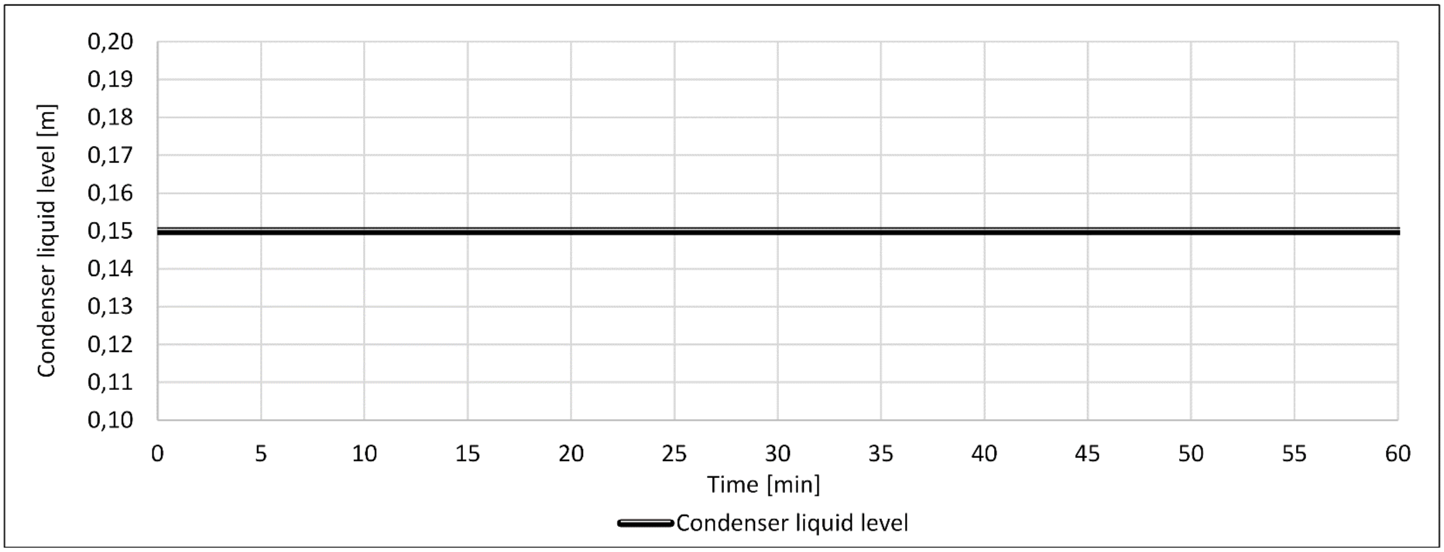

Fig. 9 Condenser liquid level of scenario b)

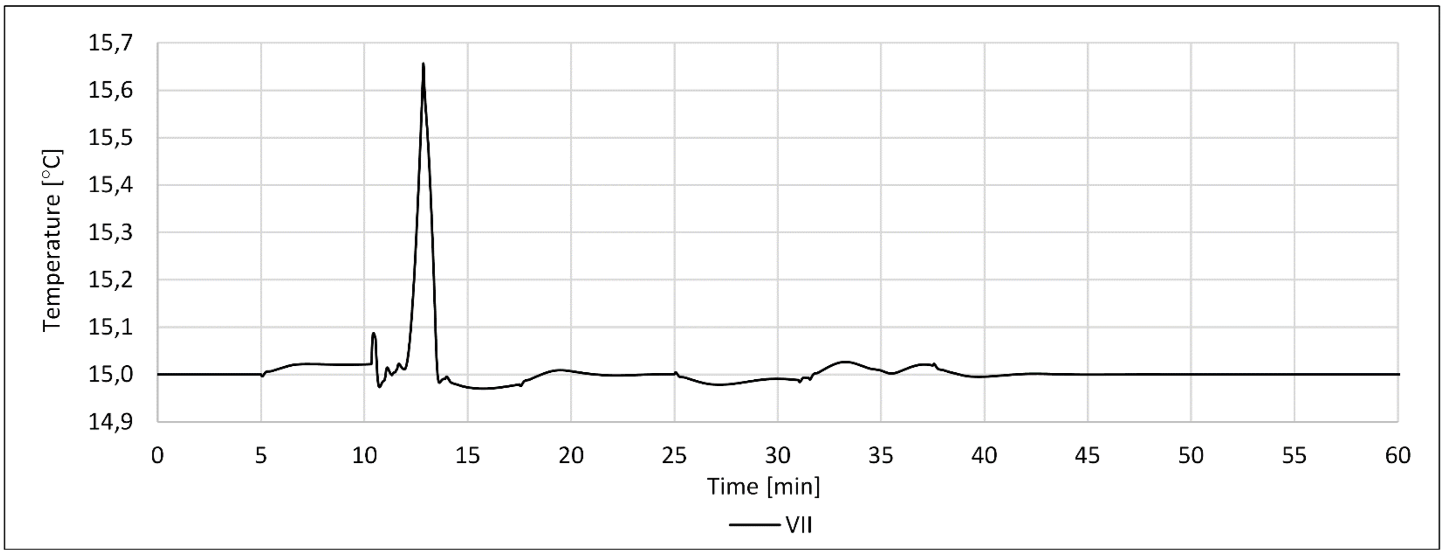

Fig. 10 Condenser temperature of scenario b)

Summing up, the simulation results suggest, that the proposed P2A cycle and its control architecture can handle a start-up scenario, large and small, smooth and sudden load changes as well as a shutdown event and, in each case, bring the system to a stable steady state. However, visible peaks, over- and undershoots as well as oscillating and undulating behavior in the displayed curves suggest, that the controls can be tuned further. In particular, but not exclusively, judging from scenario b), the parameters of the recycle valve PC 3, the purge PC 2, the condenser level LC and temperature TC 5 controls should be improved.

However, a simultaneous tuning of controls is not possible in Aspen Plus Dynamics®. One way forward would be an iterative successive tuning of all controls until the displayed unwanted behaviors are diminished to an acceptable level. In addition, another tuning rule than the chosen Ziegler-Nichols method could be tried. The problem of an iterative procedure could be, and was encountered partially when setting up the controls and trying to optimize them, that the optimization is going in circles, as always the same changes of one control lead to the same changes in the other controls and so forth. A way to counter this would be a change in the order of controls that are being tuned. Alternatively, in particular for the closely connected pressure controls PC 2 and PC 3, other control structures available in Aspen Plus Dynamics® could be explored, e.g. a cascaded control, where PC 3 is the dominant control loop for the high-pressure section and PC 2 is only being activated, when PC 3 reaches its limits.

5. CONCLUSIONS

The results of the dynamic simulations suggest, that the proposed P2A cycle and its control architecture can handle all simulated cases, i.e. a start-up, various load changes and a shutdown scenario.

Nevertheless, unwanted behaviors such as oscillations occur, which lead to believe, that the parameters of the different cycle controls should be optimized in order to minimize these unwanted behaviors.

Regarding the optimization, three ways forward, an iterative approach, using another tuning rule and a cascaded control approach are suggested.

The experimental data coming from the test campaign in 2023 will be used, first, to validate the dynamic model and second, to improve the control parameters of the model. Moreover, model adaptions will take place if necessary.

The thus developed and tuned dynamic P2A model and modeling approach can be used, not only to predict the behavior of this cycle, but also in the design of similar small-scale P2A processes.

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