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On-line estimation of inlet and outlet composition in catalytic partial oxidation

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ABSTRACT

An estimation strategy is presented for determining inlet and outlet composition of catalytic partial oxidation (CPOX) of methane over rhodium catalyst using simple, fast measurements: temperature, and thermal conductivity. A 1-D high fidelity simulation model for CPOX studied in Ref. [1] for a portable fuel cell application is developed and enhanced for transient experiments. Process dynamics are analysed to demonstrate how solid temperatures along the axes of the reactor reflect the endothermic/exothermic interplay of reactions during a process upset. Model reduction is then used to obtain a low complexity model suitable for use in a moving horizon estimator with update rates faster than 0.02 s. System theoretic observability analysis is then conducted to predict the suitability of different measurement designs and the best locations for temperature measurements for estimating both inlet and outlet gas mole fractions for all species. Finally, a Moving Horizon estimator is implemented and simulation experiments are conducted to verify the accuracy of the estimator.

1. Introduction

Catalytic partial oxidation reforming of methane is an efficient process used to produce syngas (H_2 and CO) using a fuel mixture that contains methane CH_4 and oxygen O_2. CPOX reforming is a compact size low-capital cost reactor that is suitable for portable applications as in fuel cells. CPOX is also being considered as a potential process for large scale production of syngas in view of its economic and environmental advantages over steam reforming [2].

Fast and accurate measurement of both inlet and outlet gas mole fractions is essential for process reliability and to effectively maintain the quality specifications on syngas. Fuel cells, for example, require varying inlet H_2 concentrations in the stack depending on load demands while maintaining low CO content to avoid poisoning the cell. Furthermore, polymer electrolyte membrane (PEM) fuel cells require low CO_2 concentrations. Restrictions on H_2O content can also be present. Different fuel cell and fuel processing control strategies can make use of accurate measurements of species mole fractions of the gas coming in and out from the CPOX reactor to enable feed-forward temperature control of the reactor, prevent excess H_2 generation, prevent fuel cell stack starvation and/or prevent CPOX clogging [3,4].

In this paper an estimator for inferring both inlet and outlet gas mole fractions in real time is developed. The developed state estimator can be used in portable fuel cell applications

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for monitoring and/or control. It can also be used in case the main composition measurement device is off-line and a substitute is needed to enhance reliability. The estimator design uses a single output measurement, such as thermal conductivity or gas density, that is combined with temperature measurements along the reactor and nominal input flows. In order to obtain well defined input and output composition estimates, these measurements are reconciled against a reactor model using a moving horizon estimator.

Previous work towards the development of a nonlinear observer for the CPOX process was given in Ref. [5]. A simple lumped parameter model was used that relied on one temperature measurement and one gas species composition measurement at the outlet to infer the remaining outlet gas species compositions at the outlet. The model used, however, was based on only two global reactions; partial and total oxidation and did not account for steam and dry reforming reactions. Further work in Ref. [6] was made for estimating inlet gas CH_4/O_2 ratio in the context of biogas reforming. Also, a simple lumped parameter model of a continually stirred reactor model was used but combined with a detailed reaction mechanism. In both models, spatial variations in temperature along the reactor were not accounted for, not to mention other important mass and energy transport effects present in the CPOX reforming process.

This paper is an extension of these two studies in multiple directions. First, a high fidelity 1-D model for CPOX process, originally studied in Ref. [1] and experimentally verified in Ref. [7], is developed and enhanced for transient simulation experiments. The high fidelity model captures the possible transport and kinetic effects in the lateral direction, assuming homogeneity in the radial and angle coordinates. A detailed analysis of process dynamics is conducted to determine the important measurements suitable for state estimation. The analysis revealed that solid temperatures across the reactor foam monolith have different dynamics and are highly correlated with the disturbances in the C/O ratio of the inlet gas. The different temperature dynamics are associated with the exothermic/endothermic interplay of reactions along the reactor.

Second, several transient simulation experiments with random variations in the inlet C/O ratio were conducted. The collected simulation data was then used to fit a high order state space model using linear subspace identification techniques [8]. The resulting high order state space model is then reduced in size using balanced truncation with matched DC gain. The state space model is then transformed into a descriptor model that is suitable for unknown input estimation and can incorporate the consistency condition in which the sum of mass fractions in the outlet gas stream must equal to one. A descriptor system observability analysis is performed to evaluate different measurement designs that guarantee numerical stability and uniqueness of the estimates. Local observability analysis of the low complexity model indicated that three temperature measurements spread apart combined with either a density or thermal conductivity measurement of the outlet gas stream allows a well conditioned and stable estimator to be designed.

Third, a moving horizon state estimator that incorporates the low complexity descriptor model, best measurement design, known inequality constraints of the CPOX process is then developed. State estimator performance in terms of mean square error is then verified via simulation. The estimation accuracy, in terms of mean square error values, was in the order of $O(10^{-5})$ with very good performance for inlet gas O_2, CH_4 and outlet gas H_2, CO and Ar species mole fractions and marginal accuracy for other variables due to unaccounted non-linearities.

The linearized system identification/model reduction strategy used in this study provided solution times of less then 0.02 s per iteration which are adequate for the CPOX process time scales but with some compromise in estimation accuracy. Another advantage is that no quasi-steady state assumptions were needed and the time scales of the original high fidelity model are retained in the low complexity model. Finally, the solution strategy is implementable on a stand alone microprocessor using custom C code generated from CVXGEN available in Ref. [9] which can speed implementation even further.

The organization of the paper is as follows: Section 2 will present the detail model equations of the CPOX reactor used in this study. Section 3 will describe the efforts used to accelerate transient simulations of the model followed by an analysis of process dynamics. Section 4 will discuss the process of extracting a low complexity model using subspace identification techniques combined with model reduction. Section 5 will formulate the desired state estimation problem to be solved by forming a descriptor model of the process followed by local observability analysis for different proposed measurement designs. Section 6 will present the Moving Horizon State Estimator algorithm for descriptor systems that will be used and finally Section 7 will present the results obtained followed by a discussion in Section 8.

2. Model description

The CPOX reformer model adapted in this work was developed in Ref. [1] and was also validated via experiments in Ref. [7] in the context of biogas fuel reforming. The system consists of a reactor tube made from a catalyst-loaded Al_2O_3 ceramic foam installed inside a furnace as depicted in Fig. 1. Feed flows of CH_4, O_2, and Ar are metered with mass-flow controllers and

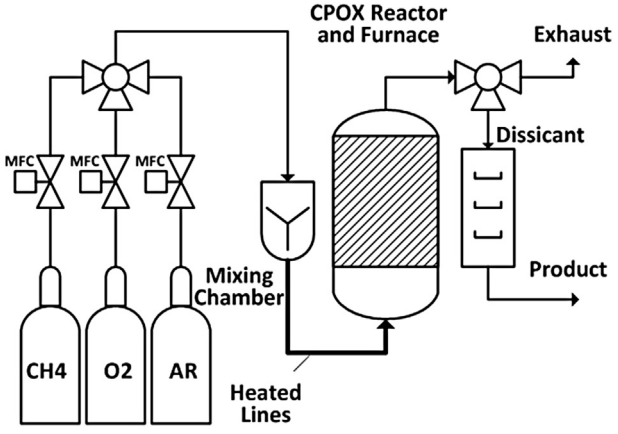

Fig. 1 – Process flow diagram.