Optimal Control Policies for Tubular Reactors Experiencing Catalyst Decay

Part I. Single Bed Reactors

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The optimal control problem is formulated, and a weak maximum principle for such distributed parameter systems is derived. From this principle an efficient computational algorithm is presented which allows the simultaneous solution of the problems of both catalyst distribution (along the reactor) and optimal control. Detailed numerical examples are worked for isothermal reactors, adiabatic reactors, and adiabatic reactors with both a catalyst distribution problem and an optimal control problem. For irreversible reactions constant conversion policies are found to be optimal a significant portion of the time.

The phenomenon of both reversible and irreversible catalyst decay is very common in catalytic chemical reactors. The decay may be due to poisons (when impurities are contained in the reactant feed), to sintering (when the catalyst surface area is reduced by physical deterioration due to high operating temperatures), or to fouling (when coke or high polymeric material is deposited on the catalyst). Mathematical models which represent the time and spatial history of this decay have been proposed by Voorhies (*1*), Froment and Bischoff (*2*), Jackson (*3*), Szepe and Levenspiel (*4*), Osawa and Bischoff (*5*), Butt and Rohan (*6*), and others.

Among the numerous commercial processes in which catalyst decay is an important factor are: the cracking, reforming, and desulfurization of petroleum to produce gasoline (*7*); the dehydrogenation reactors for production of styrene and butadiene (*8*); the hydrogenation reactor for production of ethylene, ethane, and cyclohexylamine (*9*); the isomerization reaction for production of isopentane; and the production of vinyl chloride monomer from acetylene. For this reason the optimal control of chemical reactors experiencing catalyst decay is of some commercial interest.

Particular aspects of this optimization problem have been previously treated. Szepe and Levenspiel (*4, 10*) as well as Chou *et al.* (*11*) treated the isothermal tubular reactor problem where the catalyst decay is independent of the extent of the main reaction through the use of the Bolza form of the calculus of variations. Ogunye and Ray (*12*) later treated this special class of problems using the maximum principle. In a more general treatment, Volin and Ostrovskii (*13, 14, 15*) and Jackson (*3, 16, 17*) derived distributed maximum principles and proposed computational algorithms for the optimal control synthesis when the control is a distributed parameter. For example, the temperature at each spatial point in the reactor as a function of time $T(z, t)$ and the yield of a single component are to be maximized. The only computational experience reported is by Jackson (*16*) who used a steepest ascent control vector iteration method and found convergence problems frequently encountered with such first-order methods.

In this paper, a very general (weak) maximum principle is derived to treat the above results as special cases and to allow the solution of a much wider, more realistic set of problems. Some other problems solvable by this formulation are the selection of the optimal catalyst distribution along the reactor and optimal length of the catalyst bed, the selection of the optimal feed composition throughout the on-stream time of the reactor, and the inclusion in the profit function of the raw material costs, catalyst costs, and heating or cooling costs as well as product values so that a more realistic objective can be maximized.

We shall first discuss the reactor mathematical model used throughout and then treat the industrial optimization problem. Next, the most general optimal control problem will be considered and the special cases of isothermal or adiabatic operation noted. A computational procedure for applying the maximum principle will be described, and a number of examples will be worked to demonstrate the power of the optimization procedure and to indicate the character of the optimal policy for some commonly encountered problems.

CHEMICAL REACTOR MODEL

For our mathematical model of the reactor, we shall make three almost classical assumptions: that the reactor is in plug flow with negligible axial and infinite radial dispersion, that the reactor tube has enough catalyst particles per unit volume for an adequate continuum representation, and that the reactor time scale is much shorter than the time scale for catalyst decay so that the quasi-steady state approximation is valid.* Under these assumptions the extent of reaction ξ_i for each of the $\hat{r}$ reactions taking place is

$$\frac{\partial \xi_i}{\partial z'} = f_i'(\xi_j, \Psi_k, T, P) \qquad \xi_i(0, t') = \xi_{i0}(t') \qquad (1)$$

$$0 \leq z' \leq L, \ 0 \leq t' \leq \theta \qquad i = 1, 2, \ldots \hat{r}$$

If the catalyst bed is fixed, the rate of change of the k^{th} catalyst activity is

$$\frac{\partial \Psi_k}{\partial t} = g_k'(\xi_i, \Psi_j, T, P) \qquad \Psi_k(z', 0) = \Psi_{k0}(z') \quad (2)$$

$$0 \leq z' \leq L, \ 0 \leq t' \leq \theta \qquad k = 1, 2, \ldots \hat{q}$$

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* This assumption has been shown (*18*) to be adequate except for very long tubes filled with very rapidly decaying catalyst.

Because a catalyst may have more than one type of catalytic agent on it, and thus more than one catalytic function, we allow a separate activity for each catalytic function.

In addition, if the reactor temperature is a result of heat transfer through the wall, we have another equation

$$\frac{\partial T}{\partial z'} = f'_{\hat{r}+1}(\xi_j, \Psi_k, T, P, \mathbf{u}) \qquad T(0, t') = T_0(t') \quad (3)$$

$$0 \leq z' \leq L, \ 0 \leq t' \leq \theta$$

where u is a control action, such as coolant flowrate, output of heating coils, etc. In a similar way, an equation for pressure

$$\frac{\partial P}{\partial z'} = f'_{\hat{r}+2}(\xi_j, \Psi_k, T, P, \mathbf{u}) \qquad P(0, t') = P_0(t') \quad (4)$$

$$0 \leq z' \leq \mathrm{L}, \ 0 \leq t' \leq \theta$$

or other state variable can also be related to controls.

It is therefore both convenient and easy to generalize our model to the equations

$$\frac{\partial x_i}{\partial z} = f_i(\mathbf{x}, \mathbf{y}, \mathbf{u}) \qquad x_i(0, t) = v_i(t) \quad (5)$$

$$0 \leq z \leq 1, \ 0 \leq t \leq 1 \qquad i = 1, 2, \ldots r$$

and

$$\frac{\partial y_j}{\partial t} = g_j(\mathbf{x}, \mathbf{y}, \mathbf{u}) \qquad y_j(z, 0) = w_j(z) \quad (6)$$

$$0 \leq z \leq 1, \ 0 \leq t \leq 1 \qquad j = 1, 2, \ldots q$$

where Equation (5) is the most general form of Equations (1), (3), and (4), and Equation (6) is the most general form of Equation (2). The independent variables have been made dimensionless so that they lie between zero and unity.

OPTIMIZATION PROBLEM

In our reactor we would like to maximize the profits* produced over the lifetime of the catalyst while taking into consideration the lack of production while the reactor is down for catalyst regeneration or replacement.† Roberts (19) has posed this problem as maximizing A, the average profit over an operation and regeneration cycle, such as dollars/calendar day, where

$$A(\theta, \mathbf{x}, \mathbf{y}, \mathbf{u}, \mathbf{v}, \mathbf{w}) = \frac{\int_0^\theta \int_0^L G(\mathbf{x}, \mathbf{y}, \mathbf{u}, \mathbf{v}, \mathbf{w}, t', z)\, dt'dz', - C_R}{\theta + \theta_R} \quad (7)$$

where the symbols are defined in the Notation.

Thus, our problem is to pick the control variables v, w, u over

$$0 \leq t' \leq \theta,$$

$$0 \leq z' \leq L$$

and the length of the operating life θ so that A is a maximum.

If the operating life is fixed in advance by other considerations and cannot be left free, then θ becomes a constant in Equation (7) and the problem reduces to the maximization of

$$I = \int_0^1 \int_0^1 G(\mathbf{x}, \mathbf{y}, \mathbf{u}, \mathbf{v}, \mathbf{w})\, dzdt \quad (8)$$

subject to the satisfaction of Equations (5) and (6).

Thus, a procedure for finding the optimal θ, and thus the maximum of A, can be determined:

1. Fix $\theta = \theta_1$ and maximize I in Equation (8) subject to Equations (5) and (6).
2. Compute A from Equation (7) and use a univariate search technique (20) to correct θ, $\theta_{i+1} = \theta_i + \delta\theta_i$.
3. Keep iterating until a suitably accurate approximation to the optimal θ is found.

This technique should require only a few iterations to give the desired result.

WEAK MAXIMUM PRINCIPLE

We have seen that the optimization problem in Equation (7) can always be reduced to an optimal control problem of the form

$$\max_{\mathbf{u}, \mathbf{v}, \mathbf{w}} \left\{ I = \int_0^1 \int_0^1 G(\mathbf{x}, \mathbf{y}, \mathbf{u}, \mathbf{v}, \mathbf{w})\, dtdz \right\} \quad (9)$$

subject to Equations (5) and (6) and

$$\mathbf{v}_* \leq \mathbf{v} \leq \mathbf{v}^*, \ \mathbf{u}_* \leq \mathbf{u} \leq \mathbf{u}^*, \ \mathbf{w}_* \leq \mathbf{w} \leq \mathbf{w}^* \quad (10)$$

We must now develop a maximum principle for such systems (See Appendix I).*

Theorem

If the controls u, v, w are to be optimal controls for the problem given by Equations (5), (6), (9), and (10), it is necessary [for all $u_k(z, t)$] that

$$\frac{\partial H}{\partial u_k} = \frac{\partial G}{\partial u_k} + \lambda_i \frac{\partial f_i}{\partial u_k} + \mu_j \frac{\partial g_j}{\partial u_k} \quad (11)$$

vanish almost everywhere on $0 \leq z \leq 1$, $0 \leq t \leq 1$ when u_k is unconstrained and that

$$H = G + \lambda_i f_i + \mu_j g_j \quad (12)$$

be a maximum with respect to u_k when u_k is constrained by Equation (10). For u_k a function of z alone, the conditions

$$\int_0^1 \frac{\partial H}{\partial u_k}\, dt = 0 \quad (13)$$

for unconstrained u_k and $\int_0^1 H\, dt$ taking a maximum with respect to u_k [for u_k constrained by (10)] must hold. Similarly for u_k a function of t alone, the conditions

$$\int_0^1 \frac{\partial H}{\partial u_k}\, dz = 0 \quad (14)$$

for unconstrained u_k and $\int_0^1 H\, dz$ taking a maximum with respect to u_k [for u_k constrained by (10)] must hold.

In addition, for $v_i(t)$ to be optimal, it is necessary that

$$\frac{\partial H_1}{\partial v_i} = \int_0^1 \frac{\partial G}{\partial v_i}\, dz + \lambda_i(0, t) = 0 \quad (15)$$

for unconstrained v_i, and that

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* Or at least some profitlike function.

† As the problem is mathematically the same whether we replace or regenerate, the term regeneration will be used.

* The convention that a repeated subscript denotes a sum over that index is used throughout.

$$H_1 = \int_0^1 G\,dz + \lambda_j(0,t)v_j(t) \qquad (16)$$

to be a maximum with respect to v_i for v_i constrained by Equation (10). For the case where v_i is independent of t, the condition

$$\int_0^1 \frac{\partial H_1}{\partial v_i}\,dt = 0 \qquad (17)$$

is required for v_i unconstrained and $\int_0^1 H_1\,dt$ must be a maximum with respect to v_i at a constraint.

Finally, for $w_j(z)$ to be optimal, it is necessary that

$$\frac{\partial H_2}{\partial w_j} = \int_0^1 \frac{\partial G}{\partial w_j}\,dt + \mu_j(z,0) = 0 \qquad (18)$$

for unconstrained w_j and that

$$H_2 = \int_0^1 G\,dt + \mu_i(z,0)w_i(z) \qquad (19)$$

to be a maximum with respect to w_j constrained by (10). For the case where w_j is independent of z, the condition

$$\int_0^1 \frac{\partial H_2}{\partial w_j}\,dz = 0 \qquad (20)$$

is required for w_j unconstrained and $\int_0^1 H_2\,dz$ must be a maximum with respect to w_j at a constraint.

The adjoint variables in Equations (11), (12), (15), (16), (18), and (19) are defined by

$$\frac{\partial \lambda_i}{\partial z} = -\left[\frac{\partial G}{\partial x_i} + \lambda_k\frac{\partial f_k}{\partial x_i} + \mu_j\frac{\partial g_j}{\partial x_i}\right] \quad \begin{matrix} 0 \le z \le 1 \\ 0 \le t \le 1 \end{matrix}$$

$$\lambda_i(1,t) = 0 \qquad i = 1, 2, \ldots r \qquad (21)$$

$$\frac{\partial \mu_j}{\partial t} = -\left[\frac{\partial G}{\partial y_j} + \lambda_i\frac{\partial f_i}{\partial y_j} + \mu_k\frac{\partial g_k}{\partial y_j}\right] \quad \begin{matrix} 0 \le z \le 1 \\ 0 \le t \le 1 \end{matrix}$$

$$\mu_j(z,1) = 0 \qquad j = 1, 2, \ldots q \qquad (22)$$

for the case where **x** and **y** are left unspecified at $z = 1$, $t = 1$, respectively. Although the case for x_i or y_j, specified at $z = 1$ or $t = 1$, is easily treated theoretically, the free end-point problem is the easiest to treat computationally, and thus we will assume free end points from here on.

COMPUTATIONAL ALGORITHM

The maximum principle of the last section provides the basis for a large number of algorithms for synthesizing the optimal control policy. We chose, however, to use a control vector iteration method based on a gradient technique. The algorithm (see Appendix II) is as follows:

1. Guess the controls **u**, **v**, **w**.
2. Solve the state Equations (5) and (6) forward in t and z. Store the results.
3. Solve the adjoint Equations (21) and (22) backwards in t and z.
4. Calculate a correction $\delta\mathbf{u}$, $\delta\mathbf{v}$, $\delta\mathbf{w}$ to the controls based on the gradients of the functions H, H_1, H_2 (see Appendix II).
5. Go back to Step 2 and iterate until convergence to the optimum is attained.

Steps 2 and 3 should be carried out by a numerical technique which is both accurate and efficient because these steps are repeated many times in the iteration. The Equations (5), (6), (21), and (22), are coupled, hyperbolic, partial differential equations which can be solved numerically either by finite difference methods or by the method of characteristics (*21*). The characteristic curves for these equations are parallel to the coordinate axes due to the quasi-steady state approximation, so that the mesh is rectangular no matter which numerical scheme is used. However, the method of characteristics (*21*) permits much greater accuracy with the same mesh size than do finite difference methods. For our computational procedure, therefore, we chose the method of characteristics with the fourth-order Runge-Kutta method being used between characteristic line mesh points.

Jackson (*3, 16*) has done computations with a similar gradient method, but experienced convergence problems as the optimum was approached. Although few convergence problems were encountered in our examples, the possibility that a second-order method might speed convergence where first-order methods fail will be discussed in a later paper.

SOME EXAMPLES

To illustrate the power of this very general computational algorithm and to exhibit the optimal control policy for a few simple cases, a number of examples are worked in detail. In all the examples we will assume that there is but one catalyst activity and that it has the form suggested by Szepe (*10*) and others:

$$\frac{\partial y_1}{\partial t} = -\rho \exp\left\{-\frac{1}{p\tau}\right\}(y_1)^2 = g_1 \quad \begin{matrix} 0 \le t \le 1 \\ 0 \le z \le 1 \end{matrix} \qquad (23)$$

$$y_1(z,0) = w_1(z)$$

where $w_1(z)$, constrained by $0 \le w_1 \le 1$, is the initial catalyst activity in the bed, and τ is the dimensionless reactor temperature. Initially, we will assume $w(z) = 1$ and solve only the optimal control problem, but later we will solve the combined optimal control, catalyst distribution problem.

In the method of characteristics for these examples, the spatial direction $0 \le z \le 1$ was divided into 10 intervals. Because catalyst decay was very rapid for small t, 22 mesh points were normally used in the interval $0 \le t \le 0.11$ and 89 mesh points in the remaining interval $0.11 \le t \le 1$. For the few cases where the catalyst decay was very rapid, the number of mesh points had to be doubled in the t direction.

The gradient methods in Appendix II were used in the examples. In cases where there was any question of convergence, duplicate runs were made from an entirely different starting point. In all cases, convergence to the same policy was found.

Isothermal Reactor

For the isothermal reactor, the control variable is reactor temperature and it is independent of the spatial dimension. To demonstrate the algorithm we shall consider the reaction $A_1 \rightarrow A_2 \rightarrow A_3$ discussed by Bilous and Amundson (*22*) and others. The state equations for the extent of reaction are

$$\frac{\partial x_1}{\partial z} = y_1\beta_1 \exp\{-(u_1)^{-1}\}(1 - x_1) = f_1 \qquad (24)$$

$$x_1(0,t) = 0$$

$$\frac{\partial x_2}{\partial t} = y_1\beta_2 \exp\left\{-\left(\frac{u_1}{p_1}\right)^{-1}\right\}(\gamma_2 + x_1 - x_2) = f_2 \qquad (25)$$

where $x_i = \xi_i/c_{10}$ are dimensionless extents of reaction, and $u_1 = \tau$ is the dimensionless reactor temperature. If

we desire the policy which maximizes the production of A_2 subject to the maximum temperature constraint

$$u_1 \leq u_1^* \tag{26}$$

then our objective is to maximize

$$I = \int_0^1 \int_0^1 [f_1 - f_2]\, dzdt \tag{27}$$

Because u_1 depends only on t, our modified gradient method moves in the direction

$$\delta u_1 = \epsilon \int_0^1 \frac{\partial H}{\partial u_1}\, dz \tag{28}$$

at every ascent. The results of using our algorithm are shown in Figure 1. The optimal policy is a rising temperature policy followed by maximum temperature. The determination of this policy required only 4 ascents and 6 min. on an IBM 360/75. The optimal value of the objective function I was 0.5485, an improvement of 2.4% in I over the policy of constant maximum temperature.

Adiabatic Reactor

Many industrial reactors having catalyst decay problems are operated adiabatically. The optimal control policies for adiabatic reactors therefore are of great practical interest. For the adiabatic reactor we have the happy relationship between temperature and extents of reaction

$$\tau = \tau_0 + \sum_{i=1}^{\hat{r}} [J_i(\xi_i - \xi_{i0})] \tag{29}$$

so that, if the inlet temperature and extents of reaction are known, the reactor temperature is determined. For this reason, most adiabatic reactors are controlled by adjusting inlet temperature τ_0. This will be our control variable in the examples given. As the reactor is nonisothermal, the catalyst activity given by Equation (23) depends on position in the bed. Some typical profiles at a fixed time point are shown in Figure 2. As would be expected, for exothermic reactions decay is more rapid at the end of the bed (where temperature is the highest), while decay is most rapid at the front of an endothermic reactor.

Irreversible Reactions. Let us first consider the irreversible reaction $A_1 \rightarrow A_2$, where the state equations become

$$\frac{\partial x_1}{\partial z} = y_1\beta_1 \exp\{-(u_1 + J_1x_1)^{-1}\}(1 - x_1) = f_1 \tag{30}$$

$$x_1(z, 0) = 0$$

for $x_1 = \xi_1/c_{10}$, the dimensionless extent of reaction, and $u_1 = \tau_0$, the dimensionless inlet temperature. If we desire the policy which maximizes the production of A_2 subject to a maximum inlet temperature constraint, our objective is

$$I = \int_0^1 \int_0^1 f_1\, dzdt \tag{31}$$

Again, because u_1 depends only on t, our modified gradient method moves in the direction given by Equation (28) at each ascent.

A number of cases were run to determine the effect of some of the parameters on the optimal policy (*26*). The first-order accelerated gradient procedure was used. It worked well in every case, with the optimum being found in less than 5 ascents and the computing time being 3 to 5 min. on the IBM 360/75.

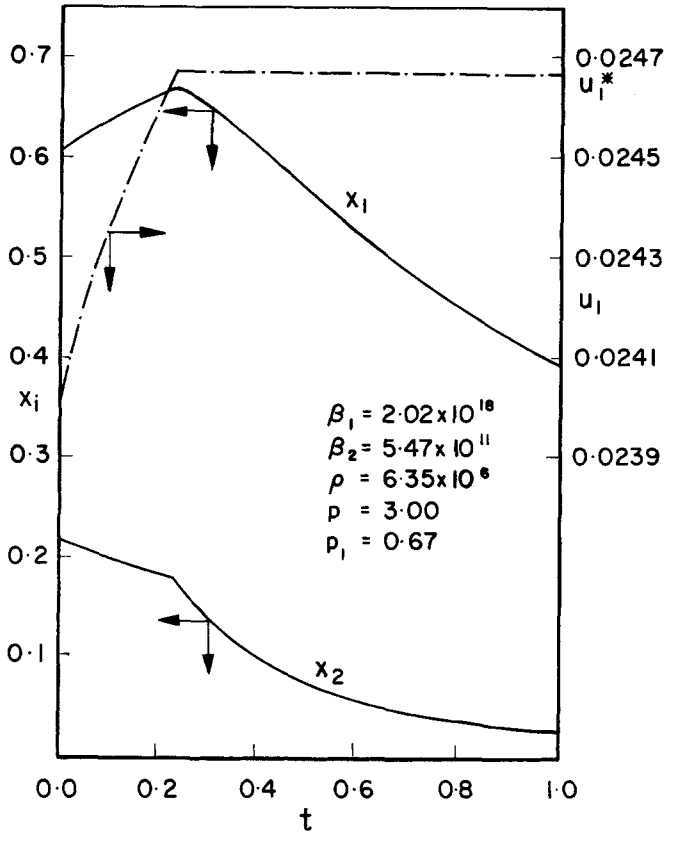

Fig. 1. The isothermal reactor for $A_1 \rightarrow A_2 \rightarrow A_3$.

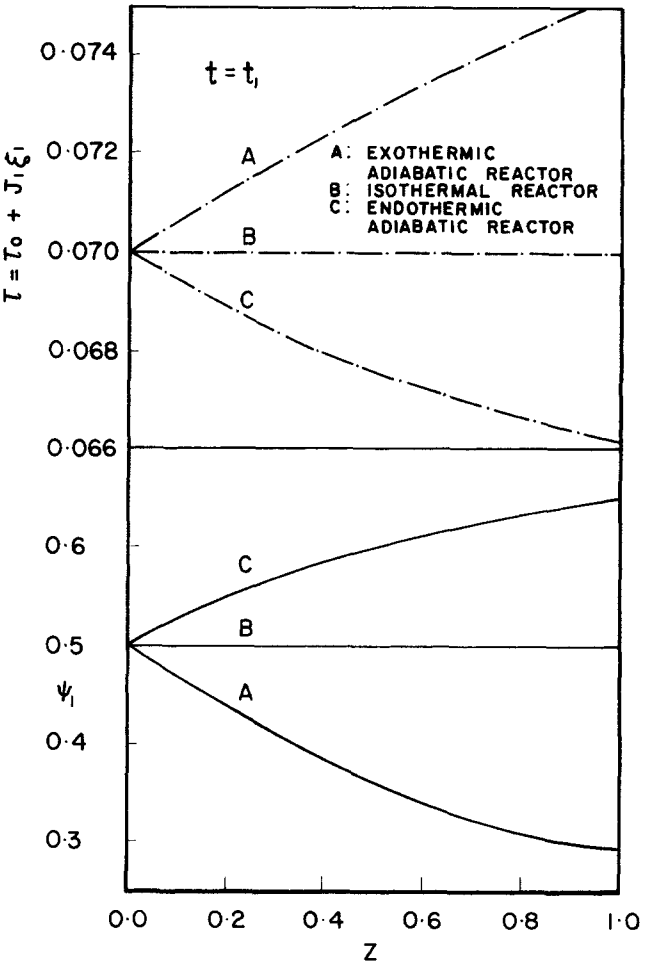

Fig. 2. Typical temperature, catalyst activity profile for irreversible reaction.

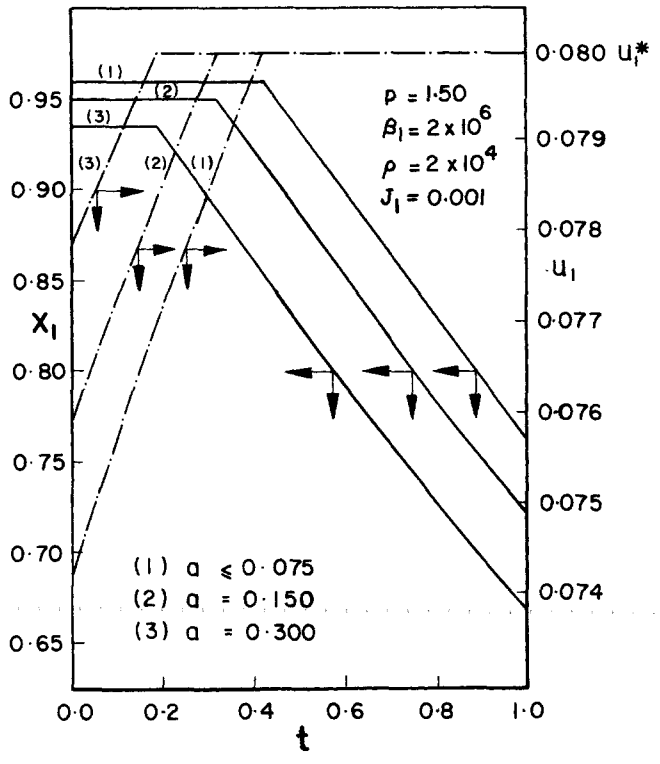

Fig. 3. Optimal control policies for catalyst distribution problem.

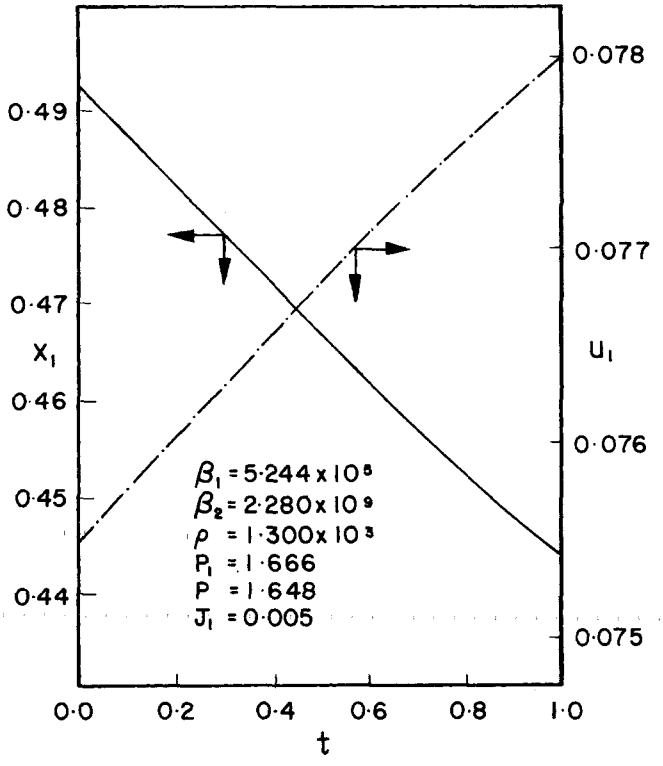

Fig. 4. Reversible exothermic reaction in an adiabatic reactor.

The optimal policy was composed of a constant conversion policy followed by maximum temperature. This was to be expected because Aris (*11*) showed that the extremal policies are constant conversion for any nonisothermal reactor with this type of kinetics. Only the level of conversion and the length of time at constant conversion must be determined for each case.

A typical case is shown in Figure 3, curve 1. The effects of the system parameters can be summarized [cf. (*26*) for more detail] as follows:

1. The effect of the heat of reaction J_1 for both the exothermic and endothermic case is to make the constant conversion portion longer, the larger J_1 becomes. This is what one might expect because as J_1 increases, the average temperature in the reactor increases and τ_0 can stay below τ_0^* a larger portion of the time.

2. Doubling the operating life ρ decreases both the constant conversion level and the fraction of time at constant conversion. This might be expected, for longer run life suggests gentler initial treatment of the catalyst, but more time at an upper temperature constraint.

3. Cutting the reactor length β_1 by half reduces the constant conversion level and the fraction of time at constant conversion.

4. Changing the temperature sensitivity of the catalyst decay rate relative to the main reaction p increases the constant conversion level and decreases the fraction of time at constant conversion as p increases. This is to be expected because at high p, the coking reaction is less sensitive to temperature and one can more profitably operate at higher temperatures.

5. Decreasing the maximum allowable inlet temperature τ_0^* for both exothermic and endothermic reactors decreases the constant conversion level as well as the fraction of time spent at constant conversion.

The trends noted in this parametric study are important because they not only agree with qualitative physical intuition but also may suggest operating policies for complex reaction systems that may have approximate models in the form of Equations (23) and (30). Even if an explicit model is unavailable, the trends may suggest "evolutionary operational" policy changes.

Reversible Reactions. For a first-order reversible reaction $A_1 \rightleftarrows A_2$, the state equation becomes

$$\frac{\partial x_1}{\partial z} = y_1 \{\beta_1 \exp\{-(u_1 + J_1 x_1)^{-1}\}(1 - x_1) - \beta_2 \exp\{-(p_1[u_1 + J_1 x_1])^{-1}\}(\gamma_2 + x_1)\} = f_1 \quad (32)$$

$$x_1(0, t) = 0$$

For the objective function (31) and maximum inlet temperature constraints, the optimal policy can be found as before.

The computing effort required to illustrate the optimal policy for reversible reactions was comparable to that of the irreversible case, but the conjugate gradient method of ascent (*25*) was found to be more efficient for the exothermic case where the optimal policy was unconstrained everywhere. For the exothermic case shown in Figure 4, the optimal policy was completely unconstrained with a slightly rising temperature profile, similar to the isothermal reactor results found earlier (*12*).

In Figure 5, the endothermic reaction cases are identical except that curve 2 has 10 times the operating life of curve 1. For the longer running time, the optimal policy is that of rising temperature and conversion followed by constant temperature. For the shorter running time (curve 1), the policy is simultaneous maximum temperature and constant conversion. This arises because the reactor is of sufficient length and the upper temperature constraint is so low that the equilibrium conversion is attained at all times. The decrease in catalyst activity is not enough to keep the reaction from being equilibrium limited.

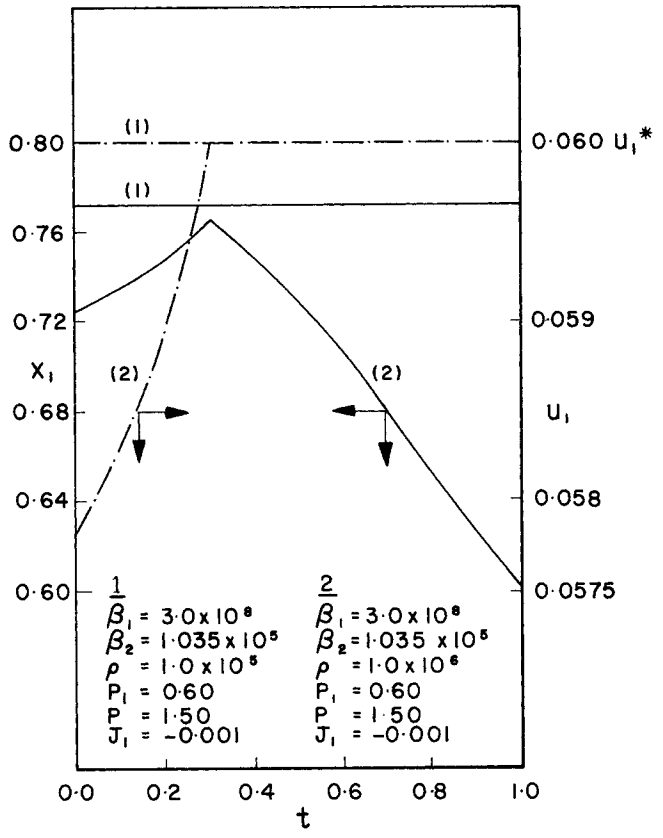

Fig. 5. Reversible endothermic reaction in an adiabatic reactor.

Consecutive Reactions. The case of the consecutive reactions $A_1 \rightarrow A_2 \rightarrow A_3$ was also treated for an adiabatic reactor. The state equations are the same as Equations (24) and (25) except that Equation (29) is used to calculate the reactor temperature. For the maximum inlet temperature constraint and objective function (27), the example shown in Figure 6 was worked. The conjugate gradient method was used as the optimal policy was found to be unconstrained everywhere. The unconstrained temperature policy was seen to be very nearly constant with time. The addition of a second state variable, which did not seem to affect seriously the convergence properties of the algorithm, increased the computing time per ascent by approximately 50%.

Catalyst Distribution Problem

To illustrate the computational technique applied to a catalyst distribution-optimal control problem, the optimal control for an irreversible adiabatic reactor was found simultaneously with the optimal catalyst distribution. This problem allows the choice of mixing inexpensive inert packing with the catalyst or decreasing the amount of catalytic agent on the catalyst to reduce the initial caalyst activity in the bed. For very expensive catalysts such as platinum, substantial savings could result. The state equations are Equations (23) and (30), but now the boundary condition on the catalyst decay Equation (23) is a control rather than at pure catalyst loading $w_1(z) = 1$, as before. If we specify that the average catalyst cost over the lifetime of operation is a, then our objective function becomes

$$I = \int_0^1 \int_0^1 [f_1 - a w_1(z)]\, dz dt \qquad (33)$$

The gradient $\partial H_2/\partial w_1$ used for determining the optimal catalyst loading is given by

$$\frac{\partial H_2}{\partial w_1} = \mu_1(z, 0) - a \qquad (34)$$

so that for $a \leq u_1(z, 0)$, pure catalyst $w_1(z) = 1$ is optimal.

For the optimal policy found previously (Figure 3, curve 1) when $w_1(z) = 1$, $\min_z \{\mu_1(z, 0)\} = 0.075$; thus it is obvious that for $a \leq 0.075$, pure catalyst is the optimal loading. Trials for $a = 0.15$ and $a = 0.30$ are shown in Figures 3 and 7. Increasing the cost of the catalyst causes a nearly uniform reduction in catalyst concentration along the bed, shortens the length of time spent at constant conversion, and reduces the constant conversion level. Because the optimal catalyst concentration is essentially spatially uniform, the optimal policy is very easily implemented.

The number of ascents needed to generate optimal policies for this combined problem (9 ascents and 8 min. of IBM 360/75 time) were somewhat more than the requirements for the simple optimal control problem. Faster convergence could probably be obtained through more efficient programming or the use of second-order ascent methods.

CONCLUSIONS

A very general method for calculating optimal control policies for catalytic reactors having catalyst decay has been developed. A number of examples have been done to demonstrate the great power of this method for catalyst decay problems.

Other practical cases that can easily be treated are optimal cooling jacket control of hot-spot reactors, optimal control of feed composition over the operating life of the reactor, and optimal reactor tube cross-sectional area for optimal pressure drop over the life of the reactor. Because many industrial reactors are in multiple beds with interbed feeds, a future paper will extend the present results to such systems.

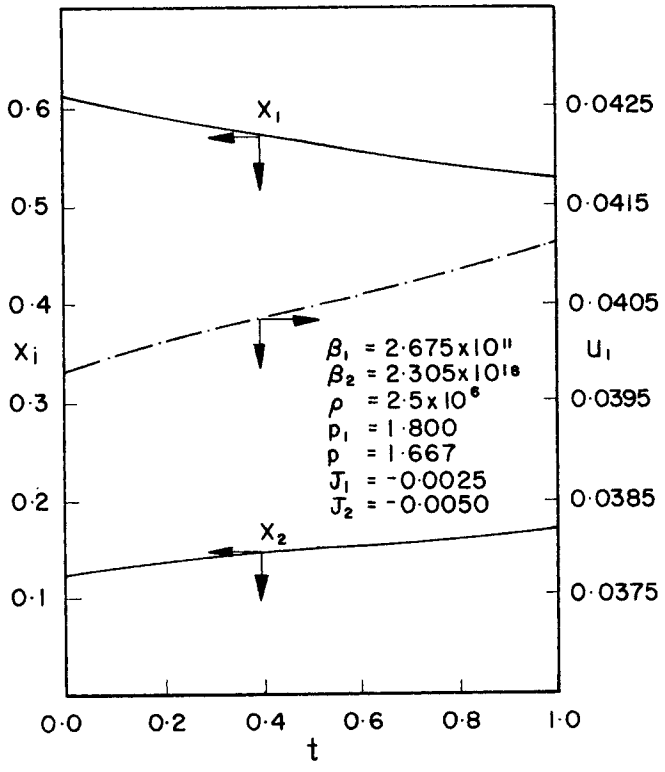

Fig. 6. Consecutive endothermic reactions in an adiabatic reactor.

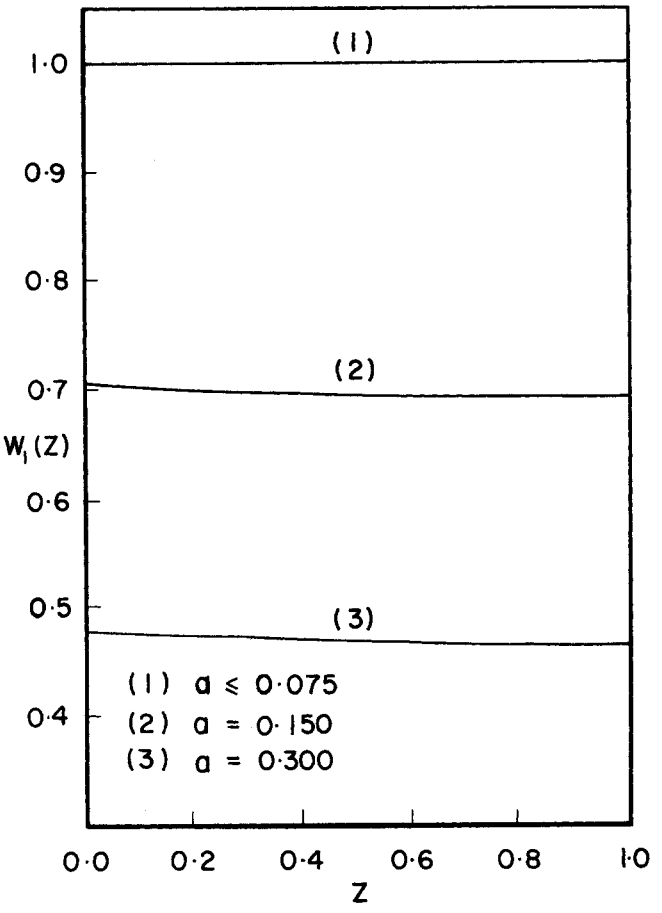

Fig. 7. Optimal catalyst distribution.

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NOTATION

a = average cost of catalyst over the lifetime of operation
A = average profit over an operation and regeneration cycle
A_0 = pre-exponential factor for fouling reaction rate constant
A_{i0} = pre-exponential factors of main reaction rate constants
c_i = concentration of the i^{th} chemical component
C_p = specific heat of reaction mixture
C_R = cost of regeneration
E = activation energy of fouling reaction rate constant
E_i = activation energies of main reaction rate constants
F = instantaneous profit not including regeneration costs
f_i' = rate of the i^{th} chemical reaction
G = integrand of objective function
g_k' = rate of the k^{th} fouling reaction
H, H_1, H_2 = Hamiltonians defined by Equation (I.11) in Appendix I
ΔH_i = heat of reaction of the i^{th} chemical reaction
I = objective function
$J_i = \left(\frac{-\Delta H_i}{C_p}\right)\left(\frac{R}{E_1}\right)$
L = total reactor residence time
P = pressure
$p = E_1/E$
$p_1 = E_2/E_1$
T = temperature
t' = time
$t = t'/\theta$
u = control variable
v(t) = inlet reactant concentration
w(z) = initial catalyst activity along the reactor length
x, y = state variables
z' = residence time in the reactor
$z = Z'/L$ (dimensionless distance along reactor length)

Greek Letters

$\beta_1 = A_{10} \cdot L$
$\beta_2 = A_{20} \cdot L$
$\gamma_2 = C_{20}/C_{10}$
ϵ = scalar constant
θ = total on-stream time
θ_R = regeneration time
λ_i, μ_i = adjoint variables defined by Equations (21) and (22)
ξ_i = extent of the i^{th} reaction
$\rho = A_0\theta$
$\tau = RT/E_1$
Ψ_k = instantaneous catalyst activity for the k^{th} catalytic agents

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APPENDIX I.

A Maximum Principle

The optimization problem to maximize Equation (9), subject to constraints in the form of Equations (5), (6) and (10), leads to the following maximum principle.

Let us consider the nominal controls $\bar{u}$, $\bar{v}$, $\bar{w}$ producing the nominal state trajectories $\bar{x}$, $\bar{y}$. Small variations about these trajectories δu, δv, δw, δx, δy can be approximated by linearization of Equations* (5), (6), and (9).

$$\frac{\partial(\delta x_i)}{\partial z} = \frac{\partial f_i}{\partial x_j}\,\partial x_j + \frac{\partial f_i}{\partial y_k}\,\delta y_k + \frac{\partial f_i}{\partial u_m}\,\delta u_m \qquad (Ia)$$

$$\delta x_i(0,t) = \delta v_i(t) \qquad i = 1, 2, \ldots r$$

$$\frac{\partial(\delta y_j)}{\partial t} = \frac{\partial g_j}{\partial x_i}\,\delta x_i + \frac{\partial g_j}{\partial y_k}\,\delta y_k + \frac{\partial g_j}{\partial u_m}\,\delta u_m \qquad (Ib)$$

$$\delta y_j(z,0) = \delta w_j(z) \qquad j = 1, 2, \ldots q$$

$$\delta I = \int_0^1 \int_0^1 \left[\frac{\partial G}{\partial x_i}\,\delta x_i + \frac{\partial G}{\partial y_j}\,\delta y_j + \frac{\partial G}{\partial u_k}\,\delta u_k + \frac{\partial G}{\partial v_i}\,\delta v_i + \frac{\partial G}{\partial w_j}\,\delta w_j \right] dz\,dt \qquad (Ic)$$

If we multiply Equation (Ia) by the r vector $\boldsymbol{\lambda}$ and multiply Equation (Ib) by the q vector $\boldsymbol{\mu}$, integrate the result with respect to both z and t, and add to Equation (Ic), we obtain

$$\delta I = \int_0^1 \int_0^1 \left[\left\{ \frac{\partial G}{\partial x_i} + \lambda_j \frac{\partial f_j}{\partial x_i} + \mu_k \frac{\partial g_k}{\partial x_i} \right\} \delta x_i + \left\{ \frac{\partial G}{\partial y_j} + \lambda_i \frac{\partial f_i}{\partial y_j} + \mu_k \frac{\partial g_k}{\partial y_j} \right\} \delta y_j + \left\{ \frac{\partial G}{\partial u_k} + \lambda_i \frac{\partial f_i}{\partial u_k} + \mu_j \frac{\partial g_j}{\partial u_k} \right\} \delta u_k + \left\{ \frac{\partial G}{\partial v_i}\,\delta v_i + \frac{\partial G}{\partial w_j}\,\delta w_j - \lambda_i \frac{\partial(\delta x_i)}{\partial z} - \mu_j \frac{\partial(\delta y_j)}{\partial t} \right\} \right] dz\,dt \qquad (Id)$$

When the last two terms are integrated by parts and $\boldsymbol{\lambda}$ and $\boldsymbol{\mu}$ are defined by

$$\frac{\partial \lambda_i}{\partial z} = -\left[\frac{\partial G}{\partial x_i} + \lambda_k \frac{\partial f_k}{\partial x_i} + \mu_j \frac{\partial g_j}{\partial x_i} \right]; \quad \lambda_i(1,t) = 0 \qquad (Ie)$$

$$i = 1, 2, \ldots r$$

$$\frac{\partial \mu_j}{\partial t} = -\left[\frac{\partial G}{\partial y_j} + \lambda_i \frac{\partial f_i}{\partial y_j} + \mu_k \frac{\partial g_k}{\partial y_j} \right]; \quad \mu_j(z,1) = 0 \qquad (If)$$

$$j = 1, 2, \ldots q$$

Equation (Id) yields

$$\delta I = \int_0^1 \int_0^1 \left[\left\{ \frac{\partial G}{\partial u_k} + \lambda_i \frac{\partial f_i}{\partial u_k} + \mu_j \frac{\partial g_j}{\partial u_k} \right\} \delta u_k \right] dz\,dt + \int_0^1 \left[\left\{ \int_0^1 \frac{\partial G}{\partial v_i}\,dz + \lambda_i(0,t) \right\} \delta v_i(t) \right] dt \qquad (Ig)$$

$$+ \int_0^1 \left[\left\{ \int_0^1 \frac{\partial G}{\partial w_j}\,dt + \mu_j(z,0) \right\} \delta w_j(z) \right] dz$$

In order for the nominal controls $\bar{u}$, $\bar{v}$, $\bar{w}$ to be the optimal ones, $\delta I \leq 0$ must hold for all small variations δu, δv, δw. This can hold only if the quantities in brackets in Equation (Ig) vanish for unconstrained variations.

$$\frac{\partial H}{\partial u_k} = \frac{\partial G}{\partial u_k} + \lambda_i \frac{\partial f_i}{\partial u_k} + \mu_j \frac{\partial g_j}{\partial u_k} = 0 \qquad (Ih)$$

$$k = 1, 2, \ldots n$$

$$\frac{\partial H_1}{\partial v_i} = \int_0^1 \frac{\partial G}{\partial v_i}\,dz + \lambda_i(0,t) = 0 \qquad (Ii)$$

$$i = 1, 2, \ldots r$$

$$\frac{\partial H_2}{\partial w_j} = \int_0^1 \frac{\partial G}{\partial w_j}\,dt + \mu_j(z,0) = 0 \qquad (Ij)$$

$$j = 1, 2, \ldots q$$

must hold where H, H_1, H_2 are defined by

$$H = G + \lambda_i f_i + \mu_j g_j$$

$$H_1 = \int_0^1 G\,dz + \lambda_i(0,t)\,v_i(t) \qquad (Ik)$$

$$H_2 = \int_0^1 G\,dt + \mu_j(z,0)\,w_j(z)$$

Simply, if any of the controls u_k, v_i, w_j are constrained by (10) at the optimum, the appropriate Hamiltonian H, H_1, or H_2 should be a maximum with respect to that control on the constraint.

Where one or more of the u_k are functions of t alone, then condition (Ih) becomes $\int_0^1 \frac{\partial H}{\partial u_k}\,dz = 0$ and $\int_0^1 H\,dz$ should be a maximum at a constraint on u_k. Similarly, for a u_k only a function of z, then (Ih) becomes $\int_0^1 \frac{\partial H}{\partial u_k}\,dt = 0$ and $\int_0^1 H\,dt$ should be a maximum at a constraint. Again, if $v_i(t)$ is independent of t, then (Ii) becomes $\int_0^1 \frac{\partial H_1}{\partial v_i}\,dt = 0$ and $\int_0^1 H_1\,dt$ should be a maximum at a constraint. Also, if $w_j(z)$ is independent of z, then (Ij) becomes $\int_0^1 \frac{\partial H_2}{\partial w_j}\,dz = 0$ and $\int_0^1 H_2\,dz$ should be a maximum at a constraint.

APPENDIX II.

Gradient Methods

A control vector iteration scheme is easily developed from the maximum principle of Appendix I.

Modified or Accelerated Gradient Method. This method, very analogous to the well-known technique (23) developed for lumped parameter systems, has the following steps:

1. Guess a set of controls $\bar{u}$, $\bar{w}$, $\bar{v}$.
2. Integrate the state equations forward and the adjoint equations backward (using the method of characteristics). This produces the current value $I = \bar{I}$.

* A repeated subscript denotes summation over that index.

3. Calculate $\frac{\partial H}{\partial u_k}, \frac{\partial H_1}{\partial v_i}, \frac{\partial H_2}{\partial w_j}$ and make a correction in the control given by*

$$\delta u_k = \epsilon \frac{\partial H}{\partial u_k} \qquad \text{(IIa)}$$

$$\delta v_i = \epsilon \frac{\partial H_1}{\partial v_i} \qquad \text{(IIb)}$$

$$\delta w_j = \epsilon \frac{\partial H_2}{\partial w_j} \qquad \text{(IIc)}$$

Notice that from Equation (Ig), this guarantees δI increasing with each iteration if ϵ is picked small enough.

4. Integrate the state equations and get a new value of I.

5. If $I > \bar{I}$ (a success), then double ϵ, and go back to (3). If $I < \bar{I}$ (a failure), then divide ϵ by 4 and go back to (3). If after at least one success, we get a failure, then fit a quadratic in ϵ to I to find the optimal ϵ in the direction given by (IIa) to (IIc). Then go back to (2).

6. Terminate the procedure when it is felt the optimum has been reached. This method has been found to be more efficient than simple steepest ascent, and it has the property that constraints of the type in Equation (10) are no problem. When a constraint is reached, it is followed and the basic properties of the algorithm are unchanged.

Conjugate Gradient Method. This method, based on the ideas of Fletcher and Reeves (*24*) for discrete systems, has been developed for lumped parameter optimization by Lasdon *et al.* (*25*). As in the case of discrete systems, a requirement for the method to work is that the controls be unconstrained. Our use was therefore limited to the steps in the iteration where the controls remained unconstrained. In these cases the method was noticeably more efficient than the modified gradient method. Our application of it was for the adiabatic reactor examples where v_i and w_j were fixed in advance, in which case the method is identical to that described by Lasdon *et al.* (25).

* These corrections will be modified if u_k is a function of only one independent variable or if v_i and w_j are constants as discussed in Appendix I.

Heat and Mass Transfer in the Wall Region of Turbulent Pipe Flow

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Models for the laminar sublayer of turbulent pipe flow are reviewed. The observations of the sublayer by Popovich and Hummel and the experimental fluctuation frequencies obtained by Shaw and Hanratty are used with the models to evaluate experimental heat and mass transfer data. The eddy diffusivity model and a parallel conductance model appear to be consistent with the experimental data.

Heat and mass transfer in the wall region for turbulent flow has been a subject of continuing interest. Several models have been proposed to represent the rate of transport from the wall to a turbulent fluid. These must obviously represent the behavior of the fluid in the immediate vicinity of the wall. The fluid velocity increases from a very small value near the wall to a large value for the region of free turbulence in the core. A thin nonturbulent layer has been postulated next to the wall which has been designated as the laminar sublayer. The mechanism of momentum, heat, and mass transport would thus be molecular rather than by eddy motion. Fage and Townend (*1*) examined the motion of small dust particles in water under the ultramicroscope very near the wall and found no evidence of the existence of a region possessing rectilinear motion. Lin et al. (*2*) obtained data for concentration profiles with mass transfer at Schmidt numbers of about 900 and concluded that there is no laminar film corresponding to molecular diffusion. Subsequent mass transfer data have been obtained at Schmidt numbers up to 100,000, and these data show that the observed rate is much greater than could occur with a fully developed laminar film and molecular diffusion at the pipe wall. Popovich and Hummel (*3*) have used a flash photolysis method to study the wall region for turbulent flow in a square smooth pipe at a Reynolds number of 13,100. This work shows that there is a layer $y^+ = 1.6 \pm 0.4$ in which there is always a linear velocity gradient but the slope of