Quasilinearization in Optimization: A Numerical Study

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The quasilinearization technique is used to obtain numerical answers for optimization problems formulated by the calculus of variations and the maximum principle. The optimum temperature profile in a tubular reactor, together with its initial optimum pressure, is obtained by this technique. Various variations of this problem such as control variable inequality constraints and the presence of pressure drop in the reactor are also discussed.

The two-point boundary value difficulties limit the use of both the calculus of variations and the maximum principle in obtaining numerical answers in optimization. In a recent paper (*6*) it has been shown that the quasilinearization technique can be used to overcome these difficulties when the control variable can be eliminated from the Euler-Lagrange equations. It is shown in this paper that the quasilinearization technique is equally effective when the control variable cannot be eliminated from the Euler-Lagrange equations or when the maximum principle is used. In the former case a trial and error procedure is used to solve numerically the algebraic and ordinary differential equations. In the latter case a search routine is added to the trial and error procedure. Only continuous systems are discussed; discrete systems involving complex structures will be discussed in other papers.

Another purpose of this paper is to show how a systematic approach can be used to optimize the control variables and system parameters simultaneously. By system parameters we mean a set of constant values which must be chosen before the process begins and which appear in the profit function of the process. The usual procedure for designing such a system is to choose several parameters, optimize the system under these different choices, and then select the most promising combination. The present procedure considers these parameters as additional state variables. The unknown initial conditions for these additional state variables can be obtained by applying the free boundary conditions or the transversality conditions. It has been shown that, due to the effectiveness of the quasilinearization technique in treating boundary value problems, the added state variable does not significantly increase the amount of work in obtaining numerical solutions.

The variational equations are first outlined with emphasis on practical applications instead of theoretical development. The optimum temperature profile in a tubular reactor with pressure as a parameter is then obtained by the combined use of the variational equations and the quasilinearization technique.

VARIATIONAL EQUATIONS

Calculus of Variations

Let us consider the following variational problem: Find that function

$$z(t) \tag{1}$$

and that set of constant parameters

$$a_1, \ldots, a_q \tag{2}$$

such that the set of functions

$$x_1(t), \ldots, x_n(t) \tag{3}$$

given by the differential equations

$$\dot{x}_i = f_i(x_1, \ldots, x_n, a_1, \ldots, a_q, t, z), \quad i = 1, \ldots, n \tag{4}$$

and end conditions

$$\psi_j(t_1, x_1(t_1), \ldots, x_n(t_1), t_2, x_1(t_2), \ldots, x_n(t_2), a_1, \ldots, a_q) = 0, \quad j = 1, \ldots, p \leq 2n + 2 \tag{5}$$

minimize a function of the form

$$J = g(t_1, x_1(t_1), \ldots, x_n(t_1), t_2, x_1(t_2), \ldots, x_n(t_2), a_1, \ldots, a_q) \tag{6}$$

where the expression $\dot{x}$ represents the first differential dx/dt. The variables $x_1(t), \ldots, x_n(t)$ are the state variables and the variable $z(t)$ is within our control and thus is called the control variable. The variable t can be considered as the time or length coordinate. The problem formulated above is essentially the problem of Mayer (*2*), except for the presence of the unknown constant parameters. However, this difference can be eliminated if we consider the constant parameters as functions of t and treat them as state variables. The following differential equations can be established (*11*):

$$\dot{a}_h(t) = 0, \quad h = 1, \ldots, q \tag{7}$$

The a's in Equations (5) and (6) now become $a_1(t_1), \ldots, a_q(t_1)$. The number of differential equations now becomes $(n + q)$. The unknown initial conditions $a_h(t_1)$, which are also the constant parameters a_h, can be obtained from the free boundary or transversality conditions during the process of solution.

To simplify notation, let $y_1, \ldots, y_{n+q}$ represent the state variables $x_1, \ldots, x_n, a_1, \ldots, a_q$. Following the classical treatment in the calculus of variations, let us introduce the set of Lagrange multipliers:

$$\lambda_i(t), \quad i = 1, \ldots, n, \ldots, (n + q) \tag{8}$$

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and the set of constant multipliers

$$\nu_j, \quad j = 1, \ldots, p \tag{9}$$

Define the functions

$$F(t, \bar{y}, \dot{\bar{y}}, z, \bar{\lambda}) = \sum_{i=1}^{n+q} \lambda_i (\dot{y}_i - f_i(t, z, \bar{y})) \tag{10}$$

$$G(t_1, \bar{y}(t_1), t_2, \bar{y}(t_2)) = g(t_1, \bar{y}(t_1), t_2, \bar{y}(t_2)) + \sum_{j=1}^{p} \nu_j \psi_j(t_1, \bar{y}(t_1), t_2, \bar{y}(t_2)) \tag{11}$$

where the vectors $\bar{y}$, $\dot{\bar{y}}$, and $\bar{\lambda}$ represent $y_1, \ldots, y_{n+q}$; $\dot{y}_1, \ldots, \dot{y}_{n+q}$; and $\lambda_1, \ldots, \lambda_{n+q}$, respectively. The Euler-Lagrange equations are

$$\frac{d}{dt}\frac{\partial F}{\partial \dot{y}_i} - \frac{\partial F}{\partial y_i} = 0, \quad i = 1, \ldots, n+q \tag{12}$$

$$\frac{\partial F}{\partial z} = 0 \tag{13}$$

The equation

$$\left[\left(F - \sum_1^{n+q} \dot{y}_i \frac{\partial F}{\partial \dot{y}_i}\right)dt + \sum_1^{n+q} \frac{\partial F}{\partial \dot{y}_i} dy_i\right]\Bigg|_{t_2} - \left[\left(F - \sum_1^{n+q} \dot{y}_i \frac{\partial F}{\partial \dot{y}_i}\right)dt + \sum_1^{n+q} \frac{\partial F}{\partial \dot{y}_i} dy_i\right]\Bigg|_{t_1} + dG = 0 \tag{14}$$

must hold at t_1 and t_2 for every choice of dy_1, dy_2, dt_1, and dt_2. In other words, if dy or dt or both do not vanish, their corresponding coefficients must vanish:

$$\left[-F + \sum_1^{n+q} \dot{y}_i \frac{\partial F}{\partial \dot{y}_i}\right]\Bigg|_{t_1} + \frac{\partial G}{\partial t}\Bigg|_{t_1} = 0,$$

$$\left[F - \sum_1^{n+q} \dot{y}_i \frac{\partial F}{\partial \dot{y}_i}\right]\Bigg|_{t_2} + \frac{\partial G}{\partial t}\Bigg|_{t_2} = 0 \tag{15}$$

$$\frac{\partial G}{\partial y_i}\Bigg|_{t_1} - \frac{\partial F}{\partial \dot{y}_i}\Bigg|_{t_1} = 0, \quad \frac{\partial G}{\partial y_i}\Bigg|_{t_2} + \frac{\partial F}{\partial \dot{y}_i}\Bigg|_{t_2} = 0, \quad i = 1, \ldots, n+q \tag{16}$$

where the symbol $|_{t_1}$ means that the expression at left is evaluated at t_1. Equation (14) or Equations (15) and (16) are known as the transversality condition. Equations (12) to (14) form a necessary condition for the optimization problem and have been called the multiplier rule by Bliss. Equation (12) can be reduced to

$$\dot{\lambda}_i = \frac{\partial F}{\partial y_i}, \quad i = 1, \ldots, n+q \tag{17}$$

For most chemical engineering problems, the values of t_1 and t_2 are known and thus Equation (15) no longer holds. If the end values of a certain state variable, say $y_r(t_1)$ and $y_r(t_2)$, do not appear in Equation (11), then Equation (16) for this variable can be reduced to

$$\frac{\partial F}{\partial \dot{y}_r}\Bigg|_{t_1} = 0, \frac{\partial F}{\partial \dot{y}_r}\Bigg|_{t_2} = 0 \tag{18}$$

which is known as the free boundary condition (3).

Our system now is composed of $2(n+q)$ differential equations [Equations (4), (7), and (12)], $2(n+q)+2$ transversality conditions [Equations (15) and (16)], one equation for the control variable [Equation (13)], and p end conditions [Equation (5)], to determine $(n+q)$ state variables and $(n+q)$ Lagrange multipliers, $2(n+q)+2$ end values t_1, t_2, $y_i(t_1)$, $y_i(t_2)$, $i = 1, \ldots, n+q$, one control variable, and p constant multipliers, ν_j, $j = 1, \ldots, p$.

Since the boundary conditions are not all given at the initial point t_1, the above system forms a two-point boundary value problem. The differential equations which are useful for engineering applications are generally nonlinear and cannot be solved analytically. The problem of finding numerical answers for this nonlinear boundary value problem is not simple and has limited the use of the calculus of variations. It will be shown by actual example that the quasilinearization is fairly effective to overcome these boundary value difficulties (*11*).

The above problem is formulated in the form which is most frequently encountered in chemical engineering applications. Some generalizations can be made easily. If there are m control variables, $z_1(t), \ldots, z_m(t)$, then Equation (13) becomes

$$\frac{\partial F}{\partial z_i} = 0, \quad i = 1, \ldots, m \tag{19}$$

Instead of Equation (6), the optimization of the following functional

$$J = g(t_1, \bar{y}(t_1), t_2, \bar{y}(t_2)) + \int_{t_1}^{t_2} f_o(\bar{y}, z, t)\, dt \tag{20}$$

can also be treated. The function F now becomes

$$F(t, \bar{y}, \dot{\bar{y}}, z, \bar{\lambda}) = f_o(\bar{y}, z, t) + \sum_1^{n+q} \lambda_i (\dot{y}_i - f_i(t, \bar{y}, z)) \tag{21}$$

The other equations remain the same except that Equation (17) no longer holds. The optimization of Equation (20) is known as the problem of Bolza. If the function g is equal to zero in Equation (20), we obtain the more familiar Lagrange problem and obviously the above equations are still applicable.

Calculus of Variations with Control Variable Inequality Constraint

Let us now assume that the decision variable $z(t)$ is chosen subject to the following inequality constraint:

$$\phi(\bar{y}, z) \leq 0 \tag{22}$$

Introducing the multiplier $\mu(t)$ and treating the product $\mu\phi$ in the same way as the product $\lambda_i(\dot{y}_i - f_i)$, we change Equation (10) to

$$F_1(t, \bar{y}, \dot{\bar{y}}, \bar{\lambda}, \mu, z) = \mu\phi(\bar{y}, z) + \sum_1^{n+q} \lambda_i (\dot{y}_i - f_i) \tag{23}$$

Equation (23) is defined only when the constraint is violated. The Euler-Lagrange equations remain the same except that F is replaced by F_1. In terms of F, Equations (12) and (13) become

$$\frac{d}{dt}\frac{\partial F}{\partial \dot{y}_i} = \frac{\partial F}{\partial y_i} + \mu \frac{\partial \phi}{\partial y_i}, \quad i = 1, \ldots, n+q \tag{24}$$

$$\frac{\partial F}{\partial z} + \mu \frac{\partial \phi}{\partial z} = 0 \qquad (25)$$

When the constraint is not violated, $\mu = 0$ and Equations (24) and (25) are reduced to Equations (12) and (13). However, when the constraint is violated, the control variable z is determined by the equality in Equation (22) and the extra Equation (25) is used to determine the newly introduced variable μ. The transversality conditions remain unchanged.

The problem with decision variable inequality constraints was first investigated by Valentine (*8, 10*). Obviously the results can be extended easily to several control variables with several inequality constraints. It should be pointed out that in order for the method to be valid, the control variable must appear in the inequality constraint. Inequalities involving only state variables are more difficult because of the basic difference between state and control variables. The control variable is, in a sense, more independent and is not subject to differential equation constraints. State variables are not independent. A trial and error or iterative procedure is needed when the inequality constraint involves state variables only.

Note that if the constraint involves the control variable only, then $\partial\phi/\partial y_i = 0$ and Equation (24) is reduced to Equation (12). Thus, if only the control variable is in the constraint equations, such as $\phi = z - z_0 \leq 0$, the numerical solution procedure is the same as that of the unconstrained case, except that whenever the constraint is violated, the control variable is obtained from the equality in the constraint equation. Since the independent variable t can always be treated as a state variable by establishing

$$\dot{x}_{n+1} = 1 \qquad (26)$$

the above equation can also be used when the constraint equation involves the variable t explicitly.

Maximum Principle

The maximum principle is a very powerful tool for obtaining analytical solutions of linear optimization problems with control variable inequality constraints. However, when the problem is nonlinear and an analytical solution cannot be obtained, the maximum principle has the same boundary value difficulties as the calculus of variations in obtaining numerical answers. It can be shown that the unbounded maximum principle is equivalent to the Weierstrass necessary condition in the classical calculus of variations (*8*). An unbounded control variable belongs to an open set or is subject to no inequality constraints. With the development of the calculus of variations to include control variable inequality constraints, the advantages of the maximum principle over the calculus of variations are almost completely lost when analytical solution cannot be obtained. In order to preserve space and maintain generality and consistency with the previous discussions, the maximum principle will be obtained, formally, from the Weierstrass necessary condition. Another advantage of this approach is that the results show clearly the similarity between the maximum principle and the calculus of variations with bounded control variables.

Let us consider the same problem represented by Equations (1) to (7) and add the following inequality constraint on the control variable.

$$\phi(z) \leq 0 \qquad (27)$$

The Weierstrass necessary condition is (*2*):

$$E \equiv F_1(t, \bar{y}, \bar{Y}, Z, \bar{\lambda}, \mu) - F_1(t, \bar{y}, \bar{\dot{y}}, z, \bar{\lambda}, \mu) - \sum_{i=1}^{n+q} (\dot{Y}_i - \dot{y}_i) \frac{\partial F_1}{\partial \dot{y}_i} \geq 0 \qquad (28)$$

where

$$F_1 = \sum_{i=1}^{n+q} \lambda_i (\dot{y}_i - f_i(t, z, \bar{y})) + \mu\phi(z) \qquad (29)$$

and λ_i and μ have the same meaning as before. The $\bar{Y}$ and Z are nonoptimal but permissible values of $\bar{y}$ and z. In other words, the $\bar{Y}$ and Z represent perturbations from the optimum. The E is known as the Weierstrass excess function or E function. The Weierstrass necessary condition guarantees a local minimum, while the Euler-Lagrange equation gives an extremal which may be a minimum, maximum, or even a saddle point. If Equations (4), (27), and (29) are substituted into Equation (28), the Weierstrass condition for the set of side conditions considered becomes

$$\sum_{i=1}^{n+q} \lambda_i f_i(t, \bar{y}, z) \geq \sum_{i=1}^{n+q} \lambda_i f_i(t, \bar{y}, Z) \qquad (30)$$

which is clearly equivalent to

$$\max_z H(t, z, \bar{y}, \bar{\lambda}) = \max_z \sum_{i=1}^{n+q} \lambda_i f_i(t, \bar{y}, z) \qquad (31)$$

Equation (31) is generally known as the maximum principle. The function H is known as the Hamiltonian function.

It has been shown previously that the addition of inequality constraints involving the control variable does not change Equation (12) or (17). Furthermore, it can be shown that

$$\frac{dy_i}{dt} = \frac{\partial H}{\partial \lambda_i}, \quad \frac{d\lambda_i}{dt} = -\frac{\partial H}{\partial \lambda_i}, \quad i = 1, \ldots, n+q \qquad (32)$$

The transversality conditions remain unchanged. Our problem is now reduced to the solution of Equation (32) with boundary conditions, Equation (14). At the same time, Equation (31) is maximized subject to the inequality constraint Equation (27).

Notice that Equation (13) is replaced by Equation (31) for the maximum principle approach. Equation (31) is a much stronger necessary condition than Equation (13). Equation (13) can generally be obtained by differentiating Equation (31) with respect to the control variable for most engineering problems. It can be shown easily that if we minimize

$$J = \sum_{i=1}^{n+q} c_i y_i(t_2) \qquad (33)$$

with known initial conditions

$$y_i(t_1) = y_i^o, \quad i = 1, \ldots, n \qquad (34)$$

subject to the right end constraints

$$\psi_j(\bar{y}(t_2)) = 0, \quad j = 1, \ldots, p \leq n \qquad (35)$$

The transversality conditions Equation (14) is reduced to

$$\lambda_i(t_2) = -\left[c_i + \sum_1^p \nu_j \frac{\partial \psi_j}{\partial y_i(t_2)}\right], \quad i = 1, \ldots, n+q \qquad (36)$$

Equations (31) and (32) remain the same. Equations (31) to (36), (27), (4), and (7) are in the form most frequently encountered in chemical engineering applications.

OPTIMUM TEMPERATURE GRADIENT WITH PRESSURE AS PARAMETER

To illustrate the technique outlined above for treating parameters and also to show the effectiveness of the

quasilinearization technique in obtaining numerical results for boundary value problems, the optimum temperature gradient in a tubular reactor with pressure as the parameter is obtained. The following gaseous reactions are considered

$$A \rightarrow 2B \rightarrow C \tag{37}$$

where B is the desired product. The first reaction is first order, and the desired product is transformed into C by a second-order reaction. This problem has been discussed in previous papers for the case of constant temperature (6, 7). If the reaction mixture is assumed to be ideal gases and Dalton's law is obeyed, the following equations can be obtained:

$$\frac{dx}{dt} = -2K_1P\frac{x}{A+y} \tag{38}$$

$$\frac{dy}{dt} = 4K_1P\frac{x}{A+y} - 4K_2P^2\frac{y^2}{(A+y)^2} \tag{39}$$

where x and y are the concentrations of A and B, respectively, t is the length parameter and is equal to the length divided by the mass flow rate for a reactor with unit cross-sectional area, P is the total pressure, and

$$A = 2x^o + y^o \tag{40}$$

where x^o and y^o are the known initial concentrations of x and y, respectively. In obtaining Equations (38) and (39), it has been assumed that only components A and B are present at the entrance of the reactor. The reaction rate constants K_1 and K_2 represent the first and second reactions, respectively, and

$$K_1 = K_{1o}\exp(-E_1/RT), \quad K_2 = K_{2o}\exp(-E_2/RT) \tag{41}$$

Suppose, by actual experiments, it has been found that the cost of providing a certain initial pressure $P(t_1)$ is a function of this initial pressure and can be expressed as

$$b_3\exp(P_a - P(t_1))^2, \quad P(t_1) \leq P_a \tag{42}$$

The minimum cost b_3 is obtained when $P(t_1) = P_a$. The symbol P_a can be considered as the atmospheric pressure plus pressure drop through the reactor. For the sake of simplicity, we shall neglect any other operating costs. Our problem is to find a function $T(t)$ and a constant $P(t_1)$ such that the functions $x(t)$ and $y(t)$ given by Equations (38) and (39) maximize the following expression

$$J = b_1x(t_2) + b_2y(t_2) - b_3\exp(P_a - P(t_1))^2 \tag{43}$$

The constants b_1 and b_2 can be considered as the cost of x and y, respectively, and b_3 is the minimum cost of providing the pressure to produce $(b_1 + b_2)$ dollars worth of x and y.

In the following, this problem will be solved by the equations outlined in the previous section with the aid of the quasilinearization technique.

Calculus of Variations with Pressure as a Parameter

In this section, the above problem will be solved by the calculus of variations with pressure as a constant parameter. Let

$$\dot{P}(t) = 0 \tag{44}$$

The only end conditions are the given initial conditions for x and y:

$$x(t_1) = x^o, \quad y(t_1) = y^o \tag{45}$$

Equations (10) and (11) become

$$F = \lambda\left(\dot{x} + 2K_1P\frac{x}{A+y}\right) + \mu\left(\dot{y} - 4K_1P\frac{x}{A+y} + 4K_2P^2\frac{y^2}{(A+y)^2}\right) + \theta(\dot{P}) \tag{46}$$

$$G = b_1x(t_2) + b_2y(t_2) - b_3\exp(P_a - P(t_1))^2 + \nu_1(x(t_1) - x^o) + \nu_2(y(t_1) - y^o) \tag{47}$$

Note that x, y, P, λ, μ, θ, and T were represented by x_1, x_2, a_1, λ_1, λ_2, λ_3, and z, respectively, in Equations (1) to (11).

The Euler-Lagrange equations can be obtained by Equations (12) and (13):

$$\dot{\lambda} = 2K_1P\frac{\lambda}{A+y} - 4K_1P\frac{\mu}{A+y} \tag{48}$$

$$\dot{\mu} = -2K_1P\frac{\lambda x}{(A+y)^2} + 4K_1P\frac{\mu x}{(A+y)^2} + 8K_2P^2\frac{\mu y}{(A+y)^2} - 8K_2P^2\frac{\mu y^2}{(A+y)^3} \tag{49}$$

$$\dot{\theta} = 2K_1\frac{\lambda x}{A+y} + 8K_2P\frac{\mu y^2}{(A+y)^2} - 4K_1\frac{\mu x}{A+y} \tag{50}$$

$$K_1E_1x(\lambda - 2\mu) + 2K_2E_2P\frac{\mu y^2}{A+y} = 0 \tag{51}$$

The unknown boundary conditions can be obtained by the use of the transversality condition. Since t_1 and t_2 are fixed, Equation (15) no longer holds. The initial values of x and y are also fixed. By the use of Equation (16), the unknown end conditions are

$$\text{at } t_1: \quad \theta(t_1) = 2b_3\{P_a - P(t_1)\}\exp\{P_a - P(t_1)\}^2 \tag{52}$$

$$\text{at } t_2: \quad \lambda(t_2) = -b_1, \quad \mu(t_2) = -b_2, \quad \theta(t_2) = 0 \tag{53}$$

Our problem is to find the seven unknowns, x, y, P, λ, μ, θ, and T by solving the seven equations represented by Equations (38), (39), (44), and (48) to (51). Six of these equations are differential equations with six boundary conditions represented by Equations (45), (52), and (53). Notice that they are nonlinear differential equations with two-point boundary conditions. In order to obtain numerical results, these equations first will be linearized by the quasilinearization technique (6). Consider the following system of nonlinear differential equations:

$$\frac{du_i}{dt} = f_i(u_1, \ldots, u_n, t), \quad i = 1, \ldots, n \tag{54}$$

with appropriate boundary conditions. Equation (54) can be linearized by the following vector equation:

$$\frac{du_{k+1}}{dt} = f(u_{k+1}, t) = f(u_k, t) + J(u_k)(u_{k+1} - u_k) \tag{55}$$

where u_{k+1}, u_k, and f are in vector form and represent the vectors $u_{1,k+1}, \ldots, u_{n,k+1}$; $u_{1,k}, \ldots, u_{n,k}$; and $f_1, \ldots, f_n$, respectively. The Jacobi matrix $J(u_k)$ is

$$J(u_k) = \begin{bmatrix} \frac{\partial f_1}{\partial u_{1,k}}, & \frac{\partial f_1}{\partial u_{2,k}}, & \ldots, & \frac{\partial f_1}{\partial u_{n,k}} \\ \cdots & \cdots & \cdots & \cdots \\ \frac{\partial f_n}{\partial u_{1,k}}, & \frac{\partial f_n}{\partial u_{2,k}}, & \ldots, & \frac{\partial f_n}{\partial u_{n,k}} \end{bmatrix} \tag{56}$$

If we assume u_k is the known value and is obtained from previous calculations and u_{k+1} is unknown, the right-hand side of Equation (55) will always be linear. By the use of

Equations (55) and (56), the following sequence of linear differential equations can be obtained from Equations (38), (39), and (48) to (50):

$$\frac{dx_{k+1}}{dt} = 2K_1 \frac{1}{A+y}\left(Px - P\frac{xy}{A+y} - Px_{k+1} + P\frac{x}{A+y}y_{k+1} - xP_{k+1}\right) \quad (57)$$

$$\frac{dy_{k+1}}{dt} = 4/(A+y)\left[P\left(3K_2P\frac{y^2}{A+y} + K_1\frac{xy}{A+y} - K_1x - 2K_2P\frac{y^3}{(A+y)^2}\right) + PK_1x_{k+1} + \frac{P}{A+y}\left(-K_1x + 2K_2P\frac{y^2}{A+y} - 2K_2Py\right)y_{k+1} + \left(K_1x - 2K_2P\frac{y^2}{A+y}\right)P_{k+1}\right] \quad (58)$$

$$\frac{d\lambda_{k+1}}{dt} = 2K_1\frac{1}{A+y}\left[P\left(-\lambda + \frac{\lambda y}{A+y} + 2\mu - 2\frac{\mu y}{A+y}\right) + \frac{P}{A+y}(-\lambda+2\mu)y_{k+1} + (\lambda-2\mu)P_{k+1} + P\lambda_{k+1} - 2P\mu_{k+1}\right] \quad (59)$$

$$\frac{d\mu_{k+1}}{dt} = \frac{2}{(A+y)^2}\left[2P\left(K_1\lambda x - 2K_1\mu x - 6K_2P\mu y + 12K_2P\frac{\mu y^2}{A+y} - K_1\frac{\lambda xy}{A+y} + 2K_1\frac{\mu xy}{A+y} - 6K_2P\frac{\mu y^3}{(A+y)^2}\right) + K_1P(-\lambda+2\mu)x_{k+1} + 2P\left(K_1\frac{\lambda x}{A+y} - 2K_1\frac{\mu x}{A+y} + 2K_2P\mu - 8K_2P\frac{\mu y}{A+y} + 6K_2P\frac{\mu y^2}{(A+y)^2}\right)y_{k+1} + \left(-K_1\lambda x + 2K_1\mu x + 8K_2P\mu y - 8K_2P\frac{\mu y^2}{A+y}\right)P_{k+1} - K_1Px\lambda_{k+1} + 2P\left(K_1x + 2K_2Py - 2K_2P\frac{y^2}{A+y}\right)\mu_{k+1}\right] \quad (60)$$

$$\frac{d\theta_{k+1}}{dt} = \frac{2}{A+y}\left[\left(-K_1\lambda x + 2K_1\mu x + K_1\frac{\lambda xy}{A+y} - 2K_1\frac{\mu xy}{A+y} - 12K_2P\frac{\mu y^2}{A+y} + 8K_2P\frac{\mu y^3}{(A+y)^2}\right) + K_1(\lambda-2\mu)x_{k+1} + \frac{1}{A+y}\left(-K_1\lambda x + 2K_1\mu x + 8K_2P\mu y - 8K_2P\frac{\mu y^2}{A+y}\right)y_{k+1} + 4K_2\frac{\mu y^2}{A+y}P_{k+1} + K_1x\lambda_{k+1} + 2\left(-K_1x + 2K_2P\frac{y^2}{A+y}\right)\mu_{k+1}\right] \quad (61)$$

and Equation (44) becomes

$$\frac{dP_{k+1}}{dt} = 0 \quad (62)$$

The subscript k which appears in all the unsubscripted variables x, y, P, λ, μ, and θ has been omitted in the above equations for simplicity. It is understood that the unknown variables are the variables with subscript $k+1$. All other variables are known and are calculated from the previous k^{th} iteration.

The temperature $T(t)$ can be obtained from Equation (51):

$$1/T = \frac{R}{E_1 - E_2}\left[\ln\{K_{1o}E_1x(\lambda-2\mu)\} + \ln\left(2K_{2o}E_2P\frac{\mu y^2}{A+y}\right)\right] \quad (63)$$

Since Equations (57) to (62) are linear equations, their general solution must consist of one particular solution and six homogeneous solutions:

$$\begin{aligned} x(t) &= x_p(t) + d_1x_{h1}(t) + d_2x_{h2}(t) + d_3x_{h3}(t) + d_4x_{h4}(t) + d_5x_{h5}(t) + d_6x_{h6}(t), \\ y(t) &= y_p(t) + d_1y_{h1}(t) + d_2y_{h2}(t) + d_3y_{h3}(t) + d_4y_{h4}(t) + d_5y_{h5}(t) + d_6y_{h6}(t), \\ &\text{etc.,} \qquad t_1 \leq t \leq t_2 \end{aligned} \quad (64)$$

There are similar equations for the state variable $P(t)$ and multipliers $\lambda(t)$, $\mu(t)$, and $\theta(t)$. The subscript p indicates particular solution and the subscripts $h1, \ldots, h6$ indicate homogeneous solutions. The symbols d_1 to d_6 are integration constants and are to be determined from the boundary conditions. Any seven sets of arbitrary initial conditions can be used to obtain the particular and homogeneous solutions provided they are independent and the homogeneous solutions obtained are nontrivial. The initial conditions used for obtaining these seven numerical solutions are listed in Table 1. They are chosen with the two known initial conditions in mind. It can be shown easily from the two known initial conditions and Equation (64) that the values in Table 1 make $d_5 = d_6 = 0$. Thus only four homogeneous solutions are needed. The four integration constants, d_1 to d_4, can be obtained by substituting the four boundary conditions, Equations (52) and (53), and the values in Table 1 into Equation (64). After some rearrangements, the following expressions can be obtained:

$$d_4 = \frac{v_2(v_1 - v_3) + v_4(v_5 - v_6) - (v_7v_2 - v_8v_4)\,2b_3(P_a - d_4 - 1)\exp(P_a - d_4 - 1)^2}{v_9v_2 - v_{10}v_4} \quad (65)$$

$$d_1 = 2b_3(P_a - d_4 - 1)\exp(P_a - d_4 - 1)^2 \quad (66)$$

$$d_3 = (v_6 - v_5 - v_8d_1 - v_{10}d_4)/v_2 \quad (67)$$

$$d_2 = (\lambda(t_2) - \lambda_p(t_2) - \lambda_{h1}(t_2)d_1 - \lambda_{h3}(t_2)d_3 - \lambda_{h4}(t_2)d_4)/\lambda_{h2}(t_2) \quad (68)$$

where

$$
\begin{aligned}
v_1 &= \lambda(t_2)\mu_{h2}(t_2) - \mu(t_2)\lambda_{h2}(t_2),\\
v_2 &= \mu_{h3}(t_2)\theta_{h2}(t_2) - \theta_{h3}(t_2)\mu_{h2}(t_2),\\
v_3 &= \lambda_p(t_2)\mu_{h2}(t_2) - \mu_p(t_2)\lambda_{h2}(t_2),\\
v_4 &= \lambda_{h3}(t_2)\ \mu_{h2}(t_2) - \mu_{h3}(t_2)\lambda_{h2}(t_2),\\
v_5 &= \mu_p(t_2)\theta_{h2}(t_2) - \theta_p(t_2)\mu_{h2}(t_2),\\
v_6 &= \mu(t_2)\theta_{h2}(t_2) - \theta(t_2)\mu_{h2}(t_2),\\
v_7 &= \lambda_{h1}(t_2)\mu_{h2}(t_2) - \mu_{h1}(t_2)\lambda_{h2}(t_2),\\
v_8 &= \mu_{h1}(t_2)\theta_{h2}(t_2) - \theta_{h1}(t_2)\mu_{h2}(t_2),\\
v_9 &= \lambda_{h4}(t_2)\mu_{h2}(t_2) - \mu_{h4}(t_2)\lambda_{h2}(t_2),\\
v_{10} &= \mu_{h4}(t_2)\theta_{h2}(t_2) - \theta_{h4}(t_2)\mu_{h2}(t_2),
\end{aligned}
\tag{69}
$$

where the v's are known functions and they are used only for the simplicity in representation. The only unknowns in the above equations are the integration constants d_1 to d_4. Since d_4 is implicit in Equation (65), some trial and error procedure must be used. To ensure convergence, the method of successive substitutions or simple iterations and the Newton-Raphson method are used alternatively to solve Equation (65) (5). In general, five iterations by the Newton-Raphson method are allowed first. If the desired convergence is not obtained, five iterations by the method of successive substitutions are allowed next. The two methods are alternately used in this manner until the following desired accuracy in d_4 is obtained:

$$|d_4(m+1) - d_4(m)| < 0.1 \times 10^{-4} \tag{70}$$

where m indicates the number of iterations. With an initially assumed value of $d_4(m=0) = 0.1$, it has been found that if the first method, say, successive substitution didn't converge in five iterations, the second method will converge to the desired accuracy in about two to three iterations for the results discussed in this work.

Ideally, the control variable T should be solved and expressed explicitly as in Equation (63). However, for many practical problems, the control variable is in an implicit form in Equation (13) and cannot be cancelled from the differential equations. To test the effectiveness of the method in the implicit case, the above set of equations is solved in the following two ways:

1. (a) A reasonable set of numerical functions is assumed:

$$
\begin{aligned}
&x_k(t) = x_o(t), \quad y_k(t) = y_o(t), \quad P_k(t) = P_o(t),\\
&\lambda_k(t) = \lambda_o(t), \quad \mu_k(t) = \mu_o(t), \quad \theta_k(t) = \theta_o(t),\\
&\qquad t_1 \leq t \leq t_2
\end{aligned}
\tag{71}
$$

(b) The control variable $T_k(t) = T_o(t)$ is obtained from Equation (63) with the initial approximation, Equation (71).

(c) The values for the $(k+1)^{st}$ iteration x_{k+1}, y_{k+1}, P_{k+1}, λ_{k+1}, μ_{k+1}, and θ_{k+1} are obtained from the known k^{th} approximation as follows. A particular solution and four homogeneous solutions are first obtained with Equations (57) to (62) with the initial conditions listed in Table 1. The integration constants d_1 to d_4 are then obtained by the use of Equations (65) to (69). Finally these results are combined by the use of Equation (64).

Table 1. Initial Conditions Used for Obtaining Homogeneous and Particular Solutions in the Numerical Example

	Particular solution	Homogeneous solutions					
		1	2	3	4	5	6
$x(t_1)$	x^o	0.0	0.0	0.0	0.0	0.0	1.0
$y(t_1)$	y^o	0.0	0.0	0.0	0.0	1.0	0.0
$P(t_1)$	1.0	0.0	0.0	0.0	1.0	0.0	0.0
$\lambda(t_1)$	1.0	0.0	0.0	1.0	0.0	0.0	0.0
$\mu(t_1)$	1.0	0.0	1.0	0.0	0.0	0.0	0.0
$\theta(t_1)$	0.0	1.0	0.0	0.0	0.0	0.0	0.0

(d) The values for $T_{k+1}(t)$ are obtained by the combined use of Equation (63) and the results obtained in c.

(e) Steps c and d are repeated until the following required accuracies are obtained:

$$
\begin{aligned}
&|T_{k+1}(t) - T_k(t)| < \epsilon_T, \quad t_1 \leq t \leq t_2,\\
&|x_{k+1}(t) - x_k(t)| < \epsilon_x, \quad t_1 \leq t \leq t_2,\\
&\text{etc.,}
\end{aligned}
\tag{72}
$$

Similar criteria are used for y, P, λ, μ, and θ.

2. (a) Same as a in 1.

(b) A reasonable set of numerical values is assumed arbitrarily for the control variable $T_k(t) = T_o(t)$.

(c) Same as c in 1.

(d) Step c is repeated until the required accuracy for the state variables and multipliers is obtained with fixed value $T_k(t)$. The required accuracy is defined by Equation (72).

(e) An improved $T_{k+n}(t)$ is obtained from Equation (51) by a trial and error procedure. The subscript n represents the number of iterations carried out in steps c and d.

(f) Steps c to e are repeated until the required accuracy as defined by Equation (72) is obtained.

Notice that method 2 is used only when T cannot be solved explicitly. In this case, an improved T is obtained only when no more improvement for the state variables and Lagrange multipliers can be obtained.

The Runge-Kutta integration scheme is used in obtaining the particular and homogeneous solutions from Equations (57) to (62) in step c. The numerical values used are

$$
\begin{aligned}
&K_{1o} = 0.2 \times 10^9 \text{ g.-moles/(liter)(min.)(atm.)}\\
&K_{2o} = 0.63 \times 10^{16} \text{ g.-moles/(liter)(min.)(atm.)}^2\\
&E_1 = 0.18 \times 10^5 \text{ cal./mole}\\
&E_2 = 0.3 \times 10^5 \text{ cal./mole}\\
&R = 2 \text{ cal./(mole)(°K.)}\\
&x^o = 0.01 \text{ g.-moles/g.}\\
&t_1 = 0.0\\
&t_2 = 8.0 \text{ (liter)(min.)/g.}\\
&\Delta t = 0.025, \quad 0 \leq t \leq 1.0\\
&\Delta t = 0.1, \quad 1.0 < t \leq 8.0\\
&b_1 = 0.1\\
&b_2 = 1.0\\
&b_3 = 0.001\\
&P_a = 1.5
\end{aligned}
\tag{73}
$$

where Δt is the integration step size used for the Runge-Kutta integration scheme. Except for P, the boundary values are assumed arbitrarily as the numerical functions for the initial approximation in Equation (71):

$$
\begin{aligned}
&x_o(t) = x^o = 0.01, \quad y_o(t) = y^o, \quad \lambda_o(t) = \lambda(t_2) = -b_1,\\
&\mu_o(t) = \mu(t_2) = -b_2, \quad \theta_o(t) = \theta(t_2) = 0,\\
&\qquad P_o(t) = 1.0, \quad t_1 \leq t \leq t_2
\end{aligned}
\tag{74}
$$

where the values of $P_o(t)$ are assumed arbitrarily. An initial approximation for the control variable $T_k(t) = T_o(t)$, $t_1 \leq t \leq t_2$, must also be assumed for method 2. Although these values could have been estimated for the present problem from Equation (63), in order to test the method this has not been used. Instead, a constant value was assumed for $T_o(t)$, $t_1 \leq t \leq t_2$. For most of this work, this constant value has been set at 350°K. No convergence problem has been encountered. However, since both Equa-

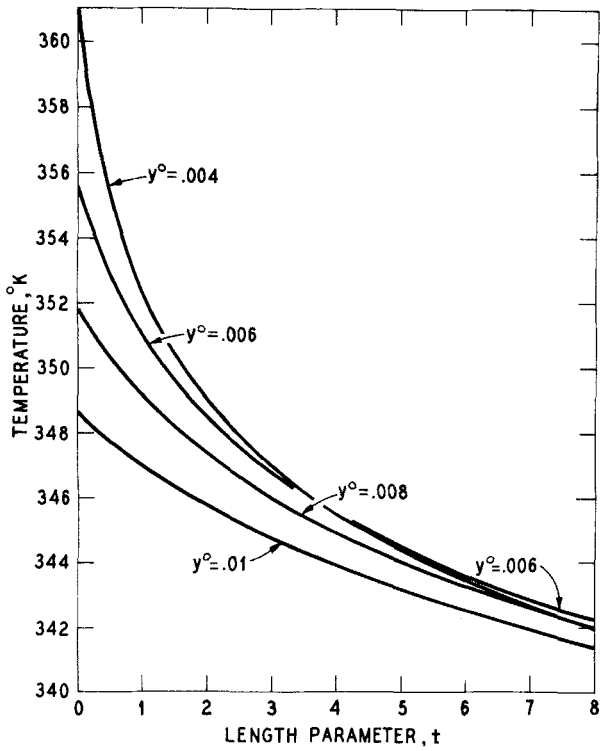

Fig. 1. Optimum temperature profiles.

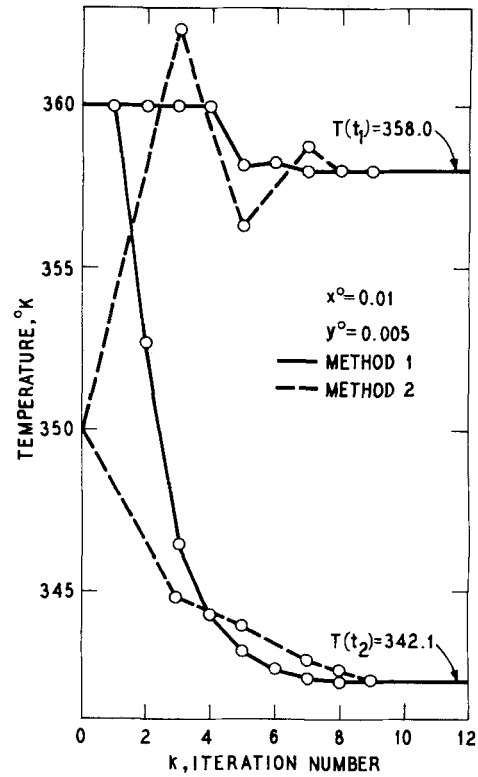

Fig. 3. Typical rates of convergence at t_1 and t_2.

tion (74) and $T_o(t)$ are assumed and far from the actual values, a faster convergence rate can be obtained if the values of $T_k(t)$ are restricted to a certain range for the first few iterations. Generally, this range is set at 20 to 30 deg. and $T_o(t)$ is assumed at the middle of this range.

The problem is solved by using both methods 1 and 2 with an IBM 7094 computer. Some of the results are shown in Figures 1 and 2. For $y^o = 0.01$, 0.008, 0.006, and 0.004, the maximum profit functions are $J = 0.013326$, 0.012085, 0.010965, and 0.009960, respectively, and the optimum constant pressures are $P = 1.196$, 1.184, 1.182, and 1.191, respectively. The required accuracy for the control variable $T(t)$ are $\epsilon < 0.01$. A much higher accuracy has been obtained for the state variables and multipliers.

As can be seen from Equation (74), the initial approximations for the state variables and Lagrange multipliers are very poor. These initial approximations can be estimated very easily from the physical problem for any engineering applications. The trial and error procedure usually used for solving boundary value problems has also been used to solve this problem. It has been found that, in order for the problem to converge, the initial approximation has to be nearly the same as the correct solution.

The convergence rates for methods 1 and 2 are compared in Figure 3 for $y^o = 0.005$. All other calculations in this work exhibit a similar convergence behavior. To simplify the illustration, only the convergence rates of the control variable T at the initial time t_1 and the final time t_2 are shown. The assumed initial value for $T_o(t)$ for method 2 is 350°K. An upper constraint of $T_{max} = 360$ is used

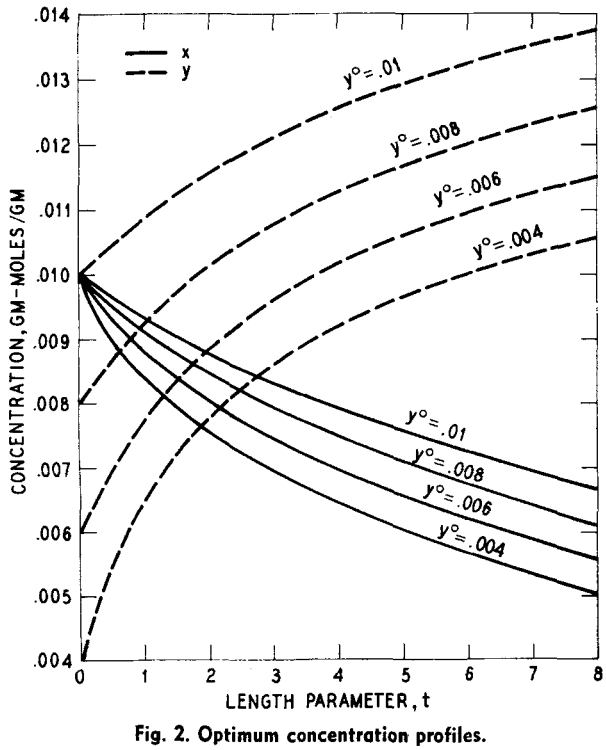

Fig. 2. Optimum concentration profiles.

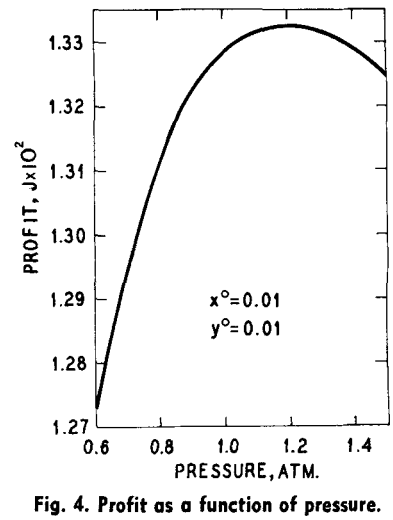

Fig. 4. Profit as a function of pressure.

for method 1. As can be seen from the figure steps c and d of method 2 are repeated three times before the temperature is first evaluated. The accuracy obtained in these three iterations are $\epsilon < 10^{-3}$ for x and y and $\epsilon < 10^{-1}$ for the multipliers. However, only two iterations are needed to obtain the same accuracy for the second and third evaluations of the temperature. Thereafter, one iteration is enough to obtain this accuracy. The accuracy for $T(t)$ obtained in ten iterations is $\epsilon < 0.04$ for method 1 and $\epsilon < 0.1$ for method 2. The accuracies for the state variables and multipliers are much higher. The significant result is that, although $T(t)$ is an unknown variable in method 2, the convergence rate for method 2 is almost as good as that of method 1. Each iteration takes approximately 4 sec. of computation time for both methods.

To prove the fact that the values obtained are indeed optimal, the above problem is solved with known and fixed values of pressure. Our problem now is to find the temperature profile $T(t)$ such that the functions of $x(t)$ and $y(t)$ given by Equations (38) and (39) maximize Equation (43). The same equations and procedures can be used as outlined above except that the differential equations for P and θ are not needed here. Consequently, the boundary conditions are simplified and Equations (52) and (65) to (69) are no longer needed. The results for $y^o = 0.01$ are shown in Figure 4. It can be seen that the optimum is indeed a maximum. An added benefit can be obtained from this calculation. Since p is fixed and known, we only have two state variables. It is shown that the number of iterations required for certain accuracy is approximately the same as that required for three-state variables. This furnishes some evidence that the quasilinearization technique is an effective tool for solving optimization problems with a fairly large number of state variables.

Calculus of Variations with Control Variable Inequality Constraint

Let us add the following inequality constraint to the problem treated above:

$$T(t) \leq 350°\text{K}. \qquad (75)$$

Since this equation does not include the state variables, the equations and procedures remain the same except that Equation (51) or (63) does not always hold. Whenever the temperature is higher than 350°, it is set at this value instead of using Equations (51) or (63). The results for $y^o = 0.005$ are shown in Figure 5. The results without the temperature constraint, as obtained in the previous section, are also shown. The optimum constant pressure and maximum profit are 1.185 and 0.010449, respectively, without the constraint, 1.239 and 0.010431, respectively, with the constraint. Since it is generally impractical to produce the rapid decrease in temperature in the first part of the reactor, these results furnish an interesting comparison.

Calculus of Variations with Pressure Drop in the Reactor

In tubular or packed-bed reactors, pressure drop is always present across the reactor. It is generally impossible to control this pressure drop or pressure profile. However, the initial pressure can always be controlled and optimized. Our problem in this section is to find the optimum initial pressure and optimum temperature profile simultaneously. Let us assume a constant pressure drop:

$$\frac{dP(t)}{dt} = P_d, \quad t_1 \leq t \leq t_2 \qquad (76)$$

Since Equation (76) is equal to a constant, it can be shown that all the other equations remain the same. With $y^o = 0.005$ and $P_d = 0.05$, the problem solved in the previous section is solved here with Equation (76) replacing Equation (62). The results are shown in Figure 6. It is interesting to note that the temperature profile decreases at first and then increases through the last part of the reactor. The maximum profit is $J = 0.0106115$.

Maximum Principle

The same problem which has been treated previously by the calculus of variations is solved by the maximum principle. The Hamiltonian function can be obtained from

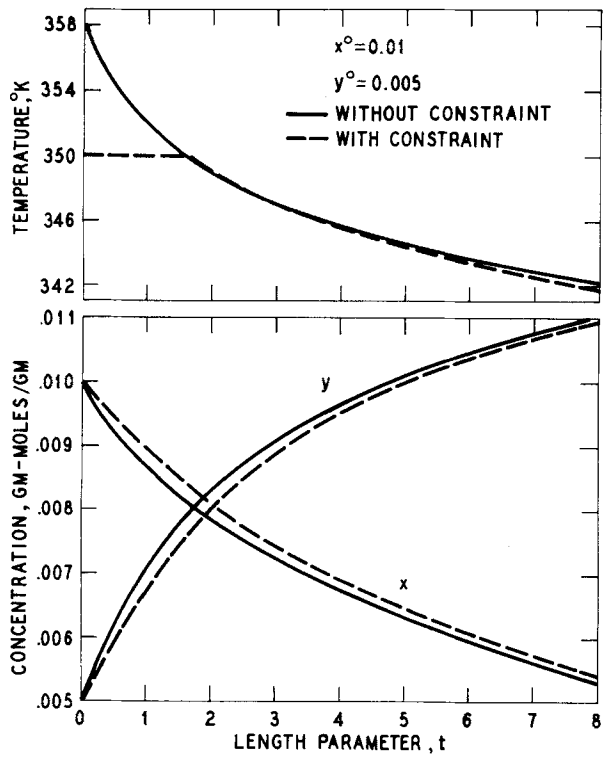

Fig. 5. Influence of constraint on optimum profiles.

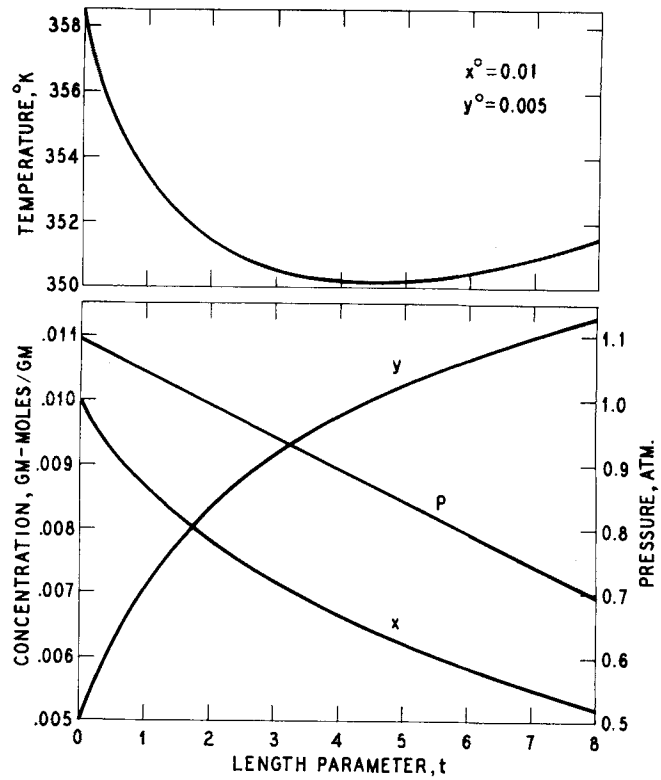

Fig. 6. Influence of pressure drop on optimum profiles.

Equations (31), (38), (39), and (44):

$$H = -2K_1P\lambda\frac{x}{A+y} + 4K_1P\mu\frac{x}{A+y} - 4K_2P^2\mu\frac{y^2}{(A+y)^2} \quad (77)$$

Our problem is to find a constant pressure P and a temperature profile $T(t)$ such that the functions x, y, P, λ, μ, and θ given by Equations (38), (39), (44), (48), (49), and (50) minimize Equation (77) for all t, $t_1 \leq t \leq t_2$. The boundary conditions remain the same. The only difference between the present problem and that given previously is that Equation (51) is replaced by Equation (77). As noted previously, Equation (77) is a much stronger necessary condition than Equation (51).

The problem can be solved by the same procedure as that listed in method 2. However, instead of solving an algebraic equation, we now must find the profile $T(t)$ which minimizes Equation (77) at all time t for $t_1 \leq t \leq t_2$. Thus, only step e in method 2 needs to be modified. Since only one control variable $T(t)$ needs to be searched for in Equation (77), any search procedure can be used to find the minimum of H. Both the Fibonaccian search procedure and the random search technique are used in the present work. The Fibonaccian method has been shown to be the optimal procedure for the search of one variable which does not give relative maximum or minimum (*1*). The random search technique has been discussed and used on the analog computer in a previous paper (*7*). The method is essentially the same as that discussed there. The random number with a Gaussian distribution is generated in the IBM 7094 computer by a random number subroutine.

The results presented in Figures 1 to 3 are also obtained by the maximum principle. Since the procedures and equations are the same as those of method 2 except that the solution of an algebraic equation is replaced by a search routine, the results and convergence rates are completely the same. However, the computation time needed for each iteration is approximately doubled due to the use of the search procedure.

DISCUSSION

The present technique appears to be an effective tool for obtaining numerical solutions of optimization problems. Although only the continuous process is discussed, the method can be extended also to discrete processes with complex structures. The results for discrete processes will be discussed in separate papers.

Obviously, the feed conditions to the reactor, x^o and y^o, can also be optimized in the same manner. Suppose the cost of obtaining a certain composition is $F(x^o, y^o)$ which may be considered as the purification cost. The term $\{-F(x^o, y^o)\}$ must now be added to the function to be maximized, Equation (43). In this way, the operating cost for obtaining a certain feed purity is also optimized simultaneously.

Instead of assuming a constant rate for the pressure drop, the momentum balance and experimental correlations can be used to obtain the actual pressure drop. For packed-bed reactors, it is known that the pressure drop is proportional to the properties of the packing material and the properties of the fluid. Since the fluid properties are functions of its temperature T, its pressure P, and its composition x and y, the pressure drop equation is

$$\frac{dP}{dt} = f(P, T, x, y, \text{packing}) \quad (78)$$

The procedure for finding the optimum with this pressure equation is the same as that discussed in the third section. The exact form of this pressure equation is discussed by Ergun (*4*) and by almost any standard textbook on chemical reactor design.

NOTATION

$a_1, \ldots, a_q$ = constant parameter
A = defined by Equation (40)
b_1, b_2, b_3 = constants as defined by Equation (43)
$d_1, \ldots, d_6$ = integration constants
E = Weierstrass excess function as defined by Equation (28)
E_1, E_2 = activation energies of reactions
F = Lagrange function as defined by Equation (10)
G = function as defined by Equation (11)
H = Hamiltonian function
J = profit function
$J(u_k)$ = Jacobi matrix
K_1, K_2 = reaction rate constants
K_{1o}, K_{2o} = frequency factor constants in Arrhenius equation
P = pressure
P_a = reference pressure
P_d = pressure drop as defined by Equation (76)
R = gas constant
T = temperature
t = independent variable
Δt = integration step size
t_1 = initial value of t
t_2 = final or terminal value of t
$v_1, \ldots, v_{10}$ = defined by Equation (69)
x, y = state variables
z = control variable
ψ = end conditions as defined by Equation (5)
ν = constant multipliers
ϕ = inequality constraint function
λ, μ, θ = Lagrange multipliers
ϵ_T, ϵ_x = required accuracies as defined by Equation (72)

Subscripts

k = k^{th} iteration
p = particular solution
h_1, h_2, etc. = homogeneous solutions

Superscripts

o = initial condition

LITERATURE CITED

1. Bellman, R., "Dynamic Programming," Princeton Univ. Press, N. J. (1957).
2. Bliss, G. A., "Lectures on the Calculus of Variations," Univ. Chicago Press, Ill. (1946).
3. Courant, R., and D. Hilbert, "Methods of Mathematical Physics," Interscience, New York (1953).
4. Ergun, S., *Chem. Eng. Progr.*, **48**, 89 (1952).
5. Hildebrand, F. B., "Introduction to Numerical Analysis," McGraw-Hill, New York (1956).
6. Lee, E. S., *Chem. Eng. Sci.*, **21**, 183 (1966).
7. ———, *AIChE J.*, **10**, 309 (1964).
8. Leitmann, G., ed., "Optimization Techniques," Academic Press, New York (1962).
9. Pontryagin, L. S., V. G. Boltyanskii, R. V. Gamkrelidge, and E. F. Mishchenko, "The Mathematical Theory of Optimal Processes," Wiley, New York (1962).
10. Valentine, F. A., in "Contributions to the Calculus of Variations, 1933-1937," Univ. Chicago Press, Ill. (1937).
11. Lee, E. S., "Quasilinearization and Invariant Imbedding: With Applications to Chemical Engineering and Adaptive Control," Academic Press, New York, in press.

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