SIMULATION OF STEADY-STATE AND DYNAMIC BEHAVIOUR OF A TUBULAR CHEMICAL REACTOR

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KEYWORDS

Tubular chemical reactor, mathematical model, steady-state, dynamics.

ABSTRACT

The paper presents some results concerning analysis and simulation of steady-state and dynamic behaviour of a tubular chemical reactor. This analysis represents a necessary condition for the reactor control design purposes. The mathematical models used in simulations together with simulation results are contained.

INTRODUCTION

Tubular chemical reactor are units frequently used in chemical and biochemical industry. From the system theory point of view, tubular chemical reactors belong to a class of nonlinear distributed parameter systems. Their mathematical models are described by sets of nonlinear partial differential equations (PDR). The methods of modelling and simulation of such processes are described eg. in (Luyben 1989; Ingham et al. 1994; Severance 2001; Babu 2004). Relations between process behaviour and their control methods can be found in (Seborg et al. 1989; Ogunnaike and Ray 1994; Marlin 1995; Corriou 2004).

It is well known that the control of chemical reactors, and, tubular reactors especially, often represents very complex problem. The control problems are due to the process nonlinearity, its distributed nature, and high sensitivity of the state and output variables to input changes. In addition, the dynamic characteristics may exhibit a varying sign of the gain in various operating points, the time delay as well as non-minimum phase behaviour. Evidently, the process with such properties is hardly controllable by conventional control methods, and, its effective control requires application some of advanced methods (e. g. Adaptive Control, Predictive Control, Robust Control or any others). However, at all events, a previous analysis of the process behaviour is obligatory.

The paper presents all mathematical models used for simulations of both steady-state and dynamic characteristics of the tubular chemical reactor together with results of some simulations. The combinations of observed variables are chosen in accordance with purposes of prospective control design.

MODEL OF THE PLANT

An ideal plug-flow tubular chemical reactor with a simple exothermic consecutive reaction $A \xrightarrow{k_1} B \xrightarrow{k_2} C$ in the liquid phase and with the countercurrent cooling is considered as shown in Fig. 1.

Figure 1: Tubular chemical reactor.

Heat losses and heat conduction along the metal walls of tubes are assumed to be negligible, but dynamics of the metal walls of tubes are significant. All densities, heat capacities, and heat transfer coefficients are assumed to be constant. Under above assumptions, the reactor model can be described by five PDRs in the form

$$\frac{\partial c_A}{\partial t} + v_r \frac{\partial c_A}{\partial z} = -k_1 c_A \quad (1)$$

$$\frac{\partial c_B}{\partial t} + v_r \frac{\partial c_B}{\partial z} = k_1 c_A - k_2 c_B \quad (2)$$

$$\frac{\partial T_r}{\partial t} + v_r \frac{\partial T_r}{\partial z} = \frac{Q_r}{(\rho c_p)_r} - \frac{4U_1}{d_1(\rho c_p)_r} (T_r - T_w) \quad (3)$$

$$\frac{\partial T_w}{\partial t} = \frac{4}{(d_2^2 - d_1^2)(\rho c_p)_w} \left[d_1 U_1 (T_r - T_w) + d_2 U_2 (T_c - T_w) \right] \quad (4)$$

$$\frac{\partial T_c}{\partial t} - v_c \frac{\partial T_c}{\partial z} = \frac{4n_1 d_2 U_2}{(d_3^2 - n_1 d_2^2)(\rho c_p)_c} (T_w - T_c) \quad (5)$$

with initial conditions

$$c_A(z, 0) = c_A^s(z), \quad c_B(z, 0) = c_B^s(z), \quad T_r(z, 0) = T_r^s(z),$$

$$T_w(z,0) = T_w^s(z), \quad T_c(z,0) = T_c^s(z)$$

and boundary conditions

$$c_A(0,t) = c_{A0}(t), \quad c_B(0,t) = c_{B0}(t), \quad T_r(0,t) = T_{r0}(t),$$

$$T_c(L,t) = T_{cL}(t).$$

Here, t is the time, z is the axial space variable, c are concentrations, T are temperatures, v are fluid velocities, d are diameters, ρ are densities, c_p are specific heat capacities, U are heat transfer coefficients, n_1 is the number of tubes and L is the length of tubes. The subscript $(\cdot)_r$ stands for the reactant mixture, $(\cdot)_w$ for the metal walls of tubes, $(\cdot)_c$ for the coolant, and the superscript $(\cdot)^s$ for for steady-state values.

The reaction rates and heat of reactions are nonlinear functions expressed as

$$k_j = k_{j0} \exp\left(\frac{-E_j}{RT_r}\right), \quad j = 1, 2 \quad (6)$$

$$Q_r = (-\Delta H_{r1})k_1 c_A + (-\Delta H_{r2})k_2 c_B \quad (7)$$

where k_0 are pre-exponential factors, E are activation energies, $(-\Delta H_r)$ are the negative considered reaction enthalpies, and R is the gas constant.

The fluid velocities are calculated via the reactant and coolant flow rates as

$$v_r = \frac{4q_r}{\pi n_1 d_1^2}, \quad v_c = \frac{4q_c}{\pi(d_3^2 - n_1 d_2^2)} \quad (8)$$

The parameter values with correspondent units used for simulations are given in Table 1.

Table 1: Parameter values

$L = 8 \text{ m}$	$n_1 = 1200$
$d_1 = 0.02 \text{ m}$	$d_2 = 0.024 \text{ m}$
$d_3 = 1 \text{ m}$	
$\rho_r = 985 \text{ kg/m}^3$	$c_{pr} = 4.05 \text{ kJ/kg K}$
$\rho_w = 7800 \text{ kg/m}^3$	$c_{pw} = 0.71 \text{ kJ/kg K}$
$\rho_c = 998 \text{ kg/m}^3$	$c_{pc} = 4.18 \text{ kJ/kg K}$
$U_1 = 2.8 \text{ kJ/m}^2\text{s K}$	$U_2 = 2.56 \text{ kJ/m}^2\text{s K}$
$k_{10} = 5.61 \cdot 10^{16} \text{ 1/s}$	$k_{20} = 1.128 \cdot 10^{18} \text{ 1/s}$
$E_1/R = 13477 \text{ K}$	$E_2/R = 15290 \text{ K}$
$(-\Delta H_{r1}) = 5.8 \cdot 10^4 \text{ kJ/kmol}$	$(-\Delta H_{r2}) = 1.8 \cdot 10^4 \text{ kJ/kmol}$

From the system engineering point of view, $c_A(L,t) = c_{A\text{out}}$, $c_B(L,t) = c_{B\text{out}}$, $T_r(L,t) = T_{r\text{out}}$ and $T_c(0,t) = T_{c\text{out}}$ are the output variables, and, $q_r(t)$, $q_c(t)$, $c_{A0}(t)$, $T_{r0}(t)$ and $T_{cL}(t)$ are the input variables. Among them, for the control purposes, mostly $q_r(t)$ and $q_c(t)$ can be taken into account as the

control variables, whereas other inputs enter into the process as disturbances. As the controlled output, next to $c_{B\text{out}}$ also the mean reactant temperature given by

$$T_m(t) = \frac{1}{L} \int_0^L T_r(z,t) dz \quad (9)$$

can be under some assumptions considered.

COMPUTATION MODELS

For computation of both steady-state and dynamic characteristics, the finite differences method is employed. The procedure is based on substitution of the space interval $z \in [0, L]$ by a set of discrete node points $\{z_i\}$ for $i = 1, \dots, n$, and, subsequently, by approximation of derivatives with respect to the space variable in each node point by finite differences. Two types of finite differences are applied, either the backward finite difference

$$\left. \frac{\partial y(z,t)}{\partial z} \right|_{z=z_i} \approx \frac{y(z_i,t) - y(z_{i-1},t)}{h} = \frac{y(i,t) - y(i-1,t)}{h} \quad (10)$$

or the forward finite difference

$$\left. \frac{\partial y(z,t)}{\partial z} \right|_{z=z_i} \approx \frac{y(z_{i+1},t) - y(z_i,t)}{h} = \frac{y(i+1,t) - y(i,t)}{h}. \quad (11)$$

Here, a function $y(z,t)$ is continuously differentiable in $[0, L]$, and, $h = L/n$ is the diskretization step.

Dynamic Model

Applying the substitutions (10), (11) in (1) – (5) and, omitting the argument t in parenthesis, PDRs (1) – (5) are approximated by a set of ODRs in the form

$$\frac{dc_A(i)}{dt} = -[b_0 + k_1(i)]c_A(i) + b_0 c_A(i-1) \quad (12)$$

$$\frac{dc_B(i)}{dt} = k_1(i)c_A(i) - [b_0 + k_2(i)]c_B(i) + b_0 c_B(i-1) \quad (13)$$

$$\frac{dT_r(i)}{dt} = b_1 Q_r(i) - (b_0 + b_2)T_r(i) + b_0 T_r(i-1) + b_2 T_w(i) \quad (14)$$

$$\frac{dT_w(i)}{dt} = b_3 [T_r(i) - T_w(i)] + b_4 [T_c(i) - T_w(i)] \quad (15)$$

$$\frac{dT_c(m)}{dt} = -(b_5 + b_6)T_c(m) + b_5 T_c(m+1) + b_6 T_w(m) \quad (16)$$

for $i = 1, \dots, n$ and $m = n - i + 1$, and, with initial conditions

$$c_A(i, 0) = c_A^s(i), \quad c_B(i, 0) = c_B^s(i), \quad T_r(i, 0) = T_r^s(i), \\ T_w(i, 0) = T_w^s(i) \text{ and } T_c(i, 0) = T_c^s(i) \text{ for } i = 1, \dots, n.$$

The boundary conditions enter into Eqs. (12) – (14) and (16) for $i = 1$.

Now, nonlinear functions in Eqs. (12) – (16) take the discrete form

$$k_j(i) = k_{j0} \exp\left(\frac{-E_j}{RT_r(i)}\right), \quad j = 1, 2 \quad (17)$$

$$Q_r(i) = (-\Delta H_{r1})k_1(i)c_A(i) + (-\Delta H_{r2})k_2(i)c_B(i) \quad (18)$$

for $i = 1, \dots, n$.

The parameters b in Eqs. (12) – (16) are calculated from formulas

$$b_0 = \frac{v_r}{h}, \quad b_1 = \frac{1}{(\rho c_p)_r}, \quad b_2 = \frac{4U_1}{d_1(\rho c_p)_r}, \\ b_3 = \frac{4d_1U_1}{(d_2^2 - d_1^2)(\rho c_p)_w}, \quad b_4 = \frac{4d_2U_2}{(d_2^2 - d_1^2)(\rho c_p)_w} \quad (19) \\ b_5 = \frac{v_c}{h}, \quad b_6 = \frac{4n_1d_2U_2}{(d_3^2 - n_1d_2^2)(\rho c_p)_c}.$$

Here, the formula for computation of T_m (9) is rewritten to the discrete form

$$T_m(t) = \frac{1}{n} \sum_{i=1}^n T_r(z_i, t) \quad (20)$$

Steady-State Model

Computation of the steady-state characteristics is necessary not only for a steady-state analysis but the steady state values $y^s(i)$ also constitute initial conditions in ODRs (12) – (16) (here, y presents some of the variable in the set (12) – (16)).

The steady-state model can simply be derived equating the time derivatives in (12) – (16) to zero. Then, after some algebraic manipulations, the steady-state model takes the form of difference equations

$$c_A^s(i) = \frac{b_0}{b_0 + k_1^s(i)} c_A^s(i-1) \quad (21)$$

$$c_B^s(i) = \frac{1}{b_0 + k_2^s(i)} \left[k_1^s(i) c_A^s(i) + b_0 c_B^s(i-1) \right] \quad (22)$$

$$T_r^s(i) = \frac{1}{b_0 + b_2} \left[b_1 Q_r^s(i) + b_0 T_r^s(i-1) + b_2 T_w^s(i) \right] \quad (23)$$

$$T_w^s(i) = \frac{1}{b_3 + b_4} \left[b_3 T_r^s(i) + b_4 T_c^s(i) \right] \quad (24)$$

$$T_c^s(m) = \frac{1}{b_5 + b_6} \left[b_5 T_c^s(m+1) + b_6 T_w^s(m) \right] \quad (25)$$

for $i = 1, \dots, n$ and $m = n - i + 1$, and, nonlinear functions accordant with a steady-state are

$$k_j^s(i) = k_{j0} \exp\left(\frac{-E_j}{RT_r^s(i)}\right), \quad j = 1, 2 \quad (26)$$

$$Q_r^s(i) = (-\Delta H_{r1})k_1^s(i)c_A^s(i) + (-\Delta H_{r2})k_2^s(i)c_B^s(i) \quad (27)$$

Here, the formula for computation T_m takes the form

$$T_m^s = \frac{1}{n} \sum_{i=1}^n T_r^s(z_i) \quad (28)$$

SIMULATION RESULTS

The combinations of the inputs and outputs in all simulations of steady-state and dynamic characteristics were considered with respect to an importance for a prospective control design.

Steady-state Characteristics

Steady-state characteristics were computed from Eqs. (21) – (27) using fixed point iterations algorithm and for $n = 200$. Typical concentration and temperature profiles along the reactor tubes computed for $c_{A0}^s = 2.85$,

$c_{B0}^s = 0$, $T_{r0}^s = 323$, $T_{c0}^s = 299$, $q_c^s = 0.35$ and

$q_r^s = 0.15$ are shown in Figs. 2, 3. The presence of a peak on the reactant B profile is given by considered reaction type. The existence of a maximum on the reactant temperature profile (so called *hot spot*) is a common property of many tubular reactors with exothermic reactions.

The concentration and reactant profiles for various values q_c^s are exhibited in Figs. 4 – 6. All simulation results document strong dependence of all profiles upon this input in the steady-state.

The dependences of some output variables upon q_c^s and for various q_r^s are shown in Figs. 7 – 9. The courses document strong sensitivity of outputs upon both flow rates which can be considered as control inputs. In this way obtained results are very important for the reactor control design.

The dependence of the component B concentration

Figure 2: Concentration profiles along the reactor.

$c_{B\text{out}}^s$ on the reactant mean temperature is presented in Fig. 10. Also this result can be important when the output concentration c_B is non-measurable and, its desired (maximum) value could be achieved only by measured reactant temperatures along the reactor.

Figure 3: Temperature profiles along the reactor.

Figure 4: Concentration c_A^s profiles for various q_c^s .

Figure 5: Concentration c_B^s profiles for various q_c^s .

Figure 6: Reactant temperature profiles for various q_c^s .

Figure 7: Dependence of $c_{A\text{out}}^s$ on q_c^s for various q_r^s .

Figure 8: Dependence of $c_{B\text{out}}^s$ on q_c^s for various q_r^s .

Figure 9: Dependence of T_m^s on q_c^s for various q_r^s .

Figure 10: Dependence of c_B^s upon T_m^s .

Dynamic Characteristics

All dynamic characteristics were computed using the Runge-Kutta method with a fixed step. All inputs and outputs were considered as deviations from their steady values. This form is frequently used in the control. The deviations are denoted as follows:

$$\begin{aligned}v_1(t) &= q_c(t) - q_c^s \\v_2(t) &= c_{A0}(t) - c_{A0}^s \\v_3(t) &= T_{r0}(t) - T_{r0}^s \\y_1(t) &= c_{Bout}(t) - c_{Bout}^s \\y_2(t) &= T_m(t) - T_m^s\end{aligned}$$

where $c_{Bout}^s = 1.345$ and $T_m^s = 337.77$ are the output steady-state values computed from the model (21) – (25) for the input steady-state values

$q_c^s = 0.25$, $q_r^s = 0.15$, $c_{A0}^s = 2.85$, $T_{r0}^s = 323$ and $T_{c0}^s = 299$.

The responses to q_c and c_{A0} step changes in Figs. 11-14 clearly document a strong nonlinearity of the process. Moreover, they show a better applicability of the reactant mean temperature as the controlled output in comparison with the output concentration c_B having in this regard very unfavourable properties.

Figure 11: Output concentration c_B step responses.

Figure 12: Mean temperature T_m step responses.

As an influence illustration of the random disturbances on the process output, the responses to random signals c_{A0} and T_{r0} are shown in Figs. 15 – 18. These simulation results document high sensitivity of both outputs to random input signals.

Figure 13: Output concentration c_B step responses.

Figure 14: Mean temperature step responses.

Figure 15: Output c_B response to random input v_2 .

Figure 16: Output T_m responses to random input v_3 .

Figure 17: Output T_m responses to random input v_2 .

Figure 18: Output concentration c_B response to random input v_3 .

CONCLUSIONS

In the paper, the mathematical model of a tubular chemical reactor with a consecutive exothermic reaction has been presented. The computer models for simulations of steady-state and dynamic characteristics in the form of sets of ordinary differential and difference equations were derived to an original model in the form of partial differential equations. The simulation experiments were chosen with a view to a prospective control of the process.

ACKNOWLEDGMENTS

This work was supported in part by the Ministry of Education of the Czech Republic under the grant MSM 7088352101 and by the Grant Agency of the Czech Republic under the grant No. 102/06/1132.

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