Control of catalytic flow reversal reactor

Kariwala, Vinay; Forbes, J. Fraser

2006

Kariwala, V., & Forbes, J. F. (2006). Control of catalytic flow reversal reactor. In 9th International Conference of Control, Automation, Robotics and Vision, (pp: 1-6).

<https://hdl.handle.net/10356/91195>

<https://doi.org/10.1109/ICARCV.2006.345070>

© IEEE. Personal use of this material is permitted. However, permission to reprint/republish this material for advertising or promotional purposes or for creating new collective works for resale or redistribution to servers or lists, or to reuse any copyrighted component of this work in other works must be obtained from the IEEE. This material is presented to ensure timely dissemination of scholarly and technical work. Copyright and all rights therein are retained by authors or by other copyright holders. All persons copying this information are expected to adhere to the terms and constraints invoked by each author's copyright. In most cases, these works may not be reposted without the explicit permission of the copyright holder.

Downloaded on 26 Apr 2023 23:21:22 SGT

Control of Catalytic Flow Reversal Reactor

Vinay Kariwala

Division of Chemical & Biomolecular Engineering
Nanyang Technological University
Singapore 637722
Email: vinay@ntu.edu.sg

J. Fraser Forbes

Department of Chemical & Materials Engineering
University of Alberta
Edmonton, Canada T6G 2G6
Email: fforbes@ualberta.ca

Abstract—This paper deals with control of catalytic flow reversal reactors (CFRR) used for oxidation of greenhouse gases like methane. This system differs from conventional reactors as it is a discrete-event or hybrid system with the only available manipulated variable being the flow direction. For maintaining high conversion rate and safety reasons, closed loop control is required. We successfully implement and compare the performances of different techniques like logic control, scheduling and predictive control for safe operation of CFRR. We find that the performances of latter two schemes are almost equivalent.

Keywords—Control applications, Discrete event systems, Logic controllers, Predictive control, Reactor control.

I. INTRODUCTION

Hybrid systems are continuous variable, continuous time systems with a phased operation. They display continuous behavior consisting of smooth flows within phases and discrete behavior consisting of abrupt transition between phases [1]. Thus, hybrid system displays both continuous and discrete behavior. Such hybrid systems arise in varied contexts in manufacturing, communication networks, auto-pilot design, computer synchronization, traffic control and chemical processes, among others [2].

This paper deals with such a hybrid system *i.e.* Catalytic flow reversal reactors (CFRR). As opposed to conventional plug flow reactors, the direction of the feed to CFRR is periodically reversed to provide self-sufficient operation [3]. CFRR were first studied by Frank-Kamenetski [4] for heterogeneous exothermic reactions. Matros and Bumynovich [5] provide a detailed review of different applications of CFRR. In this paper, we consider CFRR for catalytic combustion of greenhouse gases with large potential for global warming like methane.

In presence of various disturbances *i.e.* change in feed composition or flow rate, the performance of the CFRR (measured in terms of outlet methane concentration) degrades and closed loop control is required. The use of closed loop control is also necessary to ensure that the reactor does not extinguishes and the temperature does not violate an upper bound causing damage to the reactor. The control of CFRR has been considered earlier in [6], [7]. They used an external heating element in the central limb of the CFRR as the manipulated variable (see Figure 1). Alternatively, Balaji and Lakshminarayan [8] considered the control of CFRR by withdrawing an intermittent stream from the central limb with

the use of constant flow reversal sequence. The setup used in this paper does not have a heating element, nor is a stream withdrawn intermittently. Then, the flow reversal command sequence represents the only available manipulated variable, which also gives rise to discrete dynamics in CFRR. In this sense, the results of this paper complement the results in [8]. We compare the performances of different techniques like logic control, scheduling and predictive control. Most of these techniques perform satisfactorily and the carefully constructed scheduling-based algorithm closely matches the performance of predictive control. Due to space restrictions, the discussion is brief at places and further details are available in [9].

II. CATALYTIC FLOW REVERSAL REACTOR

Fig. 1. Catalytic Flow Reversal Reactor

A schematic diagram of the U-shaped catalytic converter is shown in Figure 1. The insulated ceramic monolith sections at the two ends of the reactor are used to store the energy of the exit flow. The heart of the reactor consists of the two sections containing the catalyst, which are connected together through an open channel. A fraction of the reaction mixture can be withdrawn through an outlet located in this open channel. The reactor's temperature is measured using thermocouples ($T_1, T_2, \dots, T_{16}$) located along the limbs. At some points, more than one thermocouple is used at different

radial positions to detect the presence of any *hot spots*. The concentration of the reaction mixture is measured at the inlet, outlet and in the open channel using gas chromatograph.

Fig. 2. Departure of Temperature profile towards Reactor's exit for uni-directional flow

When this reactor is operated in a uni-directional flow mode with no preheating of the feed stream, the temperature profile gradually approaches the reactor's exit (See Figure 2) and the reactor ultimately extinguishes. Preheating of the feed is required to keep the reactor under continuous operation. An alternate approach is to periodically reverse the flow direction through the reactor. Then the energy stored in the monolith sections can be used to preheat the feed when the flow reverses. Not only this operational strategy requires no preheating, the stream withdrawn in the open channel can be used as an energy source.

The flow in the reactor is reversed by opening and closing appropriate valves (See Figure 3). When valves 1 and 4 are opened, the feed flow from left to right across the reactor, referred as *forward flow*. On the other hand, opening of valves 2 and 3 causes the feed to flow from right to left, referred as *reverse flow*.

Fig. 3. Flow reversal in the reactor

A. Model

Liu *et al.* [3] have presented a 1-D model of this process assuming a pseudo-steady state and ignoring the radial variation of system states. The model consists of the mole and the energy balances in the fluid (gases like methane and carbon

mono-oxide) and solid phases (reactor, catalyst *etc.*). The major assumptions related to the development of the model are described by Liu *et al.* [3] and Hayes and Kolaczowski [10].

B. Simulator

Salomons *et al.* [11], [12] have developed a simulator for this system in FORTRAN. The set of partial differential equations are discretized using Finite Element Method (FEM) and the resulting algebraic equations are solved recursively using dirichlet type boundary conditions. A non-uniform mesh is used, whose grid size has been optimized. The response of this simulator has been validated against experimental results. In this work, the following assumptions are made:

- The flow rate of the reaction mixture withdrawn in the open channel is zero.
- In the experimental setup, the concentration is measured using gas chromatograph, but it is assumed to be available frequently here.

The simulation time for 1-D model is approximately 3 – 4 times larger than the real time on a Pentium® III 866MHz processor. Though a simulator based on a 2-D model is also available, the 1-D model is used here due to the involved computational expense and is considered as the system throughout this paper. It is understood that the numerical values obtained from the 1-D model contain some errors, but the trends of the system states are same as the 2-D model.

Fig. 4. Response of open-loop system for $\tau = 25$ for nominal case

III. OPEN LOOP CONTROL

We first present the system's response for the open-loop case. The term *open-loop* refers to the case, where the switching sequence is predetermined or is chosen independent of the values of system states. For simplicity, only symmetric flow reversal sequences with constant half-cycle length τ (seconds) are used. A sampling time of 1 second is used. The case, where $C_{CH_4,in} = 3.87 \times 10^{-3}$ mol % and $v(t) = 0.42$ m/s is considered as the nominal case. Here, $v(t)$ is the superficial

Fig. 5. Response of open-loop system for $\tau = 300$ for nominal case

velocity of the feed. Negative $v(t)$ represents reverse flow and *vice versa*. The temperatures indicated by thermocouples in the left and right limbs of the converter are referred as $T_1 - T_8$ and $T_9 - T_{16}$, respectively. Temperatures above 400°C can cause damage to the insulation layer in the monolith section, so T_1 and T_{16} are also monitored.

The responses of the open-loop system for $\tau = 25$ and 300 are shown in Figures 4 and 5. It is seen that with an increase in τ , the temperatures often reach close to the upper bounds. Similar trend is seen for other values of τ , $25 \leq \tau \leq 300$.

Fig. 6. Disturbances in $v(t)$ and $C_{CH_4,in}(t)$

To evaluate the effect of disturbance on the system response, we consider $\pm 20\%$ and $\pm 10\%$ variation in the feed flow rate and feed composition as compared to the nominal case. The time trajectories for $v(t)$ and $C_{CH_4,in}(t)$ are shown in Figure 6 and the system's response for $\tau = 300$ in Figure 7. Though nearly optimal for the nominal case, in presence of disturbances, the temperature constraints are violated on some occasions for $\tau = 300$. If open-loop control is used, then persisted disturbances can cause the performance to deteriorate and closed loop control is necessary.

IV. CLOSED-LOOP CONTROL

In this section, various schemes for feedback control of the CFRR is discussed. Representing the dynamics of forward

Fig. 7. Response of open-loop system for $\tau = 300$ in presence of disturbances

and reverse flows as $f_1(\cdot)$ and $f_2(\cdot)$ respectively, the system is given as

$$\dot{x}(t) = \delta(t)f_1(x(t), d(t)) + (1 - \delta(t))f_2(x(t), d(t))$$

where $\delta(t) = \text{sign}(v(t))$. Lyapunov stability analysis suggests that this process has no equilibrium points except when the reactor extinguishes [9]. Then the control objective is to maintain continuous operation with maximal conversion rate and is formally given as

$$\begin{aligned} \min_{\delta(t)} J &= \bar{C}_{CH_4,out} \\ T_1(t) &< 390^\circ\text{C} \\ T_{16}(t) &< 390^\circ\text{C} \\ \delta(t) &\in \{-1, 1\} \quad \forall t \end{aligned} \quad (1)$$

where $\bar{C}_{CH_4,out}$ denotes the average outlet methane concentration. Note that we have backed-off from the specified bounds of 400°C to maintain feasibility. The manipulate variable is updated every 5 seconds to provide sufficient time for the calculations involved for all the algorithms studied. It is assumed that all the disturbances are deterministic and extension to the stochastic case is easy by using a filter such as averaging.

A. Logic Control

Programmable Logic Controllers (PLC) are used extensively in process industries owing to their simplicity [13]. Here two simple logic-based techniques are analyzed for control of CFRR. In the first scheme, analogous to common household thermostat, the feed flow direction is changed when one of the temperature constraints is violated with no consideration given to the outlet methane concentration. As can be expected, the performance is poor as shown in Figure 8. The magnitude of constraint violation can be reduced by constraining the

Fig. 8. Response of logic control (scheme 1)

temperatures to values lower than 390°C , but this has an adverse effect on the conversion.

It is observed that at any time instant, use of only one of the flow directions results in increased conversion. Then the performance is improved by reversing the flow, whenever $C_{CH_4,out}$ reaches a local minima (slope becomes positive) or one of the temperature constraints is violated. This algorithm provides better performance but requires frequent flow reversals (*cf.* Figure 8), which may result in wear and tear of the valves.

Fig. 9. Response of logic control (scheme 2)

B. Scheduling

Gain scheduling [14] is a popular technique for control of non-linear processes. In this scheme, a number of linear controllers are designed by linearizing the system around

different operating points. As the process moves from one operating point to the other, switching between these locally optimal controllers is made based on the current operating point. The performance of the two logic-based algorithms is seen to be poor, when both of $\bar{C}_{CH_4,out}$ and frequency of flow reversals are considered. Based on an idea similar to gain scheduling, a scheduling based algorithm is considered, which gives intermediate performance as compared to the earlier algorithms.

The time required for the logic-based algorithm (Scheme 2) to reach the local minimas of the $C_{CH_4,out}$ (where its slope becomes positive) decreases successively [9]. This causes the flow reversal time for this algorithm to decrease simultaneously. Consider the case, when only a constant flow reversal time is to be used. Then, an upper bound on the optimal constant flow reversal time can be estimated by evaluating the time required for the logic based algorithm (Scheme 2) to reach the first local minima. Now, the flow reversal times can be scheduled by calculating this upper bound (τ_{ref}) for different operating conditions characterized by $v(t)$ and $C_{CH_4,in}$.

For the given system, $C_{CH_4,in}$ variation has little effect on τ_{ref} and is ignored. The variation of τ_{ref} with feed flow rate is shown in Table I and during implementation, Lagrange method is used to interpolate between these points.

v , m/s	0.30	0.34	0.38	0.42	0.46	0.50
τ_{ref} , s	480	422	370	330	305	280

TABLE I
VARIATION OF τ_{ref} WITH FEED RATE

During the implementation, the flow is reversed on constraint violation or depending on the value of $v(t)$. The closed loop response is shown in Figure 10, which is substantially better than the previous approaches.

Fig. 10. Response of scheduling based algorithm

C. Predictive Control

Model Predictive Controller (MPC) uses a model of the system to predict the system's behavior several steps into the future and then makes the control decision based on it. In its comparison, other control schemes are also referred as *myopic*. The present problem differs from the conventional MPC approach in the sense that the manipulated variable can assume only finite values. In the context of dynamic programming, a similar problem on machine replacement has been discussed by Bertsekas [15]. The details on MPC are given by Meadows and Rawlings [16] and are omitted here for the sake of brevity.

The effectiveness of MPC largely relies on the accuracy of the model used. Since the available simulator runs slower than real-time on simple PCs, a simpler model is required. Based on the distributed parameter model and some trial and error, the following structure is chosen for temperature readings

$$T_i(k+1) = T_i(k) + \left[p_i T_{i+1}(k) - (2p_i + p_{16+i} v(k)) T_i(k) + (p_i + p_{16+i} v(k)) T_{i-1}(k) \right] + q_i (-R_{CO})_H (\Delta H_R)_{CO} C_{CH_4, in}$$

$$i = \{1, \dots, 16\} \quad (2)$$

$$T_{17}(k+1) = \begin{cases} p_{33} + p_{34} T_1(k) & \text{if } \delta(k) = 1 \\ p_{35} + p_{36} T_{16}(k) & \text{if } \delta(k) = -1 \end{cases} \quad (3)$$

where $T_1 - T_{16}$ are the 16 thermocouple readings. T_0 and T_{17} are the temperatures of inlet and outlet streams respectively. Here, p_i and q_i are unknown parameters and q_i is taken to be zero, if the associated thermocouple is not in one of the catalytic sections. In (2), $(-R_{CO})_H$ and $(\Delta H_R)_{CO}$ represent homogeneous reaction rate and heat of reaction of carbon mono-oxide, respectively. Both of these parameters depend on the temperature non-linearly.

Since the concentration of methane is available only at the inlet and outlet of the reactor, it is modelled as a function of available temperature readings.

$$C_{CH_4, out}(k+1) = \begin{cases} p_{45} C_{CH_4, out}(k) + p_{46} (-R_{CH_4}(T_{12}))_S & \text{if } \delta(k) = 1 \\ p_{47} C_{CH_4, out}(k) + p_{48} (-R_{CH_4}(T_5))_S & \text{if } \delta(k) = -1 \end{cases} \quad (4)$$

where $(-R_{CH_4})_S$ is the catalytic reaction rate of methane defined as [3]

$$(-R_{CH_4})_S = 3 \times 10^9 e^{\frac{-1.59 \times 10^5}{R_g T}} \quad (5)$$

Here, R_g is the gas constant. Overall, this model has 48 unknown parameters, which are estimated by minimizing 1-step ahead prediction error. Ideally, for MPC relevant identification n_p -step ahead prediction should be minimized to ensure that good predictions are available over the entire prediction horizon, n_p . In the present case, it is deemed unnecessary as the model obtained with 1-step ahead prediction error minimization has good prediction capabilities for multi-step ahead

prediction. The predicted and actual values for temperatures and methane concentration are compared for 10-step ahead prediction in Figure 11.

(a) Temperature profile at t=1000s

(b) $C_{CH_4, out}$

Fig. 11. 10-step ahead prediction capabilities of identified model (- - Predicted)

The MPC is implemented on CFFR with prediction horizon of 10s and control horizon of 5s. To avoid excessive input usage, a penalty factor of 1×10^{-4} is imposed on the rate of change of input. Since, the manipulated variable can assume only 2 states and is updated every 5s, no optimization problem is required to be solved. At every step, the predictions are obtained for $\delta = -1$ and 1 taking it to be constant over the entire prediction horizon and the best strategy is chosen based on this. The response of the closed loop system is shown in Figure 12. It is seen that MPC gives reasonable performance with maintaining the temperatures within their bounds. It might be possible to improve its' response by varying the parameters like prediction horizon, control horizon and input penalty factor and is an issue for further research.

V. COMPARISON

In the previous section, different strategies for feedback control of the CFFR were discussed and their performance is compared in this section. For comparison purposes, the closed loop system was simulated under the disturbances sequences for v and $C_{CH_4, in}$ shown in Figure 6.

Different schemes are compared on the basis of achieved performance defined in terms of average outlet methane concentration, $\bar{C}_{CH_4, out}$ and the number of flow reversals required. To take the magnitude and duration of temperature

Fig. 12. Response of predictive control

constraint violation into account simultaneously, an index, I_C is calculated as

$$I_C = \sum_{t=1}^k \max(T_1(t) - 390, 0) + \max(T_{16}(t) - 390, 0)$$

where k is the simulation length.

	$\bar{C}_{CH_4,out}$	Reversals	I_C
Open Loop	1.68×10^{-4}	8	132.35
Logic Control # 1	1.67×10^{-4}	6	27.07
Logic Control # 2	1.44×10^{-4}	36	0
Scheduling	1.63×10^{-4}	7	2.44
Predictive Control	1.66×10^{-4}	8	0

TABLE II
COMPARISON OF CONTROL SCHEMES

The comparison results are shown in Table II. The average conversion obtained using open-loop control ($\tau = 300s$) is similar to most of the feedback control algorithms, but constraint handling is poor. Logic-based control schemes lies at the two end of the spectrum in terms of minimum number of flow reversals and average methane concentration. It is interesting to note that Logic control (scheme 2) does not violate any constraints. Though the conversion increases with temperature, the incremental rate of conversion does not, *i.e.* the incremental rate of conversion become positive before the reactor's temperatures cross safety limits. Scheduling based algorithm and MPC balance the performance with the number of flow reversals required. The performance of these two algorithms matches closely, though the former algorithm is a simpler control scheme. This happens as scheduling the flow direction based on the disturbances provides an implicit feed forward action. Taking all the factors into account, MPC and Scheduling are considered to be equivalent.

VI. CONCLUSIONS

The problem of controlling the catalytic flow reversal reactor in presence of disturbances is studied. The comparison of different techniques *viz.* logic-based control, scheduling and model predictive control is made. Considering the performance of the closed loop system defined in terms of average methane concentration in the outlet stream, flow reversals required and temperature constraint violation simultaneously, scheduling and MPC are found to be equivalent. However, MPC requires a good model of the process and online concentration measurement, which may not always be available. Based on this, we conclude that scheduling is the most efficient technique for control of this process. The future work will focus on modification of these techniques, once the scope of the simulator is extended to include the withdrawal of reaction mixture in the open channel of CFRR.

ACKNOWLEDGEMENT

The author thank Prof. Robert E. Hayes, Steven Salomons and Amika Kushwaha of University of Alberta, Canada for providing the simulator and further details regarding the reactor.

REFERENCES

- [1] A. Deshpande, "Control of hybrid systems," Ph.D. Thesis, University of California, Berkeley, May 1994.
- [2] P. Antsaklis and A. Nerode, "Hybrid control systems: An introductory discussion to the special issue," *IEEE Trans. Automat. Contr.*, vol. 48, no. 4, pp. 457–460, 1998.
- [3] B. Liu, R. Hayes, M. Checkel, M. Zheng, and E. Mirosh, "Reversing flow catalytic converter for a dual fuel engine," *Chem. Engg. Sci.*, vol. 56, no. 8, pp. 2641–2658, 2001.
- [4] D. A. Frank-Kamenetski, *Diffusion and Heat Exchange in Chemical Kinetics*. Princeton, NJ, USA: Princeton University Press, 1955.
- [5] Y. S. Matros and G. Bunimovich, "Reverseflow operation in fixed bed catalytic reactors," *Catalysis Reviews: Science and Engineering*, vol. 38, pp. 1–68, 1996.
- [6] P. Dufour and Y. Toure, "Multivariate model predictive control of a catalytic reverse flow reactor," *Computers and Chemical Engineering*, vol. 28, pp. 2259–2270, 2004.
- [7] D. Edouard, P. Dufour, and H. Hammouri, "Observer based multivariable control of a catalytic reverse flow reactor: Comparison between LQR and MPC approaches," *Computers and Chemical Engineering*, vol. 29, pp. 851–865, 2004.
- [8] S. Balaji and S. Lakshminarayanan, "Heat removal from reverse flow reactors used in methane combustion," *Canadian Journal of Chemical Engineering*, vol. 83, pp. 1–10, 2005.
- [9] V. Kariwala, "Analysis and control of catalytic flow reversal reactor," University of Alberta, Edmonton, Canada, Internal Research Report, 2004.
- [10] R. Hayes and S. Kolaczowski, *Introduction to Catalytic Combustion*. Amsterdam, Netherlands: Gordon and Breach Science Publishers, 1997.
- [11] S. Salomons, R. Hayes, M. Poirier, and H. Sapoundjiev, "Flow reversal reactor for the catalytic combustion of lean methane mixtures," *Catalysis Today*, vol. 83, pp. 59–69, 2003.
- [12] S. Salomons, "Modelling the behaviour of a reverse-flow catalytic reactor for the combustion of lean methane," M.Sc. Thesis, University of Alberta, Edmonton, Canada, 2003, Available at <http://www.ualberta.ca/~salomons/CFRR.pdf>.
- [13] M. Chidambaram, *Computer Control of Processes*. CRC Press, 2001.
- [14] H. Khalil, *Nonlinear Systems*, 2nd ed. Upper Saddle River, NJ: Prentice Hall, 1996.
- [15] D. Bertsekas, *Dynamic Programming and Optimal Control*, 2nd ed. Belmont, MA: Athena Scientific, 1995, vol. I.
- [16] E. Meadows and J. Rawlings, *Nonlinear Process Control*. Prentice Hall, 1996, ch. Model Predictive Control.