Optimal Feedforward Control of Concurrent Tubular Reactors

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Optimal control of concurrent nonlinear tubular reactors for complex reaction system is studied. The jacket side temperature is allowed to vary along the reactor length and with time. The optimal control possesses feedforward component only, and the nonlinear gain may be precomputed. For a Denbigh type of reaction system, if the heat generation is negligible, the control temperature is insensitive to feed concentration variations. With heat generation effect present, the optimal control is time dependent, and the nonlinear feedforward gain is obtained. The gain may be approximated by a linear function to construct a simple linear feedforward control loop which contains a synchronized time delay.

A number of papers dealing with feedforward control of distillation columns, continuous stirred tank reactors, and other processes have been published in the past (*1* to *17*, *26*). In these and many other chemical processes, the existence of various devices for measuring disturbances and on-line computers makes feedforward control particularly suitable and attractive.

For tubular reactors, Tinkler and Lamb (*15*) applied linear methods and frequency domain analysis to synthesize feedforward controllers. A parametrically forced isothermal plug flow tubular reactor was studied by Koppel (*16*). He discussed feedforward manipulation of control variable analytically obtained for his process. Luyben (*17*) treated the synthesis of nonlinear feedforward controllers for continuous stirred tank and batch reactors. He also studied a simplified nonisothermal plug flow tubular reactor and discussed difficulties associated with implementing the control equation. He pointed out the fact that in feedforward control of ideal tubular reactors, practical problems arise because the reactor temperature must be varied continuously with time and reactor length as required by the control temperature profile. Recently, optimal feedback control for a class of distributed parameter systems has been reported (*23*, *25*). For a linear distributed system, it was shown by Denn (*24*) that the optimal controller possesses a linear feedback-feedforward control structure with entirely precomputed gains.

We treat in this paper the optimal control of jacketed concurrent tubular reactors. The complex reaction system treated here is the one originally treated by Denbigh (*18*):

$$\begin{array}{ccc} & k_1 & & k_3 & \\ A & \rightarrow & B & \rightarrow & C \\ \downarrow k_2 & & \downarrow k_4 & & \\ D & & E & & \end{array}$$

where C is the desired product, and k_i $(i = 1, 2, 3, 4)$ depends on temperature by the Arrhenius expression. It has been shown by Denbigh that the reaction system is uniquely characterized by six ratios between the eight kinetic parameters. They are

$$w_1 = \frac{k_{20}}{k_{10}},\ w_2 = \frac{k_{30}}{k_{10}},\ w_3 = \frac{k_{40}}{k_{30}},\ q_1 = \frac{E_1 - E_2}{R},$$

$$q_2 = \frac{E_1 - E_3}{R}, \text{ and } q_3 = \frac{E_3 - E_4}{R}$$

In the treatment of two-stage stirred tank reactor problem, he assigned values

$$w_1 = 10^4,\ w_2 = 10^{-2},\ w_3 = 10^{-4},\ q_1 = -3{,}000,$$
$$q_2 = 0, \text{ and } q_3 = 3{,}000$$

Several other authors (*19* to *21*) investigated ideal steady state temperature policies in continuous stirred tank and tubular reactors for the reaction system and showed that temperatures as high as 1,000°K. were needed at the end of the optimal policies. Horn and Troltenier (*20*) included values of $w_2 = 10^{-1}$, 10^0, and 10^1 in their steady state tubular reactor optimization. The ideal yields from such ideal steady state reactors provide the theoretical upper bounds for comparison in the optimal performance of present concurrent tubular reactors.

CONTROL EQUATION

For use in the analysis of the problem, the following development is needed. We consider the optimal control problem for the following system of first-order partial differential equations:

$$\frac{\partial v}{\partial t} = f[t, x, v(t,x)] - N(t)\frac{\partial v}{\partial x} \qquad t \in [0, t_f],\ x \in [0, L] \tag{1}$$

with initial and boundary distributions

$$v(0, x) = \Phi(x) \qquad \text{at} \quad t = 0 \tag{2}$$

$$v(t, 0) = \Psi[t, u(t), d(t)] \qquad \text{at} \quad x = 0 \tag{3}$$

where the n dimensional state vector $v(t, x)$ is involved nonlinearly in f. Since N can be diagonalized, if it is not in a canonical form, by a similarity transformation $P^{-1}NP$ = diagonal matrix, it is assumed that Equation (1) is already in the canonical form. The boundary control and disturbance $u(t)$ and $d(t)$ may be distributed linearly, $\Psi = B\,u(t) + G\,d(t)$, in which case B and G are probably diagonal matrices such that $B + G = I$. The minimizing performance index is

$$J = \int_0^{t_f}\int_0^L \mathcal{G}\ dxdt \tag{4}$$

where $\mathcal{G} = \langle g(t), \frac{\partial v}{\partial x} \rangle$ in which no control cost is

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assigned.

Form a Hamiltonian function H:

$$H\left(t, x, v, \frac{\partial v}{\partial x}, p \right) = -\mathcal{G} + < p, \left[f(t, x, v) - N \frac{\partial v}{\partial x} \right] > \quad (5)$$

It can be shown (22) that H must be maximum almost everywhere, in particular, maximum at $x = 0$ for all $t \in [0, t_f]$. Then, at $x = 0$

$$H\left[t, 0, v(t, 0), \frac{\partial v}{\partial x}(t, 0), p(t, 0) \right] = < g(t), \frac{\partial v}{\partial x}(t, 0) > + < p(t, 0), [f(t, 0, v(t, 0))] - N(0) \frac{\partial v}{\partial x}(t, 0) > \quad (6)$$

where $v(t, 0)$ can be replaced from Equation (3) to obtain the optimal control

$$\bar{u}(t) = \left\{ u(t) : \underset{u(t)}{\text{Max}} \; H(t, 0, \Psi[t, u(t), d(t)], \bar{\mu}(t, 0), \bar{p}(t, 0)) \right\} \quad (7)$$

Optimal $\bar{\mu}(t, 0) \left[= \frac{\partial v}{\partial x}(t, 0) \right]$ and $\bar{p}(t, 0)$ are evaluated along $x = 0$. If $H(t, 0)$ is maximum at some interior point of the control region, then the optimal control $\bar{u}(t)$ expressed in Equation (7) is the solution of

$$\nabla_u H(t, 0) = 0 \quad (8)$$

To evaluate $\bar{p}(t, 0)$, we have to solve for $p(t, x)$. The system for $p(t, x)$ is

$$\frac{\partial p}{\partial t} = (\nabla_\mu H)_x - \nabla_v H \quad (9)$$

subject to final and boundary conditions

$$p(t_f, x) = 0, \; \nabla_\mu H(t, L) = 0 \quad (10)$$

From Equations (5) and (9), it is evident that

$$\frac{\partial p}{\partial t} = <h(t, x, v), p> - N(t) \frac{\partial p}{\partial x} \quad (11)$$

where $h(t, x, v)$ is a vector valued function of $v(t, x)$.

CONSTRUCTION OF GAIN EQUATION

The proper representation of $p(t, x)$ to seek a solution for a linear distributed system with control and disturbance distributed both in interior and on boundary has been shown by Denn (24) to be

$$p(t, x) = \int_0^L M(t; x, \eta) v(t, \eta) d\eta + Q(t, x) \quad (12)$$

where $Q(t, x)$ depends only on the disturbance. In an analogous manner, we seek a solution with $p(t, x)$ represented by

$$p(t, x) = \int_0^L K[t, x, \eta; \; v(t, \eta)] \, d\eta + Q(t, x) \quad (13)$$

where $Q(t, x)$ depends only on the disturbance on the boundary.

Introduce the characteristic derivatives

$$\frac{d}{ds_i} = \frac{\partial}{\partial t} + \nu_i \frac{\partial}{\partial x} \; (i = 1, 2, \ldots n) \quad (14)$$

where ν_i are the diagonal elements of matrix N. An equivalent expression is $dx/ds_i = \nu_i$, in which s_i can be viewed as the parameter along the i^{th} characteristic curve. Then, Equation (13) becomes

$$p_i[t(s_i), x(s_i)] = \int_{\eta(s_i)=0}^{\eta(s_i)=L} K_i[t(s_i), x(s_i), \eta(s_i); \; v(t(s_i), \eta(s_i))] \, d\eta(s_i) + Q_i \, [t(s_i), \; x(s_i)] \quad (15)$$

$$(i = 1, 2, \ldots, n)$$

In the kernel, $v[t(s_i), \eta(s_i)]$ is a vector containing elements v_m $(m = 1, 2, \ldots, n)$ which must be evaluated not only along the i^{th} characteristic curve but also along the total set of other $m = 1, 2, \ldots, n$ $(m \neq i)$ interacting characteristic curves emanated from the Cauchy boundary. This makes the evaluation of $p_i[t(s_i), x(s_i)]$ very difficult. However, if $\nu_i = \nu$ for all $i = 1, 2, \ldots, n$, then $s_i = s$ for all $i = 1, 2, \ldots, n$, and all the characteristic curves for the elements of v coincide and are given by s. In view of Equation (11), the characteristic curves of p all coincide also and are given by the same single characteristic curve represented by s:

$$p_i[t(s), x(s)] = \int_{\eta(s)=0}^{\eta(s)=L} K_i[t(s), x(s), \eta(s); \; v(t(s), \eta(s))] \, d\eta(s) + Q_i[t(s), x(s)] \quad (16)$$

Along this s, $v[t(s), \eta(s)]$ is uniquely determined by $\Phi(x)$ or $\Psi[t, u(t), d(t)]$, the Cauchy data of $v(t, x)$; thus

$$v[t(s), \eta(s)] = \begin{cases} F(\Phi(x(0))) & \text{or} \\ F(u(t(0)), d(t(0))) \end{cases} \quad (17)$$

where F is a transition matrix like function along the characteristic s. The substitution of Equation (17) into (16) shows that

$$p_i[t(s), x(s)] = F_i(t(s), \; x(s), \; u(t(0)), \; d(t(0))) + Q_i[t(s), x(s)] \quad (18)$$

where F_i is some function. Since Q_i also depends only on the disturbance alone, F_i and Q_i can be combined to give

$$p[t(s), x(s)] = \Lambda[t(s), x(s)] \quad (19)$$

where $\Lambda[t(s), x(s)]$ depends only on $u[t(0)]$ and $d[t(0)]$. This can be looked upon as the kernel $M = 0$ and $Q = \Lambda$ in Equation (12). The final and boundary conditions for Λ are identical with those of p.

By using a similar procedure applied by Denn, it can be shown that the feedback component of gain equation vanishes in view of Equation (19), and the feedforward component is given by [see Equation (23) of Denn's work]

$$u(t) = \Gamma[d(t)] \quad (20)$$

where Γ is a nonlinear function of $d(t)$. Now that the disturbance appears physically only in the entrance conditions [Equation (3)], such an arbitrary disturbance can be premeasured by a monitoring device at a position in the process upstream and hence can be treated as deterministic. Consequently, the disturbance function may be allowed to be a series of steps of random height and occurrence. The feedforward gain may be approximated by a linear function once the true nonlinear gain has been precomputed from the optimal control solution of Equation (7).

CONCURRENT REACTOR

The reactor being considered here is a jacketed concurrent plug flow tubular reactor of length L in which the above reactions with and without heat generation take place. The jacket side flow rate is of a comparable order of magnitude as that of the reactant fluid in the tube, so that the temperature in the jacket varies along the reactor length and with time. The object is to obtain the time dependent optimal inlet jacket temperature for any continual disturbance in the feed concentration such that the cumulative yield of the desired product C from the reactor over a time interval be maximized. The process dynamics describing this process is given by the following system of nonlinear partial differential equations:

$$
\begin{aligned}
\frac{\partial v_1}{\partial t} &= -(k_1 + k_2)v_1 - \nu \frac{\partial v_1}{\partial x} \\
\frac{\partial v_2}{\partial t} &= k_1 v_1 - (k_3 + k_4)v_2 - \nu \frac{\partial v_2}{\partial x} \\
\frac{\partial v_3}{\partial t} &= k_3 v_2 - \nu \frac{\partial v_3}{\partial x} \\
\frac{\partial v_4}{\partial t} &= b_1 k_1 v_1 + b_2 k_2 v_1 + b_3 k_3 v_2 + b_4 k_4 v_2 \\
&\quad + \frac{1}{\alpha}(v_5 - v_4) - \nu \frac{\partial v_4}{\partial x} \\
\frac{\partial v_5}{\partial t} &= -\frac{1}{\alpha_0}(v_5 - v_4) - \nu_0 \frac{\partial v_5}{\partial x}
\end{aligned}
\tag{21}
$$

subject to initial and boundary conditions

$$v_i(0, x) = \Phi_i(x) \quad \text{and} \quad v_i(t, 0) = \Psi_i(t) \tag{22}$$

Here $k_i = k_{i0} \exp(-E_i/Rv_4)$ $(i = 1, 2, 3, 4)$ in which v_4 is nonlinearly involved. The functions $\Phi_i(x)$ $(i = 1, 2, 3, 4, 5)$ are characterized by either optimal or nonoptimal steady state operating conditions, and the functions $\Psi_i(t)$ are either the disturbances or the boundary control variables of the system. In practice, it generally happens that the feed concentrations are disturbances and the easily manipulative variables such as temperatures are control variables. If the reactions accompany negligible heat generation, then $b_i = 0$ $(i = 1, 2, 3, 4)$; otherwise $b_i \neq 0$ in the process equations.

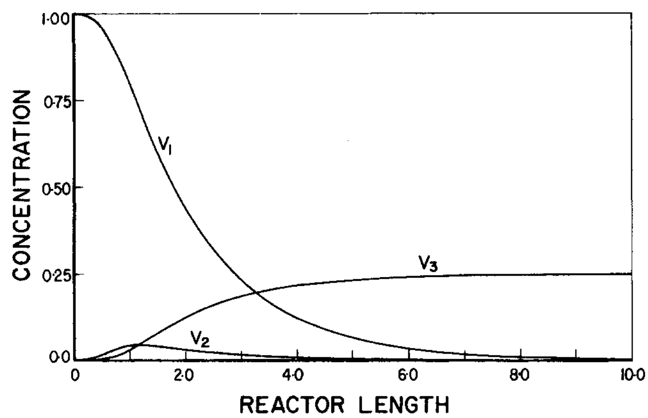

Fig. 1. Optimal steady state concentration profiles, $b_i = 0$ $(i = 1, 2, 3, 4)$.

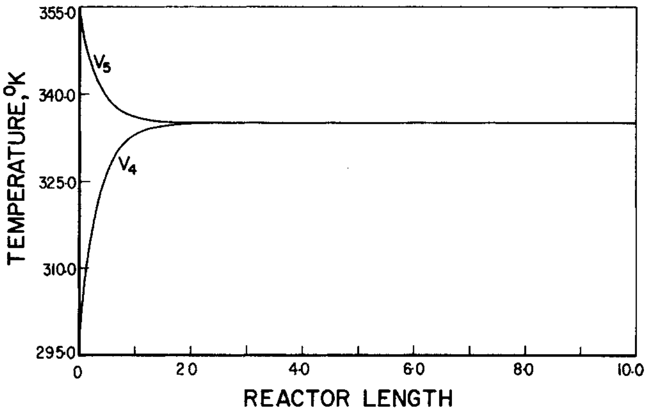

Fig. 2. Optimal steady state temperature profiles, $b_i = 0$ $(i = 1, 2, 3, 4)$.

Given a time interval $[0, t_f]$, the minimizing objective functional is then represented by

$$J = \int_0^{t_f} \int_0^L \left(-\frac{\partial v_3}{\partial x} \right) dx dt \tag{23}$$

minimization being carried out over the specified control variables $\Psi_i(t)$ $(i \neq 3)$.

Now the numerical values of kinetic parameters from the Denbigh's ratio requirements are given by

$$
\begin{aligned}
k_{10} &= 10^{15} \text{ min.}^{-1} & E_1 &= 24{,}000 \text{ cal./g.-mole} \\
k_{20} &= 10^{19} & E_2 &= 30{,}000 \\
k_{30} &= 10^{16} & E_3 &= 24{,}000 \\
k_{40} &= 10^{12} & E_4 &= 18{,}000
\end{aligned}
$$

These values are based on the ratio $w_2 = 10$. For the case without heat generation, $b_i = 0$ $(i = 1, 2, 3, 4)$, and with heat generation, $b_1 = -50$, $b_2 = 30$, $b_3 = 100$, and $b_4 = 80$. In addition, the following numerical values are introduced: $R = 2.0$, $L = 10$ units of length, $\nu = 1.0$ units of length/min., $\nu_0 = 1.0$ unit of length/min., $\alpha = 0.5$, and $\alpha_0 = 1.0$. The choice of ν and L gives the reactor residence time of 10 min.

The disturbance of the process through the boundary conditions may be allowed to be an arbitrary function, since it can be premeasured in the reactor upsrteam. However, for the sake of convenience, a sinusoidal disturbance in the feed concentration of raw material A is introduced with other feed conditions fixed at the steady state values. Then

$$
\begin{aligned}
d(t) = \Psi_1(t) &= v_{1s} + A \sin \omega t \\
\Psi_i(t) &= v_{is} \quad (i = 2, 3) \\
\Psi_4(t) &= v_{4s} \quad \text{(optimal)} \\
u(t) &= \Psi_5(t)
\end{aligned}
\tag{24}
$$

The function $u(t)$ is the control variable. Arbitrary or optimal initial conditions $\Phi_i(x)$ can also be specified. However, if the disturbance is periodic, $\Phi_i(x)$ are the functions describing the optimal state of the process resulted from the disturbance $d(t)$ of the previous period with the corresponding periodic optimal control.

METHOD OF SOLUTION

To solve this problem according to a variational technique, we construct the Hamiltonian function according to Equation (5) which is in extenso:

$$H = \frac{\partial v_3}{\partial x} + \left[-(k_1 + k_2)v_1 - \nu \frac{\partial v_1}{\partial x} \right] p_1 + \left[k_1 v_1 - (k_3 + k_4)v_2 - \nu \frac{\partial v_2}{\partial x} \right] p_2 + \left[k_3 v_2 - \nu \frac{\partial v_3}{\partial x} \right] p_3 + \left[\frac{1}{\alpha}(v_5 - v_4) + b_1 k_1 v_1 + b_2 k_2 v_1 + b_3 k_3 v_2 + b_4 k_4 v_2 - \nu \frac{\partial v_4}{\partial x} \right] p_4 + \left[-\frac{1}{\alpha_0}(v_5 - v_4) - \nu_0 \frac{\partial v_5}{\partial x} \right] p_5 \quad (25)$$

where the auxiliary system for adjoint variable p_i is by Equation (9):

$$\frac{\partial p_1}{\partial t} = (k_1 + k_2)p_1 - k_1 p_2 - (b_1 k_1 + b_2 k_2)p_4 - \nu \frac{\partial p_1}{\partial x}$$

$$\frac{\partial p_2}{\partial t} = (k_3 + k_4)p_2 - k_3 p_3 - (b_3 k_3 + b_4 k_4)p_4 - \nu \frac{\partial p_2}{\partial x}$$

$$\frac{\partial p_3}{\partial t} = -\nu \frac{\partial p_3}{\partial x}$$

$$\frac{\partial p_4}{\partial t} = \frac{1}{R v_4^2}\left[(E_1 k_1 + E_2 k_2)v_1 p_1 - (E_2 k_2 v_1 - E_3 k_3 v_2 - E_4 k_4 v_2)p_2 - E_3 k_3 v_2 p_3 - (b_1 E_1 k_1 v_1 + b_2 E_2 k_2 v_1 + b_3 E_3 k_3 v_2 + b_4 E_4 k_4 v_2)p_4\right] + \left(\frac{p_4}{\alpha} - \frac{p_5}{\alpha_0}\right) - \nu \frac{\partial p_4}{\partial x}$$

$$\frac{\partial p_5}{\partial t} = -\frac{p_4}{\alpha} + \frac{p_5}{\alpha_0} - \nu_0 \frac{\partial p_5}{\partial x} \quad (26)$$

subject to the final and boundary conditions

$$p_i\big|_{t=t_f} = 0 \text{ and } p_i\big|_{x=L} = \frac{\delta_{i3}}{\nu} \quad (i = 1, 2, 3, 4, 5) \quad (27)$$

According to the necessary condition for the optimal control, H must be maximum almost everywhere, in particular, maximum at $x = 0$ at all $t \in [0, t_f]$. Thus the variation on the objective functional for this case yields an iterative algorithm for the synthesis of optimal control:

$$\overset{(i+1)}{u}(t) = \overset{(i)}{u}(t) + \overset{(i)}{\epsilon}(t) \left[\frac{p_4(t, 0)}{\alpha} - \frac{p_5(t, 0)}{\alpha_0} \right]^{(i)} \quad (28)$$

for all $t \in [0, t_f]$. Equations (21) and (26) must be iteratively solved by using a sequence of improved controls according to Equation (28) until there is no significant improvement in $u(t)$ value for all t and in J value.

OPTIMAL CONTROL

We first find the optimal steady state. The steady state feed conditions are set at $v_{1s} = 1.0$ and $v_{is} = 0$ $(i = 2, 3)$ g.-mole/liter. Under these conditions the optimal $v_{4s} =$ 297°K. for $b_i = 0$ and 298°K. for $b_i \neq 0$, respectively. The optimal steady state temperature profiles and concentration profiles are shown in Figures 1 and 2 for $b_i = 0$. For $b_i \neq 0$, the profiles are shown in Figures 3 and 4. The profiles without heat generation change more rapidly in the initial section of the reactor than those with heat generation. In particular, the temperature profiles v_4 and v_5 for $b_i = 0$ monotonically approach an asymptotic value early in the reactor, whereas those for $b_i \neq 0$ show the heat generation effect in almost entire portion of the reactor. The optimal steady state percentage yields are 24.84 for the case with $b_i = 0$ and 25.03 for the case with $b_i \neq 0$, respectively. These values are, respectively, 0.36 and 0.17 less than the theoretically possible maximum percentage yield of 25.20 on perfectly ideal steady state reactors as reported by Horn and Troltenier (*20*) and Story (*21*).

We now compute the optimal control for the sinusoidal disturbance. The iterative computations for optimal control are carried out for oscillation periods $\tau (= 2\pi/\omega) = 10$, 15, 30, and 45 min. without $(b_i = 0)$ and with $(b_i \neq 0)$ heat generation. The amplitude of oscillation A is set at 0.50 g.-mole/liter. In the integration of the process dynamics in Equation (21) and the adjoint system in Equation (26), computations can be expedited if the characteristic derivative $d/ds \equiv \partial/\partial t + \nu(\partial/\partial x)$ is introduced. The independent variable s can be considered as the residence time of a bulk reactant in the reactor. Thus it is to be noted that $s \in [0, 10]$, while $t \in [0, \tau]$. The integration step size Δs is taken as 0.05, and the fourth-order Runge-Kutta integration scheme is used. The control iteration

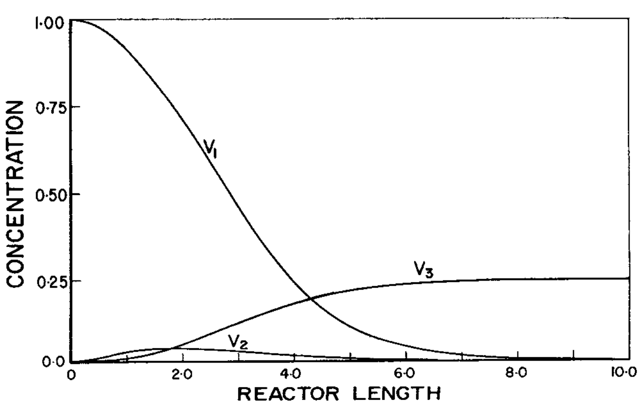

Fig. 3. Optimal steady state concentration profiles, $b_i \neq 0$.

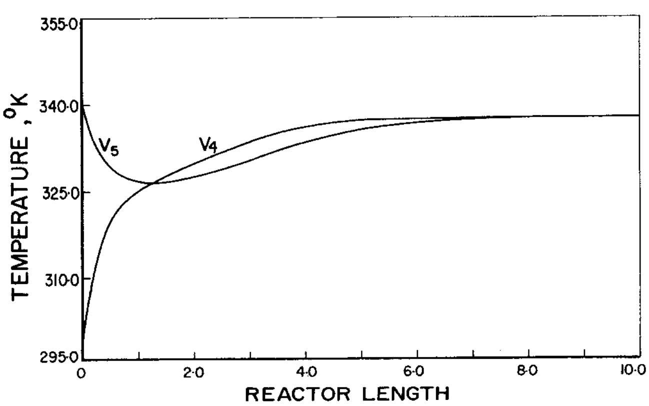

Fig. 4. Optimal steady state temperature profiles, $b_i \neq 0$.

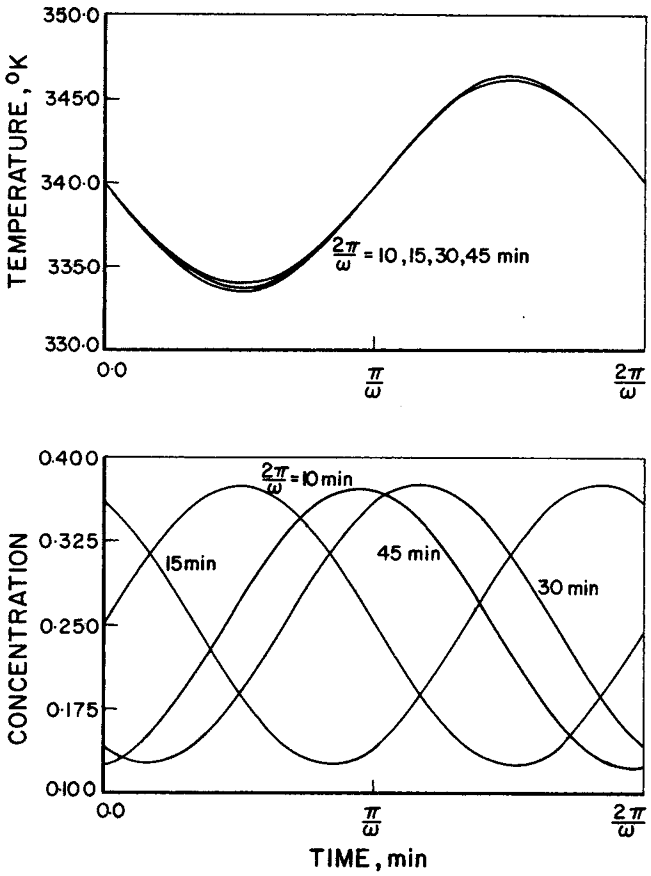

Fig. 5. Optimal control $u(t)$ **and optimal yield** $v_3(t, 10)$**,** $b_i \neq 0$**.**

step size $\epsilon(t)$ is adjusted in each iteration, depending on the success or the failure of the iteration.

Without heat generation, the optimal control for the sinusoidal disturbance turns out to be nearly constant at the value of 354°K. for all periods. This is also the optimal value for the steady state operation. The corresponding percentage yield is 24.84. This shows that the jacket temperature is insensitive to any variation in inlet feed condition of v_1 if there is negligible heat generation. But for the reaction with heat generation, the optimal controls are time dependent periodic functions for all periods as shown in Figure 5 for one period. The optimal controls are almost sinusoidal, and they all coincide with one another very well. The average percentage yield with optimal control is 25.03 for each case. If the control is maintained at the constant value of optimal steady state, then the yield is 24.79. Thus the percentage increase in the yield by applying the time dependent optimal control is 0.24.

FEEDFORWARD CONTROL

For the case with heat generation, the optimal control temperatures $u(t)$ in Figure 5 are now plotted with respect to a range of feed concentration changes $d(t) = 0.125 \sim 1.50$ g.-mole/liter. The result is shown in Figure 6. This is the nonlinear feedforward gain represented by Equation (20). As can be seen in Figure 6, the optimal gain is not strictly a straight line with respect to the disturbance. However, it can very well be approximated by a straight line, the slope being approximately −12.3 (°K.)/(g.-mole/liter). This implies that Equation (20) can be linearized for practical purposes. In view of this fact, a simple feedforward control can now be constructed. The optimal control for any arbitrary disturbance over the range of feed concentration change is now achieved approximately

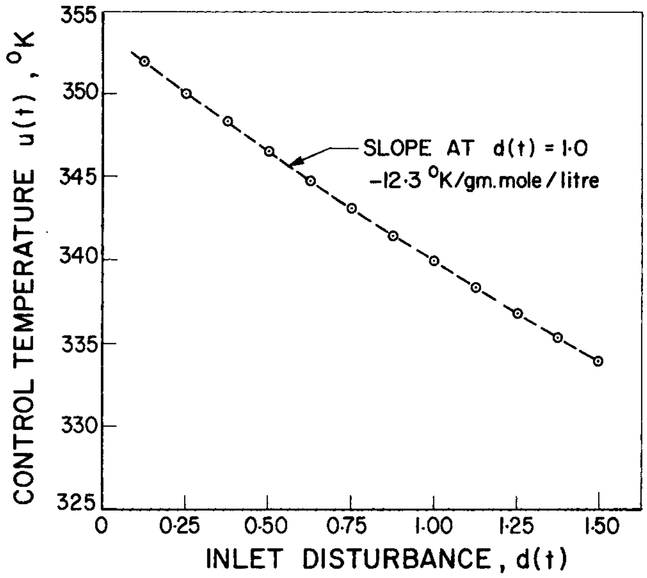

Fig. 6. Disturbance $d(t)$ **vs. optimal control** $u(t)$**,** $b_i \neq 0$**.**

by the feedforward linear control of the inlet jacket temperature $u(t)$ with a synchronized pure time delay. The time delay is to accommodate the transportation lag between the monitoring device and the reactor. The gain required for any arbitrary disturbance for such a controller is given by the slope of the optimal control −12.3 (°K.)/(g.-mole/liter).

CONCLUSIONS

The optimal control of jacketed concurrent nonlinear tubular reactors with complex reaction systems without and with heat generation allowing the jacket temperature to be a function of distance and time has been synthesized by a method derived from variational technique on a system of nonlinear first-order partial differential equations. It has been shown that the optimal control for the process contains feedforward component only, and the gains may be precomputed in the range of disturbance.

For the Denbigh type of reaction system, when the reactor is subjected to a disturbance in inlet feed concentration, without heat generation, the optimal control and the yield remain the same as in the steady state operation. With heat generation effect present, the optimal control becomes a time dependent function, and the yield increases over that with steady state average control. However, if a small loss in yield is tolerable, a steady state optimal control with a proper consideration of heat generation effect is as nearly adequate as the ideal time optimal control and indeed the control of an ideal reactor with unlimited control efforts. On the other hand, if a fractional improvement is important (expensive product or production in large quantity), this can be achieved by implementing the time dependent feedforward optimal control with precomputed gain. Furthermore, in view of the fact that the relationship between the optimal control and the inlet feed disturbance is nearly linear, the optimal feedforward control can be implemented approximately by a simple feedforward control loop with appropriate gain and synchronized pure time delay. As long as the feed disturbance can continually be measured at a position in the reactor upstream, the disturbance may be allowed to be arbitrary. If the control and disturbances are more than one-dimensional, the technique is still effective, and instead of a control curve, an optimal control hypersurface would be obtained which would lead to a multidimensional feedforward control scheme.

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NOTATION

A = amplitude of disturbance
B = matrix, $n \times n$
b_i = $\left(\frac{-\Delta H_i}{\rho \cdot C_p}\right)$ $(i = 1, 2, 3, 4)$
C_p = heat capacity of reactant fluid
C_{p0} = heat capacity of jacket fluid
d = boundary disturbance
E_i = activation energies $(i = 1, 2, 3, 4)$
F = vector valued function, n dimensional
F_i = function
f = vector valued function, n dimensional
G = matrix $n \times n$
$\mathcal{G}$ = integrand of functional
$g(t)$ = vector valued function, n dimensional
H = Hamiltonian function
$h(t)$ = vector valued function, n dimensional
$(-\Delta H_i)$ = heat of reactions $(i = 1, 2, 3, 4)$
I = identity matrix
J = objective functional
K = kernel, n dimensional
k_i = rate constants,

$$k_{i0} \exp\left(-\frac{E_i}{Rv_4}\right) \quad (i = 1, 2, 3, 4)$$

k_{i0} = pre-exponential factors $(i = 1, 2, 3, 4)$
L = total length of tubular reactor
M = kernel, n dimensional
N = diagonal matrix, $n \times n$
P = nonsingular matrix, $n \times n$
p = adjoint vector, n dimensional
Q = vector valued function, n dimensional
$q_1, q_2, q_3 = \frac{E_1 - E_2}{R}, \frac{E_1 - E_3}{R}, \frac{E_3 - E_4}{R}$
R = gas constant
r_i = inside radius of tube
r_0 = inside radius of jacket
s = parameter along characteristic curve
s_i = characteristic curve for v_i
t = time
t_f = final time
U = overall heat transfer coefficient
u = control
v = state vector, n dimensional
v_1, v_2, v_3 = concentrations of A, B, C
v_4 = reactant temperature
v_5 = jacket temperature
v_{is} = steady state value of Ψ_i $(i = 1, 2, 3, 4)$
$w_1, w_2, w_3 = \frac{k_{20}}{k_{10}}, \frac{k_{30}}{k_{10}}, \frac{k_{40}}{k_{30}}$
x = distance variable

Greek Letters

$\alpha = \frac{r_i \cdot C_p \cdot \rho}{2 \cdot U}$
$\alpha_0 = \frac{(r_0^2 - r_i^2) \cdot C_{p0} \cdot \rho_0}{2 \cdot r_i \cdot U}$
Γ = nonlinear gain vector
Δ = integration step size
δ_{i3} = Kronecker delta
$\epsilon(t)$ = iteration step size
η = distance variable
Λ = vector valued function, n dimensional
μ = vector, $\partial v/\partial x$
ν = velocity in tube
ν_i = component of N
ν_0 = velocity in jacket
ρ = density of reactant fluid
ρ_0 = density of jacket fluid
τ = period of disturbance, $2\pi/\omega$
Φ = initial distribution vector, n dimensional
Ψ = boundary distribution vector, n dimensional
ω = angular velocity of disturbance

Superscript

(i) = i^{th} iteration

Subscript

i = i^{th} component

Special Symbols

∇_μ, ∇_v = gradient operators with respect to μ and v
$<, >$ = inner product of two vectors
— = optimal values

LITERATURE CITED

1. Bollinger, R. E., and D. E. Lamb, *Chem. Eng. Progr. Symposium Ser. No. 55,* **61,** 66 (1965).
2. Luyben, W. L., and D. E. Lamb, *Chem. Eng. Progr. Symposium Ser. No. 59,* **46,** 165 (1963).
3. Rippin, D. W. T., et al., *Chem. Eng. Progr. Symposium Ser. No. 63,* **57,** 132 (1961).
4. Harris, J. T., and R. S. Schechter, *Ind. Eng. Chem. Process Design Develop.,* **2,** 245 (1963).
5. Luyben, W. L., and J. A. Gerster, *ibid.,* **3,** 374 (1964).
6. Cadman, T. W., R. R. Rothfus, and R. I. Kermode, "Systems Process Control," Am. Inst. Chem. Engrs., New York (1967).
7. Haskins, D. E., and C. M. Sliepcevich, *Ind. Eng. Chem. Fundamentals,* **4,** 241 (1965).
8. Lupfer, D. E., and J. R. Parsons, *Chem. Eng. Progr.,* **58,** 37 (1962).
9. MacMullen, E. C., and F. G. Shinskey, *Control Eng.,* **11,** No. 3, 69 (1964).
10. Luyben, W. L., *Chem. Eng. Progr.,* **61,** 74 (1965).
11. Murrill, P. W., and P. F. Woolverton, *Control Instr. Technol.,* **14,** 35 (1967).
12. Distefano, G. P., F. P. May, and C. E. Huckaba, *AIChE J.,* **13,** 125 (1967).
13. King, M. K., R. R. Rothfus, and R. I. Kermode, *Can. J. Chem. Eng.,* **45,** 114 (1967).
14. Luecke, R. H., and M. L. McGuire, *AIChE J.,* **14,** 181 (1968).
15. Tinkler, J. D., and D. E. Lamb, *Chem. Eng. Progr. Symposium Ser. No. 55,* **61,** 155 (1965).
16. Koppel, L. B., *Ind. Eng. Chem. Fundamentals,* **5,** 403 (1966).
17. Luyben, W. L., *AIChE J.,* **14,** 37 (1968).
18. Denbigh, K. G., *Chem. Eng. Sci.,* **8,** 125 (1958).
19. Aris, Rutherford, *Chem. Eng. Sci.,* **12,** 56 (1960).
20. Horn, F., and U. Troltenier, *Chemie-Ing. Tech.,* **32,** 382 (1960).
21. Storey, C., *Chem. Eng. Sci.,* **17,** 45 (1962).
22. Chang, K. S., and S. G. Bankoff, *AIChE J.,* **15,** 410 (1969).
23. Koppel, L. B., Y. P. Shih, and D. R. Coughanowr, *Ind. Eng. Chem. Fundamentals,* **7,** 286 (1968).
24. Denn, M. M., *ibid.,* 410.
25. Koppel, L. B., and Y. P. Shih, *ibid.,* 414.
26. Luyben, W. L., *ibid.,* 502.

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