c_p = heat capacity at constant pressure, L^2/t^2T
H = generalized local transport coefficient, L^3/T
h_s = inverse of surface resistance, L/t
l = thickness of stagnant film; average thickness of fluid elements, L
L = thickness of liquid layer in wetted wall column, L
m = number of capacitances or capacitors in multiple capacitances model, dimensionless
P = potential, $\rho c_p T$ in M/Lt^2 or C in M/L^3
s = frequency or fractional rate of surface renewal, t^{-1}
t = time of process on the macroscopic scale, t
T = temperature, T
z = distance from interfacial plan, L

Greek Letters

γ = h_s/κ, L^{-1}
δ = impulse function, t^{-1}
θ = contact time or age, t
κ = generalized molecular diffusivity, L^2/t
π = 3.14159 ..., dimensionless
ρ = density of fluid or particles, M/L^3
τ = mean residence time, t
ϕ = contact time (or age) distribution function, t^{-1}
ψ_i = instantaneous local transport rate, ML^2/t^3 or M/t
ψ = average local transfer rate, ML^2/t^3 or M/t
$\overline{\psi}$ = average transfer rate for any time interval, ML^2/t^3 or M/t

Subscripts

b = quantity evaluated at bulk stream
l = infinite thickness of fluid elements
0 = quantity evaluated at interface
γ = no surface resistance
∞ = infinitely many capacitors

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Studies of Singular Solutions in Dynamic Optimization: II. Optimal Singular Design of a Plug-Flow Tubular Reactor

DANIEL Y. C. KO and WILLIAM F. STEVENS

Northwestern University, Evanston, Illinois

Although much attention has been given recently to the development of methods for the determination of the optimal control of a batch reactor or the best operating conditions for a tubular reactor, a number of difficulties and uncertainties still remain, especially when the analysis involves an exothermic reversible reaction. Several investigators (*1, 5, 8*) have been concerned with the establishment of the optimum temperature profile along a tubular reactor, from which the optimal control (heat removal rate) must then be obtained. Others (*2, 10* to *12*) have studied methods suitable for direct determination of the heat flux profile, some of which resulted in the possible appearance of singular control for a portion of the reactor length. The present authors have looked further into the occurrence of such singular problems during the application of the theory of optimal control and have developed an improved approach to the determination of the optimal heat transfer coefficient distribution along a tubular reactor (*6, 7*).

This paper presents the details of an application of the method of solution presented in the companion paper (*7*) to the optimal design of a tubular reactor. It is shown that, in general, if the reactor is "sufficiently long," the optimal

Daniel Y. C. Ko is with Gulf Research and Development Company, Pittsburgh, Pennsylvania.

heat transfer coefficient distribution always ends with a singular control section. Before singular control is applied, the control may or may not switch, depending on the initial conditions and the constraints on the control variable. Numerical results are presented, and a comparison is made between the new method and a conventional gradient approach.

STATEMENT OF THE PROBLEM

Consider a first-order, reversible, exothermic chemical reaction taking place in a plug-flow tubular reactor according to the equation $A \underset{k_2}{\overset{k_1}{\rightleftarrows}} B$. Steady state differential material and energy balances in the reactor are given by

$$\frac{dC_B}{dt} = R(C_B, T) = k_1(C_{A0} + C_{B0} - C_B) - k_2 C_B \quad (1)$$

$$C_p\rho \frac{dT}{dt} = (-\Delta H)\, R(C_B, T) - h(T - T_c) \quad (2)$$

The specific reaction rates k_i follow Arrhenius' law:

$$k_1 = k_{10} \exp\,(-E_1/R_gT) \quad k_2 = k_{20} \exp\,(-E_2/R_gT)$$

where $E_2 > E_1$ for an exothermic reaction. Letting

$$x_1 = C_B \quad x_2 = T \quad J = (-\Delta H)/C_p\rho \quad u = h/C_p\rho$$

$$\gamma_1 = E_1/R_g \quad \gamma_2 = E_2/R_g \quad C_{A0} + C_{B0} = 1$$

Equations (1) and (2) can be written

$$f_1(\mathbf{x}, u) = \dot{x}_1 = R(x_1, x_2) \quad (3)$$

$$f_2(\mathbf{x}, u) = \dot{x}_2 = J\, R(x_1, x_2) - u(x_2 - T_c) \quad (4)$$

$$x_1(t_0) = x_1^0, \quad x_2(t_0) = x_2^0$$

where

$$R(x_1, x_2) = (1 - x_1)\, k_{10} \exp\,(-\gamma_1/x_2) - x_1\, k_{20} \exp\,(-\gamma_2/x_2) \quad (5)$$

The optimization problem can now be posed as follows:

Given the system state equations, (3) and (4), with the given initial conditions of state variables and a fixed "length" of reactor t_f, find the control u (subject to the constraints, $u_{\min} \leq u \leq u_{\max}$) so that the yield of the desired product B, $x_1(t_f)$, is maximized. In this case the control u is proportional to the heat transfer coefficient h, which can be controlled by manipulating the flow rate of coolant; $u_{\min} = 0$, corresponding to no cooling, while $u_{\max}$ corresponds to the maximum allowable coolant flow rate.

As in the usual application of Pontryagin's maximum principle, the Hamiltonian function is defined as

$$H = \mathbf{p}^T\mathbf{f} = p_1\, R + p_2\, [JR - u\,(x_2 - T_c)] \quad (6)$$

The adjoint variables p_1 and p_2 are given by

$$\dot{p}_1 = -\,(p_1 + Jp_2)\, R_1 \quad (7)$$

$$\dot{p}_2 = -\,(p_1 + Jp_2)\, R_2 + p_2 u \quad (8)$$

where

$$R_1 = \frac{\partial R}{\partial x_1} = -\,k_{10} \exp\,(-\gamma_1/x_2) - k_{20} \exp\,(-\gamma_2/x_2) \quad (9)$$

$$R_2 = \frac{\partial R}{\partial x_2} = \frac{1}{x_2^2}\,[k_{10}\gamma_1\,(1 - x_1) \exp\,(-\gamma_1/x_2) - k_{20}\,\gamma_2\,x_1 \exp\,(-\gamma_2/x_2)]$$

The final conditions for the adjoint variables are

$$p_1(t_f) = 1 \quad p_2(t_f) = 0$$

Considering that $H\,(\mathbf{x}, \mathbf{p}, u) = \psi(\mathbf{x}, \mathbf{p}) + u\,\phi\,(\mathbf{x}, \mathbf{p})$, then

$$\phi(\mathbf{x}, \mathbf{p}) = -\,p_2\,(x_2 - T_c) \quad (10)$$

$$\psi(\mathbf{x}, \mathbf{p}) = p_1 R + Jp_2 R \quad (11)$$

The optimal control, if it exists, is given by

$$\hat{u} = \begin{cases} u_{\max}, & \text{if } \phi(\mathbf{x}, \mathbf{p}) > 0 \\ 0, & \text{if } \phi(\mathbf{x}, \mathbf{p}) < 0 \\ \text{undetermined}, & \text{if } \phi(\mathbf{x}, \mathbf{p}) = 0 \text{ for } t_1 \leq t \leq t_2 \end{cases} \quad (12)$$

SINGULAR SURFACE AND NECESSARY CONDITIONS

The problem presented in the previous section is of fixed final time with an objective function of the Mayer form. Since it is only two-dimensional the singular surface can be constructed without the specification of additional conditions. Following the procedure outlined in the companion paper (7), there results

$$\dot{\phi}(\mathbf{x}, \mathbf{p}) = R_2(x_2 - T_c)p_1 + J\,[R_2(x_2 - T_c) - R]\,p_2 \quad (13)$$

On the singular arc, $\phi = \dot{\phi} = 0$, which represents a system of homogeneous linear equations in p_i, $i = 1, 2$. The system has a nontrivial solution for p_i if and only if the determinant of the coefficient matrix vanishes, to give $-R_2(x_2 - T_c)^2 = 0$. It can be assumed that the coolant temperature is always lower than the reaction temperature. Then the singular surface (curve) becomes the maximum rate curve with respect to temperature, along which $R_2 = 0$. The control which keeps the reactor on this curve (the singular control) is obtained by solving $d/dt\,(R_2) = 0$ for u, to give

$$u_s(t) = \frac{JR}{x_2 - T_c} + \frac{R_{21}R}{R_{22}(x_2 - T_c)} \quad (14)$$

where

$$\left.\begin{aligned} R_{21} &= \frac{\partial^2 R}{\partial x_2 \partial x_1} = -\,\frac{k_{10}\gamma_1}{x_2^2} \exp\,(-\gamma_1/x_2) - \frac{k_{20}\gamma_2}{x_2^2} \exp\,(-\gamma_2/x_2) \\ R_{22} &= \frac{\partial^2 R}{\partial x_2^2} = \frac{1}{x_2^4}\,[k_{10}(1 - x_1)\gamma_1 \exp\,(-\gamma_1/x_2)]\,(\gamma_1 - 2x_2) \\ &\quad - \frac{1}{x_2^4}\,[k_{20}\,x_1\,\gamma_2 \exp\,(-\gamma_2/x_2)]\,(\gamma_2 - 2x_2) \end{aligned}\right\} \quad (15)$$

It is seen from Equation (10) that $p_2 = \dot{p}_2 = 0$, on the singular arc. In addition, it is known (7) that $\psi(\mathbf{x}, \mathbf{p}) = k$, where k is a positive constant. Combining these conditions with the fact that $R_2 = 0$ on the singular arc, there results

$$\frac{\partial}{\partial u} \cdot \frac{d^2\phi}{dt^2} = \frac{-k(x_2 - T_c)^2\,\gamma^2}{Rx_2^4}\left[\, k_{10}(1 - x_1) \exp\,(-\gamma_1/x_2) - k_{20}x_1 \left(\frac{\gamma_2}{\gamma_1}\right)^2 \exp\,(-\gamma_1/x_2)\,\right] \quad (16)$$

From Equation (9) and the fact that $R_2 = 0$ on the singular arc, it can be shown that the quantity in the bracket is negative, since $(\gamma_2/\gamma_1) > 1$ for an exothermic reaction and $R(x_1, x_2) > 0$. Thus

$$\frac{\partial}{\partial u}\left(\frac{d^2}{dt^2}\frac{\partial H}{\partial u}\right) > 0 \qquad (17)$$

and the necessary condition for optimality of the singular arc is satisfied. Hence, singular control may become a part of the optimal control function.

On the other hand, if the reversible reaction is endothermic, with $(\gamma_2/\gamma_1) < 1$, then the singular control given by Equation (14) cannot be optimal. It is well known that the best policy for an endothermic reversible reaction is to operate at the highest allowable temperature. Similarly, in the case of an irreversible exothermic reaction, $k_2 = \gamma_2 = 0$, it can easily be shown that the necessary condition, Equation (17), is not satisfied, with the result that optimal control must be only bang-bang.

POSSIBLE OPTIMAL CONTROL FUNCTIONS

Previous analysis (7) indicates that the optimal control function may consist of (1) pure singular control, (2) pure bang-bang control, or (3) bang-bang control combined with singular control. Figure 1 gives a phase plane plot for the present problem: a first-order, reversible, exothermic reaction taking place in a plug-flow tubular reactor. The curve $R = 0$ is the equilibrium curve, and the curve S_s is the singular curve, that is, the maximum rate curve, $R_2 = 0$. Figure 1 will be used, in subsequent paragraphs, as the basis for the argument that the first two control function possibilities are unlikely to exist for the particular problem being considered.

First, assume that the initial point (the condition at the entrance to the reactor) lies in region Σ① since this is the condition of usual practice. Therefore, the optimal control cannot be purely singular, since $x(t_0)$ does not lie on S_s. In order to bring $x(t_0)$ to a point on S_s, an impulse function of heating would be required, at $t = t_0$, which is outside the constraints of the control variable.

Next, consider the allowable switching directions for bang-bang control (7). From Equation (13)

$$\left.\frac{d\phi}{dt}\right|_{\phi=0} = p_1 R_2 (x_2 - T_c)$$

However, when $\phi = 0$, $\psi = p_1 R = k$, which means that $p_1 > 0$. Therefore

$$\sigma = sgn\ R_2$$

Now, if $x(t)$ is in region Σ①, $R_2 > 0$. Hence, $\sigma = 1$, and the allowable switching direction is $\hat{u} = 0 \rightarrow \hat{u} = u_{max}$. If $u(t_0) = u_{max}$, no switching is allowed. If $x(t)$ is in region Σ②, $R_2 < 0$. Hence, $\sigma = -1$, and the allowable switching direction is $\hat{u} = u_{max} \rightarrow \hat{u} = 0$. If $u(t_0) = 0$, no switching is allowed.

Suppose that the reactor has started from point A and has passed through the curve $R_2 = 0$ in some optimal fashion. If $u = 0$ is the concluding optimal control, then

$$\left.\begin{array}{l}\phi(\mathbf{x}, \mathbf{p}) < 0 \quad \text{or} \quad p_2(t) > 0 \\ 1 > p_1(t) \geq 0, \quad \dot{p}_1(t) > 0\end{array}\right\} \quad t_f - \delta t \leq t \leq t_f \qquad (18)$$

since $p_1(t_f) = 1$. In addition

$$\dot{p}_2(t_f) = -(p_1 + Jp_2) R_2 > 0, \quad t_f - \delta t \leq t \leq t_f \qquad (19)$$

since $R_2 < 0$ in region Σ②. This means that $p_2(t)$ will continuously increase. However, $p_2(t_f) = 0$, so $u = 0$ cannot be the concluding control in region Σ② or on the curve $R_2 = 0$. Similarly, if the reactor has passed through the singular curve by $u = u_{max}$, and thus returned to region Σ①, it cannot switch from $u = u_{max}$ to $u = 0$. Therefore it can be stated that the optimal control cannot be ended by $u = 0$, providing the reactor is long enough to allow the representative point to travel from A through $R_2 = 0$.

Since the initial conditions are in region Σ①, the reactor requires heating at the entrance section to drive it toward the maximum rate curve. Therefore, adiabatic operation ($u = 0$) will be the initial optimum policy and the reactor will follow a straight line JK for a time. The allowable switching direction says that if the control switches, it must switch from $u = 0$ to $u = u_{max}$ in region Σ① before the reactor reaches S_s. Further switchings are not allowed, since $u = 0$ cannot be the concluding control. Therefore, the optimal control could be bang-bang, but only in the restricted form of $u = 0$ at the start followed by at most a single switch to $u = u_{max}$ before the reactor reaches S_s, and there is real question whether such a control would be optimal, since the trajectory never follows the maximum rate curve.

The third possible optimal control function is that which makes use of bang-bang control combined with singular control. From the previous analysis, it can be seen that if singular control appears, it will be in the concluding interval. Also, the optimal control will start with $u = 0$ and will have at most one switching. Thus there are two possible control functions before the singular control: (1) The control switches at $t = t_1$ from $u = 0$ to $u = u_{max}$, and

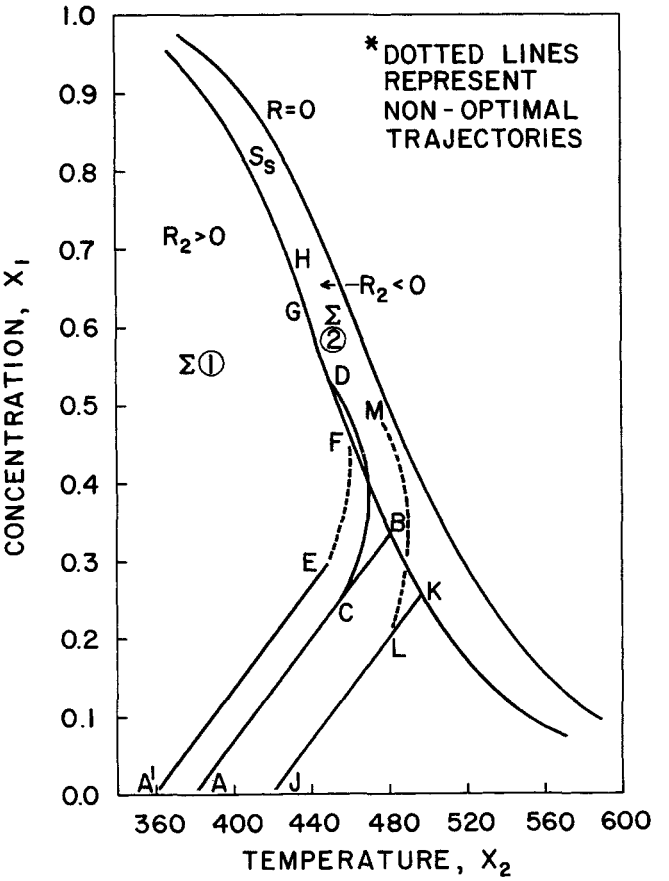

Fig. 1. Phase plane plot and possible optimal trajectories.

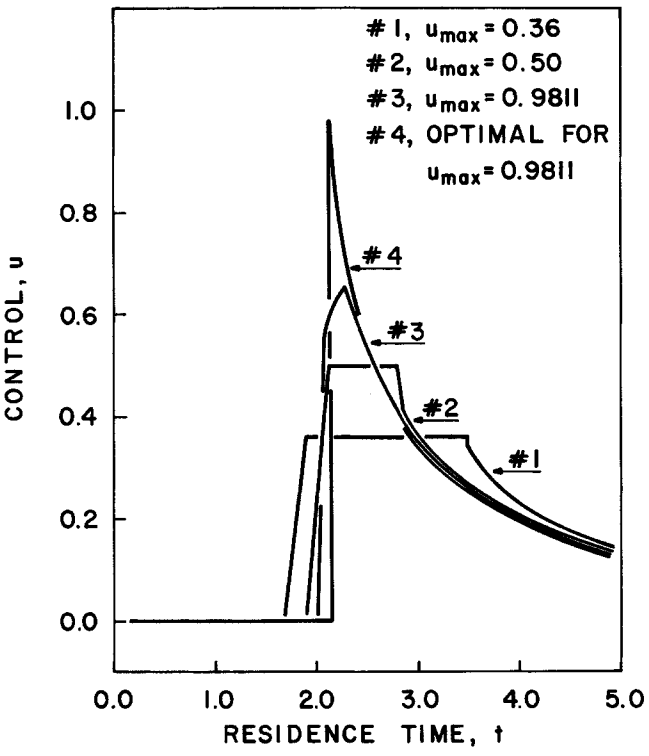

Fig. 2. Effect of u_{max} on the optimal control profile.

then at $t = t_s$ the control changes to singular control and continues to the end, $t = t_f$; or (2) the control stays at $u = 0$ from $t = t$ to $t = t_s$, where it changes to singular control and continues to the end.

For case 1, intuition leads to the assumption that the reactor can follow the path *A'EFG*. However, the following analysis rules out this possibility. On the singular curve, $\dot{p}_1 = -p_1 R_1$, for $t_s + \delta t \leq t \leq t_f$. Since $p_1(t_f) = 1$, it is true that $1 \geq p_1 > 0$, $\dot{p}_1 > 0$, and no sign change is allowed for $t_s + \delta t \leq t \leq t_f$. By continuity, and from Equation (7), it follows that

$$(p_1 + Jp_2) > 0 \quad t_s - \delta t \leq t \leq t_s + \delta t \qquad (20)$$

Assume that the full cooling curve joins the singular curve from region Σ①. For this case, $p_2(t) < 0$, $R_2(t) > 0$, and $p_2 < 0$, for $t_s - \delta t \leq t \leq t_s$, because of Equation (20). That is, p_2 should continue to decrease in region Σ① before the reactor moves onto the singular curve. It is apparent that $p_2(t_s) = 0$, which is required on the singular curve, cannot be satisfied. Thus, if p_2 cannot have a discontinuity at t_s, full cooling cannot join the curve $R_2 = 0$ from region Σ①.

On the other hand, if the full cooling curve penetrates (3) the singular curve in region Σ② and then joins the singular curve, it is easy to verify that the adjoint variable p_2 will be continuous at t_s, and hence such a path can be an optimal trajectory. See curve *ACDH* in Figure 1.

The above discussion means that when the maximum rate curve is first reached, after a switch from $u = 0$ to $u = u_{max}$, the reactor does not stay on the curve. However, as case 2 indicated, it is possible for no switching to take place before the maximum rate curve is reached. Under this mode of operation, the adiabatic operation line ($u = 0$) joins the singular curve from region Σ①. Then, $p_2(t) > 0$, $R_2(t) > 0$, and $p_2 < 0$, for $t_s - \delta t \leq t \leq t_s$, because of Equation (20). That is, $p(t_s) = 0$ can be reached. In other words, the maximum rate curve is first reached, with $u = 0$ and unchanged, the reactor stays on the curve until $t = t_f$.

It would be desirable to determine, a priori, whether or not the optimal control will switch before it reaches the singular control surface. Unfortunately, a rigorous mathematical answer is not yet available. However, numerical experience seems to indicate if the upper bound of the control variable u_{max} is greater than u_s, obtained from Equation (14) and evaluated at the intersection of the adiabatic and singular curves (point *B* in Figure 1), then no switching takes place. A plausible argument for this conclusion is that if $u_{max} < u_s$, @ $(t = t_s)$, then the singular curve cannot be reached by the adiabatic curve. Since the control required to retain the reactor on the singular arc is not allowable, the control must switch to u_{max} before the reactor reaches S_s. Once the switching is done the reactor will be required to penetrate the singular curve and return to it from region Σ②. Numerical examples to be presented later will serve to support these conclusions.

It should be emphasized that Equation (17) provides only a necessary condition for optimality of the singular arc. There is still no a priori method for selecting the true optimal control from the two possible control functions—pure bang-bang or bang-bang plus singular. The correct function must be determined by numerical methods. Even so, analysis of the possible optimal control functions gives valuable insight into the problem and prepares the designer for the possible existence of singular control. All numerical solutions obtained by the authors (6), including those presented in the next section of this paper, gave an optimal control which included a singular portion, indicating strong support for the supposition that pure bang-bang control is not likely to be encountered.

NUMERICAL RESULTS

The following data, in part due to Dyson (4), were assumed for the numerical calculations which follow:

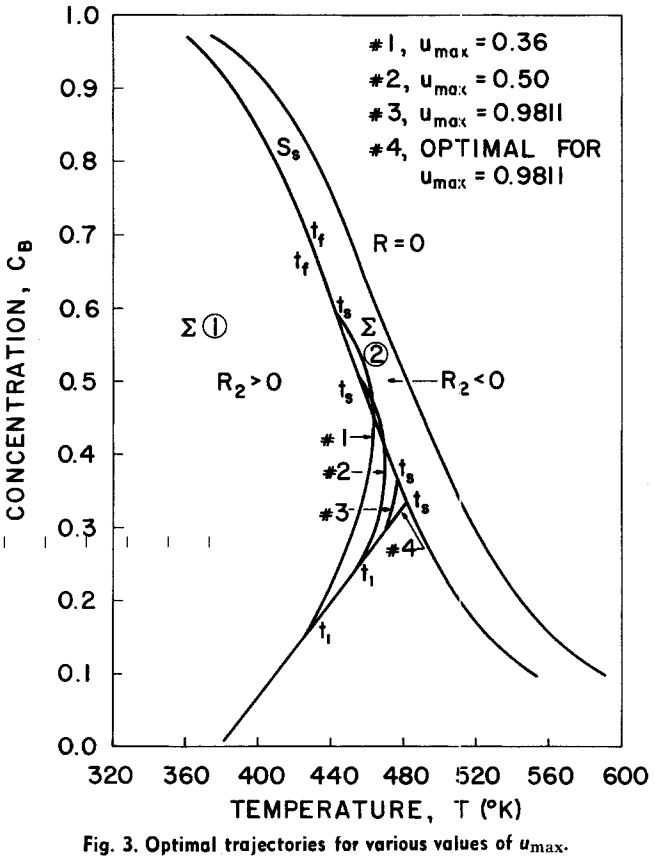

Fig. 3. Optimal trajectories for various values of u_{max}.

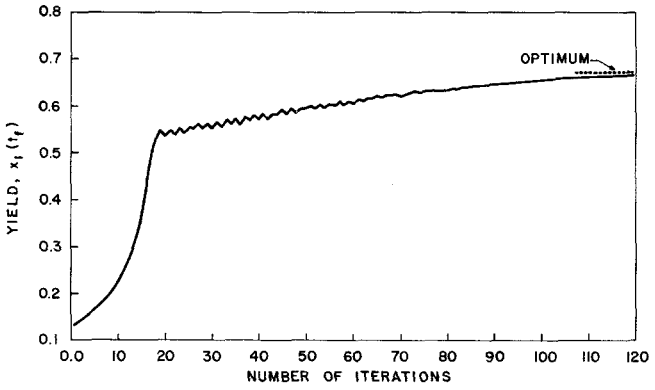

Fig. 4. Objective function history, gradient method.

$k_{10} = 1.7536 \times 10^5$ min.$^{-1}$ $k_{20} = 2.4885 \times 10^{10}$ min.$^{-1}$

$E_1 = 1.1374 \times 10^4$ cal./mole

$E_2 = 2.2748 \times 10^4$ cal./mole

$J = (300°\text{K.})\,(\text{cc.})/\text{mole}$ $T_c = 290°\text{K.}$ $t_f = 5$ min.

Figure 2 presents the effect of various values of u_{max} on the optimal control profile. For each case, $x_1(t_0) = 0.0$ and $x_2(t_0) = 380°\text{K}$. It can be seen that as the value of u_{max} increases, the section of reactor which requires full cooling becomes shorter, such that eventually no full cooling is necessary, consistent with the analysis of the previous section. Curves 1, 2, and 3 were obtained using the gradient method with the initial profile assumed close to the curves shown. More than 200 iterations were required in each instance. For the given initial conditions, $u_s(t_s) = 0.9811$, and curve 3 resulted when this value was used for u_{max}. Curve 4 was obtained by applying, a priori, the fact that optimal control will consist of a section with no cooling extending from the reactor inlet to the point at which the singular curve is reached, after which the singular control u_s, obtained from Equation (14), comes into existence. It can be seen that curve 3 is a reasonably good approximation to curve 4, as would be expected.

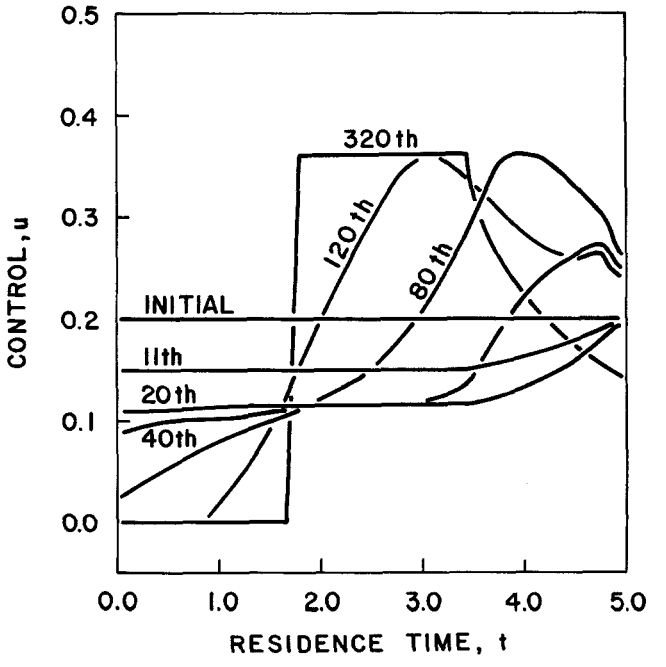

Fig. 5. Successive control profiles, gradient method.

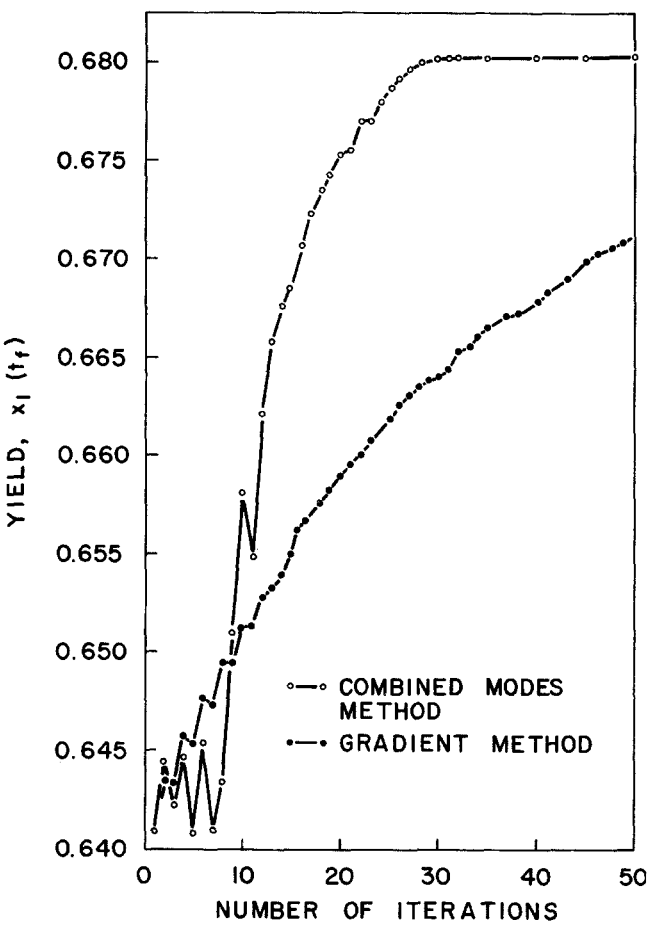

Fig. 6. Objective function history, combined mode and gradient method, case 1.

Optimal trajectories for various values of u_{max}, corresponding to the optimal controls of Figure 2, are presented in Figure 3. For each curve, $t = t_s$ is the time at which singular control begins. In addition, the control switches from $u = 0$ to $u = u_{max}$ at $t = t_1$, for curves 1 and 2. In both of these cases, the trajectory penetrates the singular curve (maximum rate curve) and then joins the singular curve from the right side, at $t = t_s$. These results substantiate the conclusions of the previous section. Also, it should be noted that curve 3, obtained by the gradient method, is an approximation to curve 4, the exact trajectory obtained by the known sequence of adiabatic and singular control. No penetration of the singular curve takes place for these two curves.

Figures 2 and 3 present the optimal solution to the problem originally posed, that of maximizing the yield of desired product from a plug-flow tubular reactor of fixed length. It is now proposed to compare the efficiency and the rate of convergence of the authors' "combined modes" method with that of a gradient procedure. The algorithms employed are presented in the companion paper (7).

One way to start the computation using the gradient method is to assume an arbitrary initial guess for the control profile. Here an initially flat control profile, $u(t) = 0.2$, was assumed, for the case of $u_{max} = 0.36$. Figure 4 shows the value of the objective function, $x_1(t_f)$, as a function of number of iterations, starting with the assumed flat profile. It can be seen that for the first 19 iterations the yield increased steadily and quickly. However, from the

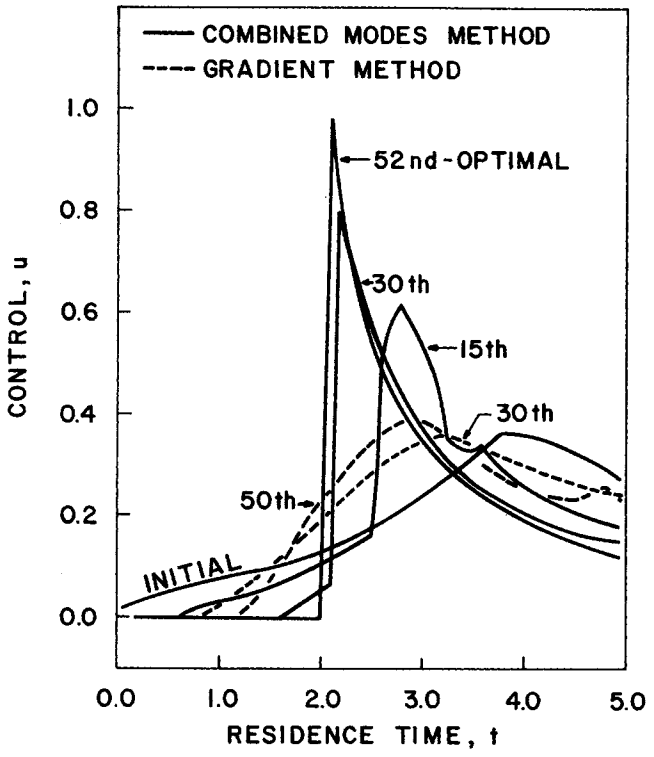

Fig. 7. Successive control profiles, combined mode and gradient method, case 1.

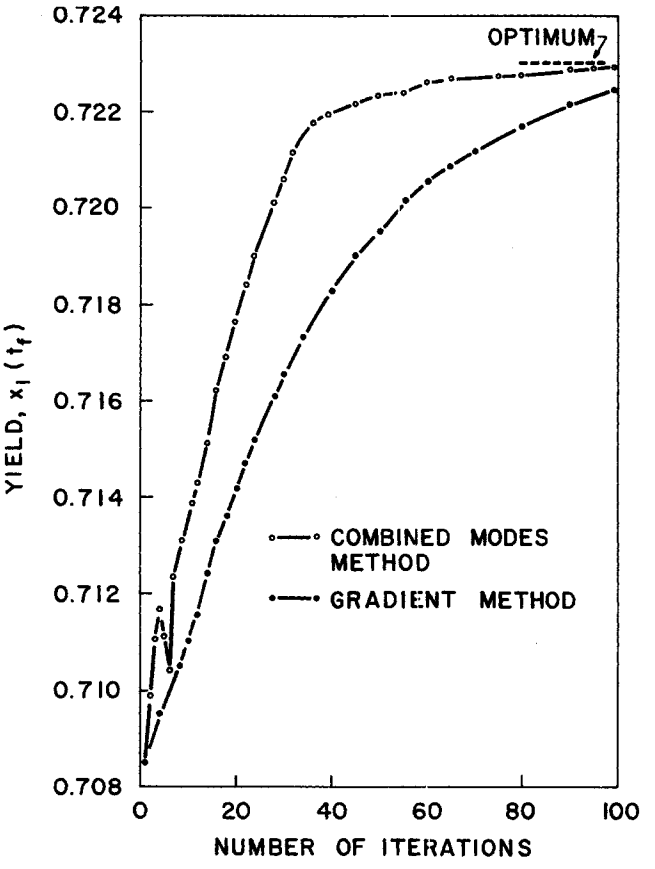

Fig. 8. Objective function history, combined mode and gradient method, case 2.

20th iteration on the curve was a sawtooth, with a slightly increasing value of $x_1(t_f)$, and after the 85th iteration it became steady but increased even more slowly. The performance shown is typical of that resulting from the gradient method when used for determining a singular solution. Between the 20th and 85th iterations, the procedure appears to have been on a "ridge," which requires regular zigzagging in order to climb, while after the 85th iteration a flat surface around the maximum seems to have been reached.

Figure 5 presents some of the control profiles determined as the gradient method proceeded. It should be noted that after 120 iterations the objective function is only 1.5% away from the known optimum value, but the control is still far from the optimal profile. Actually, 320 iterations were required to produce the optimal control curve shown in Figure 5. Thus it seems that the gradient method is a feasible method for obtaining the optimal solution, but its convergence to the actual optimal control function is extremely slow.

As discussed in detail in the companion paper (7), the combined modes method has been devised to overcome the slow convergence of the gradient procedure. It consists of the use of a new algorithm to hasten convergence, after application of the gradient method in the early stage of the solution, hence the name combined modes method. Several cases have been studied in detail, using both the gradient method and the combined modes approach. Two are reported here.

Case	$x_1(t_0)$	$x_2(t_0)$	u_{max}	ϵ
1	0.0	380.0	0.9811	0.05
2	0.0	410.0	0.5	0.10

Other small positive constants used in both cases were

$$\eta = 0.5 \quad \epsilon' = 0105 \quad \epsilon'' = 0.1$$

The yield of the reaction as a function of number of iterations of both methods are presented in Figure 6 for case 1 and in Figure 8 for case 2. Both indicate that the combined modes method causes the objective function to converge more rapidly than does the gradient method. Figures 7 and 9 give the corresponding successive control profiles for both cases. Again, it can be seen that the combined modes method approaches the optimal control profile relatively rapidly, while the gradient method fails to give the precise form of the optimal profile.

The initial curve for the control profile in Figure 7 was obtained after 85 iterations by the gradient method, while that in Figure 9 resulted after an even greater number of iterations. Limited experience indicates that if the combined modes method is employed to start the computation from an arbitrary initial guess of the control profile, convergence is not assured. Thus it seems logical to propose that the combined modes approach be used as a terminal scheme, after a start has been made using the gradient method, so as to improve significantly the rate of convergence over that obtainable with the gradient method alone.

Choices of the small positive constants ϵ, ϵ', ϵ'', and η are not especially difficult to make. After the gradient method has progressed somewhat, the magnitudes of both $|\phi|$ and $|\dot{\phi}|$ will be distinguishable and necessary modifications can be made. Furthermore, the constraint $0 \leq u_s \leq u_{max}$ serves to avoid employing the singular mode on the potential bang-bang arc. Thus whenever u_s is beyond the constraint, the gradient mode should be applied.

Finally, it has been found that the combined modes method works better when the optimal control has a long section of singular control. This is to be expected, since the

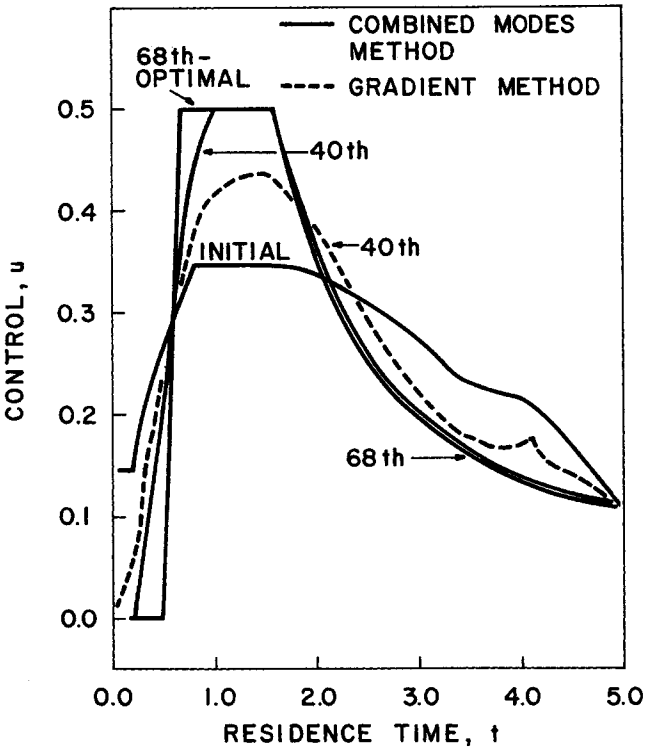

Fig. 9. Successive control profiles, combined mode and gradient method, case 2.

method was designed to overcome the effect of insensitive singular intervals.

CONCLUSIONS

The optimal design of the heat transfer coefficient distribution along the wall of a plug-flow tubular reactor in which an exothermic reversible reaction is taking place has been achieved. It has been found that optimal cooling will, in general, start with an adiabatic section and end with singular control. Whether the heat removal switches from adiabatic to full cooling depends upon the reactor entrance conditions and the maximum allowable heat transfer coefficient. If switching to full cooling occurs, penetration of the singular surface is necessary, and the bang-bang arc joins the singular arc in the region to the right of the maximum rate curve.

The gradient method (first-order variation approach) has been found feasible for obtaining an approximation to an optimal consisting of both bang-bang and singular arcs. However, it appears to give slow convergence close to the optimum, and it encounters difficulty in reaching the precise optimal control function if a singular section exists. When used as a terminal refinement scheme, the combined modes method can overcome these difficulties. Experience has shown that it is capable of rapid convergence and accurate representation of the singular control profile, providing the computation is started from an arbitrarily chosen initial profile using the gradient method. This joint use of the gradient method followed by the combined modes method is similar to the combination of first- and second-order variation approaches suggested by Luus and Lapidus (*9*).

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NOTATION

A, B = chemical species
C_A, C_B = concentration of component A or B
C_{A0}, C_{B0} = inlet concentration of component A or B
C_p = specific heat of reactants
E_i = activation energy corresponding to k_i
h = overall heat transfer coefficient
H = Hamiltonian function
$-\Delta H$ = heat of reaction
J = $(-\Delta H)/C_p\rho$
k_i = reaction rate constant of i^{th} reaction
k = positive constant
p_i = i^{th} component of adjoint vector p
R = rate of the reaction
R_g = gas constant
R_i = $\partial R/\partial x_i$
R_{ij} = $\partial^2 R/\partial x_i \partial x_j$
S_s = singular surface
t = residence time of reactant fluid
t_0 = initial time
t_f = final time
t_s = time of switch from nonsingular to singular arc
T = temperature of reactants
T_c = cooling jacket temperature
u = control variable, proportional to heat transfer coefficient
x_i = i^{th} component of state vector x
y_i = E_i/R_g

Greek Letters

ϵ = step-size parameter, gradient method
ϵ', ϵ'' = "small" positive constants
η = step-size parameter, combined modes method
ρ = fluid mass density
σ = sgn $d\phi/dt|_\phi = 0$
$\phi(\mathbf{x}, \mathbf{p})$ = switching function
$\psi(\mathbf{x}, \mathbf{p})$ = scalar function in Hamiltonian

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