Mediating Operation of Heterogeneous CSTR

Vladimir Gol'dshtein and Vadim Panfilov
Dept. of Mathematics and Computer Science, Ben Gurion University of the Negev, Beer Sheva 84105, Israel

Isaac Shreiber
Institute for Industrial Mathematics, Beer Sheva 84213, Israel

A new type of periodic operation is investigated for a simple model of a heterogeneous catalytic continuous stirred tank reactor. For the heterogeneous case, two scales of response times were derived, slow for heat processes and fast for concentration processes. Periodic operation in intermediate mode between slow and fast processes is studied by manipulation of the feed flow rate. The new approach is designated as mediating operation, which demonstrates ability to control interaction between fast and slow reactor subsystems due to desired separation of dynamic responses for temperature and concentration processes by a single input variable. The conversion improvement at lower temperature can be achieved for lower and upper stable steady states. For the system analysis, analytical investigation of the slow manifolds is used as well as numerical study.

Introduction

A new type of periodic operation for chemical reactors is presented. The approach is investigated for a simple model of a heterogeneous catalytic continuous stirred tank reactor (CSTR). CSTR is considered as a two-phase system with a gas mixture reacting on the surface of a solid catalyst. We focus on the situation with significant heat capacity of the solid catalyst in comparison with the heat capacity of the gas phase. This approach provides an opportunity to single out two-scale dynamic response times, slow for temperature processes and fast for concentration processes. Thus, a heterogeneous CSTR is considered as a multiscale system with widely separated time scales.

Periodic operation of heterogeneous CSTR is investigated as an *intermediate mode operation with respect to slow and fast reactor subsystems*. This is a new type of periodic operation that can be designated as *mediating operation*. A single forced variable—the feed flow rate—is considered as a manipulated variable. The proposed operating method presumes a unique ability to control two-scale interaction, i.e., temperature and concentration reactor processes by a single input variable; that is rather interesting to study from the control point of view.

Forced periodic operation of chemical reactors has been a general approach for modification of dynamic properties of reactor systems. The conversion improvement and the stabilized reactor operation can be noted as potential benefits of the periodic forcing approach. The classification of the basic types of periodic operation and early results in this field can be found in reviews by Bailey (1973, 1977). Bellman et al. (1983a,b) and Cinar et al. (1987a) developed the method of vibrational control by zero-average feed flow inputs. That is an open-loop control method using *fast* oscillations of system parameters. Applications of vibrational control by multiple oscillations of feed flow rate and concentrations have been studied by Cinar and coworkers (Cinar et al., 1987b; Rigopoulos et al., 1988; Shu et al., 1989). In vibrational control approach CSTR has been considered as a one-scale system with respect to fast operation mode.

This study addresses a new open-loop control approach in application to CSTR. CSTR is examined as a simple model of heterogeneous catalytic reactor with a first-order irreversible exothermic reaction. Mediating operation is analyzed for average steady-state behavior upon zero-average oscillations of feed flow rate. The zero-approximation of a reactor multiscale model is studied by using the traditional method of averaging (Bogoliubov and Mitropolsky, 1961). Some preliminary results of average conversion improvement have been obtained by Gol'dshtein and Kiselev (1987). Multiplicity of average steady states for two-step periodic forcing function has been researched by Gol'dshtein et al. (1990).

In this communication, the reactor model, based on the multiscale approach and a new type of control called mediating operation are proposed. The heart of the system analysis is the slow (integral) manifold method (see, for example, Gol'dshtein and Sobolev, 1988, 1992), supported by the averaging method and numerical investigation. The article deals with analytical study of dynamic system responses caused by mediating operation and reactor steady-state characteristics. Mediating operation of the upper steady state by two-step periodic control is examined in detail.

It is shown that mediating operation provides for control of two-scale CSTR due to desired separation of reactor responses by a single input variable. Results of numerical investigation demonstrate that mediating operation creates great possibilities for modifying the stability properties of a reactor system. As a result, for example, the conversion improvement at lower temperature can be realized by stabilized reactor operation for the lower steady state as well as for the upper one.

Reactor Model

An exothermic irreversible catalytic reaction in a well-stirred tank reactor is considered under zero-average feed flow rate inputs. In this article we draw attention to the heterogeneous CSTR defined as a two-phase system with a gas mixture reacting on the surface of a solid catalyst. As usual (see, for example, Chang and Schmitz, 1975), it is assumed that thermal equilibrium exists between different phases in the reactor at all times. For a first-order reaction the heat and mass balance equations of the CSTR are

$$(V\rho c_p + mc_s)\frac{dT}{dt'} = -\rho c_p F(T-T_f) - hA(T-T_c) + V(-\Delta H)Ck_0 \exp\left(-\frac{E}{RT}\right)$$

$$V\frac{dC}{dt'} = -F(C-C_f) - VCk_0 \exp\left(-\frac{E}{RT}\right) \quad (1)$$

Note that a CSTR model with an added heat capacity term of solid medium mc_s has been used in mathematical modeling of catalytic processes, for example, by Sheintuch and Schmitz (1977). A small influence of solid material additions has been researched (Chang and Schmitz, 1975; Cinar et al., 1987a).

We focus on the situation with significant heat capacity of the solid catalyst in comparison with the heat capacity of the gas phase, because the solid density is 10^3 times greater than the gas density. So, the ratio of gas heat capacity to the total reactor heat capacity, $\gamma = V\rho c_p/(V\rho c_p + mc_s)$ is considered as a *small parameter*. Its important role in dynamic response can be estimated as follows. Dimensionless time of temperature response is estimated as $\tau_{\text{temp.}} = (1/\gamma)\ (V/F)/t'$ under conditions of the absence of cooling ($A = 0$) as well as reaction heat ($\Delta H = 0$). As usual, dimensionless time of concentration response time is defined by $\tau_{\text{conc.}} = (V/F)/t'$, where V/F is residence time. Thus in heterogeneous CSTR, heat processes are approximately $1/\gamma$ times slower than concentration processes $\tau_{\text{conc.}} \ll \tau_{\text{temp.}}$

For reducing to a dimensionless for it would be desirable that a feed flow rate F as a control variable should be a *linear* parameter of a dimensionless system as it was in the initial system (Eq. 1). For this purpose reducing to a dimensionless form may be carried out in the way similar to Gol'dshtein et al. (1990), where $t = t'\gamma k_0\ \exp\ (-1/\beta) = (t'\gamma Da)/(V/F)$ has been chosen as a dimensionless time. As a result, the system (Eq. 1) has the following dimensionless form

$$\frac{d\theta}{dt} = -u\theta - \alpha(\theta-\theta_c) + B(1-\eta)k(\theta)$$

$$\gamma\frac{d\eta}{dt} = -u\eta + (1-\eta)k(\theta) \quad (2)$$

where θ is dimensionless temperature $=(T-T_F)/(\beta T_f)$, η is conversion (dimensionless concentration $=(C_f - C)/C_f$ are conventional variables, $u = (F/V)/[k_0 \exp(-1/\beta)$ is a dimensionless feed flow rate (control variable). For an unforced reactor, this parameter is inverse to the Damköhler number specified, for example, in Uppal et al. (1974), $u(t) \equiv u_0 = 1/Da$.

The system (Eq. 2) is a singularly perturbed system with a small parameter $\gamma \ll 1$. Further, a zero-approximation ($\gamma = 0$) of the system (Eq. 2) is considered

$$\frac{d\theta}{dt} = -u\theta - \alpha(\theta-\theta_c) + B(1-\eta)k(\theta)$$

$$0 = -u\eta + (1-\eta)k(\theta) \Rightarrow \eta = \frac{k(\theta)}{k(\theta)+u} \quad (3)$$

Type of Operation and Averaging of System

Let us consider some common types of operation, associated with regimes of forced oscillations. The early works on periodic operation have been reviewed by Bailey (1973, 1977), where the author, in particular, distinguished four families of periodic operation. Note that in all cases the object of periodic operation, namely, the chemical reactor system was considered as an entire one-scale system characterized by a single dynamic response time.

The operation of a heterogeneous catalytic CSTR seems more complicated. In this case, the selected system is the simplest type of multiscale system with widely separated time scales, ($\tau_{\text{conc.}} \ll \tau_{\text{temp.}}$). By taking into account this peculiarity, we call attention to operation in some intermediate mode with respect to response times of reactor subsystems. The approach presents the ability to use directly the nonlinear interplay between reactor processes. Indeed, the proposed operation acts as a mediator between temperature and concentration processes and influences each of these processes separately. Thus, a new type of operation called *mediating operation* is considered:

$$\text{Mediating Operation}: \tau_{\text{conc.}} \ll \tau \ll \tau_{\text{temp.}} \quad (4)$$

As usual, the control variable has been chosen as a function with constant average value. That means that it is a periodic (or quasi-periodic) control with respect to period of averaging τ

$$\hat{u} = \frac{1}{\tau}\int_0^\tau u(t)dt = \text{const} = u_0 \quad (5)$$

Taking into account the considered type of operation we can use the conventional method of averaging (Bogoliubov and Mitropolsky, 1961) to the zero-approximation of the system (Eq. 3). Note that, according to this method, *temperature can be regarded as a constant during the interval of averaging,* because temperature is a *slow* variable with respect to selected mode of control. Thus, everywhere, symbol θ can be retained for notation of an average temperature value. The averaged equations of Eq. 3 become

$$\frac{d\theta}{dt} = -\hat{u}\theta - \alpha(\theta - \theta_c) + B(1-\hat{\eta})k(\theta)$$

$$\hat{\eta} = \frac{1}{\tau}\int_0^{\tau} \frac{k(\theta)}{k(\theta)+u} dt \qquad (6)$$

We will investigate steady states of the averaged system which are defined by the relation $d\theta/dt = 0$. Finally, the examined system becomes

$$0 = -\hat{u}\theta - \alpha(\theta - \theta_c) + B(1-\hat{\eta})k(\theta)$$

$$\hat{\eta} = \frac{1}{\tau}\int_0^{\tau} \frac{k(\theta)}{k(\theta)+u} dt \qquad (7)$$

Analytical Investigation

The purpose of qualitative analysis is to determine possible dynamic response trends, caused by mediating operation for an average steady-state system (Eq. 7). For this purpose, the reactor in the presence of control ($u(t) \neq$ const) with average value $\hat{u} = u_0$ is compared with a reactor having stationary inputs of the same feed flow rate value u_0 ($u(t) \equiv u_0$). Following the methodical scheme of the slow manifold method described, for example, in Gol'dshtein and Sobolev (1988, 1992), analytical research will be held in two steps. At first, the properties of slow curve are studied separately, which are described by the second equation of the system (Eq. 7). After that, the average steady-state behavior is investigated for the full system (Eq. 7).

Let us consider separately a slow curve, namely, the average conversion behavior as a functional of feed flow rate variable. We prove now the following general statement that was claimed earlier by Gol'dshtein and Kiselev (1987): for each nonconstant $u(t)$ the average conversion is greater than conversion for constant u_0 at all fixed temperature values,

$$\hat{\eta}_u \geq \eta_{u_0} \text{ for each measurable bounded } u \geq 0 \text{ such that } \hat{u} = u_0 \qquad (8)$$

Proof

Conversion is a convex functional from u. Really, from the second equation of the system (Eq. 3) follows

$$\frac{\partial^2 \eta}{\partial u^2} = \frac{2k(\theta)}{[k(\theta)+u]^3} > 0$$

$$\text{if} \begin{cases} k(\theta) > 0 - \text{obviously, as exponential function} \\ u \geq 0 - \text{from technical restrictions} \end{cases}$$

At first we will prove this statement for simple *two-step* forcing function with period τ (see Figure 1)

$$\tilde{u}(t) = \begin{cases} u_1 & \text{for} \quad n\tau \leq t \leq (n+\nu)\tau \\ u_2 & \text{for} \quad (n+\nu)\tau < t \leq (n+1)\tau \end{cases} \qquad (9)$$

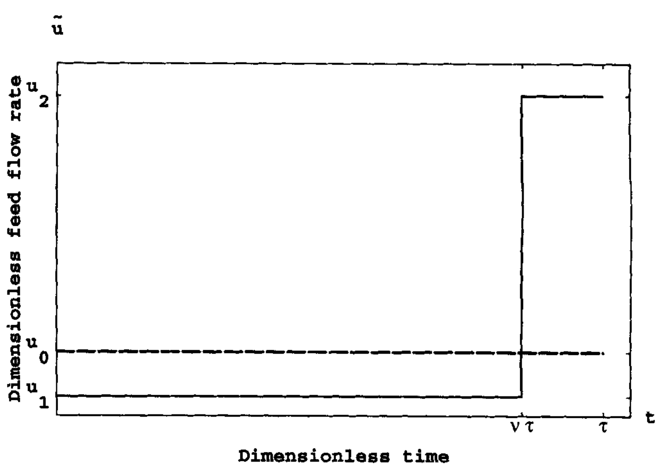

Figure 1. Two-step periodic forcing function.

Dashed line = average value u_0; parameters of forcing function = $u_1/u_0 = 0.3$; $u_2/u_0 = 5.0$.

where ν is duty fraction ($0 < \nu \leq 1$) and $n = 0, 1, 2, \ldots$.

From Eqs. 5 and 9 we have: $u_0 = 1/\tau \int_0^\tau \tilde{u}(t)dt = \nu u_1 + (1-\nu)u_2$.

Under the above assumption, temperature is regarded as a constant during the interval of averaging. So, using the well-known Jensen inequality for convex functions, we obtain

$$\eta_{u_0} = \frac{k(\theta)}{k(\theta)+u_0} = \frac{k(\theta)}{k(\theta)+[\nu u_1 + (1-\nu)u_2]}$$

$$\leq \nu\frac{k(\theta)}{k(\theta)+u_1} + (1-\nu)\frac{k(\theta)}{k(\theta)+u_2} = \frac{1}{\tau}\int_0^{\tau}\frac{k(\theta)}{k(\theta)+\tilde{u}}dt = \hat{\eta}_{\tilde{u}}$$

The same inequality is correct for all n-step functions. Every measurable bounded function can be approximated by n-step function as closely as needed. So, the inequality (Eq. 8) is correct for all these functions $u \geq 0$.

The analytical result means that *any kind of mediating operation enables desired increasing average conversion at all given temperature values*.

Now the obtained analytical result can be applied to determination of average steady-state responses with the help of a simple geometrical technique. Geometrically, average steady states are determined by intersection of θ-isocline and researched above the slow curve in the phase plane (θ, η). The θ-isocline is defined by the first equation of Eq. 7 which gives

$$\hat{\eta} = 1 - \frac{\hat{u}\theta + \alpha(\theta - \theta_c)}{Bk(\theta)} \qquad (10)$$

It is clear from Eq. 10 that the θ-isocline depends only upon the average value of feed flow rate $\hat{u}$ which is assumed to be a constant ($\hat{u} = u_0$). Hence, θ-isocline is a stationary curve in phase plane (θ, η). Its typical form is shown in Figure 2 where θ-isocline has a single minimum point separating domains of monotonic behavior.

As it was proved earlier, zero-average flow rate inputs lead to growing of an average conversion for the fixed temperature values, i.e., the slow curve defined by the second equation of Eq. 7 as a monotonic function moves above in phase

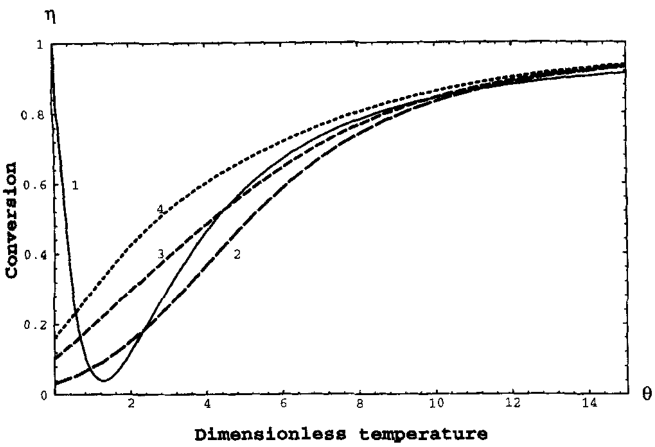

Figure 2. Steady states as intersections of θ-isocline (solid line) and slow curves (dashed lines) in phase plane (θ, η).

(1) = θ-isocline; (2) = slow curve of unforced reactor ($u(t) \equiv u_0$); (3) = slow curve for forcing function: $u_1/u_0 = 0.1$, $u_2/u_0 = 1.5$; (4) = slow curve for forcing function: $u_1/u_0 = 0.1$, $u_2/u_0 = 2.5$; $u_0 = 30.0$. Parameter values: $\alpha = 5$, $B = 15$, $\theta_c = 0$, $\beta = 0.1$.

plane (θ, η). The slow curve behavior is illustrated in Figure 2 for two-step periodic control defined by Eq. 9.

Hence for geometrical reasons (Figure 2), intersections of a stationary θ-isocline and moving up a slow curve determine the average steady-state responses upon mediating operation as follows:

For upper steady-state temperature and average conversion decrease (in different ranges, supposedly).

For lower steady-state temperature decreases while average conversion increases.

Note that these results are true for a large class of measurable forcing functions under assumption of mediating operation (Eq. 4), in particular, for *noise* too.

Simulation Results

Quantitative analysis aims at exploring the possibilities of conversion improvement for lower and upper steady states so that the desired stabilization of this effect can be achieved. Numerical simulation studies have been carried out for two-step forcing function $\tilde{u}(t)$ (see Figure 1), defined by Eq. 9. Note that the selected forcing function gives the same average output as a nonsymmetric rectangular waveform (see, for example, Rigopoulos et al., 1988). Thus by using relation $Da = 1/u_0$ the second equation of the system (Eq. 7) becomes

$$\hat{\eta} = \frac{k(\theta)[k(\theta) + u_2 + u_1 - u_0]}{[k(\theta) + u_1][k(\theta) + u_2]}$$

$$= \frac{Dak(\theta)[Dak(\theta) + n_2 + n_1 - 1]}{[Dak(\theta) + n_1][Dak(\theta) + n_2]} \quad (11)$$

Firstly, we use the traditional form of steady-state characteristics as functions of the Damköhler number ($= k_0 \exp(-1/\beta)V/F_0$ (see, for example, Uppal et al., 1974, 1976), in our case $Da = 1/u_0$. By substitution, the expression of slow curve (Eq. 11) in the equation of θ-isocline (Eq. 10), the third-order algebraic equation of variable Da for fixed value θ can obtain

$$Da^3[k(\theta)]^2\alpha(\theta - \theta_c) + Da^2k(\theta)[k(\theta)(\theta - B) + \alpha(\theta - \theta_c)(n_1 + n_2)] + Da[k(\theta)\theta(n_1 + n_2) + \alpha(\theta - \theta_c)n_1n_2 - k(\theta)Bn_1n_2] + \theta n_1n_2 = 0 \quad (12)$$

Steady-state values of average conversion are determined from Eq. 11 under the solution of Eq. 12 to Da. Average steady-state characteristics of temperature and conversion are shown in Figure 3. As predicted analytically earlier, zero-average inputs lead to decreasing temperature and average conversion for upper steady state and to decreasing temperature with increasing average conversion for lower one (Figure 3).

Mediating operation enables a stabilized operation in the vicinity of an unstable steady state of the unforced reactor, where lower stable steady state of the forced system can be obtained. Reactor operation stabilization can be achieved by moving the unstable area away (to the right side) from the steady-state profile of the unforced reactor (curve 2 in Figure 3) or by eliminating the unstable area of an S-shaped curve with full stabilization of reactor behavior (curve 3 in Figure 3). Note that a strong influence of the mediating operation on the stability properties can be expressed both in the disappearance of the unstable steady-state region and in its in-

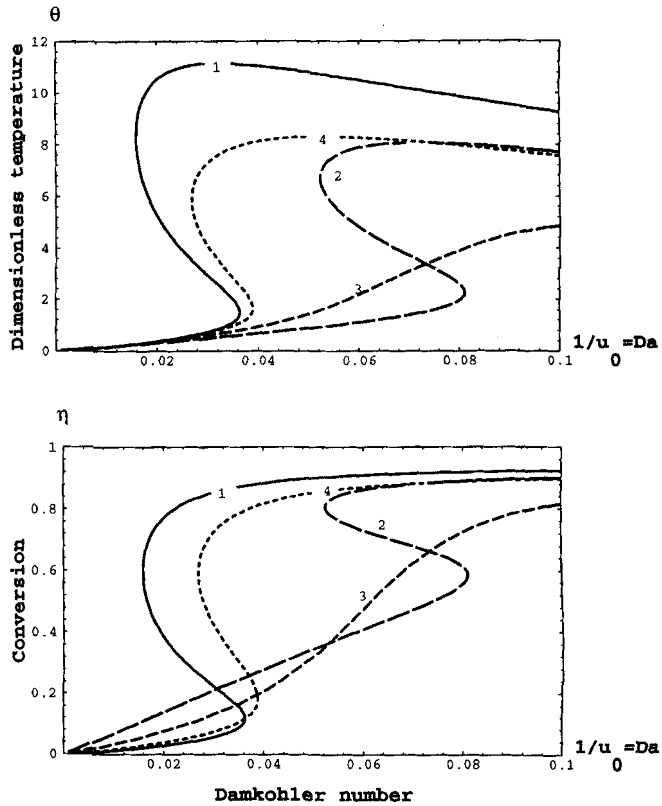

Figure 3. Average steady-state characteristics vs. Damköhler number.

(1) = unforced reactor; (2) = $u_1/u_0 = 0.1$, $u_2/u_0 = 2.5$; (3) = $u_1/u_0 = 0.3$, $u_2/u_0 = 5.0$; (4) = $u_1/u_0 = 0.7$, $u_2/u_0 = 10.0$. Parameter values are the same as in Figure 2.

crease (curves 3, 2 in Figure 3, respectively). The stabilization possibilities of mediating operation will be the topic of a further study.

One of the most desirable results of reactor control is to avoid a worsening of reactor productivity with decreasing reactor temperature in irreversible reactions. Mediating operation creates an opportunity for increasing average conversion with a lower temperature by separation of dynamic responses for conversion and temperature (see the analytical results obtained above). For lower steady-state conversion, improvement can be realized directly by mediating operation due to the temperature and conversion changes of desired directions (Figure 3). In Figure 3 curve 2 indicates the most pronounced effect of increasing conversion. Note that for lower steady-state, temperature can be changed only within a small range (Figure 3). The conversion improvement of the upper steady state arises from different ranges of the temperature and average conversion decrease (curve 4 in Figure 3) and will be considered in detail below.

Let us continue the discussion of reactor performance improvement and estimate the possibility of controlling the upper steady state. That means that we look for a domain of control parameters u_1 and u_2 where temperature can be decreased in a wide range while reactor productivity keeps its (relatively unforced reactor) high level.

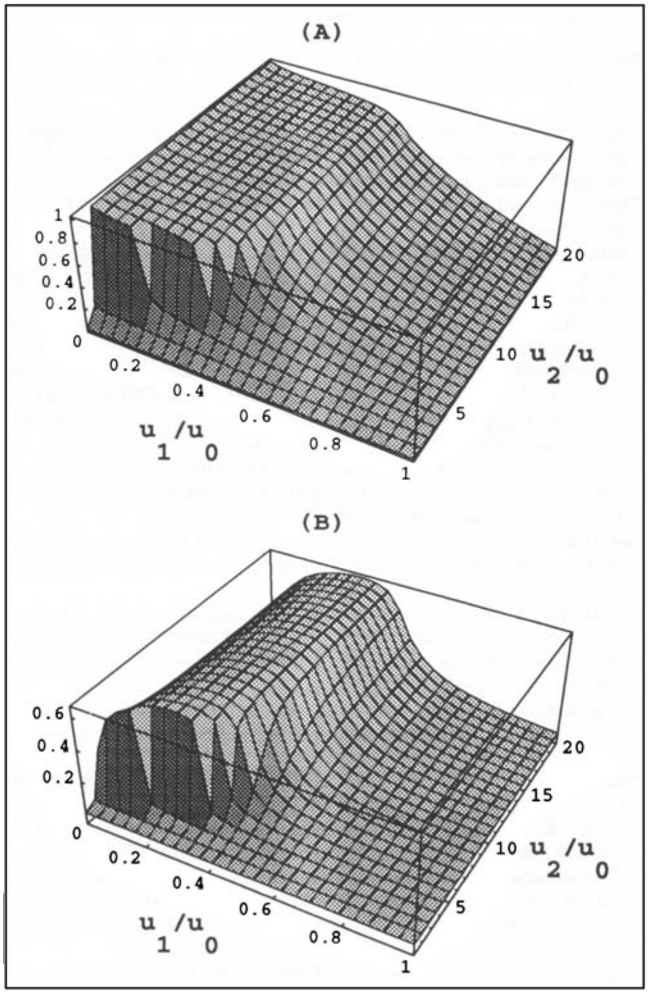

Figure 4. Upper steady-state behavior vs. parameters of two-step forcing function.

(A): relative changes of dimensionless temperature $(1-\theta_{\bar{u}}/\theta_{u_0})$; (B): relative changes of conversion $(1-\eta_{\bar{u}}/\eta_{u_0})$; $u_0 = 20.0$. Parameter values are the same as in Figure 2.

The system of Eqs. 10 and 11 has been solved numerically to upper temperature steady state and corresponding conversion as functions of two variables u_1/u_0 and u_2/u_0 with fixed value u_0. The surfaces in Figure 4 show changes of the temperature and average conversion relative to the appropriate parameters of the unforced reactor. Figure 4 indicates the existence of a *derangement* domain with a short transition of the upper steady state to a single lower one, where stabilized operation cannot be expected.

Isolines of surfaces of Figure 4 are plotted in Figure 5, which allows relative temperature changes to be compared with relative conversion changes caused by mediating operation for steady state. In Figure 5 the region of control parameters u_1/u_0, u_2/u_0 can be located where relative decrease of temperature is greater than 30%, while conversion decreases less than 10%. The region of stabilized operation (emphasized in Figure 5) is characterized, on the whole, by large amplitude of one pulse, $u_2/u_0 > 5$ and its small duty fraction ν, obviously. This type of mediating operation resulting in a stabilized effect of conversion improvement can be designated as *stroboscopic control*, which can be considered as an effective control mechanism of upper steady state,

Note that similar possibilities in shaping steady-state characteristics of system responses were obtained earlier by vibrational control with *multivariable* control strategies (Cinar et al., 1987b; Rigopoulos et al., 1988; Shu et al., 1989). Mediating operation provides different responses of the fast concentration and the slow temperature processes, so its benefit is the ability to separate the reactor process responses by only a *single* input variable.

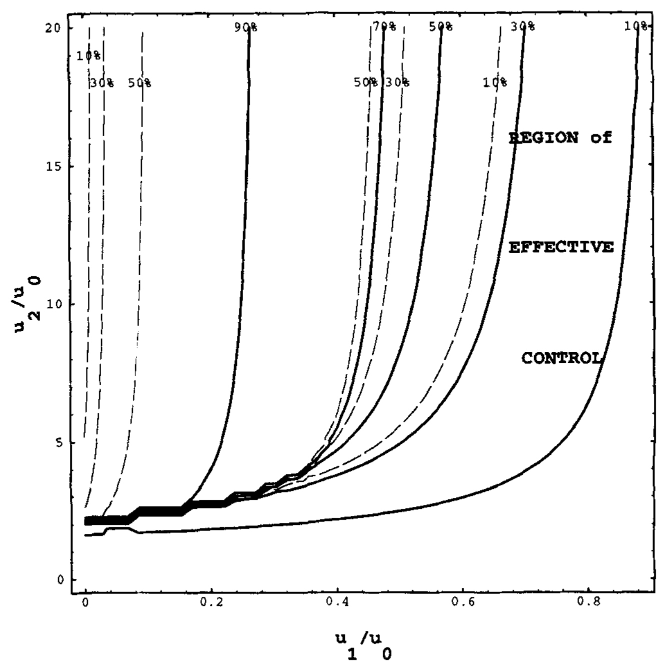

Figure 5. Isolines in percentage of surfaces in Figure 4.

Solid line = relative changes of dimensionless temperature, dashed line = relative changes of conversion.

Conclusion

A simple CSTR model of a heterogeneous catalytic reactor has been considered as a multiscale system with widely separated slow temperature processes and fast concentration ones. A new mediating operation by zero-average feed flow rate inputs has been studied for steady-state reactor behavior. Mediating operation has been analyzed as an open-loop control approach operating in intermediate mode between fast and slow reactor processes.

A mathematical CSTR model has been derived as a nonautonomous singularly perturbed system of two nonlinear ordinary differential equations, the analysis of which employs a combination of slow manifold method, averaging method, and numerical simulation study. From our point of view, this combination of qualitative analysis and the numerical methods is natural for a multiscale system investigation and we tried to demonstrate its effectiveness for the considered CSTR problem.

Due to the ability for increasing average conversion at a given temperature by mediating operation, the following results can be noted:

(1) For lower steady-state, mediating operation causes a desired separation of reactor responses—*increasing conversion* with *decreasing temperature. Stabilized lower steady state* of the forced system can be achieved in the vicinity of an unstable steady state of the unforced system.

(2) For upper steady state, a reactor response separation can be realized with *decreasing temperature* in a wide range, practically, *without worsening of reactor productivity*. Stroboscopic control can be viewed as a stabilizing mechanism for this effect.

The obtained specific results permit us to conclude that mediating operation enables providing all general control purposes for the reactor system. It is indeed shown that operation with higher reactor productivity at lower temperature can be stabilized both for lower and upper steady states.

Proposed mediating operation creates wide possibilities of control by its beneficial ability to *separate two dynamic responses* of fast and slow subsystems by a *single manipulated variable*. Mediating operation acts as an intermediary between slow and fast subsystems of a multiscale system; thus control of two-scale interaction is provided. The mediating operation approach can be viewed as a promising area of control operation for a wide spectrum of multiscale systems.

Notation

A = heat-transfer area
$B = (-\Delta H)C_f/(\rho c_p)/(\beta T_f)$
c_p, c_s = heat capacity of reacting mixture and solid phase, respectively
C, C_f = concentration, feed concentration
E = activation energy
F, F_0 = feed flow rate and its average value
h = heat-transfer coefficient
k_0 = reaction rate constant
$k(\theta) = \exp[\theta/(1+\beta\theta)]$
m = mass of solid catalyst
$n_1 = u_1/u_0$
$n_2 = u_2/u_0$
R = universal gas constant
T, T_f, T_c = reactor temperature, feed temperature, coolant temperature
u = dimensionless feed flow rate $= (F/V)/[k_o \exp(-1/\beta)]$
u_0 = average value of dimensionless feed flow rate $= (F_0/V)/[k_0 \exp(-1/\beta)] = 1/\mathrm{Da}$
V = volume of gas reaction mixture

Greek letters

$\alpha = hA/(\rho c_p)/[k_0 \exp(-1/\beta)V]$
$\beta = RT_f/E$
θ_c = dimensionless coolant temperature $= (T_c - T_f)/\beta T_f$
ρ = density of the reacting mixture
τ = dimensionless period of control variable and period of averaging

Superscripts and Subscript

$\hat{*}$ = averaged value
$\tilde{u}(t)$ = two-step periodic function, defined under Eq. 9
$u, u_0, \tilde{u}$ = state of feed flow rate

Literature Cited

Bailey, J. E., "Periodic Operation of Chemical Reactors: A Review," *Chem. Eng. Commun.*, **1**, 111 (1973).

Bailey, J. E., "Periodic Phenomena," *Chemical Reactor Theory: A Review*, L. Lapidus and N. R. Amundson, eds., Prentice-Hall, Englewood Cliffs, New York, p. 758 (1977).

Bellman, R., J. Bensman, and S. M. Meerkov, "Vibrational Control of Systems with Arrhenius Dynamics," *J. Math. Anal. Appl.*, **91**, 152 (1983a).

Bellman, R., J. Bensman, and S. M. Meerkov, "Nonlinear Systems with Fast Parametric Oscillations," *J. Math. Anal. Appl.*, **97**, 572 (1983b).

Bogoliubov, N. N., and Yu. A. Mitropolsky, *Asymptotic Methods in the Theory of Nonlinear Oscillations*, Fizmatigis, Moscow (1958). (English translation: Gordon and Breach, New York (1961)).

Chang, M., and R. A. Schmitz, "An Experimental Study of Oscillatory States in a Stirred Reactor," *Chem. Eng. Sci.*, **30**, 21 (1975).

Cinar, A., J. Deng, S. M. Meerkov, and X. Shu, "Vibrational Control of an Exothermic Reaction in a CSTR: Theory and Experiments," *AIChE J.*, **33**, 353 (1987a).

Cinar, A., K. Rigopoulos, and S. M. Meerkov, "Vibrational Control of Chemical Reactors: Stabilization and Conversion Improvement in an Exothermic CSTR," *Chem. Eng. Commun.*, **59**, 299 (1987b).

Gol'dshtein, V. M., and O. V. Kiselev, "Behavior of CSTR upon Fast External Effects: Possibility of Increasing Average Conversion," *React. Kinet. Catal. Lett.*, **34**, 349 (1987).

Gol'dshtein, V. M., and V. A. Sobolev, *Qualitative Analysis of Singularly Perturbed Systems* (In Russian), Acad. Nauk SSSR, Sib. Otd. Inst. Math., Novosibirsk (1988).

Gol'dshtein, V. M., and V. A. Sobolev, "Singularity Theory and Some Problems of Functional Analyses," *Amer. Math. Soc. Transl.*, 2, **153**, 73 (1992).

Gol'dshtein, V. M., O. V. Kiselev, A. S. Romanov, and S. A. Treskov, "Multiplicity of Steady States for CSTR Upon Fast External Effects," *Mathematical Problems of Chemical Kinetics*, (In Russian), Nauka, Novosibirsk (1990).

Rigopoulos, K., X. Shu, and A. Cinar, "Forced Periodic Control of an Exothermic CSTR with Multiple Input Oscillations," *AIChE J.*, **34**, 2041 (1988).

Sheintuch, M., and R. A. Schmitz, "Oscillations in Catalytic Reactions," *Catal. Rev. Sci. Eng.*, **15**, 107 (1977).

Shu, X., K. Rigopoulos, and A. Cinar, "Vibrational Control of an Exothermic CSTR: Productivity Improvement by Multiple Input Oscillations," *IEEE Trans. Autom. Control*, **34**, 193 (1989).

Uppal, A., W. H. Ray, and A B. Poore, "On the Dynamic Behavior of Continuous Stirred-Tank Reactors," *Chem. Eng. Sci.*, **29**, 967 (1974).

Uppal, A., W. H. Ray, and A B. Poore, "The Classification of the Dynamic Behavior of Continuous Stirred-Tank Reactors—Influence of Reactor Residence Time," *Chem. Eng. Sci.*, **31**, 205 (1976).

Manuscript received Feb. 22, 1994, and revision received Nov. 27, 1995.