

## DYNAMIC BEHAVIOUR OF A NOVEL SMALL-SCALE HYDROGEN REFORMER

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**Abstract:** This paper explores dynamics for control design for a novel small-scale hydrogen reformer based on compacted plated heat exchanger technology. Reforming is seen as an important step towards the use of renewable fuels as feedstock. For reforming to be competitive to electrolysis and trucked-in hydrogen, high operability and robustness is required. This is achieved through an understanding of the system dynamics and robust control structure design. The paper identifies important dynamic features of the reformer unit and focuses on the responses in the hydrogen production rate to changes in several candidate inputs. This work was part of EC project Hydrofueler. *Copyright © 2007 IFAC*

**Keywords:** Dynamic behaviour, gas, conversion, hydrogen, reforming.

### 1. INTRODUCTION

In this paper the dynamic behaviour of a novel small-scale hydrogen reformer has been studied. The reformer converts hydrocarbons to hydrogen and is based on new compact plated heat exchanger technology. The paper identifies important dynamic features of the reformer unit.

This work was part of the EC project, Hydrofueler. The aim of the project was to design a new hydrogen refuelling unit suitable for about 50 cars, being smaller and more compact compared to conventional technology, and to carry out necessary development of the individual stages. Reforming is seen as an important step towards the use of renewable fuels as feedstock. For reforming to be competitive to electrolysis and trucked-in hydrogen, high operability and robustness is required. This is achieved through an understanding of the system dynamics and a robust control structure design.

The main focus of this study is the dynamic responses in the hydrogen production to changes in system inputs, like the steam-to-carbon (S/C), oxygen-to-carbon (O/C) ratios and feed temperature. Steam and oxygen (air) are important independent manipulated variables and essential in the hydrocarbon conversion.

The feed temperature is an important energy input. This type of analysis, performed at an early stage, gives valuable information to control structure design as well as to further process design. The main control objective of such a process is to produce pure hydrogen at minimum cost. Reformer temperatures and flow rates must be kept at a safe level such that the reformer catalyst is not damaged. The basis for the presented analysis is mathematical models based on first principles developed in Matlab/Simulink.

The paper is organized as follows. Section 2 describes the Hydrofueler production plant, relevant control issues and the simulation model. Section 3 shows the steady state and dynamic responses to perturbations of the inputs. Section 4 contains an analysis of the simulation results and an interpretation in terms of possible control strategies. The last section contains conclusions and suggestions for further work.

### 2. SYSTEM DESCRIPTION

#### *2.1 Production plant*

The Hydrofueler production plant consists of reformer and separation units, compressor, storage tank and dispenser. The units are designed to operate

at approximately 20 bar and in the temperature range 250-800°C. Storage and dispenser units were not considered as part of the project. An overview of the Hydrofueller concept is given in Figure 1.

The Hydrofueller reformer includes both ATR (autothermal reforming) and SMR (steam methane reforming) in a novel heat integrated design, based on heat exchanger technology. ATR and SMR are among the most common technologies for converting hydrocarbons (methane) to hydrogen.

Figure 2 shows the process flow. Water and natural gas are fed to the purification unit, removing sulphur and other impurities from the feed gas. Higher order hydrocarbons are converted in the pre-reformer and at the ATR side of the reformer. The product from the pre-reformer is fed to the SMR side of the reformer. Water, air and natural gas are fed to the ATR side of the reformer. SMR also occurs at the ATR side and water gas shift (WGS) reactions occur in all reformer steps. The main conversions are given by:

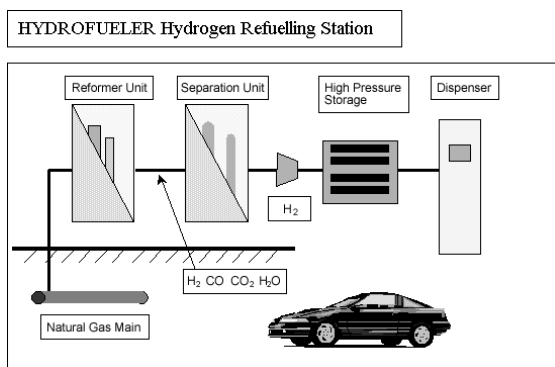
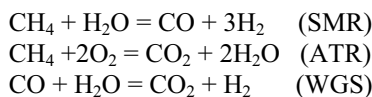


Fig. 1. The Hydrofueller concept.

### 2.2 Control issues

The main control objective is to keep the hydrogen production rate at a set point given by the production demand. There are two sub-objectives for the control; (1) set point tracking and (2) disturbance rejection.

In order for the control system to handle variations in the input disturbances at various operating points, knowledge of the response of the hydrogen production to changes in input variables is crucial.

Candidate manipulated variables in this process are:

- The steam-to-carbon-ratios (S/C) to the ATR and SMR side of the reformer.
- The oxygen-to-carbon ratio (O/C) at the ATR side of the reformer.
- The feed temperatures.
- Natural gas feed streams.

The ATR process provides the major part of the energy required for the system. Hence, the following analysis focuses on the steam feed flow (described by the S/C ratio), the oxygen feed flow (described by the O/C ratio) and the feed temperature at the ATR side as manipulated variables. Variations in the natural gas feed streams have not been considered in this study.

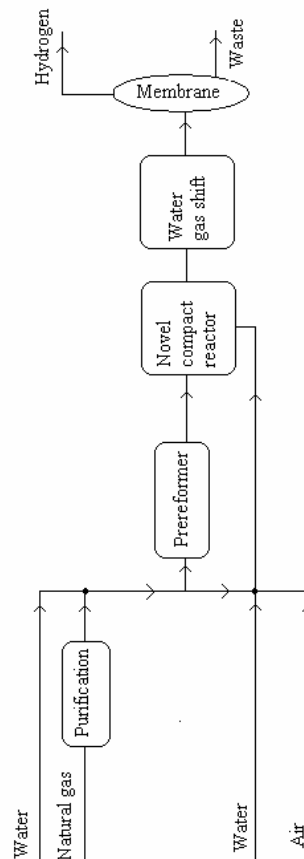


Fig. 2. Overview of the process flow.

### 2.3 Simulation model

The ATR and SMR are among the most common technologies for converting methane to carbon monoxide and hydrogen. A number of 1-d first principles based dynamic models of large scale SMR processes for refineries etc. have been developed during the past decades (Singh and Saraf, 1979; Alatiqi, *et al.*, 1989; Alatiqi and Meziou, 1991). A much referred kinetic model was made by Xu and Froment (1989). For small-scale mobile units, Springmann, *et al.* (2004) developed a distributed 1-d dynamic multiphase model for a gasoline reformer unit. This was a thermally integrated reforming unit consisting of an autothermal reformer, a heat exchanger and a high-temperature-shift-reactor. Simulation of the SMR process using generic software packages for chemical engineering applications have also been made, e.g. (Kolios, *et al.*, 2004). All these were distributed models.

In the analysis of dynamic behaviour for control purposes low order models are often sufficient. Hence, for simulation of the overall dynamics of the presented process there is a need to develop a 1-d dynamic first principles model based on mass and energy conservation. A model has been developed in a standard way as described by e.g. Thomas (1999). The following assumptions were made:

- The higher order reactions and the total oxidation reaction are totally converted and always in equilibrium.
- The overall mass balances are at steady state.
- Adiabatic process
- All gasses are ideal gasses

The steam reformation reaction and the water gas shift reaction are not always at equilibrium. They are described by one lumped reaction rate.

PRO/II was applied in the plant design process for performing steady state process simulations. Parameters for reactions and heat exchange in the dynamic model were tuned for different operation points to obtain the same steady state values for compositions, flows, temperatures, pressures as obtained in PRO/II.

### 3. SIMULATION RESULTS

The dynamic simulation model has been used to study how changes in the three candidate inputs affect the hydrogen production. The candidate inputs have been varied over a range, which is, based on process knowledge, considered a normal operation range. These ranges provide an operation region, and the model has been simulated to steady state for a set of operation points within this region. In all simulations, the natural gas feed stream was kept constant, i.e. only the relative ratios of steam and oxygen vs. this feed stream was varied.

The dynamic response in the hydrogen output to variations in the candidate inputs is of special interest for control structure design. This has been made introducing positive steps in the candidate inputs at several steady state operation points over the operation region. The steady state value has then been subtracted to give net responses, and through this provide a better basis for detecting possible changes in dynamic responses. The applied steps correspond to 10% of the variable variation range, i.e. the S/C ratio was increased by 2.4 kmol/h and the O/C ratio by 0.7 kmol/h. The feed temperature was increased by 40 °C. The steps were applied at time 100 seconds.

#### 3.1 Steady state analysis

Figure 3 shows the steady state hydrogen flow rate as function of S/C and O/C at the ATR side. The feed

temperature at the ATR is kept constant at 250 °C. The plane shows that the gain from both inputs to the hydrogen output is low for low O/C ratios, regardless of the S/C ratio. Apart from this low gain region, the gain is positive in the O/C ratio, and negative in the S/C ratio apart from at high O/C values where the sign of the gain shifts at S/C ratio 2.0. The latter is caused by the large heat integration. It appears that the gain, given by the gradient of the plane, is largest for O/C.

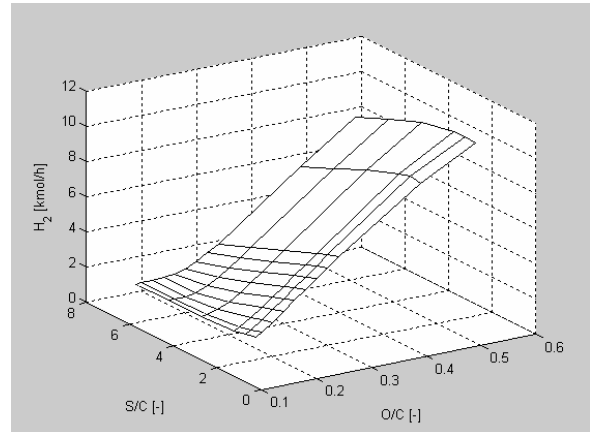


Fig.3: Steady state flow rate of produced hydrogen as function of S/C and O/C ratios at the ATR side.

Figure 4 shows the steady state hydrogen production rate as function of the feed temperature at the ATR side and O/C ratio. It appears that the gain from O/C is the highest out of these two input variables. Further, the gain from the feed temperature is low for low temperatures and low O/C ratios, but somewhat higher for higher temperatures. Moreover, the gain for high temperatures is nearly independent of the O/C ratio.

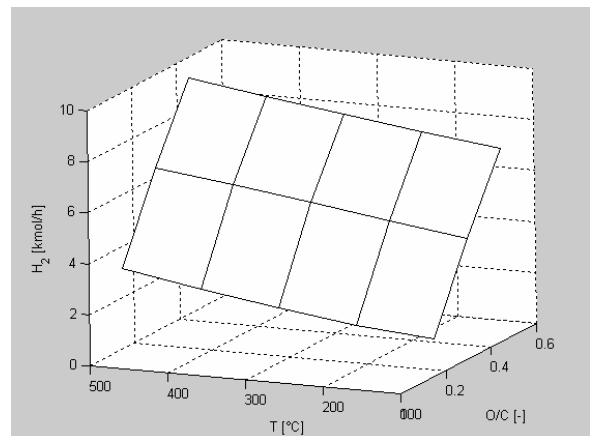


Fig.4: Steady state flow rate of produced hydrogen as function of the feed temperature and O/C at the ATR side.

#### 3.2 Analysis of dynamic behaviour

The upper part of Figure 5 shows net step responses to a step in the S/C ratio in the steady operating point at O/C=0.15. The lower set of curves correspond to O/C=0.55. The upper part of Figure 6 shows net step

responses to a step in the O/C ratio in the steady operating point at S/C=1.5, and the lower set of curves correspond to S/C=7.0. The feed temperatures were 250 °C.

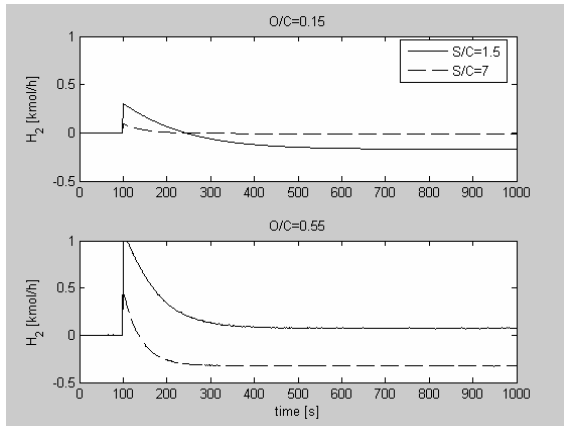


Fig.5: Net step responses in produced hydrogen to a positive step change in S/C at the ATR side.

Figure 6 shows that the dynamic characteristics do not change considerably with S/C. At very low O/C the gain is low. Also, the responses at O/C lower than 0.3 differs considerably at various S/C values, but not at higher O/C values. This means that O/C should be kept above 0.3. This is consistent with Figure 3.

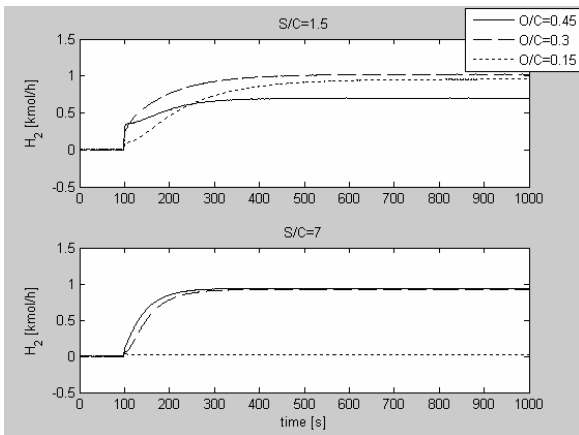


Fig.6: Net step responses in produced hydrogen to a positive step change in O/C at the ATR side.

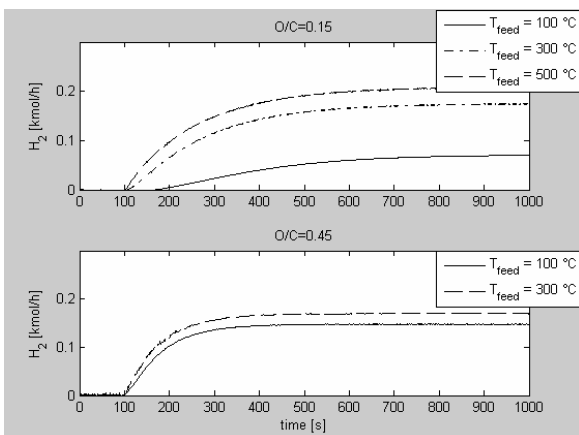


Fig.7: Step responses in produced hydrogen to a positive step change in the feed temperature at the ATR side.

The upper set of curves in Figure 7 shows net step responses to a step in the feed temperature at the ATR side in the steady operating point corresponding to O/C=0.15. The lower set of curves show net step responses in the operating point corresponding to a higher O/C ratio 0.45. The operating range has been reduced in these simulations in order to keep distance to the unstable operating region of the process.

#### 4. DISCUSSIONS

The simulations are focused on how different process inputs affect the hydrogen production. From the simulations it appears that the responses from the S/C ratio exhibit inverse characteristic, low gain and shift in the gain sign. The responses from the feed temperature at the ATR side also exhibit low gain. Similar results appear for the feed temperature at the SMR side (not shown here).

Opposed to this, the responses from the O/C ratio, for operating points above O/C=0.3, have high gain and without inverse characteristic. This favours the O/C ratio as manipulated variable for control.

Figures 5 and 7 show that the responses are faster at higher O/C ratios and at higher feed temperatures. The higher O/C ratios and feed temperatures, the shorter is the distance to the unstable operating region of the process. The dependency of the O/C ratio to the stability of the reformer is explained by the fact that the exothermic reactions in the ATR, and thereby the generated heat, increases with the oxygen feed flow. At too high flow rates, the heating of the reformer feed by the outlet flow accelerates the thermal heating in the reformer into a range where the catalyst in practice will be damaged.

This is according to Chen and Yu (2000) a well-known feature of exothermic reactor systems with heating of feed by outlet, as the Hydrofueler process. Due to the exothermic reactions, the process has positive feedback with gain given by the degree of heat exchange (energy recovery). They showed for these types of systems that the open-loop pole of the corresponding linear system approximation moves in the left half-plane towards the right-half-plane as the energy recovery increases. Similar to the critical gain in process control, there exists a critical effectiveness where the system is at the limit of stability.

However, improved external disturbance rejection and thereby better control performance is achieved when recovering more heat. Composition and temperature variations in the feed stream are typical examples of external disturbances for the Hydrofueler process. Model simulations show that the reformer

exhibit improved rejection of feed temperature disturbances at increased heat recovery (not shown here). However, simulations also show that the difference in disturbance rejection is not considerable with respect to variations in the gas composition. This means that the process has a good inherent rejection capability with respect to variations in gas composition.

The reformer in focus is based on compact plated heat exchanger technology. The simulations show that both the steady-state and dynamic behaviour of the reformer unit depends strongly on the flow rates. This is also well known feature of heat exchangers. Hence, nonlinear control strategies are often suggested for heat exchangers; see e.g. (Joshi, *et al.*, 1997). This is also relevant for the presented reformer since control of the hydrogen concentration in the outlet of the reformer is equivalent with control of outlet temperature under feed composition variations when the S/C ratio is controlled (Alatiqi, 1990).

## 5. CONCLUSIONS

This paper has analysed the dynamic behaviour of a novel small-scale hydrogen reformer, for on-site production of hydrogen in refuelling stations. The design is small-scale, compact and very energy efficient compared to conventional technology. The paper has demonstrated that the design exhibits complex dynamic behaviour, which poses challenges when designing a robust control structure with good performance for the process. The presented work is the first analysis of the dynamic behaviour of the reformer design and focuses on the responses in the hydrogen production rate to changes in several candidate inputs. Further work should include an analysis of other candidate controlled and manipulated variables, as well as the impact of various disturbances, to provide a more complete basis for control structure development.

Heat exchanger areas, feed temperatures and oxygen feed to the ATR side affect the amount of recovered energy. This quantity again balances the distance to the unstable operating region with the inherent external disturbance rejection. The more energy that is recovered, the shorter is the distance to the unstable operating region while the inherent external disturbance rejection capability is improved. This trade-off between dynamic controllability (pole location, performance) and thermal efficiency (inherent robustness for external disturbances) is an important topic in the process design. This is therefore an important topic in the further detailing of the Hydrofueler design.

## ACKNOWLEDGEMENT

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