Characteristics-based MPC of a fixed bed reactor with catalyst deactivation

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Abstract: In this work characteristics-based model predictive control (CBMPC) of a fixed bed reactor with catalyst deactivation is studied. Performance of CBMPC has been analyzed for two cases: one that incorporates the catalyst deactivation within the reactor model and another that ignores the deactivation. Simulation results show that the performance of first controller that incorporates the catalyst deactivation is better than the controller that ignores the deactivation.

Keywords: Fixed bed reactor, Catalyst deactivation, Characteristics-based model predictive control

1. INTRODUCTION

A catalyst loses its activity during operation. Catalyst deactivation can have variety of consequences. It may cause thermal instability of the reactor. It also affects the conversion and selectivity of the desired reaction. Consequently, it will affect the productivity and energy efficiency of the plant. In order to compensate for the effect of catalyst deactivation, the operating conditions are changed gradually to ensure maintaining the quality of product and the rate of production. Then designing a controller that can ensure changing optimal process operating conditions are tracked as they vary, is an important issue in operation of catalytic reactors.

Integration of the catalyst deactivation dynamics into the reaction system model results in a model that can describe the dynamical behavior of the system more precisely. By using this model in model-based algorithms, one can design a more efficient controller.

The objective of this work is to study the control of a fixed bed reactor with catalyst deactivation. In order to capture all of the main "macroscopic" phenomena (i.e., reactions, diffusion, convection, and so forth), the model of a fixed bed reactor takes the form of a mixed set of partial differential, ordinary differential, and algebraic equations. Such systems and many others (e.g., systems modeled by partial difference equations, integral equations and delay differential equations) are called distributed parameter systems (DPS) or infinite dimensional systems. Since we will consider the catalyst deactivation in the reactor's model, the resulting infinite dimensional system will be time varying.

Aksikas et al. (2009) and Aksikas et al. (2008) studied the control of the time varying infinite dimensional systems. In these works linear-quadratic controllers are developed by solution of the classical Riccati equation. This work is extended by Mohammadi et al. (2009) to cover the two-time scale property of the fixed bed reactors.

Model Predictive Control (MPC) is an optimal control technique that uses a model of the system to predict the future plant behavior and determines a sequence of control moves so that the predicted response moves to the desired set point in an optimal manner. Unfortunately, MPC algorithms for distributed parameter systems are relatively scarce. For diffusion-reaction systems, which are described by parabolic PDEs, Dubljevic et al. (2005) used modal decomposition to derive finite-dimensional systems that capture the dominant dynamics of the original PDE and are subsequently used for controller design. For the convection dominated parabolic PDEs, the modal decomposition methods result in high-order finite dimensional systems. MPC for high-order systems is computational demanding and cannot be applied on-line. For hyperbolic systems, the eigenvalues of the spatial differential operator cluster along vertical or nearly vertical asymptotes in the complex plane[Christofides (2001)], and the modal decomposition methods may not be used. Dubljevic et al. (2005) used the finite difference method to convert the hyperbolic equations to a set of ODEs and the MPC is designed for the resulting model. Using discretization methods may result in improperly capturing the dynamics of the system. More-

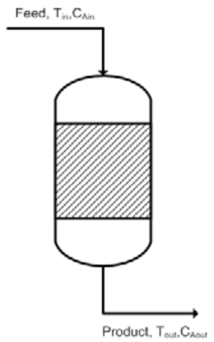

Fig. 1. Schematic diagram of Fixed-Bed reactor

over, the resulting ODEs are high-order and may result in an MPC that has high computational computational requirements.

Characteristics-based MPC is an approach for model predictive control of DPS proposed by Shang et al. (2004) and Shang et al. (2007). The method of characteristic allows controller design for linear, quasilinear, nonlinear low dimensional PDEs. In this method, partial differential equations are transformed to a set of ordinary differential equations along the characteristic curves, which exactly describe the original DPS. Then the controller design can be performed on ODEs instead of PDEs without approximation.

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. An important feature of a fixed bed reactor is the two time scale property of the system. In the other words, the dynamic behavior of the material balance is faster than the energy dynamics. Due to this property, the system has two characteristic curves. Furthermore, by incorporating the catalyst deactivation equation within reactor's model another very slow dynamic will be added to the system.

In this work the problem of controlling a fixed-bed catalytic reactor with catalyst deactivation is considered. We applied nonlinear characteristic-based MPC on-line algorithm to control the temperature of the reactor at the desired setpoint during the reactor's operation. Two cases have been considered. In the first one, the designed MPC uses a model of the system that considers the catalyst deactivation. In the second one, the catalyst deactivation is ignored for model predictive control development. Then performance of the two cases has been compared.

2. 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 one-spatial dimension model where there are no gradients in the radial direction. In the simplified system considered here, a lumped reaction kinetics equation was assumed and has the following form (see Chen et al. (2001)):

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

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

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

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

Initial and boundary conditions are:

$$\begin{aligned} C_A(0,t) &= C_{A,in},\ C_A(z,0) = C_{A0}(z), \\ T(0,t) &= T_{in}, \quad T(z,0) = T_0(z) \end{aligned} \tag{4}$$

In the equations above, $C_A, T, \epsilon, \rho_B, \rho, C_p, E, C_H, \Delta H_r, u$ denote the reactant concentration, the temperature, the porosity of the reactor packing, the catalyst density, the fluid density, the heat capacity, the activation energy,the hydrogen concentration, the enthalpy of reaction, and the superficial velocity respectively. k is the pre-exponential factor. Catalysts lose their activity with time and as a result this coefficient varies with time. The parameter k is proportional to the catalyst activity, which is a function of time and the operating conditions and can be described by an ODE (see Furimsky and Massoth (1999)). Here, we assume that the operating conditions are maintained in narrow ranges and in this case k is only a function of time, which can be described by:

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

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

3. CHARACTERISTICS-BASED MPC

The method of characteristics is a technique for solving hyperbolic partial differential equations. The idea is that every hyperbolic PDE has characteristic curves along which the dynamics evolve and as a result, the PDE can be represented as an equivalent ODE.

Consider a quasilinear system of first-order equations with two dependent variables ν_1,ν_2 and two independent variables t and x.

$$\begin{aligned} \frac{\partial \nu_1}{\partial t} + a_1\frac{\partial \nu_1}{\partial z} &= f_1(\nu_1, \nu_2, u) \\ \frac{\partial \nu_2}{\partial t} + a_2\frac{\partial \nu_2}{\partial z} &= f_2(\nu_1, \nu_2, u) \end{aligned} \tag{6}$$

if $a_1 \neq a_2$, the system has two different characteristics determined by:

$$\text{Characteristic } C_1: \quad \frac{dz}{dt} = a_1$$
$$\text{Characteristic } C_2: \quad \frac{dz}{dt} = a_2 \tag{7}$$

along these characteristics dynamic of the system is described by:

$$\frac{d\nu_1}{dt} = f_1(\nu_1, \nu_2, u) \quad \text{along characteristic } C_1$$
$$\frac{d\nu_2}{dt} = f_2(\nu_1, \nu_2, u) \quad \text{along characteristic } C_2 \tag{8}$$

Then, by using the method of characteristics, the set of partial differential equations (6) is transformed to a set of ODEs along the characteristic curves. This set of ODEs can be used to predict the future behavior of the system.

For a fixed-bed reactor which is modeled by equations (2) and (3) the characteristic curves are:

$$C_1 = \frac{dz}{dt} = \frac{u}{\epsilon} \tag{9}$$
$$C_2 = \frac{dz}{dt} = u \tag{10}$$

and the state variables C_A and T are described by the following ODEs along the characteristic curves:

$$\frac{\partial C_A}{\partial t} = -\frac{\rho_B}{\epsilon} k(t) e^{-\frac{E}{RT}} C_A^{n_1} C_H^{n_2} \tag{11}$$
$$\frac{\partial T}{\partial t} = \frac{\rho_B \Delta H_r}{\rho C_p} k(t) e^{-\frac{E}{RT}} C_A^{n_1} C_H^{n_2} \tag{12}$$

The characteristic ODEs are coupled with respect to the two characteristic curves, and the future state variables at one spatial point should be determined by simultaneous integration of both characteristic ODEs along two nonparallel characteristic curves. Fig. 2 illustrates the calculation of the future output variables using method of characteristics. This method for prediction of the future behavior is proposed by Shang et al. (2004). The idea is that at $t = t_k$ the measurements of the state variables are available at discretization points and these measurements are used to determine the value of the state variables at intersections of the characteristic curves. This algorithm provides us with the future values of the output variable. For example for point P we have:

$$C_A(P) = \int_{t(Q)}^{t(P)} f_1(Q) \tag{13}$$
$$T(P) = \int_{t(R)}^{t(P)} f_2(R) \tag{14}$$

where:

$$t(P) = \frac{a_1 t(Q) - 2a_2 t(Q) + a_2 t(R) + Z(R) - Z(Q)}{a_1 - a_2} \tag{15}$$

and a_1 and a_2 are $\frac{u}{\epsilon}$ and u respectively. The position of the point P is calculated by:

$$Z(P) = \frac{a_1 Z(R) - a_2 Z(Q) + a_1 a_2 [t(R) - t(Q)]}{a_1 - a_2} \tag{16}$$

This procedure is repeated for all nodes and then values of the future output variables are available and one can use common NMPC algorithm to compute the control action. The control action is calculated by solving the following optimization problem in receding horizon manner.

$$\min \int_t^{t+H_p} (T - T_{sp})^2 dt + \int_t^{t+H_c} \lambda (\Delta u)^2 \tag{17}$$

Where H_p is the prediction horizon, H_c is the control horizon, and λ is the weight of the input in the objective function. These parameters are tuning parameters for MPC.

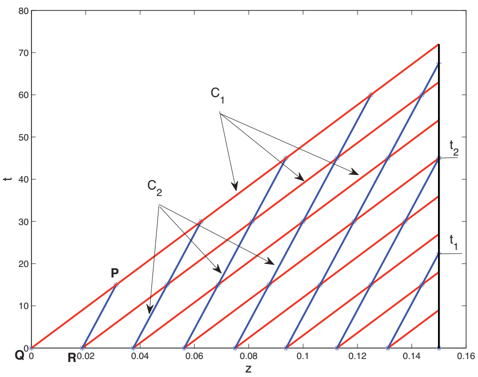

Fig. 2. Calculation of future outputs using characteristic curves

The values of T in the objective function are calculated using the method that described in this section. Since f_1 and f_2 in equations (13) and (14) are nonlinear functions, this optimization problem is a nonlinear optimization and can be solved numerically.

4. NUMERICAL SIMULATIONS

Our case study is a naphtha hydrotreating reactor. The simulation of the reactor was performed using COMSOL® Multi-physics. Using the MPC controller formulated in section 3, the control of the outlet temperature can be achieved. The manipulated variable is the superficial velocity of the feed.

To simulate the closed loop behavior of the system, we performed two cases. In the first one, we considered the deactivation equation of the catalyst and applied the MPC controller to time-varying equations.

In the second case, the model of the system that is used for MPC ignores the catalyst deactivation and assumes constant activity over operation time of the reactor.

Model parameters are given in Table 1. The objective is to control the reactor's outlet temperature at specified setpoint. The objective function is given in Equation (17).

The characteristic curves (9) and (10) are functions of the input variable. Then for the cases that the control horizon, H_c, is greater than one, the characteristic curves will not have constant slope and the calculation of the future values of the output variable will be challenging. In order to simplify the calculations, for the purpose of this example, we assumed that the control horizon is equal to 1, so the MPC problem becomes the following optimization problem:

$$\min \int_{t}^{t+H_p} (T - T_{sp})^2 dt + \lambda(\Delta u)^2 \qquad (18)$$

$$\frac{\partial C_A}{\partial t} = -\frac{\rho_B}{\epsilon} k(t) e^{-\frac{E}{RT}} C_A^{n_1} C_H^{n_2} \quad \text{along characteristic C}_1$$

$$\frac{\partial T}{\partial t} = \frac{\rho_B \Delta H_r}{\rho C_p} k(t) e^{-\frac{E}{RT}} C_A^{n_1} C_H^{n_2} \quad \text{along characteristic C}_2$$

The number of discretization points was taken to be $m = 9$. The prediction horizon is set to the residence time of the reactor, and λ is 1×10^3. The difference between the two cases is in the characteristic equations (11) and (12), which for the first case are functions of time.

This optimization problem can be solved by any optimization method for differential algebraic equations (DAE). Here we used sequential approach, which assumes piecewise constant inputs at each time interval and integrates the differential equations in each interval. This method is an easy method for solving optimization problems for MPC, but it is slower than other algorithms such as that proposed by Bock et al. (2000). The sequential algorithm is good enough for purpose of this illustration example, but for actual implementation the optimization algorithm should be improved.

Fig. 3 illustrates the performance of the CBMPC for the first and second case. This figure shows that the performance of the standard MPC algorithm for the first case is better than the second one. The second case, which considers a constant activity for the catalyst results in an steady state offset. Fig. 5 is the plot of the outlet concentration for two cases and Fig. 4 illustrates the computed control actions for two cases. As Fig. 4 shows, for the second case the input trajectory is almost constant except for first few time intervals; For the first case, due to inclusion of the time varying catalyst activity, the MPC provides more accurate control. Since we assumed

Table 1. Model Parameters

Parameter	Values	unit
ϵ	0.4	
ρ_B	700	$\text{kg}_{\text{cat}}/\text{m}^3$
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
α	0.005	
k_1	1.2384	
k_2	2.8896	

piecewise constant profiles for input variable, resulting output trajectory for the first case is non-smooth. But the fluctuations are not greater than $\pm 0.01 \times Y_{sp}$.

In order to deal with the steady state offset problem in the second case, one should implement offset elimination algorithms on standard MPC. These algorithms increase the computational demand of the MPC. Moreover the best offset elimination algorithm may achieve a performance similar to that of the first case.

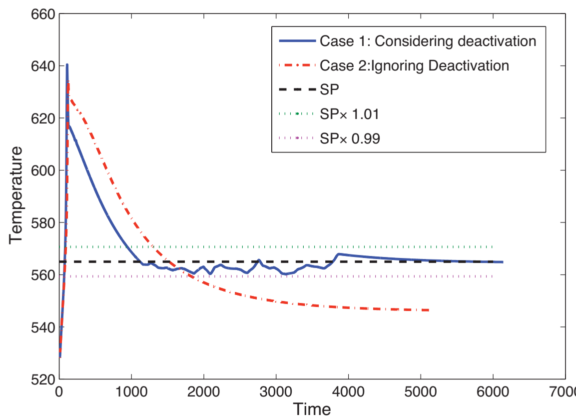

Fig. 3. Outlet temperature(Controlled variable)

Fig. 6 compares the conversion of the reactor for two cases. Although the conversion of the second case is higher at beginning, after a while the reactor's conversion decreases. Lower conversion results in decrease in the profitability of the plant.

5. CONCLUSION

In this work we studied the model predictive control of a naphtha hydrotreating reactor with catalyst deactivation. A characteristic-based MPC is developed to control the reactor. Two different case studies are studied: One that

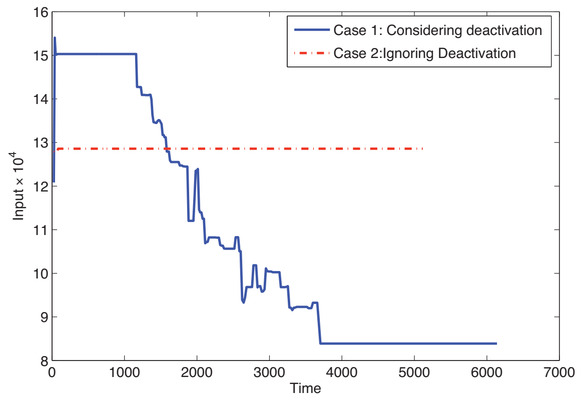

Fig. 4. Computed Input variable

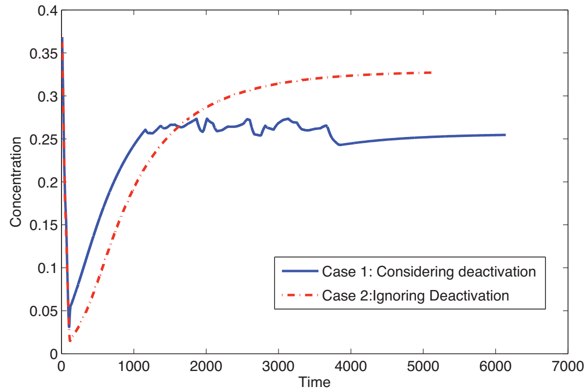

Fig. 5. Computed Input variable

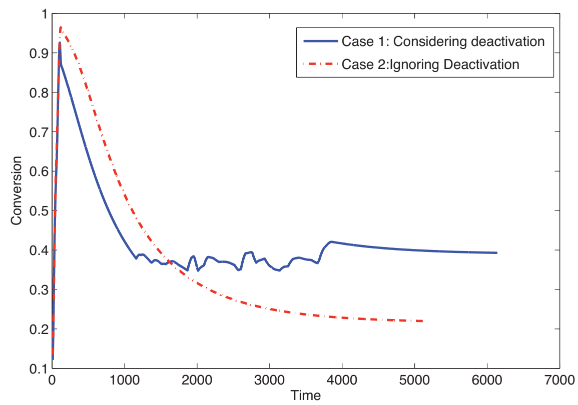

Fig. 6. Conversion at reactor outlet

incorporates the catalyst deactivation kinetics in the controller model and the second one that ignores the catalyst deactivation. The performance of two controllers are compared. The key result of this study is that integration of the catalyst deactivation kinetics with the reactor model, provides improved performance of the characteristic based MPC. This improvement in temperature control results in an improvement in conversion of the reaction, which may increase the plant profitability.

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