Journal of Process Control 23 (2013) 1508–1514

Contents lists available at ScienceDirect

Journal of Process Control

journal homepage: www.elsevier.com/locate/jprocont

Optimal control of an advection-dominated catalytic fixed-bed reactor with catalyst deactivation

I. Aksikas [a,*], L. Mohammadi [b], J.F. Forbes [b], Y. Belhamadia [c], S. Dubljevic [b]

[a] Department of Mathematics, Statistics and Physics, Qatar University, Doha, Qatar
[b] Department of Chemical and Materials Engineering, University of Alberta, Edmonton, Canada
[c] Campus Saint-Jean and Department of Mathematical and Statistical Sciences, University of Alberta, Edmonton, Canada

ARTICLE INFO

Article history:
Received 29 February 2012
Received in revised form
21 September 2013
Accepted 21 September 2013
Available online 31 October 2013

Keywords:
Fixed bed reactor
Infinite dimensional time-varying system
Linear quadratic optimal control
Catalyst deactivation

ABSTRACT

The paper focuses on the linear-quadratic control problem for a time-varying partial differential equation model of a catalytic fixed-bed reactor. The classical Riccati equation approach, for time-varying infinite-dimensional systems, is extended to cover the two-time scale property of the fixed-bed reactor. Dynamical properties of the linearized model are analyzed using the concept of evolution systems. An optimal LQ-feedback is computed via the solution of a matrix Riccati partial differential equation. Numerical simulations are performed to evaluate the closed loop performance of the designed controller on the fixed-bed reactor. The performance of the proposed controller is compared to performance of an infinite dimensional controller formulated by ignoring the catalyst deactivation. Simulation results show that the performance of the proposed controller is better compared to the controller ignoring the catalyst deactivation when the deactivation time is close to the resident time of the reactor.

1. Introduction

Catalytic fixed-bed reactors are the most widely used reactor type and play an important role in chemical industry. The temperature and concentrations in this type of reactor vary both with time and space. In order to capture the effects of reaction and transport phenomena in the reactor, the reactor model may take the form of a set of partial differential equations (PDEs). These systems are infinite dimensional in nature and are commonly referred to as distributed parameter systems (DPS). The catalyst in a fixed bed reactor deactivates during the operation for a variety of reasons (e.g., poisoning by impurities in feed, formation of coke on catalyst surface, etc). Incorporation of catalyst deactivation in the model of the reactor, results in a time-varying infinite dimensional system. Optimal operation of catalytic reactors in a chemical plant has important effect on profitability of the plant. Therefore, designing high performance controllers which are able to regulate the reactor temperature and concentrations around the optimal values during the operation time of the reactor is crucial for chemical processes.

A common approach for controlling the distributed parameter systems is to convert the set of PDEs to a set of ODEs using discretization techniques, for example finite difference or finite element methods. These discretization methods may result in models that do not capture all dynamic properties of the original system accurately. In order to have accurate finite dimensional models a very fine discretization is needed which results in high order state space systems leading to computationally demanding controllers.

In recent years, research on control of DPS has focused on methods that deal with infinite dimensional nature of these systems [1,2]. In the aforementioned work, distributed parameter systems are formulated in a state space form, similar to lumped parameter systems, by introducing a suitable infinite dimensional space setting and associated operators, which allows infinite dimensional controllers to be synthesized directly from the infinite dimensional realization of the system [1,3,4].

For diffusion–convection–reaction systems, which are described by parabolic PDEs, Christofides and co-workers studied nonlinear order reduction and control of nonlinear parabolic systems. Dubljevic et al. [5] also used modal decomposition to derive finite-dimensional systems that capture the dominant dynamics of the original PDE which are subsequently used for low dimensional controller design.

In the case of first order hyperbolic systems that model convection–reaction type of processes, the eigenvalues of the spatial differential operator cluster along vertical or nearly vertical asymptotes in the complex plane [6], which implies that the modal decomposition techniques suitable for parabolic PDEs cannot be used.

The optimal control of hyperbolic systems using spectral factorization which is based on frequency-domain description of the

* Corresponding author. Tel.: +974 44034615.
E-mail addresses: aksikas@qu.edu.qa, aksikas@ualberta.ca (I. Aksikas).

0959-1524/$ – see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.jprocont.2013.09.016

system is studied in [3,4]. As an alternative method, Aksikas et al. [7] studied the solution of LQ control problem for hyperbolic systems by solving an operator Riccati equation. Control of hyperbolic systems with time varying kinetics is studied in [8], but the approach is limited to plug flow reactors. Model of a plug flow reactor consists of a set of hyperbolic equations with identical convective terms, but in a fixed bed reactor the porosity of the bed results in two time scale system and the model of the reactor consists of a set of hyperbolic PDEs with distinct convective terms.

In this paper, the approach proposed by [8] is extended to general form of time-varying hyperbolic systems. Moreover, the conditions on the stability of these systems are explored using the Lyapunov stability. The closed loop performance of the LQ controllers is evaluated for the case of a Naphtha hydrotreating reactor with different values of rate of catalyst deactivation. Although, in a hydrotreating reactor diffusion may exist, for the purpose of this paper it is assumed that the diffusion is negligible. Existence of diffusion term results in Parabolic Partial Differential equations which represents completely deferent dynamic behaviour. Refer to [9] for solution of optimal control problem systems with significant diffusion. This example is used only for illustration purposes, and may not represent an industrial hydrotreating reactor.

2. Model description

The developments presented here are focussed on advection dominated reactors (i.e., those whose operation is characterized by large Peclet Numbers). The operation of many gas phase reactors with solid catalysts, and sufficiently high space velocities, are examples of such systems and are commonly modelled using a plug flow assumption. Some common advection dominated reactors include vehicle catalytic converters, which can contain wash coated and monolithic catalytic sections, and the reactors used in the destruction of fugitive volatile organic compounds, which often contain monolithic catalytic sections [10,11]. The dynamics of such fixed-bed reactors can be described by partial differential equations derived from mass and energy balances. To model these reactors, a plug-flow pseudo-homogeneous model in one-spatial (axial) dimension is used (i.e., no radial direction gradients).

2.1. Mathematical model

The process considered in this work is a fixed-bed, hydrotreating reactor with catalyst deactivation. In the simplified system considered here, a lumped reaction kinetics with a single olefin hydrogenation reaction was assumed and has the following form (see [12]):

$$A + H_2 \rightarrow C$$
$$r_A = k(t)\exp\left(-\frac{E}{RT}\right) C_A^{n_1} C_H^{n_2} \tag{1}$$

Given the modelling assumptions, the dynamics of the process are described by the following energy and mass balance partial differential equations (PDE's).

$$\epsilon\frac{\partial C_A}{\partial t} = -\nu\frac{\partial C_A}{\partial z} - \rho_B k(t)\exp\left(-\frac{E}{RT}\right) C_A^{n_1} C_H^{n_2}$$
$$\frac{\partial T}{\partial t} = -\nu\frac{\partial T}{\partial z} + \frac{\rho_B \Delta H}{\rho C_p} k(t)\exp\left(-\frac{E}{RT}\right) C_A^{n_1} C_H^{n_2} \tag{2}$$

with the boundary conditions given, for $t \geq 0$, by:

$$C_A(0,t) = C_{A,in},$$
$$T(0,t) = T_{in}. \tag{3}$$

The initial conditions are assumed to be given, for $0 \leq z \leq l$, by

$$C_A(z,0) = C_{A0}(z),$$
$$T(z,0) = T_0(z) \tag{4}$$

In the equations above, C_A, T, ϵ, ρ_B, ρ, C_p, E, C_H, ΔH, ν denote the reactant concentration, the temperature, the porosity of the reactor packing, the catalyst density, the fluid density, the activation energy, the concentration of hydrogen, the enthalpy of reaction, and the superficial velocity respectively. In addition, t, z and l denote the time, space and the length of the reactor, respectively. It can be assumed without loss of generality that $l = 1$. T_0 and C_{A0} denote the initial temperature and reactant concentration profiles, respectively, such that $T_0(0) = T_{in}$ and $C_{A0}(0) = C_{A,in}$. k is the pre-exponential factor. As a result of catalyst deactivation, the pre-exponential factor, k varies with time. Generally k is a function of time and operating conditions, however, here we assume that the operating conditions are maintained in narrow ranges. Therefore k is only a function of time and is assumed to be given by:

$$k = k_0 + k_1 e^{-\alpha t} \tag{5}$$

The above expression for kinetics of naphtha hydrotreating reaction is in agreement with the observations that after a rapid initial deactivation of hydrotreating catalyst there is a slow deactivation phase and finally a stabilization of catalyst activity phase [13].

2.2. Dimensionless model

Consider the following state transformation:

$$\theta_1 = \frac{T - T_{in}}{T_{in}}, \quad \theta_2 = \frac{C_{A,in} - C_A}{C_{A,in}} \tag{6}$$

Then we obtain the following equivalent representation of the model.

$$\frac{\partial \theta_1}{\partial t} = -\nu\frac{\partial \theta_1}{\partial z} + (h_0 + h_1 e^{-\alpha t})(1-\theta_2)^{n_1} e^{\frac{\mu\theta_1}{1+\theta_1}} \tag{7}$$

$$\frac{\partial \theta_2}{\partial t} = -\frac{\nu}{\epsilon}\frac{\partial \theta_2}{\partial z} + (l_0 + l_1 e^{-\alpha t})(1-\theta_2)^{n_1} e^{\frac{\mu\theta_1}{1+\theta_1}} \tag{8}$$

with the boundary conditions:

$$\theta_1(0,t) = 0, \quad \theta_2(0,t) = 0 \tag{9}$$

where

$$\mu = \frac{E}{RT_{in}}, \quad l_{0,1} = \frac{\rho_B}{\epsilon} k_{0,1} C_H^{n_2} C_{A_{in}}^{n_1} e^{-\mu}, \tag{10}$$

$$h_{0,1} = \frac{\rho_B(-\Delta H)}{\rho C_p T_{in}} k_{0,1} C_H^{n_2} C_{A_{in}}^{n_1} e^{-\mu} \tag{11}$$

2.3. Infinite-dimensional linearized model

Let us denote by θ_{ss} and u_{ss} the dimensionless profile of the model (7)–(8) at the operating point. By defining the following state variables:

$$x(t) = \theta(t) - \theta_{ss} \tag{12}$$

and new input $u(t) = \nu(t) - \nu_{ss}$, the linearization of the system (7)–(8) around its operating profile leads to the following linear time-varying infinite-dimensional system on the Hilbert space $H := L^2(0,1) \times L^2(0,1)$.

$$\begin{cases} \dot{x}(t) = A(t)x(t) + Bu(t), \\ x(0) = x_0 \in H, \\ y(t) = Cx(t). \end{cases} \tag{13}$$

where $\{A(t)\}_{t\geq 0}$ is the family of linear operators defined on their domains:

$$D(A(t)) := \left\{ x \in H : x \text{ isa.c.}, \frac{dx}{dz} \in H \text{ and } x(0) = 0 \right\} \tag{14}$$

(where a.c. means that x is absolutely continuous) by

$$A(t) = V\frac{d}{dz} + M(t,z)I \tag{15}$$

where V and M are given by:

$$V := \begin{bmatrix} v_1 & 0 \\ 0 & v_2 \end{bmatrix}, \qquad v_1 = -v_{ss}, \quad v_2 = -\frac{v_{ss}}{\epsilon} \tag{16}$$

$$M(t,z) := \begin{bmatrix} m_1(t,z) & m_2(t,z) \\ m_3(t,z) & m_4(t,z) \end{bmatrix} \tag{17}$$

and the functions m_{ij} are given by:

$$m_1(t,z) = \mu(h_0 + h_1 e^{-\alpha t})\frac{(1-\theta_{2ss})^{n_1}}{(1+\theta_{1ss})^2} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}},$$

$$m_2(t,z) = -n_1(h_0 + h_1 e^{-\alpha t})(1-\theta_{2ss})^{n_1-1} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}},$$

$$m_3(t,z) = \mu(l_0 + l_1 e^{-\alpha t})\frac{(1-\theta_{2ss})^{n_1}}{(1+\theta_{1ss})^2} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}},$$

$$m_4(t,z) = -n_1(l_0 + l_1 e^{-\alpha t})(1-\theta_{2ss})^{n_1-1} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}}.$$

The input operator $B = B_0 I \in \mathcal{L}(L^2(0,1), H)$ is the linear bounded operator where:

$$B_0 = \begin{bmatrix} b_1(z) \\ b_2(z) \end{bmatrix}, \tag{18}$$

$$b_1(z) = \frac{\partial\theta_{1,ss}}{\partial z}, \quad b_2(z) = \frac{1}{\epsilon}\frac{\partial\theta_{2,ss}}{\partial z}$$

Operator $C := C_0 I$ is the observation operator and depends on the available measurements, where C_0 is matrix.

3. Trajectory and stability analysis

This section is devoted to the trajectory and the stability analysis of the linearized fixed-bed reactor model described in the previous section. The following theorem shows the existence and uniqueness of an evolution system generated by the family of operators $\{A(t)\}_{0\leq t\leq T}$, for any $T>0$.

Theorem 1. *Consider the family of operators $\{A(t)\}_{0\leq t\leq T}$ given by (15). Then, there exists a unique evolution system $U_A(\cdot,\cdot): \{(t,s) \in R^2 : s \leq t \leq T\}$ such that*

$$\frac{\partial}{\partial t}U_A(t,s)x = A(t)U_A(t,s)x, \quad \forall x \in D(A(t)), \quad 0 \leq s \leq t \leq T.$$

Moreover, there are constants $M \geq 1$ and ω such that

$$\|U_A(t,s)\| \leq Me^{\omega(t-s)}, \quad 0 \leq s \leq t \leq T.$$

Proof. The operator $A(t)$ can be written as

$$A(t) := A_0 + M(t) = \begin{bmatrix} v_1\frac{d.}{dz} & 0 \\ 0 & v_2\frac{d.}{dz} \end{bmatrix} + \begin{bmatrix} m_1(t,z)I & m_2(t,z)I \\ m_3(t,z)I & m_4(t,z)I \end{bmatrix} \tag{19}$$

In order to prove this Theorem, it suffices to validate all the conditions in [14, Theorem 4.8, p. 145] that includes:

1 $\{A(t)\}_{0\leq t\leq T}$ is a stable family of infinitesimal generators of C_0 semigroup on H.
2 Domain of $A(t)$ is independent of t.
3 For any $x \in D(A(t))$, $A(t)x$ is differentiable.

The operator A_0 is an infinitesimal generator of a exponentially stable C_0-semigroup and $M(t)$, $t \geq 0$ is bounded linear operator, then, by using the perturbation theorem [14, Theorem 2.3, p. 132] $A(t)$ is a stable family of infinitesimal generators. Thus condition 1 holds for this system.

From Eq. (14), the domain of $A(t)$ is independent of time, and it is trivial that for any $x \in D_0$, $A(t)x$ is differentiable. Therefore all the assumption of Theorem 4.8 in [14] are satisfied. □

In order to solve the optimal control problem, the property of exponential stability has been proved in the following theorem.

Theorem 2. *Consider the family of operators $\{A(t)\}_{t\geq 0}$ as in Theorem 1. Then $\{A(t)\}_{t\geq 0}$ generates an exponentially stable evolution system.*

Proof. The sufficient condition for exponential stability is existence of a positive operator P that satisfies the following operator Lyapunov differential equation [15].

$$\dot{P}(t) + P(t)A(t) + A(t)^*P(t) + Q(t) = 0 \quad \text{with} \quad P(D(A(t))) \subset D(A(t)^*) \tag{20}$$

where

$$Q(t) = Q_0(t,z)I = \begin{bmatrix} q_1(t,z) & q_2(t,z) \\ q_2(t,z) & q_3(t,z) \end{bmatrix} \tag{21}$$

and q_i, $i=1,\ldots,3$ are arbitrary functions such that the matrix Q_0 is a nonnegative matrix. Let us assume that the solution has the following diagonal form:

$$P(t) = \mathcal{P}(t,z)I = \begin{bmatrix} p_1(t,z)I & 0 \\ 0 & p_2(t,z)I \end{bmatrix} \tag{22}$$

where I is the identity operator and p_1 and p_2 are real valued functions. Note that the adjoint operator is defined on its domain

$$D(A^*(t)) = \left\{ x \in H : x \text{ isa.c } \frac{dx}{dz} \in H \text{ and } x(1) = 0 \right\}$$

by $A^*(t) = -V\frac{d}{dz} + M^*(t,z)I$

By injecting Eq. (22) into Eq. (20), we get

$$\begin{aligned} 0 &= \dot{P}(t) + P(t)A(t) + A(t)^*P(t) + Q(t) \\ &= \dot{\mathcal{P}}(t,z)I + \mathcal{P}(t,z)\left(V\frac{d}{dz} + M(t,z)I\right) \\ &\quad + \left(-V\frac{d}{dz} + M^*(t,z)I\right)\mathcal{P}(t,z) + Q_0(t,z)I = \dot{\mathcal{P}}(t,z)I + V\mathcal{P}(t,z)\frac{d}{dz} \\ &\quad + \mathcal{P}(t,z)M(t,z)I - V\frac{d}{dz}\mathcal{P}(t,z)I + M^*(t,z)\mathcal{P}(t)I + Q_0(t,z)I \\ &= \left(\dot{\mathcal{P}}(t) - V\frac{d\mathcal{P}}{dz} + \mathcal{P}(t,z)M(t,z) + M^*(t,z)\mathcal{P}(t,z) + Q_0(t,z)\right)I \end{aligned}$$

Indeed, the last equality holds due to the following fact

$$V\mathcal{P}(t)\frac{d}{dz} - V\frac{d}{dz}\mathcal{P}(t)I = -V\frac{d\mathcal{P}}{dz}I$$

Then Eq. (20) turns into the following matrix partial differential Lyapunov equation

$$\frac{\partial \mathcal{P}}{\partial t} - V\frac{d\mathcal{P}}{dz} = -\mathcal{P}(t,z)M(t,z) - M^*(t,z)\mathcal{P}(t,z) - Q_0(t), \quad \mathcal{P}(t,1) = 0 \tag{23}$$

Note that the condition $\mathcal{P}(t,1)=0$ comes from the condition $P(D(A(t))) \subset D(A(t)^*)$. If one can prove that the solution $\mathcal{P}(t,z)$ of the matrix Lyapunov PDE (23) exists, then the operator $P = \mathcal{P}I$ will be the solution for the operator Lyapunov differential equation (20). Positiveness on $\mathcal{P}$ implies that the operator P is positive as well.

In order to prove existence of a solution for Eq. (23), let us write it explicitly as a system of equations:

$$\frac{\partial p_1}{\partial t} - \nu_1\frac{\partial p_1}{\partial z} = -2m_1(t,z)p_1(t,z) + q_1(t,z) \tag{24a}$$

$$\frac{\partial p_2}{\partial t} - \nu_2\frac{\partial p_2}{\partial z} = -2m_4(t,z)p_2(t,z) + q_3(t,z) \tag{24b}$$

$$0 = -m_2(t,z)p_1(t,z) - m_3(t,z)p_2(t,z) + q_2(t,z) \tag{24c}$$

Now we can use the method of characteristics for the two first equations (24a) and (24b). Along the characteristics given by

$$\frac{dt}{dr} = 1, \quad t(0) = 0 \quad \text{and} \quad \frac{dz}{dr} = -\nu_1, \quad z(0) = z_0$$

Eq. (24a) can be transformed into the following ordinary differential Lyapunov equation given by

$$\frac{dp_1}{dr} = -2p_1(r)m_1(r) + q_1(r) \tag{25}$$

For any given positive function q_1, Eq. (25) admits a positive solution p_1. Therefore, Eq. (24a) admits a positive solution p_1

Likewise, for any positive function q_3, Eq. (24a) admits a positive solution p_2. By using Eq. (24c), we can find the function q_2 as follow

$$q_2(t,z) = m_2(t,z)p_1(t,z) + m_3(t,z)p_2(t,z) \tag{26}$$

In order to guarantee that the matrix Q_0 is nonnegative, we can always choose q_1 and q_3 such that $q_1 \gg |q_2|$ and $q_3 \gg |q_2|$. □

4. Optimal control design

This section deals with the computation of an LQ-optimal feedback operator for the linearized fixed-bed reactor by using the corresponding operator Riccati equation. The objective of the LQ-optimal controller is to find a square integrable control $u_o \in L^2[[0,\infty); L^2(0,l)]$ for any initial state $x_0 \in H$, which minimizes the cost functional

$$J(x_0,u) = \int_0^\infty (\langle Cx(t), PCx(t)\rangle + \langle u(t), Ru(t)\rangle)dt \tag{27}$$

where $P = P_0(z)\cdot I \in \mathcal{L}(Y)$ is a positive operator and $R = r_0(z)\cdot I \in \mathcal{L}(U)$ is a self-adjoint, coercive operator in $\mathcal{L}(U)$, where P_0 is a positive 2 by 2 matrix and r_0 is a positive functions.

Based on Theorem 1, $\{A(t)\}_{t\geq 0}$ is a stable family of infinitesimal generators of C_0 semigroup and the system (13) has a unique mild solution. Therefore, the solution of the LQ-optimal control problem can be obtained by finding the positive self-adjoint operator $Q \in \mathcal{L}(H)$ which solves the equivalent operator Riccati differential equation

$$[\dot{Q}_o + A^*Q_o + Q_oA + C^*PC - Q_oBR^{-1}B^*Q_o]x = 0 \tag{28}$$

for all $x \in D(A)$, where $Q_o(D(A)) \subset D(A^*)$.

According to Theorem 2, $\{A(t)\}_{t\geq 0}$ generates an exponentially stable evolution system, then the Riccati equation (28) has a nonnegative bounded solution Q_o and this solution is minimal among all nonnegative bounded solutions of (28) (see [2, Theorem 5.2, p. 507]). Moreover, for any initial state $x_0 \in H$, the quadratic cost (27) is minimized by the unique control u_o given on $t \geq 0$ by

$$u_o(t) = -R^{-1}B^*Q_ox(t).$$

In order to solve the Riccati equation (28), the following lemma is needed.

Lemma 1. *Let us denote by* $\overline{P}_0 := C_0^*P_0C_0 = (p_{ij})_{1\leq i,j\leq 2}$ *and* $\overline{R}_0 := B_0r_0^{-1}B_0^*$. *Given two positive functions* p_{11} *and* p_{22}, *there exist two functions* r_0 *and* p_{12} *such that the matrix Riccati partial differential equation*

$$\frac{\partial \Phi}{\partial t} - V\frac{\partial \Phi}{\partial z} + M^*(t,z)\Phi(t,z) + \Phi(t,z)M(t,z) + \overline{P}_0 - \Phi(t,z)\overline{R}_0\Phi(t,z) = 0 \qquad \Phi(t,1) = 0 \quad t \in [0,\infty] \tag{29}$$

admits a diagonal nonnegative solution Φ.

Proof. Assume that the matrix Φ has the following diagonal form

$$\Phi(t,z) = \begin{pmatrix} \phi_1(t,z) & 0 \\ 0 & \phi_2(t,z) \end{pmatrix}$$

Then Eq. (29) can be written explicitly using three equations, two partial differential Riccati equations and one algebraic equation

$$\frac{\partial \phi_1}{\partial t} - \nu_1\frac{\partial \phi_1}{\partial z} = -2\phi_1(t,z)m_1(t,z) - p_{11}(t,z) + r_0^{-1}b_1^2\phi_1^2(t,z) \tag{30}$$

$$\frac{\partial \phi_2}{\partial t} - \nu_2\frac{\partial \phi_2}{\partial z} = -2\phi_2(t,z)m_4(t,z) - p_{22}(t,z) + r_0^{-1}b_2^2\phi_2^2(t,z) \tag{31}$$

$$\phi_1(t,z)b_1r_0^{-1}b_2\phi_2(t,z) = \phi_1(t,z)m_2(t,z) + m_3(t,z)\phi_2(t,z) + p_{12}(t,z) \tag{32}$$

In order to prove the existence of solution for Eq. (30), the method of characteristics will be used. Along the following characteristics

$$\frac{dt}{dr} = 1, \quad t(0) = 0 \quad \text{and} \quad \frac{dz}{dr} = -\nu_1, \quad z(0) = z_0$$

Eq. (30) can be written as a Riccati differential equation as follows

$$\frac{d\phi_1}{dt} = -2\phi_1(r)m_1(r) - p_{11}(r) + \phi_1(r)b_1r_0^{-1}b_1\phi_1(r)$$

This equation admits a positive solution ϕ_1 because $p_{11} > 0$ and $b_1r_0^{-1}b_1 > 0$ (see [16]). Likewise, Eq. (31) admits a positive solution ϕ_2. Notice that in order to solve Eqs. (30) and (31), we have to choose a weighting function r_0. Then, by using Eq. (32), we can find the weighting function p_{12}. To complete the process we need to check the positiveness of the matrix $\overline{P}_0$. If this condition is not satisfied, the procedure can be repeated with a new choice of the function r_0. □

The following theorem proves that the solution of operator Riccati equation (28) can be found by solving an equivalent matrix Riccati partial differential equation.

Theorem 3. *Consider the linear model* (13). *Assume that* p_{12} *and* r_0 *are two functions such that* $\Phi := diag(\phi_1, \phi_2)$ *is the solution of the matrix Riccati partial differential equation* (29). *Then* $Q_o := \Phi(t,z)I$ *is the unique self-adjoint nonnegative solution of the operator Riccati differential equation. Moreover, the optimal control is given by*

$$u_o(t,z) = -r_0^{-1}b_1\phi_1(t,z)x_1(t,z) - r_0^{-1}b_2\phi_2(t,z)x_2(t,z) \tag{33}$$

Proof. Let us assume that $Q_o := \Phi(t,z)I$. Our objective is to find (if possible) a matrix Φ such that Q_o is the unique self-adjoint non-negative solution of Eq. (28). Let us substitute the expression of Q_o into Eq. (28), which yields Eq. (29). Indeed,

$$0 = \dot{Q}_o + A^*Q_o + Q_oA + C^*PC - Q_oBR^{-1}B^*Q_o = \dot{Q}_o + \left(-V\frac{\partial}{\partial z}\cdot + M^*(t,z)\cdot I\right)\cdot\Phi(t,z) + \Phi(t,z)\left(V\frac{\partial}{\partial z}\cdot + M(t,z)\cdot I\right)$$

$$+ \quad C_0^*(z)P_0(t,z)C_0(z)I - \Phi(t,z)B_0(z)r_0^{-1}(t,z)B_0^*(z)\Phi(t,z)I = \frac{\partial\Phi}{\partial t}I - V\frac{\partial}{\partial z}\cdot\Phi(t,z)I + \Phi(t,z)\cdot V\frac{\partial}{\partial z}\cdot + M^*(t,z)\Phi(t,z)I + \Phi(t,z)M(t,z)I$$

$$+ \quad C_0^*(z)P_0(t,z)C_0(z)I - \Phi(t,z)B_0(z)r_0^{-1}(t,z)B_0^*(z)\Phi(t,z)I = \left(\frac{\partial\Phi}{\partial t} - V\frac{\partial\Phi}{\partial z} + M^*(t)\Phi(t) + \Phi(t)M(t) + \overline{P}_0(t,z) - \Phi(t,z)\overline{R}_0(t,z)\Phi(t,z)\right)I$$

On the other hand, Q_o should satisfy $Q_o(D(A)) \subset D(A^*)$. This condition is only valid if $\Phi(t,1)=0$ as this implies that $\Phi(t,1)x(1)=0$, $\forall x \in D(A)$ and consequently $Q_o(D(A)) \subset D(A^*)$. □

Algorithm to solve the Riccati equation (29):

Step 1: Choose p_{11}, p_{22} and r_0.

Step 2: Find ϕ_1 and ϕ_2 solutions of Eqs. (30) and (31).

Step 3: Calculate the function p_{12} using Eq. (32).

Step 4: If $\overline{P}_0$ is positive, stop the procedure. Otherwise, repeat from Step 1 by choosing a new r_0.

Remark: The assumption that is made for the proof of Theorem 3 regarding the diagonality of Φ, adds a limitation to the choice of weighting functions in the objective function; but without this assumption the conversion of the operator Riccati equation (28) to the matrix Riccati equation (29) is impossible. Note that the process generator $A(t)$, given by Eq. (19), has a special structure. Indeed, it is the sum of a diagonal transport part A_0 and non-diagonal kinetics part $M(t)$. Moreover, the transport part is the main source of distribution. In view of this structure, it seems natural to look for a diagonal state feedback, which is not possible without the weighting functions used to make a balance in the algebraic part of the system (see Eq. (32). This explanation does not mean that the solution of this control problem should be diagonal.

5. Numerical simulations

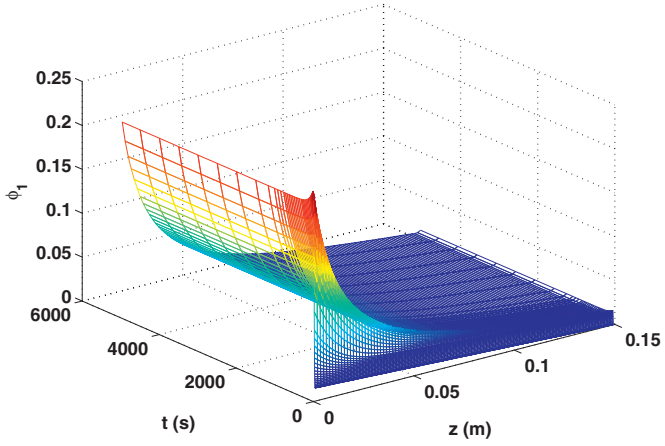
In this section the closed loop performance of the proposed controller is evaluated. Values of the model parameters are given in Table 1.

The LQ-controller discussed in the previous section was studied via a simulation that used a nonlinear model of the reactor given in Eqs. (2)–(4). The performance of the proposed controller was compared to an infinite dimensional LQ controller designed by assuming constant catalyst activity during the operation time. This LQ controller was computed using the method proposed in [7]. The solution of LQ controller based on the time-invariant model of the reactor can be computed by solving a set of ODEs instead of PDEs given by (30)–(32). Therefore, it needs less computation effort than the LQ controller proposed in this work.

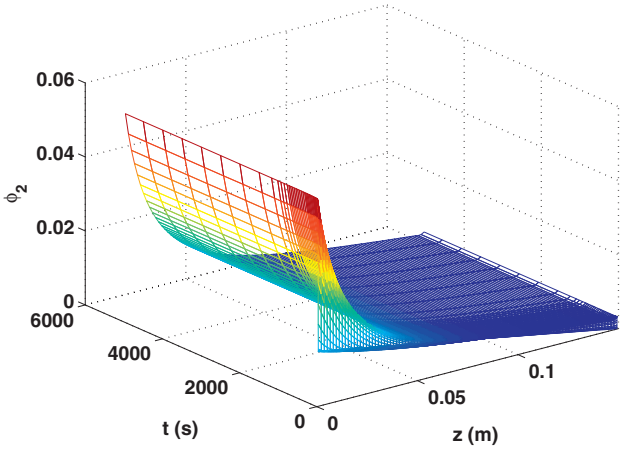
The control objective is to drive the temperature trajectory to the desired steady state profile. Using the nominal operating conditions, and the model given in Eqs. (2)–(4), the steady-state profiles of temperature and concentration were computed. Then, the nonlinear model was linearized around the stationary states and transformed to the infinite dimensional form of Eq. (13). We assumed that full state measurement is available i.e. $C=I$. Weighting function r_0 is assumed to be 50 (two different case studies). The

Table 1
Model parameters.

Parameter	Values	Unit
ϵ	0.4	
ρ_B	700	kg_{cat}/m^3
C_H	587.4437	mol/m^3
n_1	1.12	
n_2	0.85	
E	81000	J/mol
R	8.314	J/mol K
C_{A0}	0.419344	mol/m^3
C_{Ain}	0.419344	mol/m^3
T_0	523	K
T_{in}	523	K
ρ	2.7	kg/m^3
C_p	147.49	J/kg K
ΔH	101.3×10^3	J/mol
k_1	1.2384	
k_2	2.8896	
L	0.15	m



(a) φ_1



(b) φ_2

Fig. 1. Time-varying LQ-feedback functions ϕ_1 and ϕ_2, $\alpha = 1\times10^{-2}$.

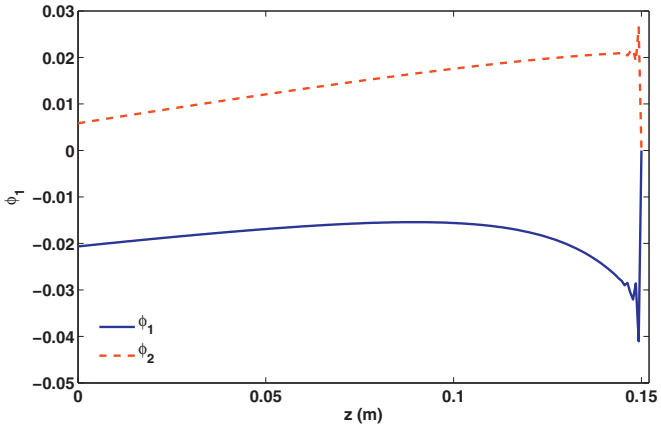


Fig. 2. Time-invariant LQ-feedback functions ϕ_1 and ϕ_2, $\alpha = 1 \times 10^{-2}$.

diagonal entries of the matrix P are chosen as follow $p_{11} = p_{22} = 0.01$. Therefore the objective function used of numerical simulation is given explicitly by.

$$J(x_0, u) = \int_0^\infty (0.01x_1^2(t) + 0.01x_2^2(t) + 2p_{12}x_1(t)x_2(t) + 50u^2(t))dt$$

The feedback functions ϕ_1 and ϕ_2 are computed by solving the set of equations given by (30)–(32). To compute the feedback functions for the second case, it is assumed that the reaction coefficient is equal to $k_0 + k_1$ during the operation time. Both controllers are applied to the original nonlinear model of the system given by Eqs. (2)–(4). Simulation are performed using UMFPACK direct solver of COMSOL®. We have used 120 discretization points both the model and controller.

In order to investigate the effect of deactivation rate on the closed loop performance of the controllers, two different values of α are considered. In the first case, $\alpha = 1 \times 10^{-2}$ the deactivation time has the same order of magnitude as residence time of the system, but in the second case, $\alpha = 1 \times 10^{-4}$, the deactivation time is much longer than the residence time of the reactor. The feedback functions ϕ_1 and ϕ_2 of two controllers for $\alpha = 1 \times 10^{-2}$ are shown in Figs. 1–6. For $\alpha = 1 \times 10^{-4}$, the feedback functions of time-varying controller have the similar trend but different values; however, the feedback functions of time-invariant controller are not functions of α, as the deactivation is ignored for developing this controller.

To assess the performance of two controllers, it is assumed that the system is initially at steady state and we are interested in maintaining the temperature profile at that steady state and cancel the

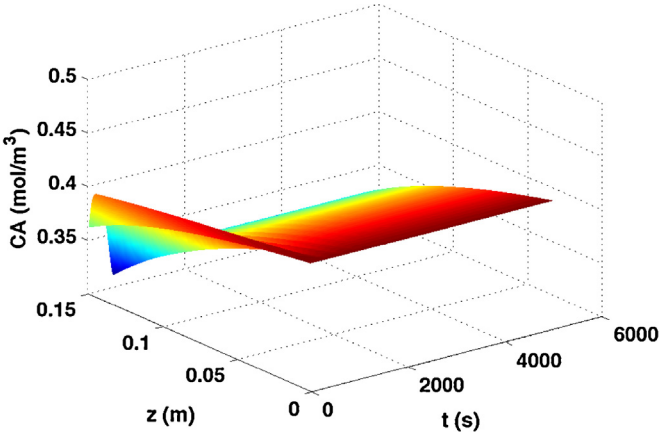


Fig. 3. Closed-loop trajectory of C_A, time-invariant controller, $\alpha = 1 \times 10^{-2}$.

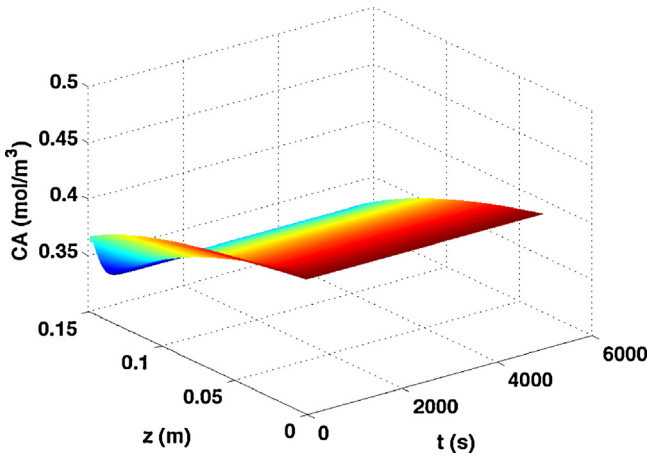


Fig. 4. Closed-loop trajectory of C_A, time-varying controller, $\alpha = 1 \times 10^{-2}$.

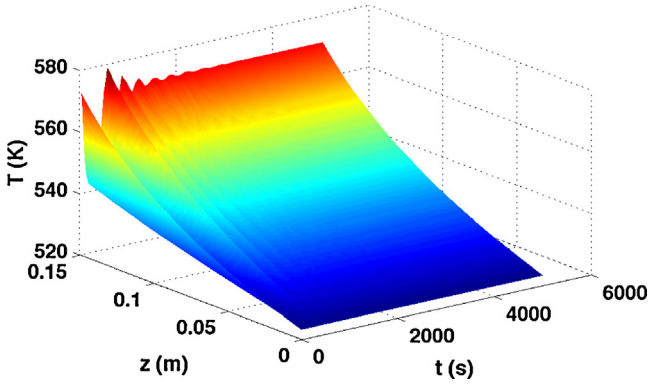


Fig. 5. Closed-loop trajectory of T, time-invariant controller, $\alpha = 1 \times 10^{-2}$.

effect of catalyst deactivation. The integral average of the temperature error is calculated for each case and is shown in Fig. 7. From Fig. 7, one can observe that for $\alpha = 1 \times 10^{-2}$, the performance of the time-varying controller is much better than the time-invariant controller. The time varying controller, regulates the temperature at the desired steady state profile with almost no error; however, the time-invariant controller results in oscillatory response with significant error. As the catalyst deactivation rate decreases the difference between performance of two controllers decreases. For $\alpha = 1 \times 10^{-4}$, the time varying controller is still better than time invariant one, but the maximum temperature error is less than 1 K which is not significant. In Fig. 8, the trajectory of manipulated

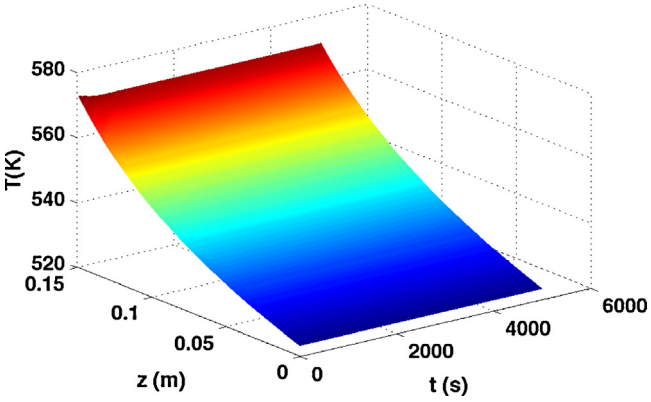
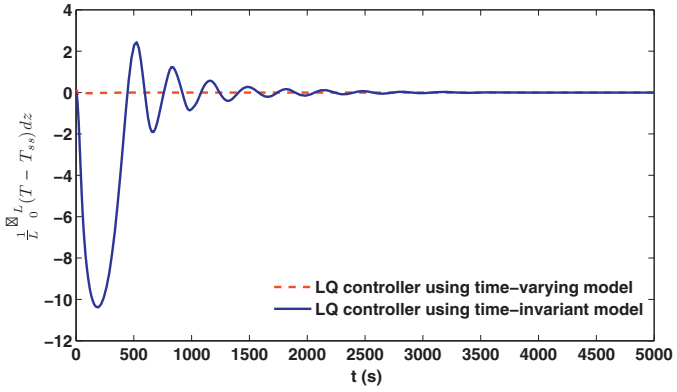
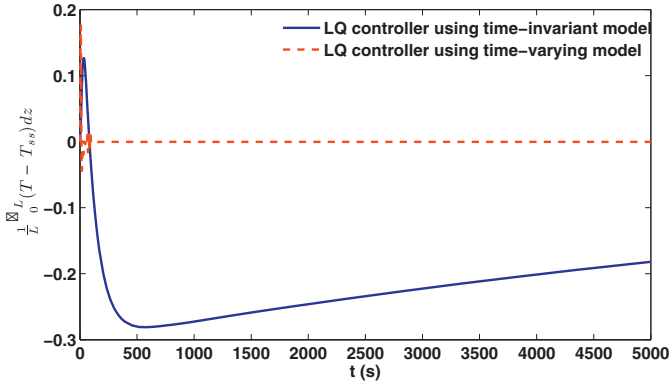


Fig. 6. Closed-loop trajectory of T, time-varying controller, $\alpha = 1 \times 10^{-2}$.



(a) $\alpha = 1 \times 10^{-2}$



(b) $\alpha = 1 \times 10^{-4}$

Fig. 7. Integral average of the error. (a) $\alpha = 1 \times 10^{-2}$. (b) $\alpha = 1 \times 10^{-4}$.

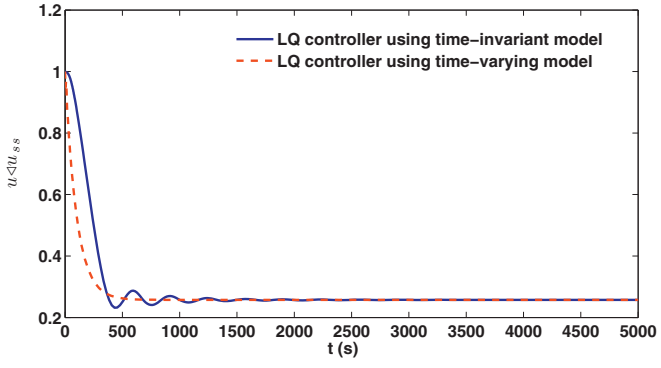


Fig. 8. Manipulated variable, $\alpha = 1 \times 10^{-2}$.

variable for two controllers are compared. It can be observed that, although the time-varying controller has significantly better performance for $\alpha = 1 \times 10^{-2}$, it does not require an aggressive input change.

6. Conclusion

In this work, a LQ-optimal controller for a time-varying set of hyperbolic equations with two time scale is formulated. It has been shown that the solution of the optimal control problem can be found by solving an equivalent matrix Riccati partial differential equation. Numerical simulations are performed to evaluate the closed loop performance of the designed controller on a hydrotreating fixed-bed reactor. The performance of the proposed controller is compared to performance of an infinite dimensional controller formulated by ignoring the catalyst deactivation. Simulation results show that the performance of the proposed controller is better than the controller ignoring the catalyst deactivation when the deactivation time is comparable with resident time of the reactor.

References

[1] R.F. Curtain, H.J. Zwart, Introduction to Infinite-Dimensional Linear Systems Theory, Springer-Verlag, 1995.
[2] A. Bensoussan, G. Da Prato, M.C. Delfour, S.K. Mitter, Representation and Control of Infinite Dimensional Systems, 2nd ed., Birkhäuser, Boston, 2007.
[3] F.M. Callier, J. Winkin, Spectral factorization and LQ-optimal regulation for multivariable distributed parameter systems, International Journal of Control 52 (1990) 55–75.
[4] F.M. Callier, J. Winkin, LQ-optimal control of infinite-dimensional systems by spectral factorization, Automatica 28 (4) (1992) 757–770.
[5] S. Dubljevic, P. Mhaskar, N.H. El-Farra, P.D. Christofides, Predictive control of transport-reaction processes, Computers and Chemical Engineering 29 (2005) 2335–2345.
[6] P.D. Christofides, Nonlinear and Robust Control of PDE Systems: Methods and Applications to Transport-Reaction Processes, Birkhäuser, 2001.
[7] I. Aksikas, A.M. Fuxman, J.F. Forbes, J.J. Winkin, LQ-control design of a class of hyperbolic PDE systems: application to fixed-bed reactor, Automatica 45 (2009) 1542–1548.
[8] I. Aksikas, F.J. Forbes, Y. Belhamadia, Optimal control design for time-varying catalytic reactors: A Riccati equation-based approach, International Journal of Control 82 (7) (2009) 1219–1228.
[9] L. Mohammadi, I. Aksikas, S. Dubljevic, J.F. Forbes, LQ-boundary control of a diffusion–convection–reaction system, International Journal of Control 85 (2012) 171–181.
[10] M. Ahmadinejad, M.R. Desai, T.C. Watling, A.P.E. York, Simulation of automotive emission control systems, Advances in Chemical Engineering 33 (2007) 47–101.
[11] A. Güthenke, D. Chatterjee, M. Weibel, B. Krutzsch, P. Kočí, M. Marek, I. Nova, E. Tronconi, Current status of modeling lean exhaust gas aftertreatment catalysts, Advances in Chemical Engineering 33 (2007) 103–211.
[12] J. Chen, Z. Ring, T. Dabros, Modeling and simulation of a fixed-bed pilot-plant hydrotreater, Industrial & Engineering Chemistry Research 40 (2001) 3294–3300.
[13] L.E. Kallinikos, G.D. Bellos, N.G. Papayannakos, Study of the catalyst deactivation in an industrial gasoil HDS reactor using a mini-scale laboratory reactor, Fuel 87 (12) (2008) 2444–2449.
[14] A. Pazy, Semigroups of Linear Operators and Applications to Partial Differential Equations, Springer-Verlag, 1983.
[15] L. Pandolfi, Existence of solutions to the Riccati equation for time-varying distributed systems, IMA Journal of Mathematical Control & Information 9 (1992) 265–273.
[16] H. Abou-Kandil, G. Freiling, V. Ionescu, G. Jank, Matrix Riccati Equations in Control and Systems Theory, Birkhäuser Verlag, Basel, 2003.