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Quasilinearization, Difference Approximation, and Nonlinear Boundary Value Problems

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A finite difference method combined with the quasilinearization technique is used to solve the nonlinear two-point boundary value problems. This method does not have the stability problem connected with the marching integration techniques. A scheme which can be used to reduce the rapid access memory requirements of digital computers is also proposed. The steady state equations resulting from mass and energy balances in a tubular reactor with axial diffusion are solved by this method. With very poor initial approximations, only three to seven iterations are needed to obtain the correct answer.

The two most frequently used methods for solving nonlinear two-point boundary value problems in ordinary differential equations are the trial and error method and the finite difference method. In the trial and error method, the values of the unknown initial conditions are assumed and are used with the given initial conditions so that the problem becomes an initial value problem. Thus, the generally used integration techniques, such as the Runge-Kutta method, can be used to obtain the solution. The final values that are obtained must agree with the given final conditions. Otherwise, some trial and error, or iterative procedure, must be devised to obtain better values for the assumed initial conditions. There are at least two difficulties associated with this procedure. The first one arises from the trial and error nature which is not suited for modern digital computers. Furthermore, actual experience has shown that the nonlinear boundary value problems encountered in engineering are frequently very sensitive to the errors of the unknown or assumed initial conditions. This difficulty becomes more severe if the problem to be solved has a large number of missing initial conditions.

The second difficulty arises from the stability problem associated with the generally used integration techniques such as the Runge-Kutta method, which is essentially a marching integration technique. To avoid this stability problem the finite difference method is used.

In the finite difference method, the original system of differential equations is approximated by a system of difference equations. There are several difficulties associated with this approach. The most important ones are the limited rapid access memory of current digital computers and the difficulty associated with the solution of a large system of nonlinear difference, or algebraic equations, that result from the original nonlinear differential equations.

In a recent paper [*1*], the quasilinearization technique was shown to be an effective tool to avoid the first difficulty associated with the trial and error procedure. This was accomplished by the combined use of linearization and the superposition principle. However, since the marching integration techniques were used in the above mentioned

paper, we still had the stability problem. The purpose of the present paper is to show that the quasilinearization technique is equally effective when the problem has the stability difficulties and the finite difference method is used. The nonlinearity difficulties associated with the finite difference method can be overcome easily by the quasilinearization technique. Furthermore, a decoupling scheme which can be used to avoid the necessity of solving the equations simultaneously are suggested. This scheme is shown to be an efficient method for solving multivariable problems.

The computational method is first introduced by solving the second-order equations resulting from a tubular reactor with axial mixing under both isothermal and adiabatic conditions. The parameters used for these problems are shown to be unstable if the marching integration technique is used. Some generalizations of the method to a system of first-order and a system of second-order simultaneous equations are then discussed.

ISOTHERMAL TUBULAR REACTOR WITH AXIAL DIFFUSION

To illustrate the difficulties associated with the marching integration techniques and to introduce the present approach, the equation describing an isothermal packed bed reactor will be considered first. This equation has been solved in a previous paper [1] with the parameter values corresponding to the conditions of an empty tubular reactor. We shall see that for a packed bed reactor the quasilinearization approach using the marching integration technique is unstable.

Consider the chemical reaction

$$A + A \rightarrow B \tag{1}$$

which is taking place in a packed bed reactor. If we assume that the packing has no influence on the reaction except its contribution to the axial mixing, the following equation can be easily established:

$$\frac{1}{N_{Pe}}\frac{d^2u}{dt^2} - \frac{du}{dt} - Ru^2 = 0 \tag{2}$$

where N_{Pe} is the Peclet group (VD_p/D), R is the reaction rate group (kD_p/V), u is the concentration of reactant A, and t is the dimensionless reactor length variable. The boundary conditions are

$$u_e = u(0) - \frac{1}{N_{Pe}}\frac{du(0)}{dt}, \qquad \text{at } t = 0 \tag{3a}$$

$$\frac{du(t_f)}{dt} = 0, \qquad \text{at } t = t_f \tag{3b}$$

where t_f is the dimensionless length (L/D_p) of the reactor and u_e represents the concentration of A before entering the reactor.

The last term of Equation (2) can be linearized by using the form

$$f(u) = f(v) + (u - v)\, f'(v) \tag{4}$$

which is obtained from Taylor series with second and higher orders neglected.

It should be noted that Equation (4) is essentially the Newton-Raphson equation used in solving algebraic equations. Just as the Newton-Raphson method, this iteration is a second-order process. If it converges, it usually converges quadratically in the sense that the error of any iteration is approximately the square of the error of the preceding iteration. More discussion of this method can be found in the literature [1, 3].

If we let $u = u_{k+1}$ and $v = u_k$, Equation (4) becomes.

$$f(u_{k+1}) = f(u_k) + (u_{k+1} - u_k)\, f'(u_k) \tag{5}$$

where the subscript k represents the kth iteration. Applying Equation (5) to the last term of Equation (2), we have

$$\frac{1}{N_{Pe}}\frac{d^2u_{k+1}}{dt^2} - \frac{du_{k+1}}{dt} - 2Ru_ku_{k+1} + Ru_k^2 = 0 \tag{6}$$

With assumed values for $u_{k=0}(t)$, $u_{k=1}(t)$ can be obtained by solving Equation (6) with the boundary conditions, Equation (3). Next, the newly obtained values of $u_1(t)$ are substituted into Equation (6) and an improved $u_2(t)$ is obtained. This iterative process is continued until the required accuracy is obtained.

If we assume that the reactor length is 48 catalyst particle diameters, the following numerical values can be used:

$$t_f = 48,\ u_e = 1,\ N_{Pe} = 2,\ R = 0.04 \tag{7}$$

With $u_{k=0}(t) = 1$ as the initial approximation and an integration interval or step size $\Delta t = 0.1$, it has been found that this problem cannot be solved by the numerical procedure listed in the above mentioned paper [1] where the superposition principle combined with the Runge-Kutta integration method was used. The numerical values of the particular and homogeneous solutions increase or decrease very rapidly as t increases during the first few iterations. Extremely large or small values have been obtained for these solutions at $t = t_f$. A reduction in step size does not help the situation.

In order to avoid these stability difficulties the finite difference method is used. Suppose now the bed length t_f is divided into N equal increments, let $u(n)$ denote the values of u at position $n\Delta t$, then the derivatives in Equation (6) can be replaced by the following difference quotients:

$$\frac{d^2u_{k+1}}{dt^2} = \frac{1}{\Delta t^2}\left[u_{k+1}(n+1) - 2u_{k+1}(n) + u_{k+1}(n-1)\right] \tag{8}$$

$$\frac{du_{k+1}}{dt} = \frac{1}{\Delta t}\left[u_{k+1}(n+1) - u_{k+1}(n)\right] \tag{9}$$

From the boundary condition $t = 0$, it follows that

$$u_e = u_{k+1}(0) - \frac{1}{N_{Pe}}\frac{u_{k+1}(1) - u_{k+1}(0)}{\Delta t} \tag{10}$$

Solving for $u_{k+1}(0)$, we obtain

$$u_{k+1}(0) = \frac{u_e + u_{k+1}(1)/(N_{Pe}\Delta t)}{1 + 1/(N_{Pe}\Delta t)} \tag{11}$$

From the boundary condition $t = N$, we have

$$u_{k+1}(N) = u_{k+1}(N-1) \tag{12}$$

Substituting Equations (8) through (12) into Equation (6) for $n = 1, 2, \ldots, (N-1)$, the following $(N-1)$ simultaneous algebraic equations can be obtained

$$\mathbf{A}u_{k+1} + \mathbf{D} = 0 \tag{13}$$

where $\mathbf{A}$ represents the following tridiagonal matrix:

$$\mathbf{A} = \begin{bmatrix} b_1 & c & 0 & \cdots & & & 0 \\ a & b_2 & c & & & & \vdots \\ 0 & a & b_3 & c & & & \vdots \\ \vdots & & \ddots & \ddots & \ddots & & \vdots \\ \vdots & & & \ddots & \ddots & \ddots & \vdots \\ \vdots & & & & \ddots & \ddots & 0 \\ & & & & a & b_{N-2} & c \\ 0 & \cdots & \cdots & & 0 & a & b_{N-1} \end{bmatrix} \tag{14}$$

and D and u_{k+1} represent the following vectors:

$$\mathbf{D} = \begin{bmatrix} Ru_k(1)^2 + \dfrac{au_e}{1 + 1/(N_{Pe}\Delta t)} \\ Ru_k(2)^2 \\ \cdot \\ \cdot \\ \cdot \\ Ru_k(N-1)^2 \end{bmatrix}$$

$$\mathbf{u}_{k+1} = \begin{bmatrix} u_{k+1}(1) \\ u_{k+1}(2) \\ \cdot \\ \cdot \\ \cdot \\ u_{k+1}(N-1) \end{bmatrix} \tag{15}$$

with

$$b_1 = g - 2Ru_k(1) + \frac{a/(N_{Pe}\Delta t)}{1 + 1/(N_{Pe}\Delta t)} \tag{16}$$

$$b_n = g - 2Ru_k(n), n = 2, 3, \ldots, (N-2) \tag{17}$$

$$b_{N-1} = g - 2Ru_k(N-1) + c \tag{18}$$

$$a = 1/(N_{Pe}\Delta t^2) \tag{19}$$

$$c = 1/(N_{Pe}\Delta t^2) - 1/\Delta t \tag{20}$$

$$g = -2/(N_{Pe}\Delta t^2) + 1/\Delta t \tag{21}$$

In Equation (13), u_{k+1} is the unknown variable. The value of u_k is considered known and is obtained from the previous iteration. Since matrix A is tridiagonal, Equation (13) can be easily solved by the Thomas method [2].

As an initial approximation, we get

$$u_0(t) = 1.0, \qquad 0 \leq t \leq t_f \tag{22}$$

The step size is $\Delta t = 0.1$.

Using the numerical values given in Equation (7), Equation (13) is solved on an IBM 7094 computer. The results are shown in Figure 1. It can be seen that only three iterations are needed to obtain the required accuracy with approximately three seconds of computation time.

A more severe reaction condition with $R = 2$ and the initial approximation

$$u_0(t) = 0.2, \qquad 0 \leq t \leq t_f \tag{23}$$

has also been solved and the results are shown in Figure 1. It should be noted that in spite of the steepness of the concentration profile and the very poor initial approximation for $u_0(t)$, only four iterations are needed to obtain the required accuracy. Since a tridiagonal matrix can be easily solved by the Thomas method without excess computations, the computation time required for the present scheme is approximately the same as that of the numerical scheme used in the previous paper [1].

It should be noted that if the linearization technique is not used and the original nonlinear equation, Equation (2), is directly approximated by difference equations, a system of nonlinear difference or algebraic equations would result. Since a large system of nonlinear algebraic equations cannot be solved easily, the finite difference method cannot be used effectively for solving nonlinear differential equations without linearization.

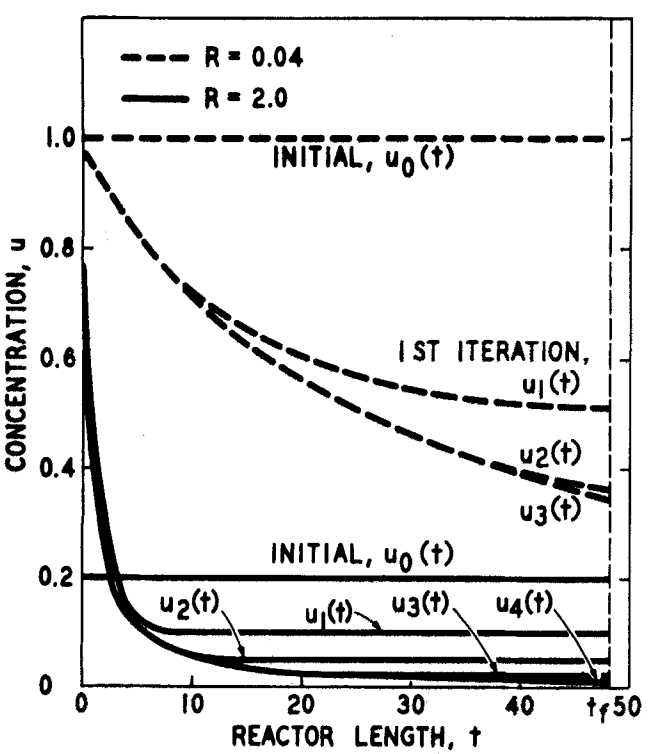

Fig. 1. Isothermal tubular reactor with axial diffusion.

ADIABATIC TUBULAR REACTOR WITH AXIAL DIFFUSION

The nonlinear equation resulting from an energy balance on a nonisothermal reactor involves the exponential temperature term which very frequently causes instability problems in obtaining a numerical solution. To test the effectiveness of the present method in overcoming this stability problem, the equations resulting from mass and energy balances are solved simultaneously. Using the same chemical reaction and assuming that the packing material plays the same role, the following two equations can be easily established:

$$\frac{1}{N_{Pe}}\frac{d^2u}{dt^2} - \frac{du}{dt} - \beta u^2 \exp(-E/T) = 0 \tag{24}$$

$$\frac{1}{N_{Pe}}\frac{d^2T}{dt^2} - \frac{dT}{dt} + Q\beta u^2 \exp(-E/T) = 0 \tag{25}$$

where

$$\beta = \frac{D_v G}{V}, \qquad Q = \frac{-\Delta H}{C_p} \tag{26}$$

In obtaining Equation (25), the mass axial diffusion coefficient D is assumed to be equal to the thermal axial diffusion coefficient.

The boundary conditions for Equation (24) are represented by Equation (3). For Equation (25), the boundary conditions are

$$T_e = T(0) - \frac{1}{N_{Pe}}\frac{dT(0)}{dt}, \qquad \text{at } t = 0 \tag{27a}$$

$$\frac{dT(t_f)}{dt} = 0, \qquad \text{at } t = t_f \tag{27b}$$

Equations (24) and (25) can be written

$$\frac{1}{N_{Pe}}\frac{d^2u}{dt^2} - \frac{du}{dt} = \beta u^2 \exp(-E/T) = f_1(T, u) \tag{28}$$

$$\frac{1}{N_{Pe}}\frac{d^2T}{dt^2}-\frac{dT}{dt}=-Q\beta u^2\exp(-E/T)=f_2(T,u) \tag{29}$$

The right hand side of Equations (28) and (29) can be linearized simultaneously by the use of the following vector equation:

$$\mathbf{f}(\mathbf{u}_{k+1})=\mathbf{f}(\mathbf{u}_k)+\mathbf{J}(\mathbf{u}_k)(\mathbf{u}_{k+1}-\mathbf{u}_k) \tag{30}$$

where $\mathbf{u}_{k+1}$, $\mathbf{f}$, and $\mathbf{u}_k$ are in vector form and represent the vectors $u_{(k+1),1}, u_{(k+1),2}, \ldots, u_{(k+1),m}$; $f_1, f_2, \ldots, f_m$; and $u_{k,1}, u_{k,2}, \ldots, u_{k,m}$ respectively. The Jacobian matrix, $\mathbf{J}(\mathbf{u}_k)$, is defined by

$$\mathbf{J}(\mathbf{u}_k)=\begin{bmatrix}\frac{\partial f_1}{\partial u_{k,1}} & \frac{\partial f_1}{\partial u_{k,2}} & \cdots & \frac{\partial f_1}{\partial u_{k,m}}\\ \cdots & \cdots & \cdots & \cdots\\ \frac{\partial f_m}{\partial u_{k,1}} & \frac{\partial f_m}{\partial u_{k,2}} & \cdots & \frac{\partial f_m}{\partial u_{k,m}}\end{bmatrix} \tag{31}$$

The symbols u and T in Equations (28) and (29) correspond to the symbols $u_{(k+1),1}$ and $u_{(k+1),2}$ in Equation (30). If we let u_k and T_k represent the kth iteration, Equations (28) and (29) can be linearized into the following form:

$$\frac{1}{N_{Pe}}\frac{d^2u_{k+1}}{dt^2}-\frac{du_{k+1}}{dt}-\beta u_k\left(-\frac{u_kE}{T_k}-u_k+2u_{k+1}+\frac{u_kE}{T_k^2}T_{k+1}\right)\exp(-E/T_k)=0 \tag{32}$$

$$\frac{1}{N_{Pe}}\frac{d^2T_{k+1}}{dt^2}-\frac{dT_{k+1}}{dt}+Q\beta u_k\left(-\frac{u_kE}{T_k}-u_k+2u_{k+1}+\frac{u_kE}{T_k^2}T_{k+1}\right)\exp(-E/T_k)=0 \tag{33}$$

where the variables with subscript $(k+1)$ are the unknown variables. The variables with subscript k are considered known. It should be noted that Equations (32) and (33) are linear differential equations.

Using the difference quotients, Equations (8) and (9), for Equation (32) and a similar set of difference quotients for Equation (33), the above two equations can be reduced into a set of simultaneous difference equations. The boundary conditions for Equation (33) become

$$T_e=T_{k+1}(0)-\frac{1}{N_{Pe}}\frac{T_{k+1}(1)-T_{k+1}(0)}{\Delta t} \tag{34}$$

and

$$T_{k+1}(N)=T_{k+1}(N-1) \tag{35}$$

Using the same procedure as that used in obtaining Equation (13), the following $2(N-1)$ simultaneous algebraic equations can be obtained

$$\mathbf{A}_1\mathbf{u}_{k+1}+\mathbf{B}_1\mathbf{T}_{k+1}+\mathbf{D}_1=0 \tag{36}$$

$$\mathbf{A}_2\mathbf{T}_{k+1}+\mathbf{B}_2\mathbf{u}_{k+1}+\mathbf{D}_2=0 \tag{37}$$

where $\mathbf{A}_1$ and $\mathbf{A}_2$ are the following tridiagonal matrices:

$$\mathbf{A}_1=\begin{bmatrix}h_1 & c & 0 & \cdots & 0\\ a & h_2 & c & & \vdots\\ 0 & a & h_3 & c & \vdots\\ \vdots & \ddots & \ddots & \ddots & \vdots\\ 0 & \cdots & 0 & a & h_{N-1}\end{bmatrix} \tag{38}$$

$$\mathbf{A}_2=\begin{bmatrix}h'_1 & c & 0 & \cdots & 0\\ a & h'_2 & c & & \vdots\\ 0 & a & h'_3 & c & \vdots\\ \vdots & \ddots & \ddots & \ddots & \vdots\\ 0 & \cdots & 0 & a & h'_{N-1}\end{bmatrix} \tag{39}$$

and $\mathbf{B}_1$ and $\mathbf{B}_2$ are the diagonal matrices:

$$\mathbf{B}_1=\begin{bmatrix}S_1 & & & & 0\\ & S_2 & & & \\ & & S_3 & & \\ & & & \ddots & \\ 0 & & & & S_{N-1}\end{bmatrix}\qquad \mathbf{B}_2=\begin{bmatrix}S'_1 & & & & 0\\ & S'_2 & & & \\ & & S'_3 & & \\ & & & \ddots & \\ 0 & & & & S'_{N-1}\end{bmatrix} \tag{40}$$

with

$$h_1=g-2\beta u_k(1)\exp(-E/T_k(1))+\frac{a/(N_{Pe}\Delta t)}{1+1/(N_{Pe}\Delta t)} \tag{41}$$

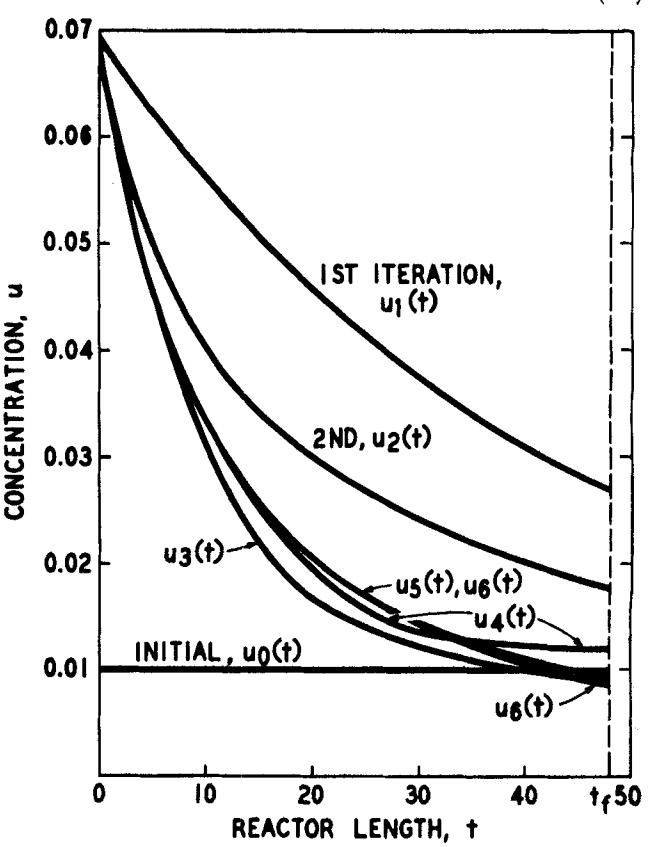

Fig. 2. Adiabatic tubular reactor wth axial diffusion, concentration profile.

$$h_n = g - 2\beta u_k(n) \exp(-E/T_k(n)),\ n = 2, 3, \ldots, N-2 \quad (42)$$

$$h_{N-1} = g - 2\beta u_k(N-1) \exp(-E/T_k(N-1)) + c \quad (43)$$

$$h'_1 = g + Q\beta E(u_k(1)/T_k(1))^2 \exp(-E/T_k(1)) + \frac{a/(N_{Pe}\Delta t)}{1 + 1/(N_{Pe}\Delta t)} \quad (44)$$

$$h'_n = g + Q\beta E(u_k(n)/T_k(n))^2 \exp(-E/T_k(n)),\ n = 2, 3, \ldots, (N-2) \quad (45)$$

$$h'_{N-1} = g + Q\beta E(u_k(N-1)/T_k(N-1))^2 \exp(-E/T_k(N-1)) + c \quad (46)$$

$$S_n = -\beta E(u_k(n)/T_k(n))^2 \exp(-E/T_k(n)),\ n = 1, 2, \ldots, (N-1) \quad (47)$$

$$S'_n = 2Q\beta u_k(n) \exp(-E/T_k(n)),\ n = 1, 2, \ldots, (N-1) \quad (48)$$

The column vectors $\mathbf{u}_{k+1}$, $\mathbf{T}_{k+1}$, $\mathbf{D}_1$ and $\mathbf{D}_2$ are

$$\mathbf{u}_{k+1} = \begin{bmatrix} u_{k+1}(1) \\ u_{k+1}(2) \\ \cdot \\ \cdot \\ \cdot \\ u_{k+1}(N-1) \end{bmatrix}, \qquad \mathbf{T}_{k+1} = \begin{bmatrix} T_{k+1}(1) \\ T_{k+1}(2) \\ \cdot \\ \cdot \\ \cdot \\ T_{k+1}(N-1) \end{bmatrix} \quad (49)$$

$$\mathbf{D}_1 = \begin{bmatrix} \beta(u_k(1))^2(E/T_k(1) + 1) \exp(-E/T_k(1)) + \dfrac{au_e}{1 + 1/(N_{Pe}\Delta t)} \\ \beta(u_k(2))^2(E/T_k(2) + 1) \exp(-E/T_k(1)) \\ \cdot \\ \cdot \\ \cdot \\ \beta(u_k(N-1))^2(E/T_k(N-1) + 1) \exp(-E/T_k(N-1)) \end{bmatrix} \quad (50)$$

$$\mathbf{D}_2 = \begin{bmatrix} -Q\beta(u_k(1))^2(E/T_k(1) + 1) \exp(-E/T_k(1)) + \dfrac{aT_e}{1 + 1/(N_{Pe}\Delta t)} \\ -Q\beta(u_k(2))^2(E/T_k(2) + 1) \exp(-E/T_k(2)) \\ \cdot \\ \cdot \\ \cdot \\ -Q\beta(u_k(N-1))^2(E/T_k(N-1) + 1) \exp(-E/T_k(N-1)) \end{bmatrix} \quad (51)$$

Equations (36) and (37) are coupled and they must be solved simultaneously. This complicates the problem considerably. In order to simplify the calculations, the values of $\mathbf{T}_{k+1}$ and $\mathbf{u}_{k+1}$ in the second terms of Equations (36) and (37) are approximated by $\mathbf{T}_k$ and $\mathbf{u}_k$ respectively. Equations (36) and (37) now become

$$\mathbf{A}_1 \mathbf{u}_{k+1} = -(\mathbf{B}_1 \mathbf{T}_k + \mathbf{D}_1) = \mathbf{d}_1 \quad (52)$$

$$\mathbf{A}_2 \mathbf{T}_{k+1} = -(\mathbf{B}_2 \mathbf{u}_k + \mathbf{D}_2) = \mathbf{d}_2 \quad (53)$$

where

$$\mathbf{T}_k = \begin{bmatrix} T_k(1) \\ T_k(2) \\ \cdot \\ \cdot \\ \cdot \\ T_k(N-1) \end{bmatrix}, \qquad \mathbf{u}_k = \begin{bmatrix} u_k(1) \\ u_k(2) \\ \cdot \\ \cdot \\ \cdot \\ u_k(N-1) \end{bmatrix} \quad (54)$$

Since the variables with subscript k are known, Equations (52) and (53) can be solved independently by the Thomas method.

The problem was solved on an IBM 7094 computer. The numerical values used are

$t_f = 48$	$\beta = 0.5 \times 10^8$ liter/moles
$N_{Pe} = 2$	$u_e = 0.07$ moles/liter
$T_e = 1{,}250°\text{R}$	$Q = 1000$ liter-°F./mole
$E = 22{,}000°\text{R}$	$\Delta t = 0.1$

As an initial approximation, let

$$T_0(t) = 1{,}250, \quad 0 \leq t \leq t_f$$
$$u_0(t) = 0.01, \quad 0 \leq t \leq t_f$$

The results are shown in Figures 2 and 3. With the very poor initial approximation, the correct results are obtained with seven iterations. Each iteration requires approximately a little over one second of computing time. As can be seen from Figure 3, the temperature fluctuates widely during the second and third iterations. The maximum temperature for the second iteration is approximately 1,562°R while the minimum for the third iteration is approximately 610°R. These fluctuations are caused by the highly nonlinear Arrhenius equation. Owing to these fluctuations, the convergence rate of this numerical example is much slower than that of the first one. The approximations in obtaining Equations (52) and (53) also reduce the convergence rate.

SYSTEMS OF DIFFERENTIAL EQUATIONS

The approach can be generalized easily to systems of differential equations. Consider first the following system of nonlinear second-order differential equations

$$\frac{d^2\mathbf{u}}{dt^2} + \mathbf{a}(t)\frac{d\mathbf{u}}{dt} = \mathbf{f}(\mathbf{u}, t) \quad (55)$$

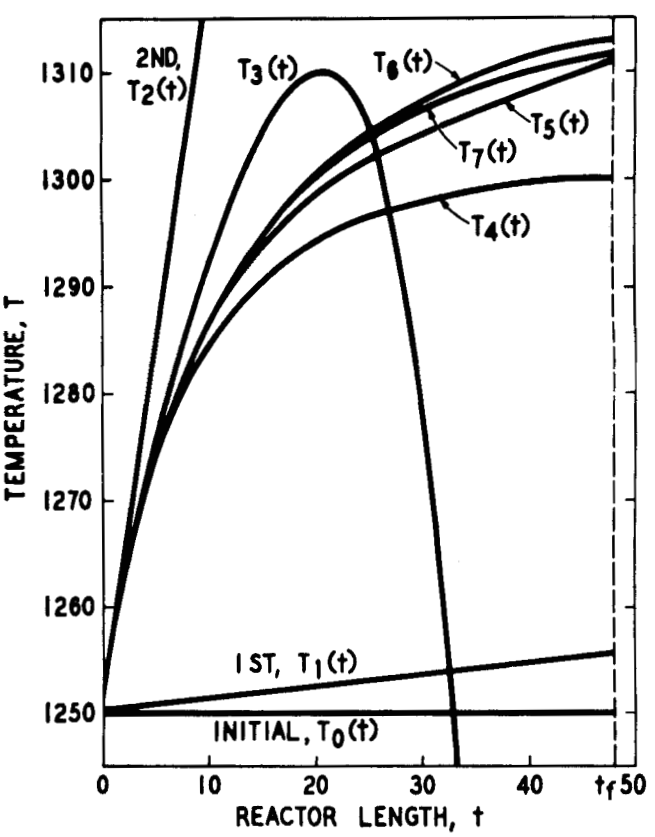

Fig. 3. Adiabatic tubular reactor with axial diffusion, temperature profile.

with boundary conditions

$$\mathbf{u}(0) = \mathbf{u}^o$$
$$\mathbf{u}(t_f) = \mathbf{u}^f \tag{56}$$

where u and f are m-dimensional column vectors and $\mathbf{a}(t)$ is an m-dimensional row vector. The right-hand side of Equation (55) can be linearized easily by Equation (30). From these linearized equations, a system of m simultaneous matrix equations similar to Equations (36) and (37) can be obtained. The same trick which is used to obtain Equations (52) and (53) from Equations (36) and (37) can be used to make the m matrices equations independent of each other. Since the matrices which are coefficients of the unknown vectors are tridiagonal matrices, the m matrix equations can be solved independently by the Thomas method.

Now let us consider the more general system of second-order nonlinear differential equations, such as

$$\frac{d^2\mathbf{u}}{dt^2} = \mathbf{f}\left(\frac{d\mathbf{u}}{dt}, \mathbf{u}, t\right) = \mathbf{f}(\mathbf{u}', \mathbf{u}, t) \tag{57}$$

with the same boundary conditions given by Equation (56). In order to obtain the recurrence relation, the derivative u′ can be considered as another function parallel to u and the right-hand side of Equation (57) can be linearized as

$$\frac{d^2\mathbf{u}_{k+1}}{dt^2} = \mathbf{f}(\mathbf{u}'_k, \mathbf{u}_k, t) + J(\mathbf{u}_k)(\mathbf{u}_{k+1} - \mathbf{u}_k) + J_{u'}(\mathbf{u}_k)(\mathbf{u}'_{k+1} - \mathbf{u}'_k) \tag{58}$$

where $J_{u'}(\mathbf{u}_k)$ is defined the same way as $J(\mathbf{u}_k)$ except that the differentiation is carried out with respect to $\mathbf{u}'_k$, not $\mathbf{u}_k$. Note that Equation (58) is a linear matrix differential equation in the unknown variable $\mathbf{u}_{k+1}$. Using difference quotients similar to Equations (8) and (9), the following $m(N-1)$ simultaneous algebraic equations can be obtained from Equation (58):

$$\mathbf{A}_i\,\mathbf{u}_{k+1,i} + \sum_{\substack{j=1\\ j\neq i}}^{m} \mathbf{B}_{i,j}\,\mathbf{u}_{k+1,j} + \mathbf{D}_i = 0, \quad i = 1, 2, \ldots, m \tag{59}$$

where the second subscript of u is used to indicate the particular variable. The matrices $\mathbf{A}_i$ are tridiagonal matrices and $\mathbf{B}_{i,j}$ are matrices in which only the main diagonal and the diagonal directly above the main diagonal are different from zero. If we use d to indicate nonzero elements, the matrices $\mathbf{B}_{i,j}$ can be represented as

$$\mathbf{B} = \begin{bmatrix} d & d & & & & \\ & d & \ddots & & & \\ & & \ddots & \ddots & & \\ & & & \ddots & \ddots & \\ & & & & d & d \end{bmatrix} \tag{60}$$

$\mathbf{u}_{k+1,i}$ is the following column vector

$$\mathbf{u}_{k+1,i} = \begin{bmatrix} u_{k+1,i}(1) \\ u_{k+1,i}(2) \\ \vdots \\ u_{k+1,i}(N-1) \end{bmatrix} \tag{61}$$

Note that the vector $\mathbf{u}_{k+1}$ is a function of t and is completely different from the vector $\mathbf{u}_{k+1,i}$. $\mathbf{D}_i$ are $(N-1)$-dimensional column vectors.

If we use $\mathbf{u}_{k,j}$ to approximate $\mathbf{u}_{k+1,j}$ in the second term of Equation (59), the following m independent matrix equations are obtained

$$\mathbf{A}_i\,\mathbf{u}_{k+1,i} + \sum_{\substack{j=1\\ j\neq i}}^{m} \mathbf{B}_{i,j}\,\mathbf{u}_{k,j} + \mathbf{D}_i = 0, \quad i = 1, 2, \ldots, m \tag{62}$$

The vectors $\mathbf{u}_{k,j}$ are known vectors and are defined as

$$\mathbf{u}_{k,j} = \begin{bmatrix} u_{k,j}(1) \\ u_{k,j}(2) \\ \vdots \\ u_{k,j}(N-1) \end{bmatrix} \tag{63}$$

The only unknown vectors in Equations (62) are $\mathbf{u}_{k+1,i}$. Since $\mathbf{A}_i$ are tridiagonal matrices, Equation (62) again can be solved by the Thomas method.

Obviously, the system of second-order nonlinear differential equations:

$$\mathbf{f}(\mathbf{u}'', \mathbf{u}', \mathbf{u}, t) = 0 \tag{64}$$

can also be linearized as

$$\mathbf{f}(\mathbf{u}''_k, \mathbf{u}'_k, \mathbf{u}_k, t) + J(\mathbf{u}_k)(\mathbf{u}_{k+1} - \mathbf{u}_k) + J_{u'}(\mathbf{u}_k)(\mathbf{u}'_{k+1} - \mathbf{u}'_k) + J_{u''}(\mathbf{u}_k)(\mathbf{u}''_{k+1} - \mathbf{u}''_k) = 0 \tag{65}$$

With assumed values for $\mathbf{u}_{k+1}(t)$, Equation (65) can be solved using the same procedure discussed above.

The above procedure can also be extended to a system of first-order nonlinear differential equations. Since an nth-order nonlinear differential equation can be reduced to n first-order simultaneous differential equations, the following nonlinear system of first-order equations is widely applicable.

$$\frac{du_i}{dt} = f_i(u_1, u_2, \ldots, u_m, t), \qquad i = 1, 2, \ldots, m \tag{66}$$

In vector notation, Equation (66) becomes

$$\frac{d\mathbf{u}}{dt} = \mathbf{f}(\mathbf{u}, t) \tag{67}$$

which can be linearized as

$$\frac{d\mathbf{u}_{k+1}}{dt} = \mathbf{f}(\mathbf{u}_k, t) + J(\mathbf{u}_k)(\mathbf{u}_{k+1} - \mathbf{u}_k) \tag{68}$$

If Equation (68) is unstable when the marching integration technique such as the Runge-Kutta method is used, the finite difference method can be used. Using difference quotients similar to Equation (9), Equation (68) can be reduced to m simultaneous matrix equations. If m is not large, these m matrix equations can be solved simultaneously. Otherwise, they can be solved independently, using the approximation technique used to obtain Equations (52) and (53).

Note that it is not necessary to make each of the matrix equations independent of all the others. In order to increase the rate of convergence the m matrix equations can be separated into small groups. Each group of equations is independent of the other groups and each group can be solved simultaneously. Thus, the first group of matrix equations may consist of the first five equations with $i = 1, 2, \ldots, 5$. The only unknown variables allowed in these five equations are $u_{k+1,j}$ $i = 1, 2, \ldots, 5$. The other unknown variables $u_{k+1,i}$, $i = 6, 7, \ldots, m$ in these five equations can be approximated by $u_{k,i}$. These five simultaneous matrix equations can be inverted by matrix partitioning. Since we are not solving all the matrix

equations simultaneously, the requirement on the number of rapid access memory of the computer can be reduced. However, since we are solving a small group of matrix equations simultaneously and thus the number of approximations made is considerably reduced, a reasonably fast convergence rate still may be obtained.

DISCUSSION

The procedure discussed can be generalized in many ways. Instead of the difference expressions, Equations (8) and (9), higher order difference expressions which can represent the derivatives more accurately also can be used. However, the numerical solution of the resulting algebraic equations would be much more difficult.

Since only the first-order difference is used, the numerical results obtained are not as accurate as those obtained by the Runge-Kutta integration method. Comparison of the present results with those obtained by the Runge-Kutta method shows that the present results only have a three digit accuracy. This accuracy can be increased by using a smaller integration step size Δt.

If there is no stability problem connected with the marching integration technique, the superposition principle approach discussed in the previous paper [*1*] has approximately the same convergence rate as that of the finite difference approach. However, since in general the Runge-Kutta method is more accurate, the superposition principle approach should be used unless the problem is unstable. Since the finite difference method is an implicit method, there is no stability problem connected with this method for two point boundary value problems.

Both the superposition principle approach and the finite difference method are much more powerful than the conventional trial and error numerical integration technique. Actual computational experience has shown that if the iterative process converges, the rate of convergence is quadratic. Furthermore, for a large number of problems convergence can be obtained even with very unreasonable initial approximations. On the other hand, with the trial and error technique very good initial approximations must be used in order to obtain convergence and, in general, the convergence rate is extremely slow.

Another advantage of the present procedure is that the coefficients of the resulting linear algebraic equations form a tridiagonal matrix whose inverse can be obtained easily by the Thomas method. Thus, although 479 simultaneous linear algebraic equations are solved in the numerical examples, the computational time and the rapid access memory requirements for solving these equations are much less than those needed for solving a general system of 479 linear equations.

There are various ways to linearize a nonlinear equation. However, the linearized equation is in general so approximate to the original nonlinear equation that it is unsatisfactory for many purposes. The quasilinearization technique not only linearizes the original nonlinear equation, but even more important, it provides a sequence of functions which converge to the solution of the nonlinear equation.

The quasilinearization technique also has several disadvantages. The first possible disadvantage is the problem of ill-conditioning. Both the superposition principle approach and the finite difference method can have this ill-conditioning difficulty. It is clear that the matrices in the difference equations such as Equation (13) must be well-conditioned. If the matrix is ill-conditioned, the inverse of this matrix would give erroneous results. In the superposition principle approach, this ill-conditioning problem is usually connected with the algebraic equations which are used to obtain the integration constants.

A second disadvantage lies in the fact that in order to retain the feature of quadratic convergence, the partial derivatives in the Taylor series expansion must be determined accurately. Since numerical differentiation is a very inaccurate process, these derivatives must be evaluated analytically. For a large complex problem this analytical differentiation can be quite tedious and subject to error. Wengert [*4*] has recently devised an analytical technique for computer evaluation of the partial derivatives. This technique appears to be quite useful in eliminating the laborious work of obtaining analytical expressions for the partial derivatives in the quasilinearization technique.

Finally, the present approach appears to be an effective tool for handling the Arrhenius exponential type nonlinearity. This type of nonlinearity has caused considerable stability problems in chemical engineering problems. To the author's knowledge, no other effective technique has been devised to handle this nonlinearity difficulty.

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NOTATION

$\mathbf{A}, \mathbf{A}_1, \mathbf{A}_2, \mathbf{B}_1, \mathbf{B}_2$ = matrices
C = specific heat of reaction mixture
D = effective axial diffusion coefficient
D_p = diameter of catalyst particle
E = activation energy of reaction divided by gas constant
G = frequency factor constant in Arrhenius equation
ΔH = heat of reaction
k = chemical reaction rate constant
L = reactor length
N = $t_f/\Delta t$
N_{Pe} = Peclet group = $(V D_p)/D$
Q = $\dfrac{-\Delta H}{C\rho}$
R = reaction rate group = (kD_p/V)
t = dimensionless reactor length variable or independent variable
T = temperature
Δt = integration interval or step size
t_f = L/D_p = dimensionless reactor length
T_e = temperature of reaction mixture before entering the reactor
u = concentration of reactant A or dependent variable
u_e = concentration of reactant A before entering the reactor
V = velocity
β = (D_pG/V)
ρ = density of reaction mixture

Subscripts

k = kth iteration
0 = initial approximation for the variables

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