



Studies on Nonlinear Dynamics and Control of a Tubular Reactor with Recycle [★]

Charalambos Antoniadis and Panagiotis D. Christofides

*Department of Chemical Engineering, University of California, Los Angeles, CA
90095, pdc@seas.ucla.edu*

Abstract

This paper presents a study on the dynamics and control of a tubular reactor with recycle loop where an exothermic reaction of the form $A \rightarrow B$ takes place. Using a nonlinear distributed parameter model for the process which accounts for diffusion, convection and chemical reaction, and recycle loop dead-time, it is found that the use of the recycle loop reduces the maximum temperature inside the reactor, maintains the desired rate of production of the species B , and introduces a feedback mechanism into the process which renders the open-loop steady state unstable. To stabilize the process, a nonlinear output feedback controller is implemented on the process. The nonlinear controller is synthesized based on an approximate model obtained through application of Galerkin's method with approximate inertial manifolds (nonlinear Galerkin's method) to the detailed process model. The performance of the controller for different values of the recycle ratio is successfully tested through simulations and is found to be superior to the one achieved by nonlinear controllers synthesized based on approximate models obtained from linear Galerkin's method and nonlinear controllers which do not account for the presence of dead-time in the recycle loop.

1 Introduction

Tubular reactors are widely used for the production of a variety of industrial products and are characterized by strong coupling of diffusive, convective and reactive mechanisms. This coupling is the source of the rich open-loop dynamic behavior exhibited by tubular reactors including multiple steady states, traveling waves, and periodic, quasi-periodic and chaotic behavior; the reader may refer to [1,2] and the classic paper [3] for results and references in this area.

In tubular reactors where highly exothermic reactions take place, a typical feature is the occurrence of a small region inside the reactor where the temperature is very high (usually referred to as 'hot-spot'). One way to reduce the

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'hot-spot' temperature is to use a recycle loop around the reactor that returns the unreacted reactant back to the reactor. The use of recycle loop typically results in a significant reduction of the 'hot-spot' temperature, while it allows maintaining the reactor conversion at the desired level. This is possible because the recycle loop reduces the fresh material fed into the reactor while increasing the residence time of the fresh material inside the reactor. Unfortunately, the use of recycle loop introduces a feedback mechanism into the process and brings the reactor closer to the verge of unstable behavior; thereby implying the need to operate tubular reactors with recycle under feedback control. The dynamics of tubular reactors with recycle in which the diffusive phenomena are negligible compared to the convective ones and a highly exothermic reaction takes place have been extensively studied in a series of papers written by Berezowski [4–9] (see also [10,11]) and the occurrence of multiple steady states and chaotic behavior has been established.

When diffusive and convective phenomena are equally important and the recycle loop dead-time is explicitly included in the process model, tubular reactors with recycle are modeled by systems of parabolic partial differential difference equations (PDDEs). The main feature of such systems is that the eigenspectrum of the parabolic spatial differential operator can be partitioned into a finite-dimensional slow one and an infinite-dimensional stable fast complement, which implies that the dominant dynamics of these systems can be approximately described by a small number of degrees of freedom. Motivated by this, we recently proposed a general methodology [12] for the synthesis of nonlinear output feedback controllers that guarantee stability and enforce output tracking in the closed-loop system. The key step of this method is the use of a nonlinear model reduction scheme which is based on combination of Galerkin's method with the concept of approximate inertial manifold is employed for the derivation of differential difference equation (DDE) systems that describe the dominant dynamics of the PDDE system. These DDE systems are then used as the basis for the explicit construction of nonlinear output feedback controllers through combination of geometric and Lyapunov techniques. The controllers enforce the stability and tracking in the closed-loop system independently of the size of the state delay.

This paper presents a study on the dynamics and control of a tubular reactor with recycle loop where an exothermic reaction of the form $A \rightarrow B$ takes place. Initially, a detailed mathematical model for the process which consists of two nonlinear PDDEs and accounts for diffusion, convection and chemical reaction, as well as for dead-time in the recycle loop is presented. Then, the dynamics of the process for different values of the recycle ration is studied and is found that the introduction of the recycle loop reduces the 'hot-spot' temperature inside the reactor, while maintaining the desired rate of production of the species B . In addition, the use of the recycle loop introduces a feedback mechanism into the process which renders the open-loop steady state unstable. To stabilize the

process, we use the method developed in [12] to synthesize a nonlinear output feedback controller based on an approximate DDE model obtained through application of nonlinear Galerkin's method to the detailed process model. The performance of the controller for different values of the recycle ratio is successfully tested through simulations and is found to be superior to the one achieved by nonlinear controllers synthesized based on approximate models obtained from linear Galerkin's method and nonlinear controllers which do not account for the presence of dead-time in the recycle loop.

2 Tubular reactor with recycle: Description and modeling

We consider a non-isothermal tubular reactor without catalyst packing, shown in Figure 1, where an irreversible first-order reaction of the form $A \rightarrow B$ takes place. The reaction is exothermic and a cooling jacket is used to remove heat from the reactor. The outlet of the reactor is fed into a separator where the

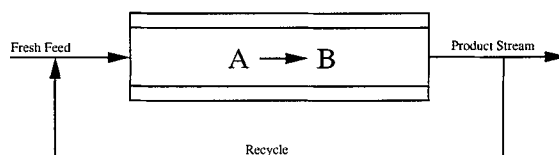


Fig. 1. A tubular reactor with recycle

unreacted species A is separated from the product B . The unreacted amount of species A is then fed back to the reactor through a recycle loop. Under the standard assumptions of constant density (ρ) and heat capacity (c_p) of the reacting fluid, and constant axial fluid velocity (v), the dynamic model of the process can be derived from mass and energy balances and takes the form:

$$\begin{aligned} \frac{\partial T}{\partial t} &= -v \frac{\partial T}{\partial \bar{z}} + \frac{k}{\rho c_p} \frac{\partial^2 T}{\partial \bar{z}^2} + \frac{-\Delta H}{\rho c_p} \exp\left(-\frac{E}{RT}\right) C_A - \frac{h A_s}{\rho c_p} (T - T_c) \\ \frac{\partial C_A}{\partial t} &= -v \frac{\partial C_A}{\partial \bar{z}} + D_A \frac{\partial^2 C_A}{\partial \bar{z}^2} - k_o \exp\left(-\frac{E}{RT}\right) C_A \end{aligned} \quad (1)$$

where T and C_A denote the temperature and concentration of species A in the reactor, respectively, k and D_A are the thermal conductivity and mass diffusivity of the reacting fluid, respectively, k_o , E and $(-\Delta H)$ represent the pre-exponential constant, activation energy, and the heat of the reaction, respectively, h is the heat transfer coefficient between the reactor and the cooling jacket, A_s is the surface area of the reactor walls, and T_c is the jacket temperature. Assuming negligible reaction in the recycle loop and instantaneous mixing of fresh feed and recycle feed at the reactor inlet, the boundary conditions of the system of Eq.1 take the form:

$$\begin{aligned} \bar{z} = 0 : \quad \frac{\partial T}{\partial \bar{z}} &= \frac{\rho c_p v}{K} (T - (1-r)T_f - rT(L, t - \alpha)), \\ \frac{\partial C_A}{\partial \bar{z}} &= \frac{v}{D_A} (C_A - (1-r)C_{Af} - rC_A(L, t - \alpha)) \end{aligned}$$

$$\bar{z} = L : \quad \frac{\partial T}{\partial \bar{z}} = 0, \quad \frac{\partial C_A}{\partial \bar{z}} = 0 \quad (2)$$

where T_f and C_{Af} denote the inlet temperature and concentration of species A in the reactor, L is the length of the reactor, r is the recycle ratio and α is the recycle loop dead time. Note that r varies from zero to one, with one corresponding to total recycle and zero fresh feed and zero corresponding to no recycle. In order to simplify the presentation of our results, we introduce the following dimensionless variables:

$$\begin{aligned} t &= \frac{\bar{t}v}{L}, \quad z = \frac{\bar{z}}{L}, \quad Pe_1 = \frac{\rho c_p v L}{k}, \quad Pe_2 = \frac{vL}{D_A}, \\ x_1 &= \frac{T - T_0}{T_0}, \quad x_2 = \frac{C_A - C_{A0}}{C_{A0}}, \quad x_{1f} = \frac{T_f - T_0}{T_0}, \quad x_{2f} = \frac{C_{Af} - C_{A0}}{C_{A0}}, \\ u &= \frac{T_c - T_0}{T_0}, \quad \gamma = \frac{E}{RT_0}, \quad \beta_T = \frac{hA_s L}{C_{A0}}, \\ B_T &= \frac{-(\Delta H)C_{A0}}{\rho c_p T_0}, \quad B_C = \frac{k_0 \exp(-\frac{E}{RT_0})L}{v} \end{aligned} \quad (3)$$

to write the system of Eqs.1-2 in the following form:

$$\frac{\partial x_1}{\partial t} = -\frac{\partial x_1}{\partial z} + \frac{1}{Pe_1} \frac{\partial^2 x_1}{\partial z^2} + \beta_T (u(z, t) - x_1) B_T B_C \exp\left(\frac{\gamma x_1}{1 + x_1}\right) (1 + x_2)$$

$$\frac{\partial x_2}{\partial t} = -\frac{\partial x_2}{\partial z} + \frac{1}{Pe_2} \frac{\partial^2 x_2}{\partial z^2} - B_C \exp\left(\frac{\gamma x_1}{1 + x_1}\right) (1 + x_2) \quad (4)$$

$$z = 0, \quad \frac{\partial x_1}{\partial z} = Pe_1 (x_1 - (1 - r)x_{1f} - rx_1(1, t - \alpha)),$$

$$\frac{\partial x_2}{\partial z} = Pe_2 (x_2 - (1 - r)x_{2f} - rx_2(1, t - \alpha)); \quad (5)$$

$$z = 1, \quad \frac{\partial x_1}{\partial z} = 0, \quad \frac{\partial x_2}{\partial z} = 0$$

Furthermore, in order to simplify the computation of the eigenvalues and eigenfunctions of the spatial differential operator which will be used in our calculations, we insert the non-homogeneous part of the boundary conditions of Eq.5 into the differential equation and obtain the following parabolic partial differential difference equation (PDDE) model for the process:

$$\begin{aligned} \frac{\partial x_1}{\partial t} &= -\frac{\partial x_1}{\partial z} + \frac{1}{Pe_1} \frac{\partial^2 x_1}{\partial z^2} + B_T B_C \exp\left(\frac{\gamma x_1}{1 + x_1}\right) (1 + x_2) \\ &\quad + \beta_T (u(z, t) - x_1) + \delta(z - 0) ((1 - r)x_{1f} + rx_1(1, t - \alpha)) \end{aligned}$$

$$\frac{\partial x_2}{\partial t} = -\frac{\partial x_2}{\partial z} + \frac{1}{Pe_2} \frac{\partial^2 x_2}{\partial z^2} - B_C \exp\left(\frac{\gamma x_1}{1+x_1}\right)(1+x_2) + \delta(z-0)((1-r)x_{2f} + rx_2(1, t-\alpha)) \quad (6)$$

where $\delta(\cdot)$ is the standard Dirac function, subject to the homogeneous boundary conditions:

$$z = 0, \quad \frac{\partial x_1}{\partial z} = Pe_1 x_1, \quad \frac{\partial x_2}{\partial z} = Pe_2 x_2, \quad z = 1, \quad \frac{\partial x_1}{\partial z} = 0, \quad \frac{\partial x_2}{\partial z} = 0 \quad (7)$$

Finally, we present the solution to the eigenvalue problem of the spatial differential operator of the process i.e.,:

$$\mathcal{A}x = \begin{bmatrix} \mathcal{A}_1 x_1 & 0 \\ 0 & \mathcal{A}_2 x_2 \end{bmatrix} = \begin{bmatrix} \frac{1}{Pe_1} \frac{\partial^2 x_1}{\partial z^2} - \frac{\partial x_1}{\partial z} & 0 \\ 0 & \frac{1}{Pe_2} \frac{\partial^2 x_2}{\partial z^2} - \frac{\partial x_2}{\partial z} \end{bmatrix} \quad (8)$$

The solution of the eigenvalue problem for \mathcal{A}_i , where $i = 1, 2$, can be obtained by utilizing standard techniques from linear operator theory (see, for example, [13]) and is of the form:

$$\lambda_{ij} = \frac{\bar{a}_{ij}^2}{Pe_i} + \frac{Pe_i}{4}, \quad \phi_{ij}(z) = B_{ij} e^{Pe_i \frac{z}{2}} \left(\cos(\bar{a}_{ij} z) + \frac{Pe_i}{2\bar{a}_{ij}} \sin(\bar{a}_{ij} z) \right), \quad (9)$$

$$\bar{\phi}_{ij}(z) = e^{-Pe_i z} \phi_{ij}(z), \quad i = 1, 2, \quad j = 1, \dots, \infty$$

where λ_{ij} , ϕ_{ij} , $\bar{\phi}_{ij}$, denote the eigenvalues, eigenfunctions and adjoint eigenfunctions of \mathcal{A}_i , respectively. B_{ij} , \bar{a}_{ij} can be calculated analytically from the following formulas:

$$B_{ij} = \left\{ \int_0^1 \left(\cos(\bar{a}_{ij} z) + \frac{Pe_i}{2\bar{a}_{ij}} \sin(\bar{a}_{ij} z) \right)^2 dz \right\}^{-\frac{1}{2}}, \quad (10)$$

$$\tan(\bar{a}_{ij}) = \frac{Pe_i \bar{a}_{ij}}{\bar{a}_{ij}^2 - \left(\frac{Pe_i}{2}\right)}, \quad i = 1, 2, \quad j = 1, \dots, \infty$$

Remark 1: It is important to point out that our study focuses on tubular reactors in which the diffusion and convection phenomena in the axial direction are both important, and thus, the model of Eqs.6-7 constitutes a *parabolic* PDDE system. This feature, together with the explicit modeling of the dead-time associated with the recycle loop, distinguishes our work from previous studies on the analysis of the dynamics of tubular reactors with recycle [4–9].

3 Effect of recycle ratio on open-loop dynamics

The objective of this section is to study the effect of the recycle ratio on the behavior and dynamics of the tubular reactor. To this end, we use standard Galerkin's method to derive an accurate high-order discretization of the PDDE system of Eqs.6-7. Specifically, the first 200 eigenfunctions of Eq.9 were used as basis functions in Galerkin's method to discretize the process model in space and reduce it into a large set (400 equations) of differential difference equations (DDEs). The time-integration of the resulting set of DDEs was performed by utilizing explicit Euler. It was verified that further increase in the number of eigenfunctions and reduction in the step of time-integration results in negligible improvements in the accuracy of the computed solution. The following values for the process parameters were used in our calculations:

$$Pe_1 = 7.0, \quad Pe_2 = 7.0, \quad B_C = 0.1, \quad B_T = 2.5, \quad (11)$$

$$\beta_T = 2.0, \quad \gamma = 10.0, \quad r = 0.5$$

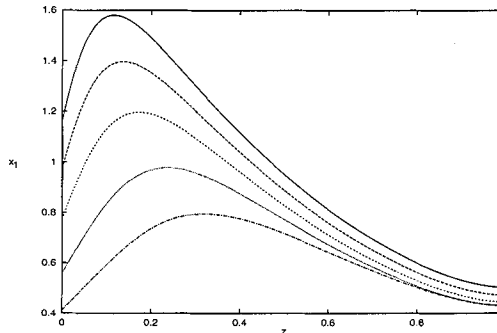


Fig. 2. Steady state profile of x_1 for different r . $r = 0.0$ (solid line), $r = 0.1$ (dashed line), $r = 0.2$ (small-dashed line), $r = 0.3$ (dotted line), and $r = 0.38$ (dashed-dotted line).

Since our objective is to study the effect of recycle ratio on open-loop dynamics and the behavior of the closed-loop system, the above parameters were chosen so that when the recycle loop is not used (i.e., $r = 0$), the open-loop system possesses a unique globally asymptotically stable spatially non-uniform steady state; this fact was verified both analytically and numerically through extensive simulations of the open-loop system for different initial conditions. Furthermore, for the above values of the process parameters, the diffusive and convective phenomena are equally important. Figure 2 (solid line) shows the resulting steady state profile of x_1 ; note the occurrence of a 'hot-spot' close to the inlet of the reactor. Subsequently, we introduced the recycle-loop to the process; the flow rate of the fresh feed to the process is reduced depending on the recycle flow rate so that the total flow rate to the reactor remains constant. Figure 2 shows the steady state profile of x_1 for various recycle ratios, $r = 0.1$

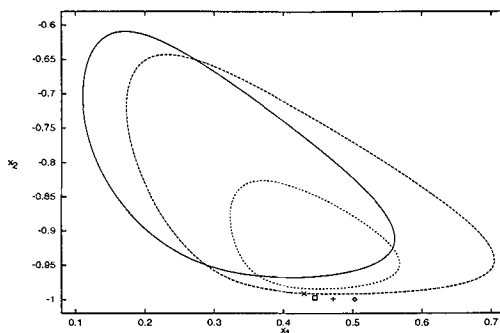


Fig. 3. Profile of $x_1(1, t)$ versus $x_2(1, t)$ for different values of r : $r = 0$ (\diamond), $r = 0.1$ ($+$), $r = 0.2$ (\square), $r = 0.3$ (\times) (stable operating points), $r = 0.4$ (solid line), $r = 0.45$ (solid line), $r = 0.5$ (small-dashed line), $r = 0.6$ (dashed line), and $r = 0.7$ (solid line) (stable limit cycles).

(dashed line), $r = 0.2$ (small-dashed line), $r = 0.3$ (dotted line), and $r = 0.38$ (dashed-dotted line), for which the process is stable.

As the recycle ratio increases, the amount of fresh feed entering the reactor decreases, and thus, the temperature of the ‘hot-spot’ inside the reactor decreases and the location of the ‘hot-spot’ moves towards the center of the reactor. We also observe that even though the ‘hot-spot’ temperature decreases, the use of the recycle loop allows maintaining the production rate of the process constant; a very important benefit from a practical standpoint. Unfortunately, as the recycle ratio increases, the stable steady state of the open-loop process gets closer to the instability limit. In particular, when the recycle ratio is equal to $r = 0.39$, the spatially non-uniform steady state becomes unstable and a globally asymptotically stable periodic (limit cycle) spatially non-uniform state appears. Figure 3 shows $x_1(1, t)$ versus $x_2(1, t)$ for different values of the recycle ratio. Clearly, when $r > 0.39$ the reactor moves to the periodic spatially non-uniform state; this can be also seen in Figure 4 where the open-loop spatiotemporal profile of x_1 for $r = 0.5$ is shown. The open-loop instability of the steady state that yields a very high production rate with relatively low ‘hot-spot’ temperature motivates implementing a nonlinear feedback controller on the reactor to enforce stability and achieve operation for $r > 0.39$.

4 Nonlinear control of a tubular reactor with recycle

In this section, we synthesize and implement a nonlinear output feedback controller on the tubular reactor with recycle. The controller is synthesized on the basis of the process model and guarantees exponential stability in the closed-loop system and enforces the controlled output to asymptotically follow the reference input, independently of the size of the recycle loop dead-time. Specifically, the following methodology is used for controller synthesis: Initially, a nonlinear model reduction scheme which is based on combination of Galerkin’s method with approximate inertial manifolds is employed for the derivation of differential difference equation (DDE) systems that describe the

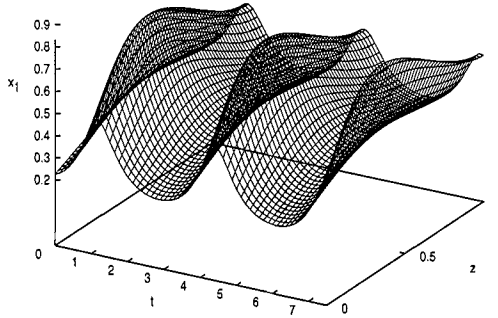


Fig. 4. Open-loop spatiotemporal profile of x_1 , $r = 0.5$.

dominant dynamics of the PDDE system of Eqs.6-7. Then, these DDE systems are used as the basis for the explicit construction of nonlinear output feedback controllers through combination of geometric and Lyapunov techniques. All the details of the model reduction and controller synthesis results including rigorous proofs are given in [12] and will be omitted in this note for brevity.

The control problem is to manipulate the jacket temperature to stabilize the reactor at a spatially-nonuniform steady state where the production of species B is desirable and the 'hot-spot' temperature is acceptable, for values of r for which the open-loop system is unstable. To achieve this control objective, the controlled output was defined as $y(t) = \int_0^1 e^{-Pez} \phi_{11}(z) x_1 dz$, and the manipulated variable was chosen as $u(z, t) = b(z)u(t)$ where the actuator distribution function was taken to be $b(z) = 1$ (uniform wall temperature in space). Furthermore, since the objective is stabilization at an unstable steady state, we assumed that there is available a large number of point measurements of the temperature throughout the reactor so that $y(t)$ is known with sufficient accuracy.

Two sets of simulation runs were performed to evaluate a) the improvement on the performance of the controller which is achieved when the controller is synthesized on the basis of DDE models derived from combination of Galerkin's method with approximate inertial manifolds, and b) compare the performance of the proposed nonlinear controllers with nonlinear controllers which are designed on the basis of approximate ODE models which do not account for the time delay in the recycle loop, for $r = 0.45$ and $r = 0.7$ (in both cases the open-loop system exhibits unstable (oscillatory) behavior). In all the simulation runs, the process was initially ($t = 0$) assumed to be at the unstable steady state, the time delay of the recycle loop to be $\alpha = 0.5$, and the reference input value was set at $v = 0.12$ (this is a value of $y(t)$ which corresponds to a steady state with the desired characteristics). Moreover, in order to perform meaningful comparisons, the same tuning parameters are used in the various controllers implemented on the reactor for $r = 0.45$ and $r = 0.7$. Following

the presentation in our previous work [12] and in order to simplify the presentation of the following results, we will denote the dimension of the DDE system used for controller design with, m , and the dimension of the approximate inertial manifold used to improve the accuracy of this model with \bar{m} . In

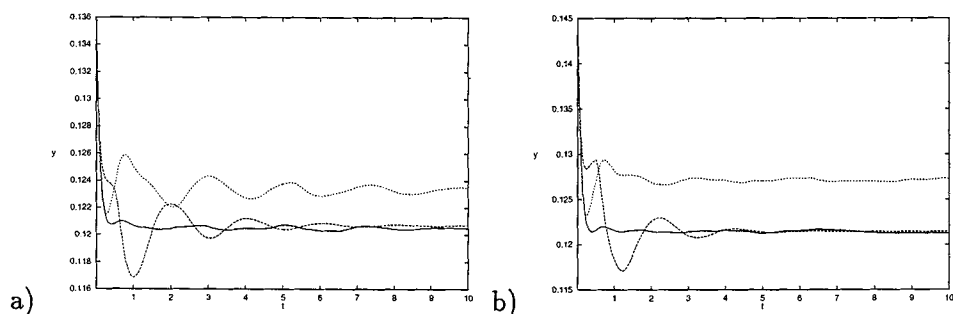


Fig. 5. Closed-loop output profiles of the process with a) $r = 0.45$ and b) $r = 0.7$, under a controller with $m = 22$, $\bar{m} = 24$ and $\alpha = 0.5$ (solid line), a controller with $m = 22$, $\bar{m} = 0$ and $\alpha = 0.5$ (short dashed line), and a controller with $m = 22$, $\bar{m} = 24$ and $\alpha = 0$ (long dashed line).

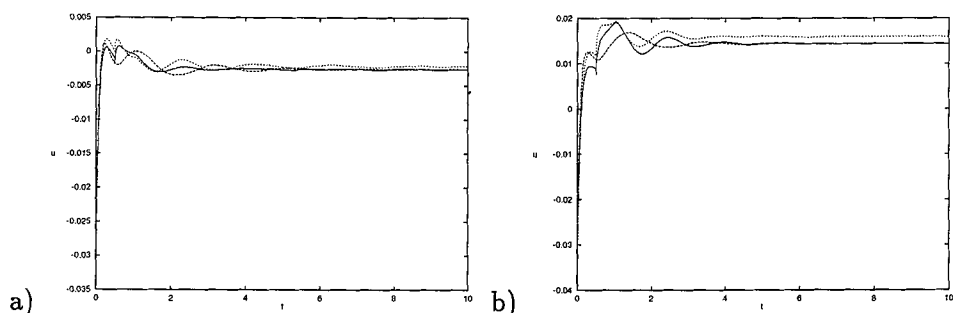


Fig. 6. Manipulated input profiles under a controller with $m = 22$, $\bar{m} = 24$ and $\alpha = 0.5$ (solid line), a controller with $m = 22$, $\bar{m} = 0$ and $\alpha = 0.5$ (short dashed line), and a controller with $m = 22$, $\bar{m} = 24$ and $\alpha = 0$ (long dashed line); a) $r = 0.45$ and b) $r = 0.7$.

the first set of simulations, we pick $r = 0.45$ which corresponds to an unstable open-loop state. In order to provide a basis for comparing the performance of controllers designed based on DDE models derived by using combination of Galerkin's method with approximate inertial manifolds, we first implemented on the process a nonlinear output feedback controller designed on the basis of a DDE model obtained using Galerkin's method with $m = 22$ and $\bar{m} = 0$ (we need a 22-th order DDE model in order to capture the dynamics of the system). Figures 5a, 6a and 7a (short dashed lines) show the evolution of the output of the closed-loop system, the profile of the manipulated input and the closed-loop steady state profile of x_1 , respectively. This controller exhibits a poor performance, stabilizing the system away from the desired steady state. Then, we used a 24-th order approximation for the approximate inertial manifold to improve the accuracy of the 22-th order DDE model obtained from

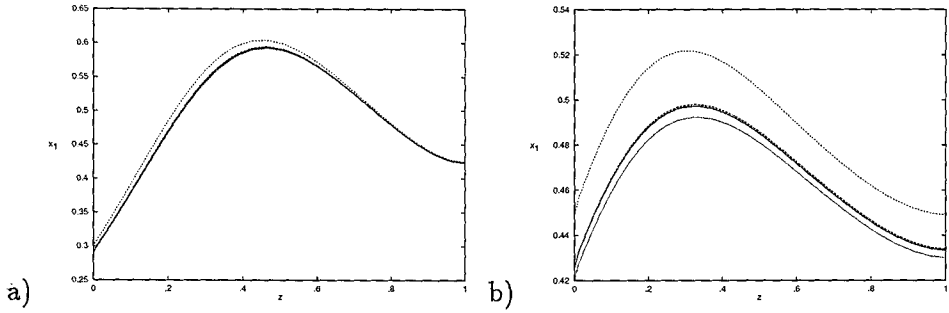


Fig. 7. Steady state profile of x_1 under a controller with $m = 22$, $\bar{m} = 24$ and $\alpha = 0.5$ (solid line), a controller with $m = 22$, $\bar{m} = 0$ and $\alpha = 0.5$ (short dashed line), a controller with $m = 22$, $\bar{m} = 24$ and $\alpha = 0$ (long dashed line), and profile of the desired operating steady state (dotted line); a) $r = 0.45$ and b) $r = 0.7$.

Galerkin's method (i.e., $m = 22$, $\bar{m} = 24$) and used the resulting model for the synthesis of a nonlinear output feedback controller. Figures 5a, 6a and 7a (solid lines) show the profiles of the controlled output, manipulated input and x_1 at steady state, respectively and Figure 8a shows the spatiotemporal evolution of x_1 . This controller clearly drives quickly the output very close to the reference input value, exhibiting a very good transient response. To

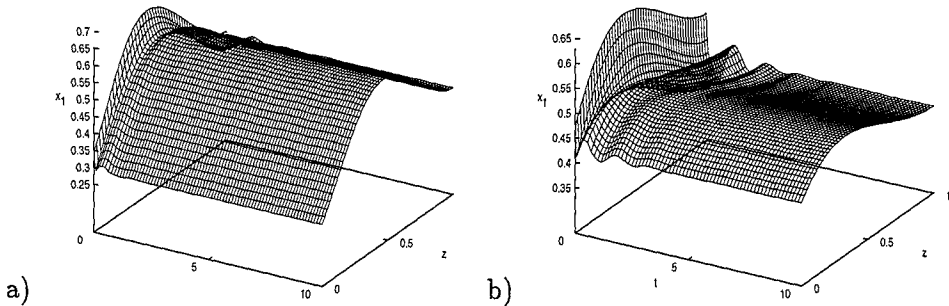


Fig. 8. Spatiotemporal evolution of x_1 under a controller with $m = 10$, $\bar{m} = 12$ and $\alpha = 0.5$; a) $r = 0.45$ and b) $r = 0.7$.

demonstrate the effect of the time delay on the closed-loop performance, we implemented on the process the nonlinear output feedback controller used in the previous simulation run with $\alpha = 0$ (i.e., the time-delay is not accounted for in the controller; the other controller parameters are the same as in the previous run). Figures 5a, 6a and 7a (long dashed lines) show the profiles of the controlled output, the manipulated input, and x_1 at steady state, respectively. This controller yields a very poor transient response before driving the output close to the new reference input value.

In the second set of simulations, we changed the recycle ratio of the process to $r = 0.7$; this value also corresponds to oscillatory open-loop behavior. As in the previous case, we first implemented on the process a nonlinear output feedback controller designed on the basis of a DDE model obtained using

Galerkin's method with $m = 22$ and $\bar{m} = 0$. Figures 5b, 6b and 7b (short dashed lines) show the evolution of the output of the closed-loop system, the profile of the manipulated input and the closed-loop steady state profile of x_1 , respectively. Again, this controller exhibits a poor performance, stabilizing the system away from the desired steady state. Then, we used a 24-th order approximation for the approximate inertial manifold to improve the accuracy of the 22-th order DDE model obtained from Galerkin's method (i.e., $m = 22$, $\bar{m} = 24$) and used the resulting model for the synthesis of a nonlinear output feedback controller. Figures 5b, 6b and 7b (solid lines) show the profiles of the controlled output, manipulated input and x_1 at steady state, respectively and Figure 8b shows the spatiotemporal evolution of x_1 . This controller clearly drives quickly the output very close to the reference input value, exhibiting a very good transient response. To demonstrate again the effect of the time delay on the closed-loop performance, we implemented on the process the nonlinear output feedback controller used in the previous simulation run with $\alpha = 0$ (i.e., the time-delay is not accounted for in the controller; the other controller parameters are the same as in the previous run). Figures 5b, 6b and 7b (long dashed lines) show the profiles of the controlled output, the manipulated input, and x_1 at steady state, respectively. This controller yields a very poor transient response before driving the output close to the new reference input value. From the simulation results, it is evident that the combination of Galerkin's method with approximate inertial manifolds leads to the synthesis of high-performance controllers based on low-order DDE systems, as well as the importance of accounting for the effect of time-delays in the controller design. We finally note that we have found similar results for many other values of $r > 0.39$; these results are not shown here for brevity.

Remark 2: Regarding the effect of the recycle-loop dead time on the open-loop dynamics and the performance of the closed-loop system, we found through simulations that: a) the stability of the spatially non-uniform steady state for $r \leq 0.39$ and the stability of the periodic state of the open-loop system for $r \geq 0.39$ are independent of the value of the dead time in the recycle-loop, α , b) the recycle ratio, $r = 0.39$, for which the globally stable open-loop steady state becomes unstable is independent of α , and c) the closed-loop response as the α increases becomes significantly more sluggish, which means that re-tuning of the controller is needed to achieve the desired rate of convergence to the steady state in the closed-loop system for larger α .

Remark 3: We note that the approach followed here for the construction of low-order DDE approximations of the model of Eqs.6-7 and the synthesis of nonlinear feedback controllers is not directly applicable to the case of tubular reactors in which the diffusive phenomena are negligible compared to the convective phenomena (like the reactors studied in [4–9]). The reason is that tubular reactors with negligible convective phenomena are modeled by first-order hyperbolic PDE systems in which the eigenvalues of the spatial differential operator cluster along vertical or nearly vertical asymptotes in the complex plane, and therefore, the controller synthesis problem has to

be directly addressed on the basis of the PDE system (see [14] and [15] for results on nonlinear control of hyperbolic PDEs and ‘hot-spot’ suppression in convection-reaction processes, respectively).

References

- [1] R. B. Root, R. A. Schmitz, An experimental study of steady state multiplicity in a loop reactor, *AIChE J.* 15 (1969) 670–679.
- [2] R. B. Root, R. A. Schmitz, An experimental study of unstable states in a loop reactor, *AIChE J.* 16 (1970) 356–358.
- [3] K. F. Jensen, W. H. Ray, The bifurcation behavior of tubular reactor, *Chem. Eng. Sci.* 37 (1982) 199–222.
- [4] M. Berezowski, A sufficient condition for the existence of single steady states in chemical reactors with recycle, *Chem. Eng. Sci.* 45 (1990) 1325–1329.
- [5] M. Berezowski, Method for analysing global stability of pseudohomogeneous chemical reactors with recycle, *Chem. Eng. Sci.* 46 (1991) 1781–1785.
- [6] M. Berezowski, Dynamic profiles of chemical reactors with recycle, *Chem. Eng. Sci.* 48 (1993) 2799–2806.
- [7] M. Berezowski, Stabilization of unstable steady states of adiabatic tubular reactors with recycle, *Chem. Eng. Sci.* 50 (1995) 1989–1996.
- [8] S. Subramanian, V. Balakotaiah, Classification of steady-state and dynamic behaviour of distributed reactor models, *Chem. Eng. Sci.* 51 (1996) 401–421.
- [9] M. Berezowski, Chaotic dynamics in homogeneous tubular reactors with recycle, *Chem. Eng. Sci.* 53 (1998) 4023–4029.
- [10] Y. V. Z., A. B. Rovinsky, M. Menzinger, Absolute stability of tubular packed bed reactor with recycle, *Chem. Eng. Sci.* 50 (1995) 1591–1593.
- [11] Y. V. Z., A. B. Rovinsky, M. Menzinger, Convective instability induced by differential transport in the tubular packed bed reactor, *Chem. Eng. Sci.* 50 (1995) 2853–2859.
- [12] C. Antoniadis, P. D. Christofides, Nonlinear feedback control of parabolic partial differential difference equation systems, *Int. J. Contr.* 73 (2000) 1572–1591.
- [13] W. H. Ray, *Advanced Process Control*, McGraw-Hill, New York, 1981.
- [14] P. D. Christofides, P. Daoutidis, Feedback control of hyperbolic PDE systems, *AIChE J.* 42 (1996) 3063–3086.
- [15] I. Karafyllis, P. Daoutidis, Control of hot spots in tubular reactors, in: *AIChE Annual Meeting*, paper 193i, Los Angeles, CA, 1997.