2009 American Control Conference
Hyatt Regency Riverfront, St. Louis, MO, USA
June 10-12, 2009

Optimal Control of a Time-Varying Catalytic Fixed Bed Reactor With Catalyst Deactivation

L. Mohammadi, I. Aksikas and J. F. Forbes

Abstract— **The paper deals with 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 by 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 show the performance of the designed controller on the fixed-bed reactor.**

Index Terms— **Fixed bed reactor, infinite dimensional time-varying system, linear quadratic optimal control, catalyst deactivation.**

I. Introduction

Catalytic fixed-bed reactors are the most widely used reactor type for gas phase reactants and play an important role in chemical industries. Interesting control problems arise due to nonlinear and distributed behavior [1]. The process considered in this work is a catalytic hydrotreating reactor. hydrotreating is the conventional means for removing sulfur from petroleum fractions. A schematic diagram of this reactor is shown in Fig.1. The hydrotreating catalyst deactivates during the operation for a variety of reasons (e.g., poisoning by impurities in feed, formation of coke on catalyst surface, and so forth). Deactivation of catalyst results in time-varying reaction rate. Then this system will behave as a time-varying infinite dimensional system. The control objective is to maintain the reactor's temperature and concentration at desired setpoint during the reactor's operation.

Linear-Quadratic (LQ) optimal control plays an important role in the control literature. Solution of the LQ-optimal control problem for infinite dimensional systems can be obtained by solving an operator Riccati equation (see e.g. [2]). Spectral factorization which is based on frequency-domain description is an alternative method for solving the LQ problem (see [3] and [4]).

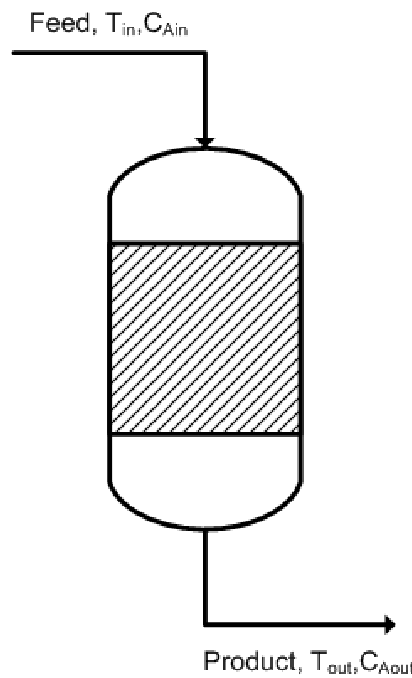

Fig. 1. Schematic diagram of Fixed-Bed reactor

In this paper, the Linear Quadratic (LQ) optimal control problem is studied for a model of catalytic fixed-bed reactor using its nonlinear infinite dimensional Hilbert state space description. Recently, the same problem was studied in [5] for a time-varying plug flow reactor by using the known Riccati equation approach. The convective terms in the model equations of a plug flow reactor have identical coefficients, while in the present case (i.e fixed-bed reactor) the coefficients of the convective terms are not necessarily identical. The objective of this work is to extend the Riccati equation approach into this framework and apply it to the case of a time varying fixed bed reactor.

II. Model description

The dynamics of a fixed-bed reactor can be described by partial differential equations derived from mass and energy balances. To model the reactor, a plug-flow pseudo-homogeneous model is considered. Moreover, we consider a

Paper submitted for presentation as an invited paper at the ACC'09.

L. Mohammadi is with Department of Chemical and Materials Engineering , University of Alberta, Edmonton, Ab, T6G 2G6, Canada leily@ualberta.ca
I. Aksikas is with Department of Chemical and Materials Engineering , University of Alberta, Edmonton, Ab, T6G 2G6, Canada {aksikas@ualberta.ca
J. F. Forbes is with Department of Chemical and Materials Engineering , University of Alberta, Edmonton, Ab, T6G 2G6, Canada fraser.forbes@ualberta.ca

one-spatial dimension model where there are no gradients in the radial direction.

A. 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 equation was assumed and has the following form (see [6]):

$$r_A = k(t)e^{\left(-\frac{E}{RT}\right)}C_A^{n_1}C_H^{n_2} \tag{1}$$

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

$$\epsilon\frac{\partial C_A}{\partial t} = -\nu\frac{\partial C_A}{\partial z} - \rho_B k(t)e^{-\frac{E}{RT}}C_A^{n_1}C_H^{n_2} \tag{2}$$

$$\frac{\partial T}{\partial t} = -\nu\frac{\partial T}{\partial z} + \frac{\rho_B\Delta H_r}{\rho C_p}k(t)e^{-\frac{E}{RT}}C_A^{n_1}C_H^{n_2} \tag{3}$$

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

$$\begin{array}{rcl} C_A(0,t) & = & C_{A,in}, \\ T(0,t) & = & T_{in}, \end{array} \tag{4}$$

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

$$\begin{array}{rcl} C_A(z,0) & = & C_{A0}(z), \\ T(z,0) & = & T_0(z) \end{array} \tag{5}$$

In the equations above, $C_A, T, \epsilon, \rho_B, \rho, C_p, E, C_H, \Delta H_r, \nu$ denote the reactant concentration, the temperature, the porosity of the reactor packing, the catalyst density, the fluid density, the activation energy, the enthalpy of reaction, and the superficial velocity respectively. In addition, t, z and l denote the time and space independent variables, and the length of the reactor, respectively. 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. Catalysts lose their activity with time and as a result this coefficient varies with time. Generally k is a function of time and operating conditions; but here we assume that the operating conditions are maintained in narrow ranges.Then k is only a function of time and is assumed to be given by:

$$k = k_0 + k_1 e^{-\alpha t} \tag{6}$$

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.

The corresponding steady-state equations of the PDE model (2) are given by the following ordinary differential equations:

$$\begin{cases} \nu_{ss}\frac{\partial C_{Ass}}{\partial z} = -\rho_B k_0 e^{-\frac{E}{RT_{ss}}}C_{Ass}^{n_1}C_H^{n_2} \\ \nu_{ss}\frac{\partial T_{ss}}{\partial z} = \frac{\rho_B\Delta H_r}{\rho C_p}k_0 e^{-\frac{E}{RT_{ss}}}C_{Ass}^{n_1}C_H^{n_2} \\ T_{ss}(0) = T_{in}, \quad C_{Ass}(0) = C_{A,in}. \end{cases} \tag{7}$$

B. Dimensionless model

Let us consider the following state transformation:

$$\theta_1 = \frac{T - T_{in}}{T_{in}}, \quad \theta_2 = \frac{C_{A,in} - C_A}{C_{A,in}} \tag{8}$$

Then we obtain the following equivalent representation of the model.

$$\frac{\partial\theta_1}{\partial t} = -\nu\frac{\partial\theta_1}{\partial z} + \left(h_0 + h_1 e^{-\alpha t}\right)(1-\theta_2)^{n_1}\,e^{\frac{\mu\theta_1}{1+\theta_1}} \tag{9}$$

$$\frac{\partial\theta_2}{\partial t} = -\frac{\nu}{\epsilon}\frac{\partial\theta_2}{\partial z} + \left(l_0 + l_1 e^{-\alpha t}\right)(1-\theta_2)^{n_1}\,e^{\frac{\mu\theta_1}{1+\theta_1}} \tag{10}$$

with the boundary conditions:

$$\theta_1(0,t) = 0, \quad \theta_2(0,t) = 0 \tag{11}$$

where:

$$\mu = \frac{E}{RT_{in}}, \quad l_{0,1} = \frac{\rho_B}{\epsilon}k_{0,1}C_H^{n_2}C_{A_{in}}^{n_1}e^{-\mu}, \tag{12}$$

$$h_{0,1} = \frac{\rho_B(-\Delta H)}{\rho C_p T_i n}k_{0,1}C_H^{n_2}C_{A_{in}}^{n_1}e^{-\mu} \tag{13}$$

C. Infinite-dimensional linearized model

Let us denote by θ_{ss} and u_{ss} the dimensionless profile of the model (9)-(10) at the operating point. Let us consider the following state definition:

$$x(t) = \theta(t) - \theta_{ss} \tag{14}$$

and new input $u(t) = \nu(t) - \nu_{ss}$. Then the linearization of the system (9)-(10) around its operating profile leads to the following linear time-varying infinite-dimensional system on the Hilbert space $H := L^2(0,l) \times L^2(0,l)$.

$$\begin{cases} \dot{x}(t) = A(t)x(t) + Bu(t), \\ x(0) = x_0 \in H, \\ y(t) = Cx(t). \end{cases} \tag{15}$$

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

$$D(A(t)) := x \in H : x \,\text{is a.c.}, \frac{dx}{dz} \in H \,\text{and}\, x(0) = 0 \tag{16}$$

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

$$A(t) = V.\frac{d.}{dz} + M(t,z).I \tag{17}$$

where V and M are given by:

$$V := \begin{bmatrix} v_1 & 0 \\ 0 & v_2 \end{bmatrix} \tag{18}$$

where

$$v_1 = -\nu_{ss}, \quad v_2 = -\frac{\nu_{ss}}{\epsilon} \tag{19}$$

$$M(t,z) := \begin{bmatrix} m_{11}(t,z) & m_{12}(t,z) \\ m_{21}(t,z) & m_{22}(t,z) \end{bmatrix} \tag{20}$$

where the functions m_{ij} are given by:

$$\begin{aligned} m_{11} &= \mu(h_0 + h_1 e^{-\alpha t})\frac{(1-\theta_{2ss})^{n_1}}{(1+\theta_{1ss})^2} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}}, \\ m_{12} &= -n_1(h_0 + h_1 e^{-\alpha t})(1-\theta_{2ss})^{n_1-1} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}}, \\ m_{21} &= \mu(l_0 + l_1 e^{-\alpha t})\frac{(1-\theta_{2ss})^{n_1}}{(1+\theta_{1ss})^2} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}}, \\ m_{22} &= -n_1(l_0 + l_1 e^{-\alpha t})(1-\theta_{2ss})^{n_1-1} e^{\frac{\mu\theta_{1ss}}{1+\theta_{1ss}}}. \end{aligned}$$

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

$$\begin{aligned} B_0 &= \begin{bmatrix} B_1 \\ B_2 \end{bmatrix}, \\ B_1 &= \frac{\partial \theta_{1,ss}}{\partial z}, \quad B_2 = \frac{1}{\epsilon}\frac{\partial \theta_{2,ss}}{\partial z} \end{aligned} \tag{21}$$

III. **Trajectory and stability analysis**

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

Theorem 1: Let $T > 0$. Consider the family of operators $\{A(t)\}_{0\le t\le T}$ given by (17) . Then, there exists a unique evolution system $U_A(\cdot,\cdot) : \{(t,s) \in R^2 : s \le t \le T\}$ such that

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

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

$$\|U_A(t,s)\| \le M e^{\omega(t-s)},\ 0 \le s \le t \le T.$$

Proof: First it can shown by using the perturbation theorem (see [7, Theorem 2.3, p. 132]) that $A(t)$ is a stable familty of operator in the sense of [7, Definition 2.1, p. 130]. The rest of the proof is based on [7, Theorem 4.8, p. 145] and it suffices to validate its assumptions. □

Now we are in a position to state a theorem on the exponential stability of the linearized model.

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

Proof: In order to prove the exponential stability, it suffices to use the Lyapunov approach : See [8].

IV. **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. First let us define an output function y(.) given by

$$y(t) = Cx(t)$$

and

$$C = C_0 I, \quad C_0 := \begin{bmatrix} w_1 & w_2 \end{bmatrix} \tag{22}$$

where $w_1, w_2 : [0,l] \to R$ are continuous functions. These functions can be interpreted as weighting factors for estimates of the distance between the initial model state and the chosen equilibrium profile. Now let us consider the LQ-optimal control problem: for any initial state $x_0 \in H$, find a square integrable control $u_o \in L^2[[0,\infty); L^2(0,l)]$ which minimizes the cost functional

$$J(x_0,u) = \int_0^\infty (\langle Cx(t), PCx(t)\rangle + \langle u(t), Ru(t)\rangle)dt \tag{23}$$

where $P = P_0.I \in \mathcal{L}(Y)$ is a positive operator and $R = R_0.I \in \mathcal{L}(U)$ is a self-adjoint, coercive operator in $\mathcal{L}(U)$, where P_0 and R_0 are two positive functions. The solution of this problem can be obtained by finding the positive self-adjoint operator $Q_0 \in \mathcal{L}(H)$ which solves the operator Riccati differential equation,viz.

$$[\dot{Q} + A^*Q + QA + C^*PC - QBR^{-1}B^*Q]x = 0 \tag{24}$$

for all $x \in D(A)$, where $Q_0(D(A)) \subset D(A^*)$.

It is known that for an infinite-dimensional state-space system of the form (15) and the cost function (23) such that the following conditions hold:

(i) There exists an evolution system generated by $A(t)$ such that $U_A(s,s)x = x, \forall x \in D(A(t)), \quad 0 \le s \le t \le T$ and

$$\frac{\partial}{\partial t} U_A(t,s)x = A(t)U_A(t,s)x,$$

(ii) (A,B) is C-stabilizable.

Then the Riccati equation (24) has a nonnegative bounded solution Q. This solution is minimal among all nonnegative bounded solutions of (24): see e.g. [9, Theorem 5.2, p.507].

The following lemma is an immediate consequence Theorems 1 and 2:

Lemma 1: Consider the linearized catalytic fixed-bed reactor model (15), with control operator B given by (21) and observation operator given by (22). Then the corresponding operator Riccati equation (24) has a nonnegative bounded solution Q_o and for any initial state $x_0 \in H$, the quadratic cost (23) is minimized by the unique control u_o given on $t \geq 0$ by

$$u_o(t) = -R_0^{-1} B^* Q_o x(t).$$

Now we are in a position to state the main theorem of this section, which gives an expression of the optimal state feedback in term of the solution of a matrix Riccati partial differential equation:

Theorem 3: Let us consider the linear model (15). Assume that V is given by (18). Let us consider P_0 a positive matrix and R_0 a self-adjoint coercive matrix such that $\Phi := diag(\phi_1, \phi_2)$ is the solution of the matrix Riccati partial differential equation:

$$\begin{aligned} \frac{\partial \Phi}{\partial t} &= -V \frac{\partial \Phi}{\partial z} + M^* \Phi + \Phi M \\ &\quad + C_0^* P_0 C_0 - \Phi B_0 R_0^{-1} B_0^* \Phi, \quad (25) \\ \Phi(t, l) &= 0, \quad t \in [0, \infty] \end{aligned}$$

Then $Q_0 := \Phi(t, z) I$ is the unique self-adjoint nonnegative solution of the operator Riccati differential equation. Moreover, the optimal control is given by ([10])

$$u_o(t, z) = -R_0^{-1} B_1 \phi_1(t, z) x_1(t, z) - R_0^{-1} B_2 \phi_2(t, z) x_2(t, z) \quad (26)$$

Proof: It is assumed that Φ is diagonal, then V commutes with Φ and since

$$-\frac{d.}{dz} \Phi I + \Phi \frac{d.}{dz} = \frac{d\Phi}{dz} I \quad (27)$$

Then the operator Riccati equation can be written as follows:

$$\frac{\partial \Phi}{\partial t} = -V \frac{d\Phi}{dz} + M^* \Phi + \Phi M + C_0^* P_0 C_0 - \Phi B_0 R_0^{-1} B_0^* \Phi$$

Comment 1: Let us assume that M, V, B, C are given by (20), (18), (21), (22), respectively. The matrix Riccati partial differential equation (25) can be written as three set of partial differential and algebraic equations given as follows (see [11, Comment 3.1] and [12, Comment 3.1]):

$$\begin{cases} \frac{\partial \phi_1}{\partial t} = -\nu_1 \frac{d\phi_1}{dz} + 2m_{11}\phi_1 + \bar{c}_{11} - \bar{b}_{11}\phi_1^2, \\ \frac{\partial \phi_2}{\partial t} = -\nu_2 \frac{d\phi_2}{dz} + 2m_{22}\phi_2 + \bar{c}_{22} - \bar{b}_{22}\phi_2^2, \\ 0 = m_{21}\phi_2 + \phi_1 m_{12} + \bar{c}_{12} - \phi_1 \bar{b}_{12}\phi_2, \\ \phi_1(l) = 0, \\ \phi_2(l) = 0 \end{cases} \quad (28)$$

TABLE I

Model Parameters

Parameter	Values
ϵ	0.4
ρ_B	700
C_H	587.4437
n_1	1.12
n_2	0.85
E	81000
R	8.314
C_{A0}	0.419344
C_{Ain}	0.419344
T_0	523
T_{in}	523
ρ	2.7
C_p	147.49
ΔH	101.3×10^3
α	0.005
k_1	1.2384
k_2	2.8896

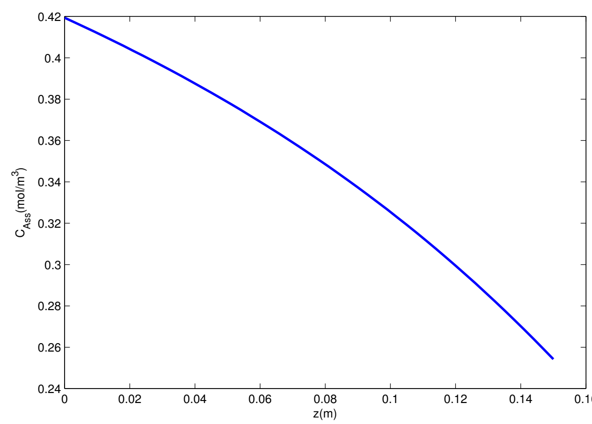

Fig. 2. Steady State Concentration Profile

where the functions $m_{ij}, \bar{c}_{ij}$ and $\bar{b}_{ij}$ are the entries of the matrices $M, C_0^* P_0 C_0$ and $B_0 R_0^{-1} B_0^*$, respectively. In the equations above, we have two unknown functions ϕ_1, ϕ_2, then we cannot solve the set of equations; however, it can be solved if we complete the number of unknown functions from the entries of the matrices P_0 and R_0 (we need one more function).

V. Numerical Simulations

To show the control performance of the closed-loop system, the formulated LQ controller is used for a hydrotreating reactor. Model parameters are given in Table I. Using the nominal operating condition,

$$u = 8.333 \times 10^{-4} m/s$$

and the model given in Eqs. (2), the stationary state was computed, see Figs (2)-(3).

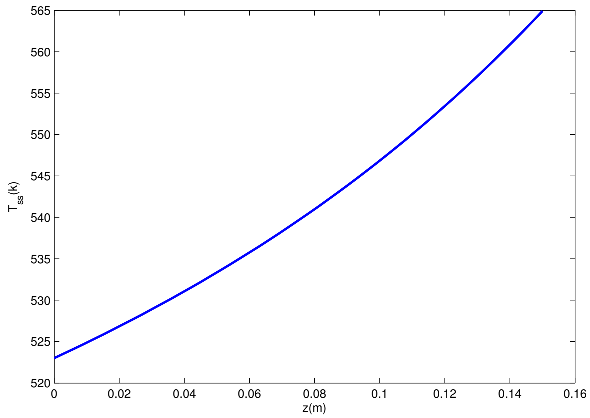

Fig. 3. Steady State Temperature Profile

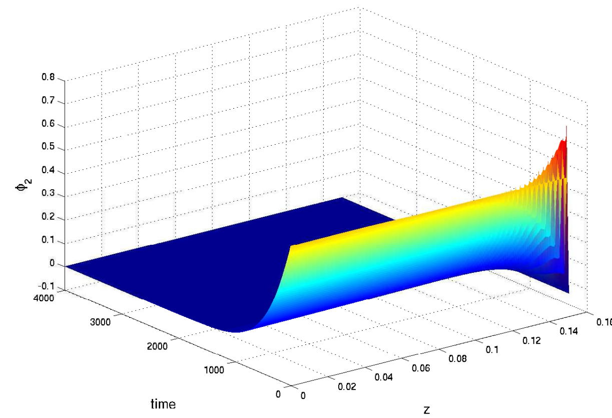

Fig. 5. LQ-Feedback function ϕ_2

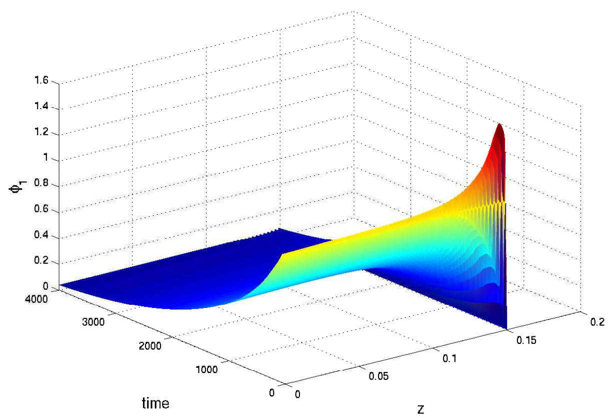

Fig. 4. LQ-Feedback function ϕ_1

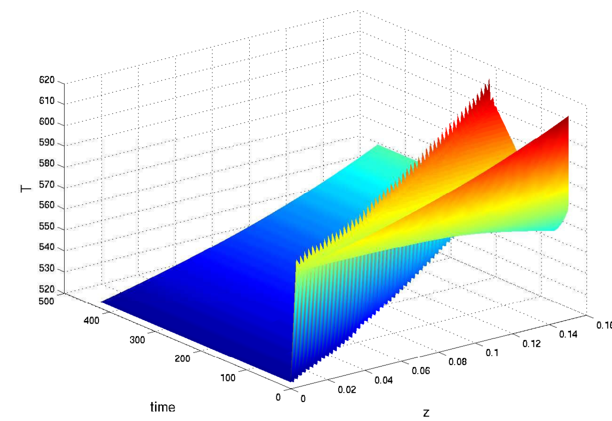

Fig. 6. Closed-loop temperature distribution

With the choice of weighting functions $w_1(z) = 1$, $w_2(z) = 1$, the LQ-feedback controller was computed using the linearized model, Eq. (15). Linearization was performed around the steady state profile. A total of 50 discretization points were used as the spatial locations where the outputs are measured. The LQ feedback functions that results from solving the system of Eq. (28) are given in Figs. (4)-(5).

The optimal control given by equation (26) is a distribution of the fluid flow velocity along the axis of the reactor. Although manipulation of the fluid velocity at a large number of points gives the best achievable control performance, it is not practical for real operation. Then we approximate the optimal spatial distribution of the manipulated variable by averaging its value as follows:

$$u_{sub}(t) = \frac{1}{L}\int_0^L u_o(t,z)dz \tag{29}$$

Using the equation (29) as input variable, the closed loop response of the system from an initial state that is not the stationary state is computed. The closed loop temperature and concentration responses are shown in Figures (6)-(7) respectively. It can be observed that the state converges to the desired equilibrium profile.

The trajectory of the manipulated variable, $u_{sub}(t)$, is shown in Figure (8).

VI. Concluding Remarks

In this paper, the linear quadratic optimal control problem has been studied for a fixed bed hydrotreating reactor with catalyst deactivation. An LQ-control feedback has been computed by using an operator Riccati differential equation, whose solution can be obtained via a related matrix Riccati partial differential equation.

The computed control algorithm has been applied to the linear model of the system and it is shown that the regulation can be achieved quickly.

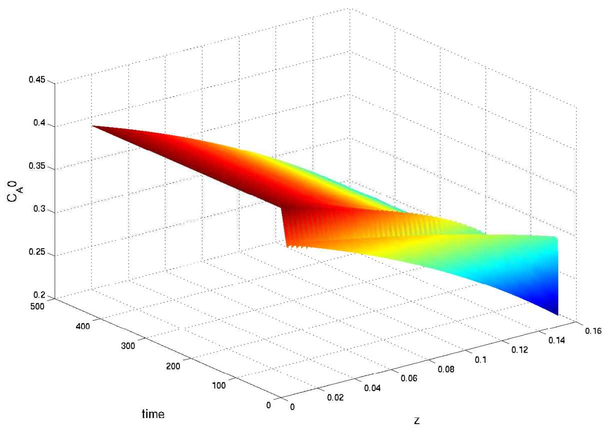

Fig. 7. Closed-loop concentration distribution

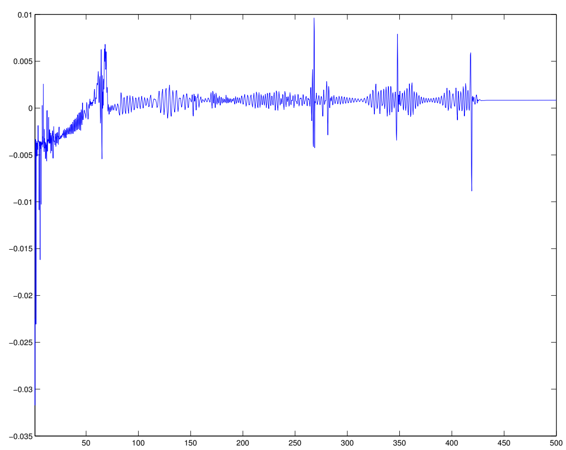

Fig. 8. Manipulated variable

REFERENCES

[1] A. Kremling and F. Allgower, "Robust control of a catalytic fixed-bed reactor," *Proceeding of the 33rd CDC*, 1994.

[2] R. Curtain and H. Zwart, *Introduction to Infinite-Dimensional Linear Systems Theory*. Springer-Verlag, 1995.

[3] F. Callier and J.Winkin, "Spectral factorization and lq-optimal regulation for multivariable distributed parameter systems," *International Journal of Control*, vol. 52, pp. 55–75, 1990.

[4] F. Callier and J. Winkin, "Lq-optimal control of infinite-dimensional systems by spectral factorization," *Automatica*, vol. 28:4, pp. 757–770, 1992.

[5] I. Aksikas, A. M. Fuxman, and J. F. Forbes, "Control of Time Varying Distributed Parameter Plug Flow Reactor by LQR," in *Proceedings of the IFAC World Congress, Seoul Korea*, 2008.

[6] J. Chen, Z. Ring, and T. Dabros, "Modeling and simulation of a fixed-bed pilot-plant hydrotreater," *Ind. Eng. Chem. Res.*, vol. 40, pp. 3294–3300, 2001.

[7] A. Pazy, *Semigroups of Linear Operators and Applications to Partial Differential Equations*. Springer-Verlag, 1983.

[8] L. Pandolfi, "Existence of solutions to the riccati equation for time-varying distributed systems," *IMA Journal of Mathematical Control & Information*, vol. 9, pp. 265–273, 1992.

[9] A. Bensoussan, G. D. Prato, M. C. Delfour, and S. K. Mitter, *Representation and Control of Infinite Dimensional Systems*, 2nd ed. Birkhäuser, Boston, 2007.

[10] I. Aksikas, F. J. Forbes, and Y. Belhamadia, "Optimal Control Design for Time-Varying Catalytic Reactors: A Riccati Equation-Bases Approach," *International Journal of Control*, accepted.

[11] I. Aksikas, A. Fuxman, J. F. Forbes, and J. J. Winkin, "LQ-Control Design of a Class of Hyperbolic PDE Systems: Application to Fixed-Bed Reactor," *Automatica*, accepted.

[12] I. Aksikas and J. F. Forbes, "State LQ-Feedback Control of Hyperbolic PDE's Systems : Application to a Fixed-bed Reactor," in *Proceedings of the European Control Conference, ECC 07, kos, Greece*, 2007, pp. 5595–5601.