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Destruction of volatile organic compounds by the optimal multivariable control of a catalytic reverse flow reactor

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Summary

This paper aims to compare 2 optimal control laws for a catalytic reverse flow reactor (RFR), which topic has not often been tackled. The RFR aims to destruct volatile organic compounds (VOCs), which are noxious products and are the source of a lot of problems: pollution, acid rains, woods wasting, green house effect and health hazards. Due to the public regulations, the VOCs discharge in the atmosphere becomes therefore more and more limited. The RFR is characterized by a periodic cycle of treatment of the VOC flow, allowing trapping the heat of reaction inside the core of the RFR. This allows using the RFR in an autothermal mode. The control problem tackled here is to maintain the RFR in this mode, such that the RFR extinction is avoided, and such that the catalyst elements are not destroyed. Effects of the influencing stochastic variations of the inlet pollutant concentration (the input disturbance) have to be accounted for by the optimal constrained tuning of the dilution rate and the internal electric heating. Optimal closed-loop performances of a linear quadratic regulator (LQR) and a model predictive controller (MPC) are compared through simulations.

Keywords

Destruction of COV, reverse flow reactor, optimal control.

INTRODUCTION

The catalytic RFR aims to destruct volatile organic compounds. These VOCs are noxious products and they are the source of a lot of environmental problems: pollution, acid rains, woods wasting, greenhouse effect and health hazards. The catalytic reverse flow reactor used here allows high temperatures in catalyst bed whereas the inlet and outlet gas stream temperatures are close to ambient temperature. Indeed, through periodic flow reversal, the heat released by the reaction is first trapped in the packing and is then used to heat up the feed when the flow direction is reversed. Because of the high efficiency of heat exchange between gas and solid phases, autothermal operation is possible even for a feed with a low adiabatic temperature rise (below 15 K). These features make therefore RFR highly competitive for VOCs combustion.

The RFR is modeled by a nonlinear dynamic partial differential equation (PDE) system

characterized by complex nonlinearities in the spatial domain (Edouard *et al.*, 2004). Explicitly, transport reaction phenomena with significant diffusive and convective phenomena are typically characterized by severe nonlinearities and spatial variations, and are naturally described by partial differential equations. Examples of such processes include tubular reactors, packed bed reactors, absorption columns, drying or curing processes.

In control theory, due to the complexity of the problem, relatively few studies are devoted to the control of processes explicitly characterized by a PDE model, especially in the nonlinear case and even with only one spatial dimension. Even if various methods are proposed to control such distributed parameter systems, there is no general framework yet. In order to implement, with a computer, a low order model based controller, the original PDE model is usually simplified into an ordinary differential equation (ODE) model. Such a finite dimensional approximation is based on the finite differences method, the finite volume method, the orthogonal collocation method or the Galerkin's method. Other works utilized properties of the initial PDE system before finite dimension controller synthesis. In a previous work, the single input single output (SISO) control of the RFR was treated (Dufour *et al.*, 2003a). The aim was to control the RFR such that the outlet gas concentration released in the atmosphere was maintained below a maximum level fixed by public regulations. This control strategy was based on a parabolic PDE model, an internal model control (IMC) structure and a MPC framework (Dufour *et al.*, 2003b). According to various regimes, it was shown that the proposed controller was able to tune correctly the control action, i.e. the heating power at the core of the reactor. In the meantime, for another particular regime (rich feed), the temperature inside the reactor was exceeding a threshold temperature that reflects the deterioration of the catalytic elements. Moreover, the input disturbance was assumed to be relatively constant and measured, whereas unmeasured and large stochastic variations need to be accounted for in reality. The aim of this paper is to provide a multivariable control framework to solve all of these problems. This requires modifying the PDE model and the control problem to account for a new second manipulated variable: the cooling action.

In this paper, two model-based control strategies for the constrained optimal control of the RFR are compared: LQR and MPC which both use a high-gain observer. The LQR is used here in a classical framework (Levine, 1996). MPC was developed for ordinary differential equation models and is well dedicated to solve a constrained problem (Morari and Lee, 1999). The IMC-MPC approach (Dufour *et al.*, 2003b) is used here. The paper is structured as follows: in Section 2, the RFR and its PDE model are briefly presented. Section 3 aims to remind the observer previously designed. Section 4 deals with the LQR and MPC strategies used here. Simulation results given allow comparing the performances of the LQR and the MPC for this process.

PROCESS DESCRIPTION

A medium-scale RFR (Nieken *et al.*) is considered (Figure 1). Cordierite monoliths of square cross sections with channels of 1*1 mm are packed in the reactor. Monolith in the core region is catalytically active and is inert in both end sections. A blower located downstream of the RFR keeps aspiration of the pollutant at a constant flow rate. In the core region, an electric heater maintains ignition temperature, while the temperature in

the catalytic layer is decreased by fresh air dilution. The packed layer is adiabatic, except in the core region where heat loss is inevitable due to both the installation for air dilution and the high temperature in this region. High temperatures exist in catalyst bed whereas the inlet and outlet gas stream have ambient temperature. Indeed, through periodic flow reversal, heat released by reaction is first trapped in the packing and then used to heat up the feed.

The model considered here for control purpose is obtained from a counter current pseudo-homogeneous model (Edouard *et al.*, 2004), accounts for mass transfer limitation and periodic frequency correction: it features one nonlinear parabolic PDE, two algebraic equations, and nonlinear boundary conditions. The nonlinearities are due to the cooling action. The advantages of this model are that it is more accurate and faster to compute than a previous model used for control (Dufour *et al.*, 2003a).

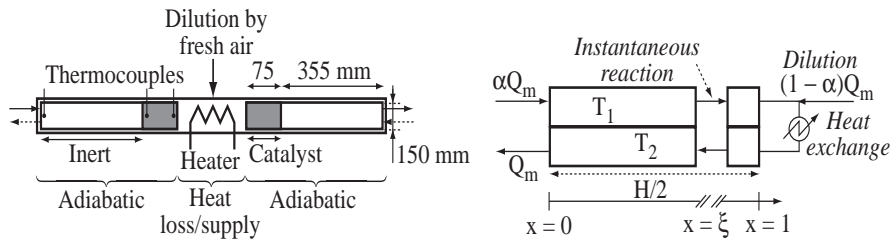


Figure 1: RFR description (left) and RFR counter current model (right).

This countercurrent reactor model is a heterogeneous model. In order to homogenize and simplify this model, the following model has been introduced by (Edouard *et al.*, 2004). It is based on the method described in (Balakotaiah and Dommeti, 1999) and it assumes that the kinetic reaction can be neglected under strong mass transfer limitation. It allows obtaining the following pseudo-homogeneous model: it features one PDE, two algebraic equations to account for mass transfer limitation, and a periodic frequency correction. Normalizing some variables, we obtain:

$$\begin{cases} \tau \frac{\partial T_s(x,t)}{\partial t} = \left(\frac{1}{P_{ax_\theta}} + \frac{1+\alpha^2}{2P_\theta} \right) \frac{\partial^2 T_s(x,t)}{\partial x^2} + \left(\frac{1-\alpha}{2} \right) \frac{\partial T_s(x,t)}{\partial x} + P_\theta \Delta T_{ad}(t) \psi(x,t) \\ \alpha \frac{\partial \omega_1(x,t)}{\partial x} + P_\theta \omega_1(x,t) = 0 \\ - \frac{\partial \omega_2(x,t)}{\partial x} + P_\theta \omega_2(x,t) = 0 \end{cases} \quad (1)$$

Where $x = \frac{z}{H/2}$ is the normalized space variable, t is the time variable, T_s is the temperature inside the solid parts, w_1 and w_2 are the VOC mass fraction in the upstream and downstream monoliths. Some model parameters are also parameterized by the period of flow reversal θ (assumed constant):

$$\begin{cases} P_\theta = \frac{ha_c H}{2\rho_0 \mu_{v0} c_{pmg}} \left(1 - \frac{\theta}{2\tau}\right) \\ \frac{1}{P_{ax_\theta}} = \frac{2\lambda_{ax}^s}{(H\rho_0 \mu_{v0} c_{pmg}) \left(1 - \frac{\theta}{2\tau}\right)} \\ \tau = \frac{(1-\varepsilon)\rho_s c_{ps} H}{2\rho_0 \mu_{v0} c_{pmg}} \end{cases} \quad (2)$$

And the non constant coefficients depend on the VOC mass fraction $\omega_{10}(t)$ fed into the RFR:

$$\begin{cases} \Delta T_{ad}(t) = \frac{\Delta H}{Mc_{pmg}} \omega_{10}(t) \\ \psi(x,t) = \varphi(x) \frac{\omega_1(x,t) + \omega_2(x,t)}{2\omega_{10}(t)} \end{cases} \quad (3)$$

where $\varphi(x)$ accounts for the type of monoliths: $\varphi(x)=0$ in the inert monoliths ($x < \xi_\theta$) and $\varphi(x)=1$ in the catalytic monoliths ($x \geq \xi_\theta$).

The boundary conditions for (1) are at $x=0$:

$$\begin{cases} \omega_1(x,t) = \omega_{10}(t) \\ T_{g1}(x,t) = T_s(x,t) - \frac{\alpha}{P_\theta} \frac{\partial T_s(x,t)}{\partial x} = T_0 \end{cases} \quad (4)$$

and at $x=1$:

$$\begin{cases} (1+N)(T_{g2}(x,t) - T_0) = \alpha(T_{g1}(x,t) - T_0) + \frac{Q_j}{S\rho_0 \mu_{v0} c_{pmg}} \\ T_{g1}(x,t) = T_s(x,t) - \frac{\alpha}{P_\theta} \frac{\partial T_s(x,t)}{\partial x} \\ T_{g2}(x,t) = T_s(x,t) + \frac{1}{P_\theta} \frac{\partial T_s(x,t)}{\partial x} \\ \alpha\omega_1(x,t) = \omega_2(x,t) \end{cases} \quad (5)$$

The initial conditions for (1) are at $t=0$:

$$T_{g1}(x,t) = T_{g2}(x,t) = T_s(x,t) = T_0 \quad (6)$$

In the above equations, T_0 is the ambient temperature and the feed temperature and T_{g1} and T_{g2} are upstream and downstream gas temperatures respectively. More details about the model parameter may be found in (Edouard *et al*, 2005). The cooling manipulated variable α is accounted for at any x , and introduces a strongly non-linear behavior. The dilution rate $(1-\alpha)$ is the percentage of fresh air in downstream flow. The heating power manipulated variable Q_j (in watts) is accounted for in the boundary condition at $x=1$.

OPTIMAL CONTROL PROBLEM

According to the operating conditions, various problematic behaviors can take place during the operation of the RFR:

- When the feed of pollutant concentration $\omega_{10}(t)$ (or $\Delta T_{ad}(t)$ as seen in (3)) is too rich, the release of heat due to the reaction produces thermal overheating that deteriorates the catalysts. The temperature inside the reactor has therefore to be maintained under the maximum temperature specified as 600 K (Ramdani *et al.*, 2001).
- If the feed of pollutant concentration $\omega_{10}(t)$ is too lean, low heat released during the reaction leads to the extinction of the reactor. The temperature inside the reactor has therefore to be maintained over the lowest temperature specified as 450 K (Ramdani *et al.*, 2001).
- Ideal operation of such reactor is finally an operation without control. This is possible when the feed concentration is such that the reaction temperature falls within an envelope outlined by the two previous boundary temperatures. This ideal case is called autothermal operation and no control action is required.

Except for the autothermal case, the controller to design has to compensate the influence of the input disturbance $\Delta T_{ad}(t)$ while preserving the stability of the system. On the other side, most of the reaction takes place at the inlet of the catalytic monoliths and is instantaneous (Nieken *et al.*, 1995; Ramdani *et al.*, 2001). Therefore, instead of the full temperature profile, only T_s at the entry z_1 of the catalyst zone is constrained:

$$450K < T_s(z_1, t) < 600K \quad \forall t \quad (7)$$

Concerning the input disturbance $\Delta T_{ad}(t)$, it varies between 0 and 30 K in real industrial use. But in order to prevent any accident, worst cases have to be evaluated. It is therefore assumed to vary randomly between 0 and 115 K (Fig. 2) and if no control is applied to the RFR, the hot-spot temperature cannot be maintained between both temperature limits (450 and 600 K). This clearly justifies the need for closed loop control in order to maintain the hot spot temperature within these 2 boundaries. Moreover, the stochastic input disturbance handled here has a much more realistic stochastic behavior than in the previous RFR control studies (Budman *et al.*, 1996, Dufour *et al.* 2003). This disturbance and the temperature profile in the RFR are estimated on-line using a high gain observer based on three temperatures measurements (Edouard *et al.*, 2004). The entire estimate state is injected in the LQR, whereas the MPC is only based on the estimated input disturbance. Simulation results allow comparing the closed-loop performances obtained with LQR and MPC.

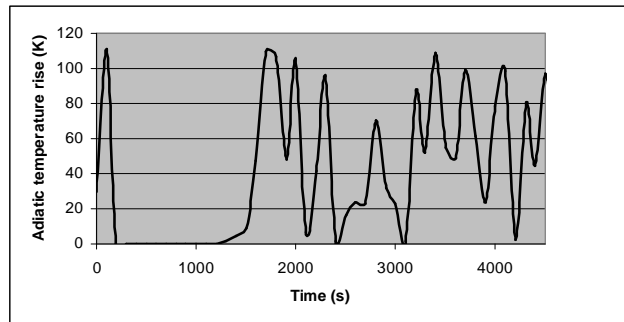


Figure 2: Adiabatic temperature rise $\Delta T_{ad}(t)$

COMPARISON OF TWO OPTIMAL CLOSED LOOP CONTROL APPROACHES

Since the hot spot temperature is constrained to be kept inside a specified region, some heating action may sometimes be required to avoid the RFR extinction. Some over times, some dilution may be required to avoid the over-heating of the catalyst elements, which would destroy them. From an optimal point of view, if constraints (7) are satisfied, no heating and no dilution are required.

LQR case

Approach used: The LQR is used here in a classical framework (Levine, 1996) where the two cost functions to minimize are given under the following:

$$J = \frac{1}{2} \sum_{k=0}^{\infty} [x'(k)Qx(k) + u'(k)Ru(k)] \quad (8)$$

with x is the process state vector, and u the manipulated variable vector. Q and R are the tuning parameters. The multivariable problem is decoupled here: when the feed of pollutant concentration is too rich, the solution of a first LQR is used for the minimum tuning of the dilution rate and no heating is possible. If the feed of pollutant concentration is too lean, the solution of another LQR is used to heat the system as less as possible, while no dilution takes place. Otherwise, no dilution and no heating are applied.

Simulation results: The output constraints (7) are satisfied at any time (Figure 3). Between 500s and 1550s, the low input disturbance (Figure 2) leads to a decrease of the temperature inside the RFR (Figure 3). LQR correctly tunes the internal heating (Figure 4) such that the temperature stays above the extinction temperature. No dilution is taking place until this time. After 1550s, rich feed (Figure 2) induces an increase of temperature inside the RFR. LQR then tunes the dilution rate such that the temperature is maintained below the maximum temperature and there is no more heating.

MPC case

Approach used: a special MPC was used here for such complex nonlinear PDE model and (Dufour *et al.*, 2003b). The constrained optimal control problem to solve is under the form (k is the actual time, j is the future time index, Np is the prediction horizon):

$$\min_{u(k)} J = \sum_{j=k+1}^{j=k+Np} (f(y_p(j), u)) \quad (9)$$

under some constraints :

$$u_{\min} \leq u \leq u_{\max}$$

$$y_{\min} \leq y_p \leq y_{\max}$$

Compared to the LQR case, J is here specified as a real multivariable cost function since it aims to minimize, at the same time, both dilution and heating action under the input/output constraints (7).

Simulation results: The output constraints (7) are also satisfied at any time (Figure 5). Between 0s and 1300s, ΔT_{ad} is small (Figure 2) and extinction of the process is avoided feeding electrical power into the reactor (Figure 6). In the meantime, there is no cooling action and the maximum amount of gas is therefore treated as expected in these conditions. After 1500s, ΔT_{ad} becomes important and overheating of the process is avoided (see the upper bound constraint on Figure 5) due to the correct use of the cooling action. The drawback is that the controller may sometimes require both heating and cooling actions at the same time (at 3180s e.g.), which should not happen.

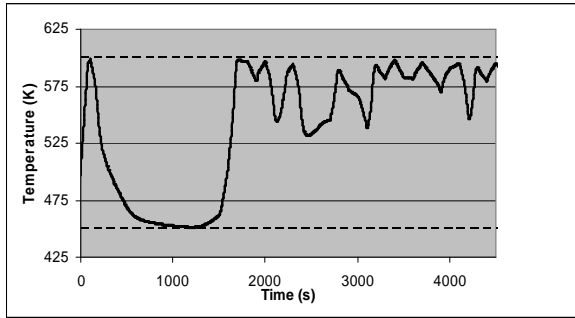


Figure 3: Hot spot temperature inside the RFR (LQR case).

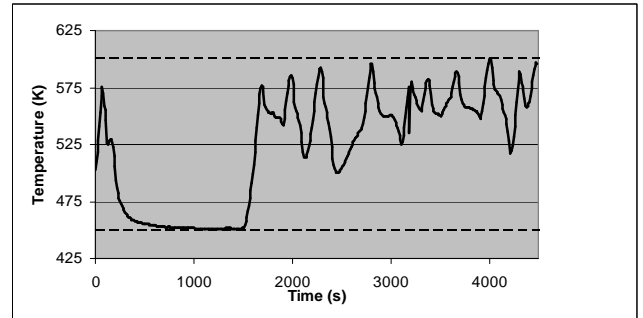


Figure 5: Hot spot temperature inside the RFR (MPC case).

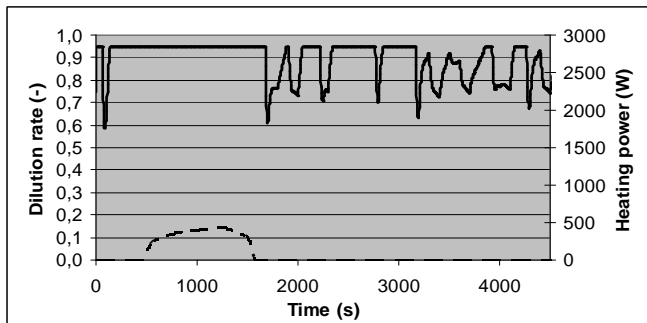


Figure 4: Cooling action by dilution (top) And heating action (bottom) (LQR case).

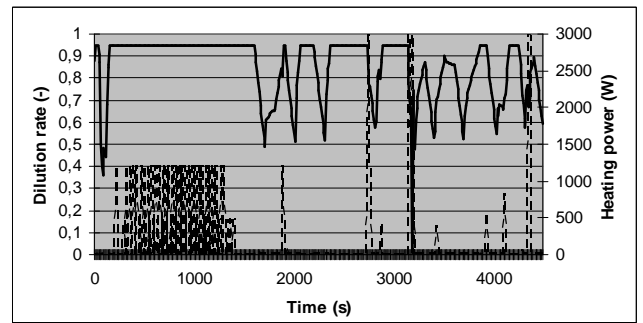


Figure 6: Cooling action by dilution (top) And heating action (bottom) (MPC case).

CONCLUSIONS ET PERSPECTIVES

In spite of a large influencing input disturbance due to the feed concentration, the temperature can be maintained by both observer based controllers inside the specified temperature envelope. Concerning the optimization performances, LQR leads here to better results than MPC since it requires less heating action while treating more gas: $\bar{Q}=83.4\text{W}$ and $\bar{\alpha}=0.894$ for LQR, $\bar{Q}=274.6\text{W}$ and $\bar{\alpha}=0.849$ for MPC (these mean values are calculated from $t=0\text{s}$ to $t=4500\text{s}$). This difference is mostly due to the impact of the stochastic variations of the input disturbance over MPC. Indeed, the estimation of the disturbance ΔT_{ad} is directly used in the MPC, where it is assumed constant in the future. This may force the MPC to over evaluate the need for heating and cooling. Combining this issue with the non minimum phase behavior of the process, the horizons tuning of the MPC is uneasy. In the meantime, a switching control structure is used for the LQR: therefore, heating and cooling action are explicitly decoupled and can not act at the same time as expected. The drawback is that this decoupling

introduces a severe nonlinearity for stability analysis whereas the explicit multivariable MPC used here is more suitable than the LQR for general MIMO control problems. Moreover, in the MPC, output constraints are explicitly handled. Perspectives are concerned with the use of such tools for a new coupled thermal-catalytic process located in the LMSPC, using new catalyst products. Observation issue will also be tackled.

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