Optimal flexible operation of an AICR for P2A

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Abstract.  
In this paper, we propose a strategy for optimal and safe operation of an adiabatic indirect cooled reactor (AICR) for Power-to-Ammonia (P2A). The intermittent nature of renewable energies requires P2A plants to operate over a wide operating window between 30% to 130% of the nominal load. We formulate a rigorous transient model of a three-bed AICR in an ammonia synthesis loop. The AICR operating conditions are optimized over the entire operating window. We set up a simple MIMO control structure for flexible operation of the AICR. This uses decentralised PI loops for regulatory control with setpoints from a supervisory optimisation over the operational window. The control structure is tested under extreme load variations and displays good performance.

Keywords: P2A, modelling, flexible operation, control.

* 1. Introduction  
     Power-to-X (P2X) technologies receive widespread recognition as one of the fundamental pillars of a future CO2-neutral society based on renewable energy sources. Of the P2X technologies especially green ammonia from power-to-ammonia (P2A) is perceived as one of the most promising species for energy storage, and decarbonizing fertilizer production and maritime transportation. The intermittent nature of renewable energy sources requires P2A plants to operate over a wide operating window between 30% to 130% of the nominal load. The traditional production of ammonia via the Haber Bosch process relies on a stable supply of reactants (nitrogen and hydrogen). Therefore, several papers describe optimal steady state operation of ammonia reactors (Khademi & Sabbaghi, 2017; Shamiri & Aliabadi, 2021). Few papers describe the transient behaviour of ammonia reactors, but dynamic simulations of an Adiabatic Qunch Cooled Reactor (AQCR) are presented in Morud & Skogestad (1998). In Rosbo et al. (2023b) we present a dynamic model of an AQCR and propose an optimal operating strategy specifically for P2A with a simple regulatory control structure. However, today modern ammonia plants are equipped with AICRs, which yield a higher reactor conversion but also a more complicated design compared with the AQCR. This paper aims to identify the optimal operation strategy for an AICR over the entire P2A operational window. Moreover, we aim to establish a robust control structure capable of regulating the reactor safely between optimal operating points under varying loads from renewable sources.
  2. AICR model and the case study   
     Figure 1 shows a schematic illustration of the ammonia synthesis loop considered in this work. The ammonia is produced via the Haber-Bosch process in an AICR. A relatively large recycle (stream 3) is used as the single-pass conversion of the reactor is around 30% due to equilibrium limitations. A fraction of the recycle is purged (Stream 8) to avoid accumulating inert gases (argon).

A diagram of a process

Description automatically generated

Figure 1: Synthesis loop of a P2A plant with an adiabatic indirect cooled reactor (grey box), compressors (orange boxes), and separator (blue box). The AICR is equipped with two internal heat exhangers (iHex 1 and iHex 2) and three valves for feed split. The AICR feed is pre-heated over the external feed-effluent heat exchanger eHex1. The streams are numbered from 1 to 12.

* + 1. AICR model   
       The governing equations of the units in the AICR system (catalytic fixed beds and heat exchangers) are presented in Rosbo et al. (2024). In this paper, we add a dynamic term to the heat exchanger model only described with static equations in Rosbo et al. (2024),

Where and are respectively the cold and hot side heat exchanger outlet temperatures, and and are the corresponding steady state temperature found via an effectiveness -model (Saari, 2011). is the heat exchanger time constant based on the thermal inertia of the heat exchanger.

* + 1. The case study: 100 MW P2A plant

The case study is based on the P2A case presented in Rosbo et al. (2024) defining a plant connected to a 250 MW renewable energy source with a capacity factor (CF) of 0.4. This corresponds to an average power supply of 100 MW to the P2A plant. The ammonia reactor volume is dimensioned based on 120% of the average power input to accommodate operation at periods with a power supply above the average load.

Tabel 1: Dimensions of the AICR beds, heat exchangers and nominal feed flow and compositions.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Bed dimensions** | | **Nominal reactor flow** | | **Heat exchangers** | |
| Porosity | 0.33 | Total flow, | 5846 | , | 89.0 |
| Volume, | 6.63 |  | 23.8 % | , | 66.4 |
| bed 1, | 0.998 |  | 71.4 % | , | 120 |
| bed 2, | 2.13 |  | 4.15 % |  | 90 |
| bed 3, | 3.50 |  | 0.60 % |  |  |

Rosbo et al. (2024) describe dimensioning of the heat exchangers to accommodate flexible operation. Table 1 summarizes the dimensions of the beds, heat exchangers, and nominal reactor flow for the AICR. Rosbo et al. (2023a) found that 91% of the total power input is consumed for the hydrogen production. Thus, the reactor load (RL) is defined based on the hydrogen flow to the reactor,

where is the reactor feed flow of hydrogen and the nominal flow. The reactor is operated at stoichiometric conditions, . The argon flow in the reactor feed stream is assumed constant over the operating window as a larger recycle ratio yields better loop efficiency at lower load (Rosbo et al., 2023a). Assuming the separator is at constant temperature and equilibrium conditions, the mole fraction of ammonia exiting the flash drum and entering the reactor is constant,

* 1. Steady state optimal operation
     1. Optimisation algorithm

The optimal AICR configuration maximises reactor conversion. We consider the bed inlet temperatures as unbound optimisation variables. The optimization problem is formulated as a constrained minimisation in Eq. 4-5,

In which is the bed inlet temperatures. The functions and refers to the formulation of the bed model in Rosbo et al. (2023b) as a differential algebraic equations system where contains the states, contains the algebraic variables, represents the balance equations and is the algebraic equations. Eq. 4-5 is solved at nominal load via Matlab's optimiser fminsearch yielding,

Figure 2a displays the temperature and conversion profile along the reactor volume for the optimized AICR at nominal load. The AQCR data from Rosbo et al. (2023b) is added to the graphs to illustrate the conversion advantage of the AICR at around 10 % higher relative conversion. Higher conversion is achieved in the AICR as all the reacting gas is passing through all the beds and the bed inlet temperatures can be optimised independently. Contrary, the AQCR only have two degrees of freedom for manipulating the bed inlet temperatures (Rosbo et al., 2023a), Figure 2b shows the graphs for reactor conversion versus temperature along with contours of reaction rate. The equilibrium line clearly illustrates how the multi-bed reactor design facilitates further conversion by inter-bed cooling. We observe that the conversion versus temperature curve for the AICR is generally located closer to the maximum reaction rate curve compared to the curve for the AQCR.

  
 (a) (b)  
Figure 2: a) Temperature and conversion profiles along the reactor volume for the AQCR and AICR. b) Reactor conversion versus temperature curves plotted with contours of reaction rate.

* + 1. Optimal static operation over the P2A operating window

We optimize the AICR by solving Eq. 4-5 over the entire operating window from 30% to 130% of nominal load. Figure 3a) shows the reactor conversion and Figure 3b) the optimal bed inlet temperatures over the operating window. The maximum reactor conversion is higher at lower loads as relatively more catalyst mass is available per reactant gas flow (higher residence time). From Figure 2b, we observe that decreasing the bed inlet temperatures creates more room for reactor conversion reaching before the equilibrium curve. However, this can only be realized if the bed residence time is sufficient to balance the decreased reaction rate at lower temperatures. Thus, the optimal bed inlet temperatures are colder at lower loads (longer residence time) as seen in Figure 3b.

  
 (a) (b)  
Figure 3: a) Reactor conversion for the optimized reactor, and b) optimal bed inlet temperatures over the operating window.

* 1. Control of the AICR under variable load operation   
     The optimisations in Sec. 3.2 compose a supervisory control layer for the optimal setpoints over the P2A operational window. A regulatory control structure is proposed for regulating the AICR safe and efficiently between the optimal static operating points.
     1. Control structure   
        Returning to Figure 1, we observe four obvious valves for manipulating the bed inlet temperatures: The three internal reactor valves and the bypass over the external heat exchanger. We neglect the fast valve dynamics and regard the reactor feed split fractions as the manipulated variables (MVs). Thus, the degrees of freedom are reduced by one as the feed split fractions sum to 1, . To confine the case to control of the reactor system, we assume the reactor feed temperature can be controlled instantaneously by the reactor bypass valve over the external heat exchanger. This allows us to regard the reactor feed temperature as a manipulated variable. These manipulated variables are naturally strongly crosscoupled to the bed inlet temperatures (controlled variables). Thus, we employ a static decoupler to compensate for cross effects. The manipulated variables, , is given from the controller outputs, , through,

Where is a static decoupler matrix. Table 2 summarizes controlled variables (CV), controller outputs and controller parameters for the control loops. The controller parameters are determined via the SIMC tuning rules proposed by Skogestad (2004).We specify control loop 1 as a pure integral controller with a very fast time constant for the desired closed loop response, . This is advantages as has a pure gain influence on the bed 1 inlet temperature, which facilitates very aggressive integral control. Consequently, we expect the bed 1 inlet temperature to be tightly controlled, while the bed 2 and 3 inlet temperature display a more conservatively controlled response towards the set points.

Tabel 2: Control loops and parameters.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
|  | CV |  |  |  |  |
| Control loop 1 |  |  | 2 s | 0 | 0.45 s |
| Control loop 2 |  |  | 90 s |  | 120 s |
| Control loop 3 |  |  | 90 s |  | 90 s |

4.2 Closed loop reactor operation   
The controller performance is tested by simulating the closed loop response to a series of step changes in the reactor load. The supervisory control layer specifies the optimal set points for bed inlet temperatures given the reactor load.

  
Figure 4: Closed loop response of the AICR system to extreme changes in reactor load.

Figure 4 displays the closed loop response of the AICR system. The four load steps of 25-30 % over only 2 hours are both larger and more frequent than what would be experienced in a real P2A plant. Thus, this provides an extreme test case for the controllers over the entire operating window. The proposed decoupled control structure performs quite well at stabilizing the reactor and tracking the optimal operating temperatures. As intended for the controller design, the bed 1 inlet temperature is controlled tightly and reaches the set point fast. The inlet temperatures to bed 2 and 3 respond slower as intended and display a efficiently controlled path towards the optimal bed inlet temperatures. The decoupling strategy appears to work well as no significant crosscoupling interactions are observed in the closed loop response.

* 1. Conclusion  
     In this paper, we have identified the optimal operating conditions of an AICR over an operating window between 30% to 130% of nominal load relevant for P2A. A MIMO control structure with centralized PI control was proposed for safely regulating the AICR between optimal operating points under varying load. We tested the control structure for a case with extreme and frequent variations in load. Even for this demanding case, the control structure performed well at regulating the reactor between the optimal operating points.

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