On the Optimization by Boundary Control of Tubular Reactors with Catalyst Decay

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In optimization problems dealing with catalytic reactors, where the catalyst deactivates slowly with time, it is common to assume a quasi steady state in the reaction and deactivation equations. Although the difference in reactor performance, for a given control policy, between the unsteady state reactor and its quasi steady state approximation is usually negligible, a very large difference can exist in the optimal control policy for the two models. For a single irreversible reaction, where the catalyst deactivation is dependent on conversion and where the control variable is the temperature of the fluid entering a tubular reactor, it is shown numerically that the performance of the reactor for a suboptimal control policy can be significantly superior to the optimal performance predicted from a quasi steady state model formulation.

SCOPE

In tubular reactors where the catalyst activity decays slowly with time, the contact time of a fluid element in the reactor is often small (seconds or minutes) compared to the total operating time (days or months), and unless the temperature changes drastically in a short time, the rate of change of conversion with time at any given point in the reactor is in general negligible when compared to the rate of change of conversion with contact time in an element passing through the reactor. For a given control policy then, the performance of the reactor can be predicted quite accurately from this so-called quasi steady state approximation. This simplification in the model equation often results in faster algorithms for calculating optimal control policies and in many cases also allows us to derive theoretically important properties of the optimal control policies for the quasi steady state problem.

The question is addressed whether there are policies which predict a performance of an unsteady state reactor which is far superior to that predicted from the optimal policy for the quasi steady state model.

CONCLUSIONS AND SIGNIFICANCE

By modeling a tubular reactor with decaying catalyst by a quasi steady state approximation, the computations of the performance are greatly simplified for a given temperature or other control. It is shown, however, that the optimal inlet temperature control for the quasi steady state model may be far from optimal for the unsteady state model which it approximates. This is because the rate at which a temperature change travels through the reactor is in general different from the rate of travel of a concentration change. But the quasi steady state model assumes these rates to be the same.

This should make one cautious about using the quasi steady state model for optimal control studies. It also could lead one to discover new and potentially attractive structures of the control for improved performance of an unsteady state tubular reactor with decaying catalyst.

There has been much recent interest in the optimal control of catalytic reactors where the catalyst decays with time. Following Szepe (1966), the rate expression for the catalyst decay is often chosen as a product of separate functions, each of only one independent variable. For the most general case, these independent variables are temperature, concentration or degree of conversion, and catalyst activity. A wide variety of reaction systems such as reversible and irreversible reactions and isothermal and nonisothermal batch and continuous reactors have been studied, both as lumped and distributed parameter systems (Szepe and Levenspiel, 1968; Chou, Ray, and Aris, 1967; Crowe, 1970; Lee and Crowe, 1970; Crowe and Lee, 1971; Ogunye and Ray, 1971 *a*, *b*; Therien and Crowe, 1974; Crowe and Therien, 1974).

Since in most of these optimization studies the catalyst is considered to decay slowly with time, a quasi steady state model (QSSM) is commonly assumed to describe reactions which take place in fixed-bed reactors.

Variational or maximum principle techniques for lumped and distributed parameter systems are then employed either to calculate optimal control policies or to derive

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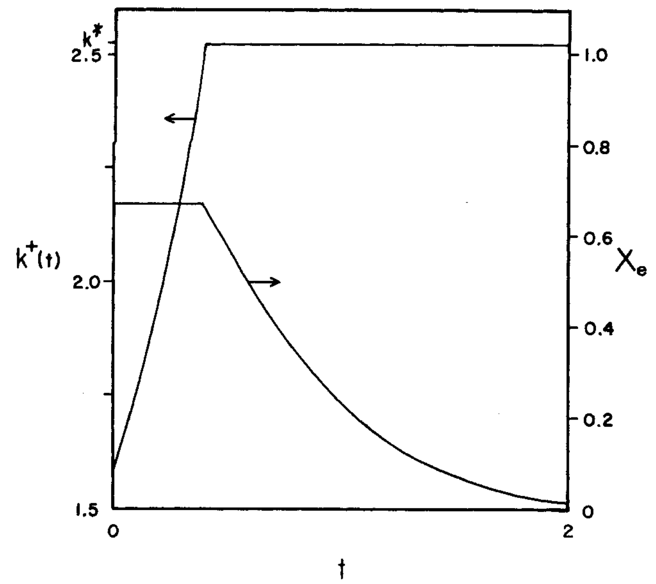

Fig. 1. Optimal control policy $k^+(t)$ and conversion $x_e(t)$ vs. time for QSSM.

properties of the optimal trajectories. Because of the complexity of such systems, the existence of an optimal solution which belongs to a specific class of functions (for example, piecewise continuous controls) is often assumed.

It will now be shown that the optimal solution, obtained from a QSSM, can be markedly inferior to a suboptimal control policy for the unsteady state operation, which the QSSM purports to approximate.

STATEMENT OF THE PROBLEM

We consider a first-order irreversible reaction, carried out in a tubular fixed-bed reactor. The unsteady state equation for reaction can then be written as

$$\frac{\partial x}{\partial t} + \nu \frac{\partial x}{\partial z} = K(T)[1 - x]\psi \tag{1}$$

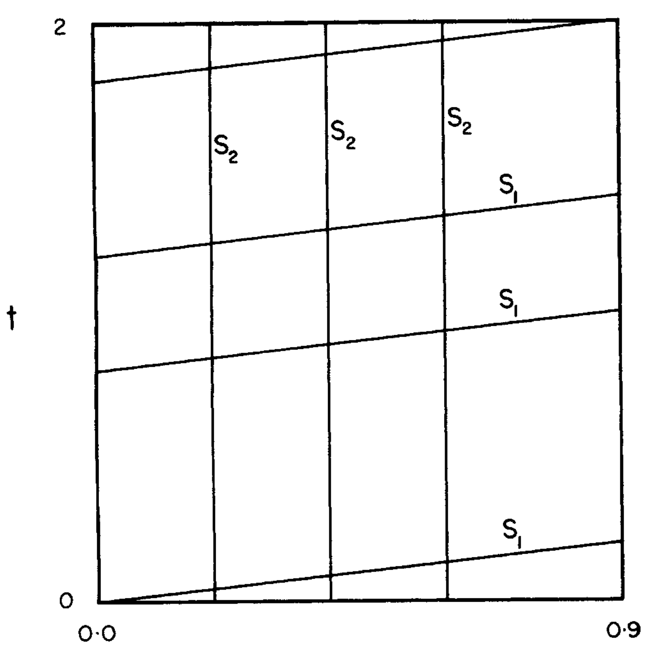

Fig. 2. Characteristic lines S_1 and S_2 for unsteady state model.

TABLE 1. VALUE OF THE OBJECTIVE FUNCTION J FOR VARIOUS POLICIES, WITH $z_f = 0.9$, $t_f = 2.0$

Model \ Policy	k^*	Optimal QSSM policy	Best bang-bang policy found.
QSSM	0.5774	0.5817	—
Unsteady state	$J^* = 0.7732$	$J_1 = 0.7762$	0.8380

where $x(z, t)$ and $\psi(z, t)$ are the distributed state variables representing the conversion and the relative catalyst activity, respectively. The independent variables z and t represent dimensionless distance along the axis of the reactor $z \epsilon [0, z_f]$ and dimensionless time $t \epsilon [0, t_f]$. The velocity factor ν is constant, and the temperature T at a given time t inside the reactor is assumed to be uniform in z. Although the temperature is not in fact uniform in actual reactors, this assumption was made to avoid unnecessary complication and to focus attention on the central aim of the paper.

The rate expression for catalyst decay depends upon the conversion and is given by

$$\frac{\partial \psi}{\partial t} = -k(T)[1 - x]^r \psi \tag{2}$$

where r is a constant parameter.

With $K(T)$ and $k(T)$ both of Arrhenius form, we have

$$K(T) = A[k(T)]^p \nu \tag{3}$$

where A is a positive constant and p is the ratio of the activation energy for reaction to that for decay. Since $K(T)$ and $k(T)$ are both strictly monotonic increasing functions of T, the boundary control variable, which is the inlet temperature into the reactor $T(t)$, can be replaced by $k(t)$.

Initial and boundary conditions for (1) and (2) are

$$\begin{aligned} x(0, t) &= x_i(t) \\ x(z, 0) &= x_o(z) \\ \psi(z, 0) &= \psi_o(z) \end{aligned} \tag{4}$$

The objective function is defined by

$$\max_{k_* \leq k(t) \leq k^*} J \quad \text{with} \quad J \equiv \int_0^{t_f} [x_e(t) - x_i(t)]dt \tag{5}$$

where k_* and k^* are lower and upper constraints on the boundary control, and $x_e(t)$ is the exit conversion from the reactor. Furthermore, the optimal control $k(t)$ is sought in the class of piecewise continuous functions.

The numerical values of the parameters used in the study of this particular problem follow and were based on a first-order reaction which has a maximum conversion of 90% at 900°K for a space time of 1 s. The decay rate constant k^* was chosen to give a loss of about 99.3% of the activity at 900°K and $x = 0$ for $t_f = 2$. The values are

$$\begin{aligned} &x_i(t) = 0; \quad x_o(z) = 1; \quad \psi_o(z) = 1; \\ &\nu = 4.5; \quad r = 0.2 \\ &A = 0.573364; \quad p = 1.5; \quad z_f = 0.9 \\ &t_f = 2; \quad k_* = 0; \quad k^* = 2.5265 \end{aligned} \tag{6}$$

QUASI STEADY STATE APPROACH

In a reaction-deactivation system where the catalyst decays slowly, and barring abrupt changes in the inlet temperature policy $k(t)$, we often have

$$\frac{\partial x}{\partial t} << \nu \frac{\partial x}{\partial z} \tag{7}$$

and the first term in Equation (1) is neglected, giving the QSSM. In the QSSM the characteristic directions associated with the state equations coincide with the directions of the coordinate axes of the partial differential Equations (1) and (2). It has been shown earlier (Gruyaert and Crowe, 1974) that for this particular optimization problem, Equations (1) to (6), with the QSSM, there exists a unique optimal boundary control policy $k^+(t)$ which belongs to the class of piecewise continuous functions. This optimal control policy was shown to be one which maintains a constant exit conversion out of the reactor over any time interval where the control was unconstrained. The optimal control policy $k^+(t)$ and the corresponding exit conversion $x_e{}^+(t)$ are illustrated in Figure 1. The values of the objective function at the optimum $k^+(t)$ and for the totally constrained control $k(t) = k^*$ are shown in Table 1.

One of the most significant features of the QSSM is that the lines along which the boundary control variable remains constant coincide with the characteristic lines associated with the state equation for the reaction. As a result of this, any control policy which remains at the lower constraint $k_* \equiv 0$ over any finite time interval can never be optimal, since over that time interval the contribution to the objective function (5) would be zero.

UNSTEADY STATE FORMULATION

For the unsteady state model, Equations (1) and (2), the set of characteristic lines S_1 associated with the reaction Equation (1) consists of straight lines which for large values of ν form angles of the order of $1/\nu$ radians with respect to the z axis (Figure 2). The set of characteristics S_2 associated with the decay Equation (2) contains lines which are still parallel to the t axis. Any line in the set S_1 is defined by a variable coordinate of position s_1 and by a constant value of the other position coordinate s_2. Any line in S_2 is analogously defined.

The set of ordinary differential equations, (1) and (2), which describes the system along the characteristic lines is then

$$\nu \frac{dx}{ds_1} = K(k)[1 - x]\psi \tag{8}$$

$$\frac{d\psi}{ds_2} = - k[1 - x]^r \psi \tag{9}$$

and is identical in form to that for the QSSM. The major difference, however, is that the boundary control $k(t)$ does not remain constant along the characteristic lines in S_1. As a result of this, the unsteady state problem cannot be transformed into a lumped parameter problem and the existence of an optimal boundary control in the class of piecewise continuous functions cannot be established by lumping the problem.

Furthermore, a control policy which remains at the lower constraint $k_* = 0$ for a finite time interval cannot be excluded from being optimal, since if this finite time interval is smaller than the space time through the reactor, the reaction is switched off in any affected fluid element for only part of its passage through the reactor.

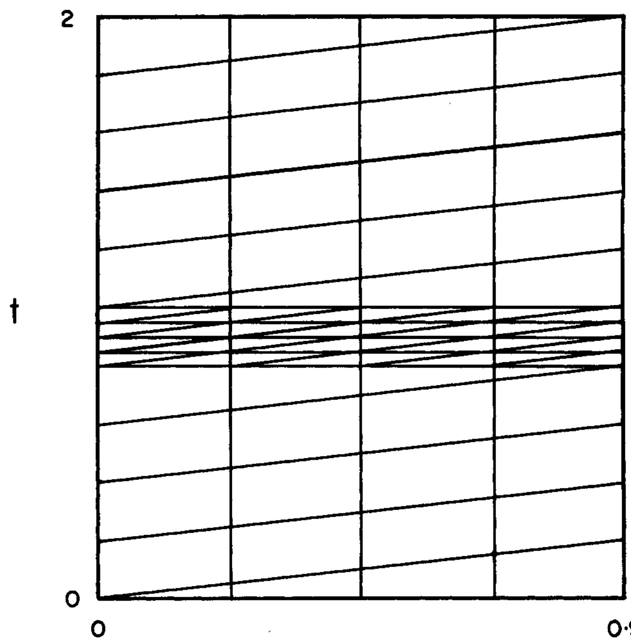

Fig. 3. Grid structure for numerical solution of unsteady state model.

Such an element then contributes a positive quantity to the objective function. As a result, relaxed or chattering controls of the problem with nonorthogonal characteristics also become feasible controllers. The discussion in Gruyaert and Crowe (1974) was incorrect insofar as it did not take this into account.

With the inclusion of relaxed controllers, however, two major difficulties arise: the proof of the existence of an optimal relaxed controller, and the impossibility of implementing a relaxed control policy in practical applications. Nevertheless, since a relaxed control function can be seen as the limit of very fast switching between two or more admissible control policies (Warga, 1962; McShane, 1967*a*, *b*; Lee and Markus, 1967), it is interesting to study the behavior of a system for a sequence of control policies which converges toward a relaxed controller.

Consider now a particular type of bang-bang control policy which is constructed as follows:

1. Let

$$k(t) = \begin{cases} k^* & \text{for } t \epsilon [t_1, t_2]; \quad t_2 > t_1 \\ k_* & \text{for } t \epsilon (t_2, t_3); \quad t_3 > t_2 \end{cases}$$

2. Let $(t_3 - t_2) = (t_2 - t_1)$.
3. Starting at $t_1 = 0$, repeat (1) over N consecutive time intervals (t_1, t_3):

$$[N < t_f/(t_3 - t_1)]$$

4. Let $k(t) = k^*$ for $t \epsilon [N(t_3 - t_1), t_f]$.

A sequence of such bang-bang control policies, defined on $[0, t_f]$ for decreasing values of $(t_2 - t_1)$ and increasing values of N, then converges to a particular relaxed controller in the limit as $(t_2 - t_1) \rightarrow 0$ and $N \rightarrow \infty$.

NUMERICAL RESULTS AND DISCUSSION

For the given unsteady state problem, Equations (1) to (6), with parameter set (7), calculations have been done for a sequence of bang-bang control policies of this type. Since the state equations are integrated along their respective characteristics, a grid has been constructed in the domain enclosed by the outermost characteristics. Since the control is a boundary control, the

TABLE 2. VALUE OF THE OBJECTIVE FUNCTION FOR A SEQUENCE OF BANG-BANG CONTROL POLICIES

$t_2 - t_1$	N	J
0.2	4	0.7773
	3	0.8012
	2	0.8101
	1	0.8031
0.15	5	0.8070
	3	0.8272
	2	0.8227
	1	0.8059
0.1	6	0.8342
	4	0.8365
	2	0.8176
0.075	10	0.8178
	7	0.8337
	4	0.8273
0.05	15	0.8226
	10	0.8378
	5	0.8235
	3	0.8066
0.025	30	0.8233
	25	0.8339
	20	0.83790
	15	0.8347
	10	0.8228
0.02	30	0.8353
	25	0.83796
0.01	70	0.8287
	60	0.8354
	50	0.83797
	49	0.83798
	48	0.83795
	40	0.8359

grid is such that its nodes lie on lines which are parallel to the z axis. This grid structure is illustrated in Figure 3 for a grid size of 4×40. In the numerical work, grid sizes of 8×80, 20×200, and 80×800 were used.

Since a bang-bang controller where $k(t) = k_* = 0$ over a time interval larger than the space time through the reactor cannot be optimal, only cases with $(t_2 - t_1) \leq 0.2$ have been considered. (The relatively high value of the space time through the reactor was chosen solely to maintain a high accuracy in the numerical results while avoiding extremely fine grids.) Numerical integration was done by a fourth-order Runge-Kutta method.

The numerical results obtained for a sequence of bang-bang control policies are listed in Table 2. The values of the objective function J^* for the totally constrained policy $k(t) = k^*$ and J_1 for the control $k^+(t)$, which had been found to be optimal for the quasi steady state problem with $z_f = 0.9$, are given in Table 1.

Since $J_1 > J^*$, the control $k(t) = k^*$ for all $t \epsilon [0, t_f]$ is not optimal. Although the relative difference between the values of J_1 and J^* is small, this conclusion was also obtained from the application of a maximum principle technique. It was found numerically that the control $k(t) = k^*$ did not satisfy the necessary conditions of a weak maximum principle for boundary control of distributed parameter systems of this type (see Gruyaert and Crowe, 1974). It should be mentioned that all attempts to find an optimal piecewise continuous control policy failed. By using a gradient method to hill climb on the boundary Hamiltonian, and by using different starting policies, the numerical search technique never converged to a stationary solution.

From the results tabulated in Table 2, the following observations can now be made:

1. All of the values of J with a suboptimal policy used for the unsteady state reactor are greater than the value J_1, obtained from the optimal QSSM policy.
2. For a given time interval $(t_2 - t_1)$, the value of the objective function shows a maximum with respect to N.
3. As $(t_2 - t_1)$ decreases, the values of this maximum J_{max} and N_{max} increase.
4. The length of the time interval over which the control switches between k^* and k_* at J_{max} seems for any value of $(t_2 - t_1)$ to remain constant. This time interval is given by $2 \times N_{max} x (t_2 - t_1)$ and lies in the neighborhood of 1.
5. The sequence of values of J_{max} as a function of $(t_2 - t_1)$ converges asymptotically towards a value J_R which would correspond to $(t_2 - t_1) = 0$.
6. The values of J_{max}, even for large time intervals $(t_2 - t_1)$, are considerably larger than J^* or J_1.

Note, however, that the relaxed controller which is the limit of the sequence of bang-bang controllers used in our calculations is not necessarily the best one for this problem. By constructing a sequence of bang-bang controllers of a different type, it is quite possible that the corresponding values of J_{max} would converge to a different value of J_R in the limit, since the sequence would converge to a different relaxed controller. These results do suggest, however, that for the optimization problem in the unsteady state formulation, there exists an optimal relaxed controller. Moreover, an optimal solution in the class of piecewise continuous control functions does not appear to exist.

The unsteady state problem is a singular perturbation of the QSSM in that the limit of a sequence of optimal controls for the unsteady state problem as $\epsilon \equiv 1/\nu \rightarrow 0$ does not coincide with the optimal control for the QSSM, where $\epsilon = 0$. The singularity lies in the fact that characteristics for reaction coincide with lines of uniform control k when $\epsilon = o$ but do not coincide for $\epsilon > 0$. However, for a fixed piecewise continuous control $k(t)$, one would reasonably expect that the limit of solutions of the unsteady state problem as $\epsilon \rightarrow 0$ coincides with the solution of the QSSM, except that $x(z, 0) = 1$ is not satisfied.

The limit of a sequence of relaxed controls as $\epsilon \rightarrow 0$, while not a feasible control for the QSSM, has been called a pseudo relaxed control (PRC) by Gruyaert (1976). This control can be represented in the QSSM by

$$k(t) = \alpha(t) k^*; \quad K(t) = \alpha(t) K^* \qquad (10)$$

and $0 \leq \alpha(t) \leq 1$. The optimal PRC is readily computed, as shown by Gruyaert (1976), to give a value of $J_{PRC} = 0.6596$, which is sharply higher than the optimal policy for the QSSM as shown in Table 1.

This again indicates that for values of ϵ close to zero there are optimal relaxed controls which are markedly superior to the optimal policy for the QSSM.

PRACTICAL CONSIDERATIONS

Since in a tubular fixed-bed catalytic reactor the rate at which a step change in inlet temperature is carried through the reactor is normally much smaller than the flow rate of the fluid stream, rapid switching of the inlet temperature should again offer an advantage over the optimal QSSM policy. Although in this case the temperature characteristics would be steeper than those for concentration, such rapid switching would cause the fluid to pass through successive hot and cold zones, as in the uniform temperature case. For practical control problems, where the frequency of switching between two control policies has a physical upper limit, one also would have to include in the state equations the inertia of the system (that is, heat transfer between solid and fluid stream).

CONCLUSION

Whereas for a given control policy the values of the objective function corresponding to a QSSM and an unsteady state model will usually differ only slightly, the structures of their respective optimal policies and hence the values of the objective function corresponding to the optimal policies can be very different. This situation can occur whenever the feasible controllers for the quasi steady state problem and for the unsteady state problem do not belong to the same class of functions. It is, therefore, possible in practice that a suboptimal control policy leads to a better performance of an unsteady state reactor than the optimum predicted from a quasi steady state optimization.

REMARKS

It can easily be seen that the particular control problem discussed above is closely related to the problem with nonorthogonal characteristics treated earlier by Gruyaert and Crowe (1974). For that latter boundary control problem with nonorthogonal characteristics, similar results for a sequence of bang-bang control policies have been found, which would again suggest that there does not exist an optimal controller in the class of piecewise continuous functions. Thus, Gruyaert and Crowe (1974) provided a counterexample to the strong maximum principle only for boundary control of those hyperbolic distributed parameter systems in which the control characteristic lines coincide with the characteristic lines of one of the state variables.

ACKNOWLEDGMENT

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NOTATION

A = constant, Equation (3)
J = objective function, Equation (5)
k = inlet temperature dependence in decay rate, also decision variable
K = inlet temperature dependence in reaction rate
N = integer value
P = ratio of activation energies for reaction over decay
r = parameter, Equation (2)
S_1 = set of characteristics for x: $S_1(t_o) = \{s_1 = z;\ s_2 = t_o\}$
S_2 = set of characteristics for ψ: $S_2(z_o) = \{s_1 = z_o;\ s_2 = t - z_o/\nu\}$
s_i = variable coordinate of position along a line in S_i $(i = 1, 2)$
t = time
t_o = starting time of a line in set S_1
T = inlet temperature
x = conversion
z = distance
z_o = starting position of a line in set S_2
α = pseudo relaxed control, Equation (10)
ϵ = $1/\nu$
ν = fluid velocity
ψ = catalyst activity

Superscripts

$*$ = value corresponding to upper constraint of the control
$+$ = value for the optimal policy

Subscripts

e = value of the conversion at $z = z_f$
f = final value for z or t
i = value at $z = 0$
o = inlet condition at $t = 0$
R = value corresponding to a relaxed controller
$*$ = value corresponding to lower constraint of the control

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