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Optimal Operation and Control of Fluidized Bed Membrane Reactors for Steam Methane Reforming

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Abstract

This work presents the optimal operation and control of a Fluidized Bed Membrane Reactor (FBMR) for Steam Methane Reforming (SMR). First, a nonlinear distributed parameter dynamic model is developed. Next, the optimal operation of the system is studied by solving a dynamic optimization problem that maximizes the conversion and separation in the reactor. Based on the optimization result, reduced order linear models are developed and used in the design of conventional and model based controllers. The performance of these controllers are tested considering the variation in the inlet concentration of the feed to the reactor and the initial conditions.

Keywords: Fluidized Bed Membrane Reactors, Steam Methane Reforming, Optimal Operation, Model Based Controllers.

1. Introduction

Hydrogen is one of the most important chemicals used in the chemical industry, and lately, it has gained particular attention due to the wide range of applications also as an energy carrier. There are several technologies available for the production of hydrogen; however, the conventional steam methane reforming (SMR) is currently still the most common and economical way to produce hydrogen. The conventional SMR process consists of two main reactions given by:

$$CH_4 + H_2O \rightleftarrows CO + 3H_2,\ \Delta H_{298\,\mathrm{K}} = +206\,\mathrm{kJmol^{-1}}$$

$$CO + H_2O \rightleftarrows CO_2 + H_2,\ \Delta H_{298\,\mathrm{K}} = -41\,\mathrm{kJmol^{-1}}$$

In order to intensify this process and enhance the yield of hydrogen and shift the reaction equilibrium in the direction of producing hydrogen, new reactor concepts have been developed and investigated. The Fluidized Bed Membrane Reactor (FBMR) is one of the most promising membrane reactors for integrated reforming/dehydrogenation, separation and purification (Rahimpour et al. 2017). In the FMBR, a bundle of hydrogen-selective membranes are immersed into the fluidized catalytic bed in order to carry out the separation and reaction steps simultaneously. Such reactor concepts can reduce capital costs and improve process efficiency, but at the cost of losing degrees of freedom in the operation. Despite the substantial amount of work done on process design based on steady state modelling of FBMR's the study of operational aspects such as controllability, stability, and operational feasibility have not been investigated as extensively. Only a limited number of works is available with the application of control theory for a fluidized bed reactor (FBR). However, these controllers cannot always be applied to an FBMR, because the intensified FBMR exhibits a quite different dynamic behaviour due to small equipment but same or larger process efficiency. Considering the impact of the intensification on the operation, the control design of a FBMR is very relevant.

In this work, we have studied the optimal operation and control of an FBMR for SMR. First, a dynamic model extending the steady-state models proposed by Medrano et al. (2018) is described. Next, the optimal operation of the system is studied. To this end, a dynamic optimization problem that maximizes conversion and separation in the reactor is proposed. Based on the result obtained from the optimization problem, and the dynamic model, linear models of the system have been developed. However, as the linear model is infinite dimensional (the dynamic model is given by a set of Nonlinear Partial Differential Equations), and control design based on these models is challenging, model reduction techniques have been applied. Finally, with the reduced linear model, low-level and Model Predictive controllers have been designed. The performance of these controllers has been tested in simulation in case of variations in the inlet composition of the feed to the reactor.

2. Dynamic Modelling of FBMR

Consider a FBMR where pure hydrogen is recovered via palladium-based membranes inserted into the fluidized bed (Gallucci et al. 2008). The dynamic model obtained in this work is an extension of the model proposed by Medrano et al. (2018) by including accumulation terms in the gas phase mass (bubble/wake and emulsion phases) and energy balances yielding the following set of Partial Differential Equations (PDEs),

$$\frac{\partial}{\partial t}\begin{bmatrix}\left(f_{bw}C_{i,bw}\right)\\ \left(f_e C_{i,e}\right)\\ (\rho C_p f_{bew}T)\end{bmatrix}=-\frac{\partial}{\partial z}\begin{bmatrix}\left(f_{bw}u_b C_{i,bw}\right)\\ \left(f_e u_e C_{i,e}\right)\\ \left(\rho C_p f_{bew}uT\right)\end{bmatrix}+\begin{bmatrix}\xi_i + r_{i,bw}f_{bw}(1-\varepsilon_{mf})+S_{mb}\\ -\xi_i + r_{i,e}f_e(1-\varepsilon_{mf})+S_{me}\\ \sum_{k=1}^{p}\sum_{i=1}^{m}(-\Delta H_{r,k})r_{i,k}f_k(1-\varepsilon_{mf})\end{bmatrix}+\begin{bmatrix}0\\ 0\\ \frac{4}{d}H_J(T_J-T)\end{bmatrix} \quad (1)$$

$\forall i=\{CH_4, CO_2, CO, H_2O, H_2\}$

where ξ_i , $S_{\{mb,me\}}$ are the mass transfer rates between the bubble and emulsion phases, and the membrane, and $r_{i,\{bw,e\}}$ are the reaction rates. The model given by Eq. (1) is subject to a set of algebraic equations which are not included in this paper due to space limitations. However, a summary of the hydrodynamics and mass transfer correlations used in this work is reported in Medrano et al. (2018).

2.1. Discrete Model

In order to solve the dynamic model, the spatial derivatives of Eq. (1) are replaced by a backward differences approximation such that the model can be written as a set of ODEs,

$$\frac{d}{dt}\begin{bmatrix}(f_{bw}C_{i,bw}^{j})\\(f_e C_{i,e}^{j})\\(\rho C_p f_{bew}T^{j})\end{bmatrix}=-\frac{1}{\Delta z}\begin{bmatrix}\Delta(f_{bw}u_b C_{i,bw}^{j})\\\Delta(f_e u_e C_{i,e}^{j})\\\Delta(\rho C_p f_{bew}uT^{j})\end{bmatrix}+M\begin{bmatrix}\xi_i^j\\S_i^j\\r_i^j\end{bmatrix}+\begin{bmatrix}0\\0\\\frac{4H_J}{d}(T_J-T)\end{bmatrix},\forall j=1,\ldots,N \quad (2)$$

where Δ is defined as $\Delta C_i^j = C_i^j - C_i^{j-1}$, N *is* the number of sections in which the reactor is divided, and M is a matrix related to the mass transfer and reaction rates. After some algebraic manipulations Eq. (2) can be expressed as: $\dot{x} = f(x,u,d),\ y = g(x,u)$, where $x=\left[C_{i,bw}^{j},C_{i,e}^{j},T^{j}\right]^T \in \mathbb{R}^{11N}$, $y=\left[X_{CH_4,out},C_{H_2,out}\right]^T \in \mathbb{R}^2$ and $u=\left[F,T_J\right]^T \in \mathbb{R}^2$ are the states, manipulated, and controlled variables respectively, with F the feed flow of the reactor, T_J the jacket temperature, and $X_{CH_4,out} = C_{CH_4,in}^{-1}(C_{CH_4,in} - C_{CH_4,out})$ the methane conversion. In addition, $d=\left[C_{i,bw}^{in},C_{i,e}^{in},T^{in}\right]^T \in \mathbb{R}^{11}$ are the disturbances of the system.

3. Optimal Operation of FBMR

The main objective of the FBMR is to maximize both the conversion of methane and the separation of the hydrogen throughout the membrane. By defining $X_{CH_4}^j \in [0,1]$ as the conversion of methane and $S_{H_2}^j \in [0,1]$ as the hydrogen separation factor along the reactor, we formulate the following optimization problem to achieve these objectives:

$$u_{opt} = \arg\min_u \int_{t_0}^{t_f}\left(\sum_{j=1}^{N}\omega\left\|1-X_{CH_4}^j\right\|_2^2+(1-\omega)\left\|1-S_{H_2}^j\right\|_2^2\right)dt \quad (3)$$
$$\text{subject to: } \dot{x} = f(x,u,d), y = g(x,u),\ \ x\in\mathbf{X},\ u\in\mathbf{U}$$

Where $\omega \in [0,1]$ is a weighting parameter. In this optimization problem, the feed flow F and the jacket temperature T_j are the decision variables. From the practical point of view, the objective function of problem (3) is selected because it takes a minimum value when the raw material and the hydrogen produced are completely consumed and separated. Finally, the problem (3) is solved by parametrizing u and converting Eq. (3) into a Nonlinear Programming problem (NLP). The results of the optimization problem are shown in Figure 1.

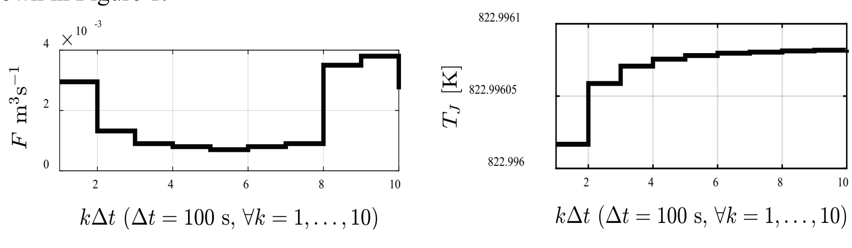


Figure 1. Optimal Trajectories for the inlet feed flow and the jacket temperature.

Notice that in Figure 1, T_J does not change significantly. That means, during the normal operation (no disturbances), this variable must remain constant around 823 K. However, it is important to clarify that the controllers can still manipulate T_J, to reject disturbances and to keep the system in the optimal trajectory.

4. Control Design for FBMR

In this paper, two control techniques are used to keep the operation of the FBMR around the optimal trajectory presented in Section 3, MPC and PID. To design these controllers, a linear model of the process must be obtained.

4.1. Linear Model and Model Reduction

The linear model of the FBMR is obtained by linearizing $\dot{x} = f(x,u,d),\ y = g(x,u)$ around the optimal operating profiles using Taylor series expansion and considering only the first order terms as follows,

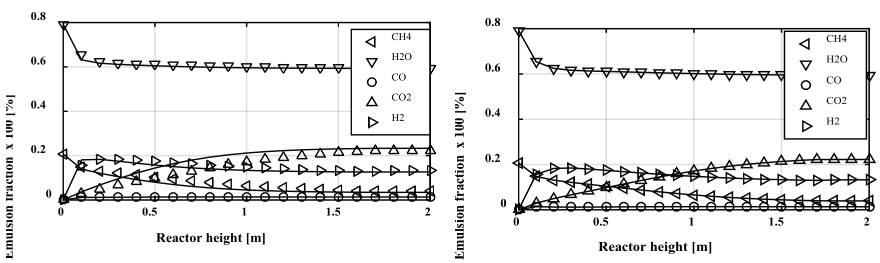
$$\begin{aligned} &\dot{x} = Ax + Bu + B_d d,\ A = f_x(x,u)\big|_{x^*,u^*},\ B = f_u(x,u)\big|_{x^*,u^*},\ B_d = f_d(x,u)\big|_{x^*,u^*} \\ &y = Cx + Du,\ C = g_x(x,u)\big|_{x^*,u^*},\ \mathrm{D} = g_u(x,u)\big|_{x^*,u^*} \end{aligned} \tag{5}$$

where $x \in \mathbb{R}^{11N}$, $y \in \mathbb{R}^2$, and $u \in \mathbb{R}^2$. In this paper, the spatial domain of the reactor is divided into 100 sections, resulting in 1100 number of states. The controllability analysis shows the system has uncontrollable states. These two facts, a large number of states and uncontrollable states, make the design and implementation of model-based controllers for the FBMR very difficult. A good solution to this issue is to eliminate the uncontrollable states. This also reduces the number of states . To this end, a model reduction technique called balance truncation (Gugercin and Antoulas, 2004) is implemented in the FBMR model, and briefly described in Table 1.

Table1: Mathematical formulation of the reduced order model

Full order Model	Reduced Order Model
$G(s): \begin{cases} \dot{x} = Ax + Bu + B_d d \\ y = Cx + Du \end{cases},\ x \in \mathbb{R}^{11N}$	$\tilde{G}(s): \begin{cases} \dot{\tilde{x}} = \tilde{A}\tilde{x} + \tilde{B}u + \tilde{B}_d d \\ y = \tilde{C}x + \tilde{D}u \end{cases},\ \tilde{x} \in \mathbb{R}^n,\ n \le 11N$
$\tilde{x} = Tx, \tilde{A} = TAT^{-1}, \tilde{B} = TB, \tilde{B}_d = TB, \tilde{C} = T^{-1}C, \tilde{D} = D$, where T is computed using reachability and observability Gramians of $G(s)$. It is important to highlight that $\tilde{x}$ does not preserve the physical meaning of x, because $G(s)$ is not unique.	

Figure 2 shows the steady state concentration profile of the nonlinear, linear and the reduced order models for CH_4, H_2O, CO, CO_2, and H_2 in the emulsion phase. In Figure 2, it can be observed that the reduced order model with only 11 states provides a good approximation of the linear and the nonlinear model.



(a) (b)

Figure 2: Models comparison: concentration of CH_4, H_2O, CO, CO_2, and H_2 in the emulsion phase in steady state for: (a) the nonlinear (–) and linear (△,▽,▷,◁,○) models. (b) Linear (–) and reduced order (△,▽,▷,◁,○) models.

4.2. PID Control

We present the PID control design in Table 2. As the PID controller is a SISO control technique, the Relative Gain Array ($\Lambda(j\omega)$) method is used in this work to calculate the best input-output pairing for the process.

Table 2: Mathematical formulation of the PID including RGA

Model	PID
$y(s)=\tilde{G}(s)u(s)$ $\Lambda(j\omega)=\tilde{G}(j\omega)*\tilde{G}(j\omega)^{-T}=\begin{bmatrix}-0.0028 & 1.0028\\ 1.0028 & -0.0028\end{bmatrix}$	$u_i(s)=\left(K_p+\frac{K_i}{s}+K_d s\right)e_i(s)$ $e_i(s)=y_{ref}-y_i(s)$

Based on the RGA the input-output pairing must be $T_J-X_{CH_4,out}$ and $F-C_{H_2,out}$. Additionally, the transfer function $\tilde{G}(s)$ is identified based on the step response of the reduced order model. The results of this controller are presented in Section 4.4.

4.3. Model Predictive Control (MPC)

MPC has been widely adopted by the industrial process control community and has been implemented successfully in many applications. MPC can handle constraints, which often have a significant impact on the quality and safety in process operations. To design predictive controllers, a discrete time model of the process must be obtained. To this end, a Zero-Order Hold discretization method is used. In Table 3, the discrete time version of the reduced order model is described.

Table 3: Mathematical formulation of the discrete time model

Reduced Order Model	Discrete Time Model
$\tilde{G}(s):\begin{cases}\dot{\tilde{x}}=\tilde{A}\tilde{x}+\tilde{B}u+\tilde{B}_d d\\ y=\tilde{C}x+\tilde{D}u\end{cases}$ $\xrightarrow{ZOH}$	$\tilde{G}(z):\begin{cases}\tilde{x}_{x+j+1}=\tilde{A}_k\tilde{x}_{k+j}+\tilde{B}_k u_{k+j}+\tilde{B}_{k,d}d_{k+j}\\ y_{k+j}=\tilde{C}_k\tilde{x}_{k+j}+\tilde{D}_k u_{k+j}\end{cases}$
The notation $k+j\triangleq(k+j)\Delta t$ where Δt is the sampling time	

The MPC control problem can be written as,

$$u_{MPC}=\arg\min_{\{u_{k+j}\}_{j=0}^{j=H_p-1}}\left\|x_{k+H_p}\right\|_P^2+\sum_{j=0}^{H_p-1}\left\|x_{k+j}\right\|_Q^2+\left\|u_{k+j}-u_{opt}\right\|_R^2$$

Subject to: (6)

$$\tilde{x}_{x+j+1}=\tilde{A}_k\tilde{x}_{k+j}+\tilde{B}_k u_{k+j}+\tilde{B}_{k,d}d_{k+j},\ y_{k+j}=\tilde{C}_k\tilde{x}_{k+j}+\tilde{D}_k u_{k+j},\ x_{k+j}\in\mathbf{X}_k,\ u_{k+j}\in\mathbf{U}_k$$

where H_p is the prediction horizon, and $\mathbf{X}_k$ and $\mathbf{U}_k$ are convex polyhedrons. The optimization problem (6) can be solved efficiently converting Eq. (6) into a Quadratic Programming (QP) problem.

4.4. MPC and PID comparison

In order to test the performance of the controllers for the FBMR, two experiments have been carried out: step changes in the disturbances of the process at t = *220s*, and different initial conditions. Figure 3 shows the comparison between the MPC and PID controllers.

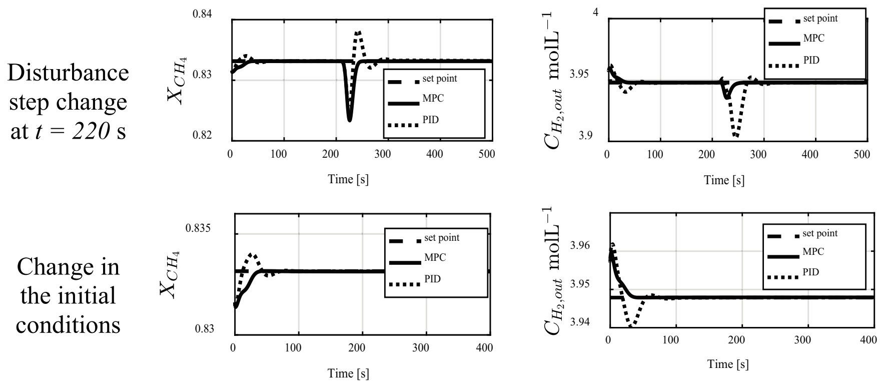


Figure 3: Controllers Comparison: Closed-loop trajectories of the outputs, MPC (—) and PID (···) controllers.

Clearly, from Figure 3, the performance of the MPC is better than the one of PID. Of course, solving an MPC problem in real time is more complex; however, the constraint handling is an advantage. In this case, the inlet flow rate should be kept above minimum fluidization velocity.

5. Conclusions

In this paper, the optimal operation and control of an FBMR for SMR are studied. A dynamic model of the process is obtained, and an offline dynamic optimization problem is proposed to maximize the conversion of methane and separation factor in the FBMR. Based on the optimal operation, and the dynamic model, a reduced order linear model is developed using balance truncation methods. Finally, PID and MPC controllers are designed, and their performance tested in simulations considering variation in the inlet concentration of the feed to the reactor and initial conditions.

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