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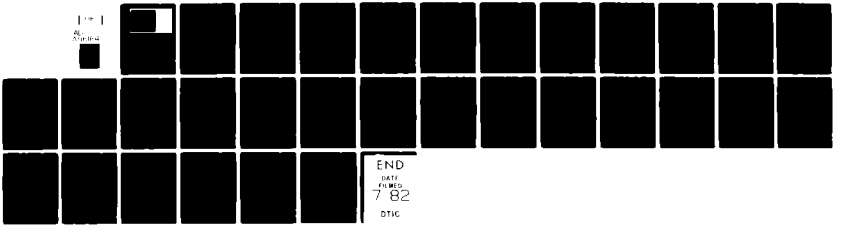

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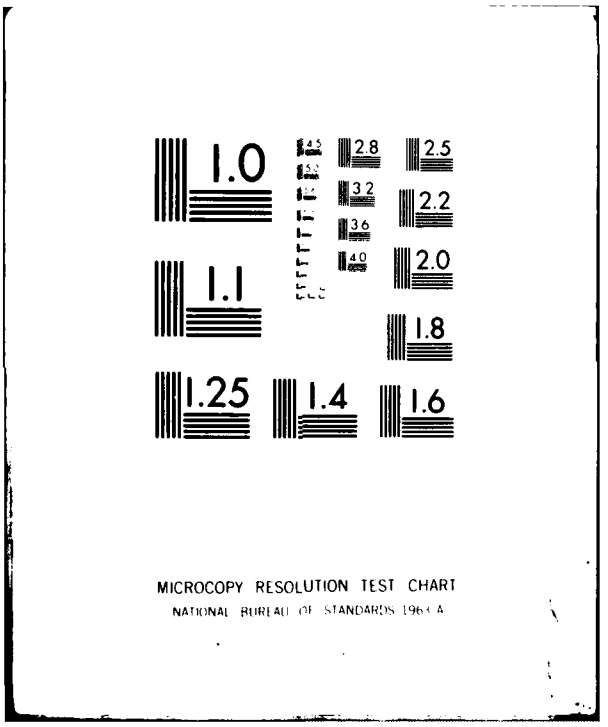
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NUMERICAL COMPUTATION OF PERIODIC SOLUTION BRANCHES AND OSCILLATORY DYNAMICS OF THE STIRRED TANK REACTOR WITH A → B → C REACTIONS

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NUMERICAL COMPUTATION OF PERIODIC SOLUTION BRANCHES AND OSCILLATORY DYNAMICS OF THE STIRRED TANK REACTOR WITH $A \to B \to C$ REACTIONS

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ABSTRACT

We present a continuation technique for branches of periodic solutions which can be applied to autonomous systems of nonlinear differential equations. Coupling the technique with Hopf bifurcation expansions enables one to compute entire periodic solution branches including those with turning points and unstable solutions. We apply these methods to a continuously stirred tank reactor with consecutive $A \to B \to C$ reactions. Our computations reveal dynamic phenomena not seen in previous reactor studies. The results include response diagrams exhibiting stable and unstable periodic branches that contain multiple turning points. The discovery of these points indicates the reactor may jump from a steady state to a periodic orbit or may jump from one periodic orbit to anther.

AMS (MOS) Subject Classifications: 80A20, 80A30, 65C20, 65L05

Key Words: Hopf bifurcation, oscillatory dynamics, chemical reactors

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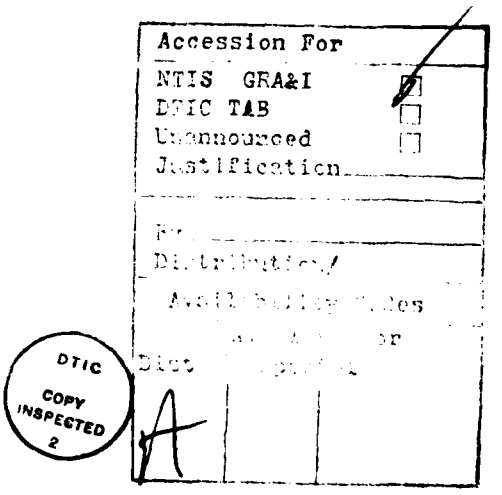
SIGNIFICANCE AND EXPLANATION

Numerical methods are developed for tracing branches of periodic solutions exhibited by mathematical methods consisting of autonomous, nonlinear differential equations. These continuation methods allow one to compute the periodic orbits which emanate from steady state solutions at the Hopf bifurcation points. It should be emphasized that the techniques permit us to compute entire periodic branches including those with turning points, unstable solutions and infinite periodicity.

We apply the methods to a mathematical model of a stirred tank reactor with an $A \rightarrow B \rightarrow C$ reaction mechanism. The reactor exhibits broad regions of multiple steady states and periodic solutions, and we discuss the effects of the intricate solution interactions on reactor operation.



NUMERICAL COMPUTATION OF PERIODIC SOLUTION BRANCHES AND OSCILLATORY DYNAMICS OF THE STIRRED TANK REACTOR WITH $A \rightarrow B \rightarrow C$ REACTIONS

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INTRODUCTION

The bifurcation analysis of time-dependent differential equations has found application in several fields of research including chemical reaction engineering [1-8], mathematical biology and biochemistry [9-16], and combustion [17-24]. The first step in analyzing the mathematical models from these fields normally consists of determining all the possible steady state solutions. In general, such computations require a continuation procedure for steady state solution branches which allows for computing past turning points without difficulty and for switching from one branch to another at points where the branches intersect. Techniques developed by Keller [25] fulfill these requirements and are used in studies of multiple solutions exhibited by tubular reactors [26], catalyst pellets [27], premixed flames [20-22] and flow between rotating cylinders and disks [28, 29].

The second step in the model analysis is often the determination of the periodic solutions. When a branch of such solutions crosses a steady state branch, the point of intersection is called a Hopf bifurcation point. In reaction engineering, Hopf theory has been used to determine all possible periodic orbits exhibited by an exothermic $A \rightarrow B$ reaction in a continuously stirred tank reactor (CSTR) [6, 7]. This local theory provides perturbational expansions at the Hopf point which determine the frequency, direction, stability and profile of the bifurcating solution without time integration. The application of the expansions is particularly useful in examining reactor dynamics and jump phenomena which accompany the stability exchange at the Hopf points. Recently these techniques were extended to distributed parameter systems and applied to the tubular reactor [8].

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Further insight into the dynamics of chemical reactors is achieved by tracing the periodic solution branches away from the bifurcation points. These computations are usually accomplished by applying initial value techniques to the model equations and performing transient calculations until the solutions converge into the periodic orbit. This strategy is capable of determining stable periodic solutions as well as unstable solutions for the special two dimensional case (e.g. the CSTR with an $A \rightarrow B$ reaction) where one can simply replace t by $-t$ in the model equations. In most other cases, initial value techniques will be difficult or impossible to use, especially when turning points occur along the periodic branches (30).

In this paper we present a simple continuation method for periodic solutions which allows the computations to proceed past such turning points as well as along unstable branches. We transform the initial value problem into a boundary value problem on the fixed interval $[0,2\pi]$ by treating the period of the solution as an unknown and imposing periodicity as a side condition. Two equations are added to regulate the step size along the solution branch and to avoid translation of the periodic solution. The resulting system of coupled, nonlinear equations is efficiently solved by Newton's method. A similar treatment for tracing branches of periodic solutions has been used by Rinzel and Miller [10] and has received extensive treatment in a recent thesis [31]. The particular formulation of the numerical scheme that we use however has great practical advantages [30]. Other numerical techniques for Hopf bifurcation are reported by Weber [32] and Langford [33].

We apply our numerical techniques to an important three dimensional problem - consecutive $A \rightarrow B \rightarrow C$ exothermic reactions in a CSTR. In an earlier paper, Hlavacek, Kubicek, and Visnak [34] give a rather complete classification of the steady state multiplicity for this system and also present isolated numerical calculations illustrating the existence of oscillatory solutions. Responding to the need pointed out by Hlavacek et al. for work in multi-dimensional systems exhibiting oscillations, Cohen and Keener [35] apply multi-time scale perturbation techniques and illustrate the utility of this method by deriving formulas for the bifurcating solutions for a few combinations of model

parameters. However, the most thorough classification of dynamics for this reactor to date is given by Halbe and Poore [36]. They apply the Hopf theory to illustrate the increased diversity of the consecutive reaction case compared to the $A \rightarrow B$ example and show that the $A \rightarrow B \rightarrow C$ CSTR may exhibit up to five steady state states and up to four Hopf bifurcation points.

None of the above studies traces the periodic branches away from the Hopf point. We present in a later section entire solution branches, containing stable and unstable orbits, which are computed using our numerical techniques. Although we do not intend to provide a complete cataloguing of the dynamic capabilities of this reactor, our computations reveal phenomena not reported in earlier CSTR studies. Of particular interest are branches which contain multiple turning points creating the possibility of jump transitions between periodic states.

MASS AND ENERGY BALANCES

We consider consecutive exothermic, first order reactions in an ideal stirred tank continuously fed by component A and cooled by a medium with constant temperature T_c. The mass balances for A and B and the energy balance are written below [34]

$$V \frac{dC_A}{dt} = v(C_{A0} - C_A) - VC_A A_1 \exp(-E_1/RT) \tag{1}$$

$$V \frac{dC_B}{dt} = -vC_B + VC_A A_1 \exp(-E_1/RT) - VC_B A_2 \exp(-E_2/Rt) \tag{2}$$

$$\rho C_p V \frac{dT}{dt} = \rho C_p v(T_0 - T) - US_A(T - T_c) + (-\Delta H_1)VC_A A_1 \exp(-E_1/RT) + (-\Delta H_2)VC_B A_2 \exp(-E_2/RT) \tag{3}$$

These equations can be written in dimensionless form

$$\frac{dy}{d\tau} = 1 - y - Dy \exp(\theta/1 + \varepsilon\theta) \tag{4}$$

$$\frac{dz}{d\tau} = -z + Dy \exp(\theta/1 + \varepsilon\theta) - D\sigma z \exp\left(\frac{\kappa\theta}{1 + \varepsilon\theta}\right) \tag{5}$$

$$\frac{d\theta}{d\tau} = -\theta - \beta(\theta - \theta_c) + DBy \exp(\theta/1 + \varepsilon\theta) + DB\alpha\sigma \exp\left(\frac{\kappa\theta}{1 + \varepsilon\theta}\right) \tag{6}$$

Here y is dimensionless concentration of A, z is dimensionless concentration of B, θ is dimensionless temperature, and τ is dimensionless time. D is Damkohler number, ε is dimensionless activation number, σ is selectivity ratio, κ is the ratio of activation energies, β is the dimensionless heat transfer coefficient, B is the adiabatic temperature rise and α is ratio of heats of reaction. It is convenient to analyze the limiting case of small ε and $\theta_c = 0$.

NUMERICAL METHODS

The numerical methods presented in this section are capable of treating nonlinear autonomous systems of ordinary differential equations:

$$\frac{du(t)}{du} = f(u(t);\lambda), \quad t > 0, \quad u,f \in R^n , \tag{7}$$

where λ is a free parameter. A more detailed presentation of these numerical procedures and computer codes can be found in [30].

Steady State Computations

The computation of the steady state solution branches is an algebraic problem consisting of the bifurcation analysis of the equation

$$f(u,\lambda) = 0 . \tag{8}$$

This is accomplished numerically by applying the continuation and switching techniques due to Keller (25). This method inflates the original problem by imposing an arc-length normalization, N, on the solution branch so that

$$\begin{pmatrix} f(u(s);\lambda(s)) \\ N(u(s),\lambda(s);s) \end{pmatrix} = 0 \qquad (9)$$

where the new unknown s (a "pseudo-arc-length") replaces λ as the continuation parameter in an Euler-Newton scheme. The main advantage of using the inflated system is the capability of computing past turning points on the solution branch, and this idea serves as a basis for the continuation techniques used to trace the periodic solutions.

Periodic Solution Branches

In computing entire branches of periodic solutions for autonomous systems both the solution $u(t)$ and the solution period ρ continuously change as λ varies. To fix the period we scale the time variable t and transform (7) into

$$\frac{du}{dt} = \frac{\rho}{2\pi} f(u(t);\lambda) \qquad t \in [0,2\pi] \qquad (10)$$

Now the unknown period $\rho(\lambda)$ appears explicitly and we seek solutions satisfying

$$u(0) = u(2\pi) \qquad (11)$$

Thus the initial value problem has been replaced by a boundary value problem with non-separated boundary conditions which allows the computation of unstable orbits. Suppose we are given a periodic solution $(v(t),\rho_0,\lambda_0)$. Our objective is to find a neighboring solution $(u(t),\rho,\lambda)$ at some distance Δs from the given solution. To this end, we solve

$$\begin{pmatrix} C(u,\rho,\lambda) \\ B(u) \\ N(u,\rho,\lambda) \\ A(u) \end{pmatrix} = 0 \qquad (12)$$

where C and B represent (10) and (11). The unknown period ρ is included in the normalization

$$N(u,\rho,\lambda) = \theta_1^2\|u - v\|_2^2 + \theta_2^2|\lambda - \lambda_0| + \theta_3(\rho - \rho_0)^2 - \Delta s^2 = 0 \qquad (13)$$

where θ_1's are fixed weights and $\|\cdot\|_2$ is the L_2 norm.

A represents an "anchor" equation and is included to insure unique periodic solutions by eliminating free translation in time. Although there are many possible choices for an appropriate anchor equation, the following is found to be extremely convenient for problems with rapidly varying solution components

$$A(u) \equiv \int_0^{2\pi} (u(t) - v(t))^T f(v(t);\lambda_0)dt = 0 \qquad (14)$$

For a discussion of the effect of choice of the anchor equation see [30].

Hopf Bifurcation

The Hopf bifurcation points are located by finding a solution $(v;\lambda_0)$ on the steady state branch where the eigenvalues of $f_u(v,\lambda_0)$ are purely imaginary. The periodic solution separating itself from the steady state branch at these points is describable by the following asymptotic expansions [37, 38]

$$u(t,\varepsilon) = v + \varepsilon\phi(t) + 0(\varepsilon^2) \qquad (15)$$

$$\rho(\varepsilon) = \rho_0 + 0(\varepsilon^2) \qquad (16)$$

$$\lambda = \lambda_0 + 0(\varepsilon^2) \qquad (17)$$

where $\pm\pi/\rho_0$ are the eigenvalues at the Hopf point and where ε is the amplitude of the orbit $\phi(t)$ is the corresponding 2π-periodic solution of the linearized problem

$$\frac{du(t)}{dt} = \frac{\rho_0}{2\pi} f_u(v;\lambda_0)\phi(t) \qquad (18)$$

(15) through (18) suggest the following procedure for switching onto the periodic solution branch. Assume the steady state branch has been computed so that the Hopf points have been located and ρ_0 and $\phi(t)$ computed. Normalize $\phi(t)$ such that $\|\phi\|_2 = 1$. Next solve

(12) but replace (14) with the nearly identical equation

$$A(u) = \int_0^{2\pi} (u(t) - v)^T \phi'(t) dt = 0 \ . \tag{19}$$

Under reasonable assumptions the modified set of equations can be shown to be well-posed near the Hopf bifurcation point, so that computational procedures can be based upon them.

Discretization

The numerical solution of (12) requires the discretization of (10) and evaluation of the integrals in (13) and (14). There are, of course, many ways to do this and to illustrate how a typical computation might proceed, we describe in some detail a specific choice of the discretization. The differential equation is approximated by the method of collocation at Gauss points with piecewise polynomials that belong to the class $C(0,2\pi)$ [39-41]. In our program the number of collocation points per mesh interval, and hence the method order, can be selected. For simplicity we describe the technique for the specific choice of two collocation points per mesh interval. Introduce a mesh $\{0 = t_0 < t_1 < \ldots < t_N = 2\pi\}$, $\Delta t_j \equiv t_j - t_{j-1}$, $(1 \leq j \leq N)$; and for each j, introduce the Lagrange basis polynomials

$$w_{j,-1}(t) = 2(t - t_{j-1/2})(t - t_j)/\Delta t_j^2 \ ,$$

$$w_{j,-1/2}(t) = -4(t - t_{j-1})(t - t_j)/\Delta t_j^2 \ ,$$

and

$$w_{j,0}(t) = 2(t - t_{j-1})(t - t_{j-1/2})/\Delta t_j^2 \ ,$$

where

$$t_{j-1/2} \equiv \frac{1}{2}(t_{j-1} + t_j) \ .$$

Relative to each subinterval $[t_{j-1},t_j]$, the Gauss points are $\xi_{j,1} = t_{j-1/2} - \Delta t_j/(2\sqrt{3})$ and $\xi_{j,2} = t_{j-1/2} + \Delta t_j/(1\sqrt{3})$. The collocation method now consists of finding

$$p_j(t) = w_{j,-1}(t)u_{j-1} + w_{j,-1/2}(t)u_{j-1/2} + w_{j,0}(t)u_j, \quad 1 \leq j \leq N \ ,$$

such that

$$p_j'(\xi_{j,i}) = \frac{\rho}{2\pi} f(p_j(\xi_{j,i});\lambda), \quad i = 1,2; \quad 1 \le j \le N , \tag{20}$$

and $p_1(0) = p_N(2\pi)$, i.e.,

$$u_0 = u_N . \tag{21}$$

With the above basis selection u_j and $u_{j-1/2}$ are to approximate the solution $u(t)$ of the continuous problem at t_j and $t_{j-1/2}$ respectively. The integral in (13) can be discretized by Simpson's rule giving

$$\theta_1^2(\lambda - \lambda_0)^2 + \theta_2^2(\rho - \rho_0)^2 + \\ + \theta_3^2 \sum_{j=1}^{N} \Delta t_j[(u_{j-1} - v_{j-1})^2 + 4(u_{j-1/2} - v_{j-1/2})^2 + (u_j - v_j)^2]/6 = s^2 , \tag{22}$$

(Accuracy is not actually important in this particular equation, since it does not affect the overall order of the numerical scheme.) The anchor condition (14) is also discretized by Simpson's rule to give

$$\sum_{j=1}^{N} \frac{\Delta t_j}{6} [(u_{j-1} - v_{j-1})^T f(v_{j-1};\lambda_0) + \\ + 4(u_{j-1/2} - v_{j-1/2})^T f(v_{j-1/2},\lambda_0) + (u_j - v_j)^T f(v_j;\lambda_0)] = 0 \tag{23}$$

except at a Hopf bifurcation point, where we replace (23) by the corresponding discretization of (19). Equations (20)-(23) are the discretized form of (12).

Application of Newton's method to equation (12) requires the solution of linear systems of the form

$$\begin{pmatrix} C_{\underline{u}}^{\nu} & C_{\rho}^{\nu} & C_{\lambda}^{\nu} \\ B_{\underline{u}}^{\nu} & 0 & 0 \\ N_{\underline{u}}^{\nu} & N_{\rho}^{\nu} & N_{\lambda}^{\nu} \\ A_{\underline{u}}^{\nu} & 0 & 0 \end{pmatrix} \begin{pmatrix} \Delta u \\ \Delta \rho \\ \Delta \lambda \end{pmatrix} = - \begin{pmatrix} C^{\nu} \\ B^{\nu} \\ N^{\nu} \\ A^{\nu} \end{pmatrix}, \quad \nu = 0,1,2,\ldots \tag{24}$$

Here $C_{\underline{u}}$ is a matrix of dimension $2Nn$ by $(2N + 1)n$, having block-diagonal structure. Each block can be subdivided into 6 square matrices of dimension n, that is,

$$w'_{j,i}(\xi_{j,k})I_n - \frac{\rho^{\nu}}{2\pi} w_{j,i}(\xi_{j,k}) f_u(p_j^{\nu}(\xi_{j,k}); \lambda^{\nu}) ,$$

where I_n is the n by n identity matrix, and

$$p_j^{\nu}(t) = w_{j-1}(t)u_{j-1}^{\nu} + w_{j,-1/2}(t)u_{j-1/2}^{\nu} + w_{j,0}(t)u_j^{\nu} .$$

C_{ρ} and C_{λ} are column vectors of dimension $2Nn$; $N_{\underline{u}}$ and $A_{\underline{u}}$ are row vectors of dimension $(2N + 1)n$; while N_{ρ} and N_{λ} are scalars.

With the Jacobian matrix partitioned, the linear system to be solved for each Newton iteration has the form

$$\begin{pmatrix} A & B \\ C & D \end{pmatrix} \begin{pmatrix} x_1 \\ x_2 \end{pmatrix} = \begin{pmatrix} h_1 \\ h_2 \end{pmatrix} .$$

An efficient solution proceeds as follows: First, decompose the matrix as

$$\begin{pmatrix} A & B \\ C & D \end{pmatrix} = \begin{pmatrix} L & 0 \\ Q & I \end{pmatrix} \begin{pmatrix} U & P \\ 0 & T \end{pmatrix} ,$$

i.e., decompose A into $A = LU$, solve

$$LP = B$$

$$U^T Q^T = C^T$$

and set

$$T = D - QP \ .$$

Then find intermediate quantities ψ_1 and ψ_2 by solving

$$L\psi_1 = h_1$$

and letting

$$\psi_2 = h_2 - Q\psi_1 \ .$$

Finally obtain the solution from

$$Tx_2 = \psi_2$$

$$Ux_1 = \psi_1 - Px_2 \ .$$

Pivoting is generally necessary to solve the next to the last equation with matrix T. Note that we can compute the LU-decomposition of A very rapidly due to its simple structure. The indicated algorithm for solving the linearized system is not necessarily the most stable although it has been used successfully in a variety of problems. Other schemes should be considered also.

In order for Newton iteration to converge to the intended solution of (24) an accurate initial approximation must be provided. In general this can be done by extrapolating from the two preceding points on the solution branch. At a Hopf bifurcation point the asymptotic expansions (15)-(17) can provide the initial approximation.

RESULTS

We now present some of the more interesting results obtained by using the above numerical methods to study the dynamics of a CSTR with $A \rightarrow B \rightarrow C$ kinetics which arise from varying the Damköhler number. These results exhibit dynamic phenomena which are much more complex than the $A \rightarrow B$ case and illustrate the utility of our techniques in tracing entire periodic solution branches containing unstable oscillations and multiple turning points.

For a particular combination of system parameters [36], we summarize the multiple steady and oscillatory states on a response curve with the Damköhler number as the abscissa

and the L_2 norm of the steady state or periodic solution as the ordinate. The first example is illustrated in Fiure 1 which corresponds to the parameter values $B = 9.0$, $\alpha = 1.0$, $\beta = 2.0$, and $\sigma = 0.01$. For all values of the Damköhler number, the steady state is unique with exchanges in stability at the four Hopf bifurcation points. The application of the switching and continuation procedures discussed in the previous section enabled us to trace one branch of periodic solutions joining the orbits bifurcating from the first and second points and another branch joining the orbits from the third and fourth points. With the notable exception of two regions of oscillatory states, this example is similar to the $A \rightarrow B$ CSTR. As D is increased the norm of the steady state solution increases until the first Hopf point ($D = 0.243$) is crossed and the reactor begins to oscillate. The oscillations cease as the second point ($D = 0.29$) is reached. Then the steady state value of $\|u\|$ increases with D and the same dynamic pattern is repeated.

The results presented in Figure 2 are certainly not comparable to those found in simpler reactors. In this case we again find a unique steady state branch containing four Hopf points, but now the orbits bifurcating from the second and third points are joined together as are the solutions from the first and fourth points. Close examination of the inner branch reveals that both solutions bifurcate to the left of the Hopf point with the lower bifurcation directed nearly straight up with a turning point located close to the steady state branch. The periodic branch joining the first and fourth Hopf points is even more complex. Again, both bifurcations occur to the left, and the enlargement in the figure shows that the branch actually contains three turning points at $D = 0.1954$, $D = 0.2003$ and $D = 0.1997$. Our techniques were able to compute all the periodic solutions (both stable and unstable) along the branch and continue around the turning points without difficulty. (Note the intersection of the periodic and steady state branches at $D = 0.246$ is not a Hopf bifurcation point but simply a point where the norms of the steady state and the periodic solutions become identical; a stablility exchange does not accompany the intersection.)

The location and direction of the bifurcations in Figure 2 agree with the asymptotic results of Halbe and Poore [36]. Their stability analysis indicates that the solutions

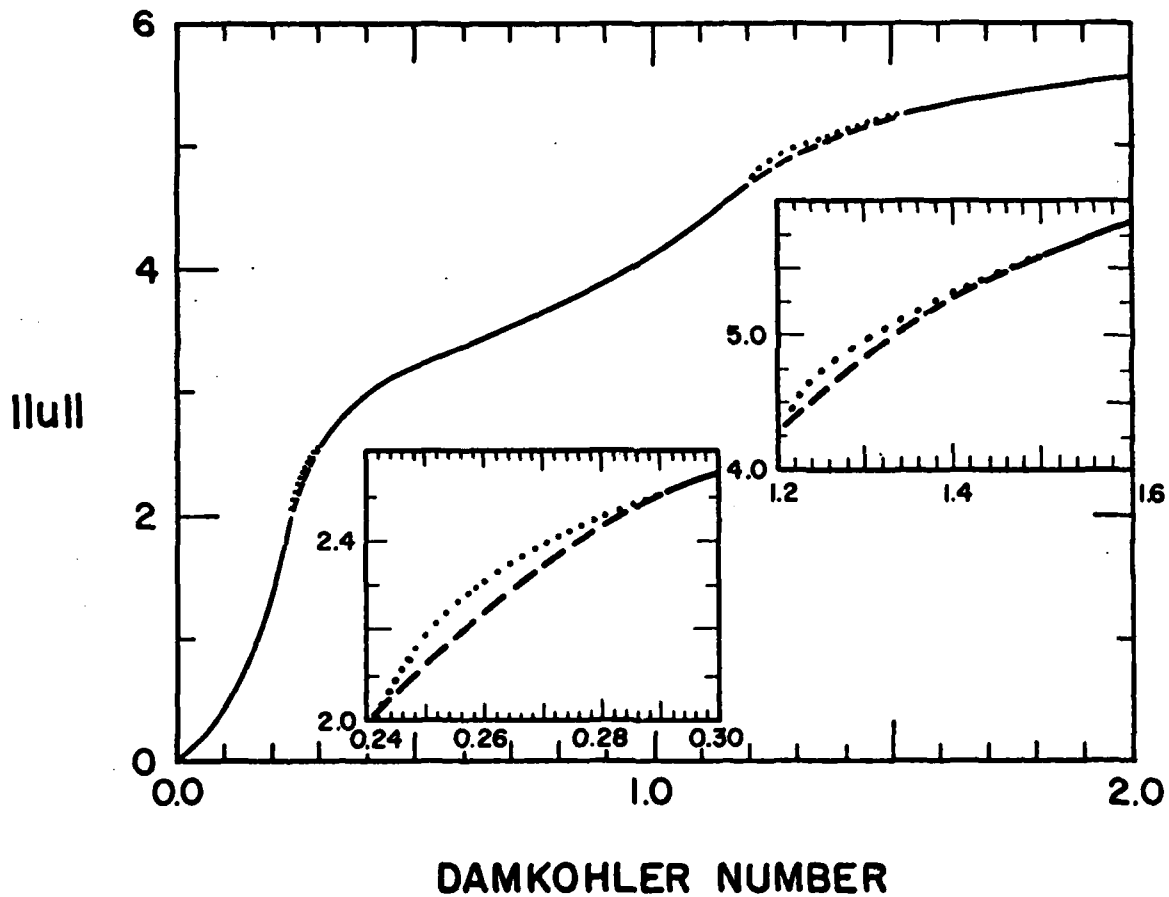

Figure 1. Computation of Entire Periodic Branches ($B = 9.0$, $\alpha = 1.0$, $\beta = 2.0$, $\sigma = .01$)

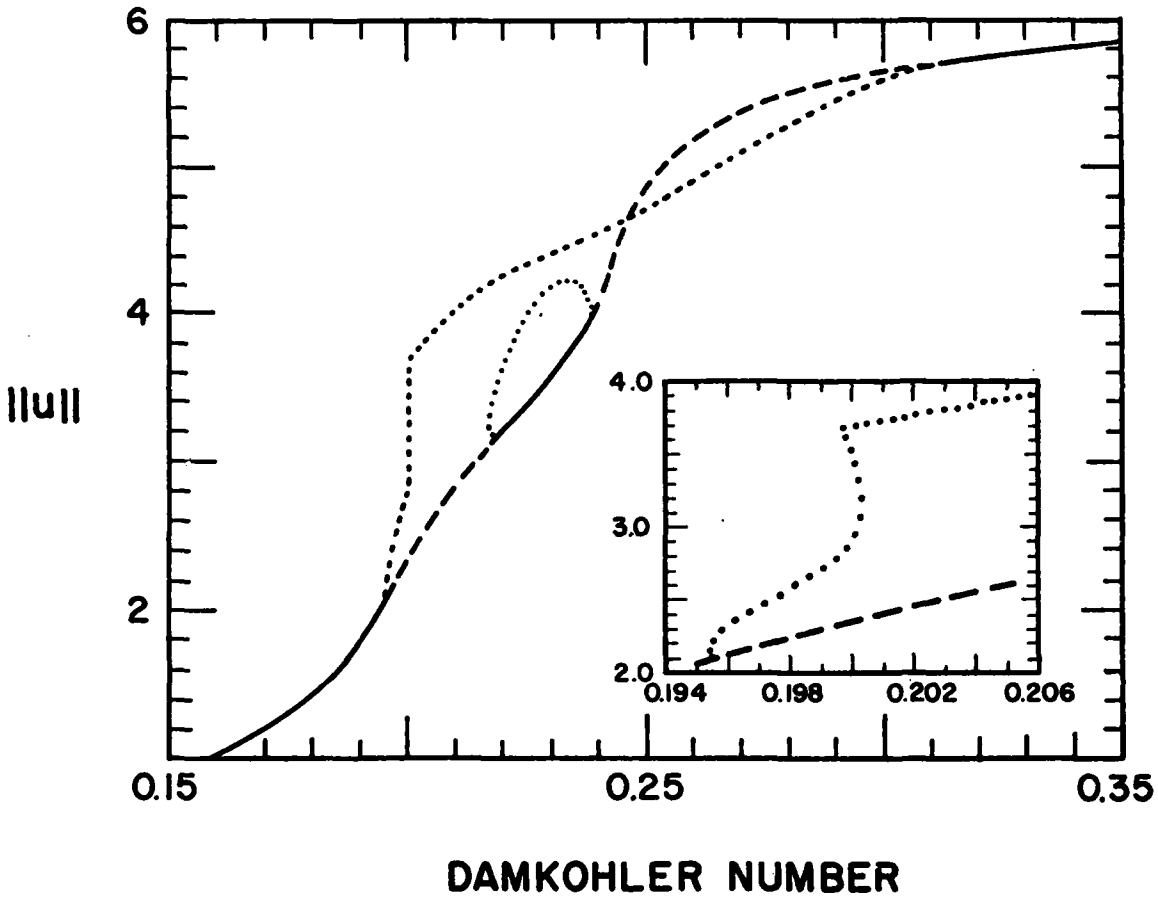

Figure 2. Response Diagram Illustrating Two Periodic Branches with Multiple Turning Points ($B = 8.0$, $\alpha = 1.0$, $\beta = 1.5$, $\sigma = 0.4$)

bifurcating from the first and third points are unstable while the solutions splitting from the other two points are stable. Given these results and assuming stability exchanges along the periodic branches occur only at the turning points (preliminary calculations confirm this assumption), we can explain the complex dynamic behavior exhibited by the reactor. Consider first the reactor operating at steady state with D = 0.23, and note that this state is surrounded by a large amplitude, stable limit cycle and a smaller amplitude, unstable limit cycle. As D is decreased, we remain on the steady state branch until the Hopf point (D = 0.2186) is crossed at which time, the reactor moves out along the periodic branch and begins to oscillate. The reactor operates in this periodic mode until the turning point at D=0.2172 is encountered, and if D is decreased below this value, the reactor must jump to the stable, larger amplitude oscillations above. Transitions from periodic states at turning points are often seen in the $A \rightarrow B$ CSTR [7] but in that case the reactor must always jump to a lower, stable steady state.

Jump transitions between periodic states can occur in a slightly different fashion along branch joining the first and fourth Hopf points. If we further decrease D from 0.217 to just below 0.1997, the reactor will drop from the upper solution to another stable orbit of smaller amplitude. The opposite effect is of course achieved by now increasing D past the turning point at 0.2003 so that the hystersis phenomena associated with multiple steady states now occurs with oscillatory states. S-shaped branches of periodic solutions are not reported in earlier studies and could explain similar periodic transitions reported in the tubular reactor with an $A \rightarrow B$ reaction [8].

The next three figures locate the concentration and temperature at various points along the solution branch and plot the variables as time ranges from 0 to 2π. As the reactor moves around the branch, the amplitude of the solutions becomes larger and the temperature oscillation becomes increasing complex.

Figure 6 illustrates a response curve containing again four bifurcation points and now a region of three steady states. The first two Hopf points are tied together by a solution branch like those in Figure 1, but the orbits emanating from the third and fourth points are not connected. In the case of the third Hopf point, the period of the oscillations

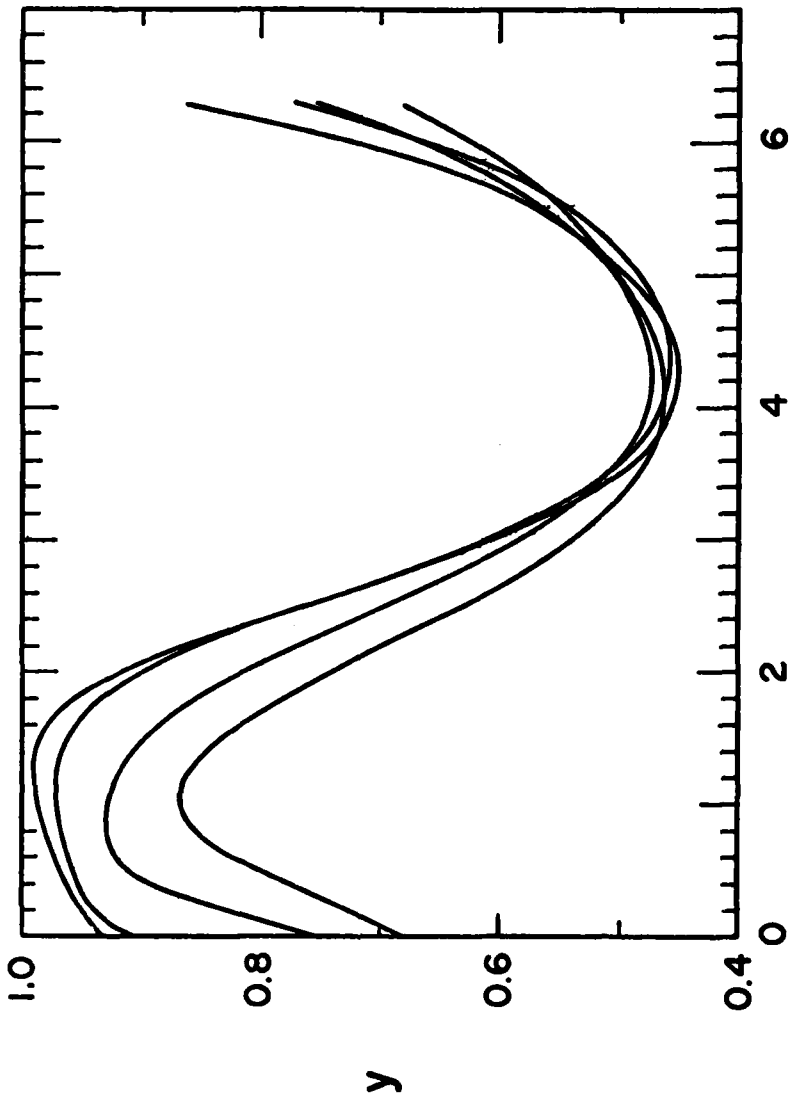

Figure 3. Concentration of A vs. Time for the Parameters in Figure 2

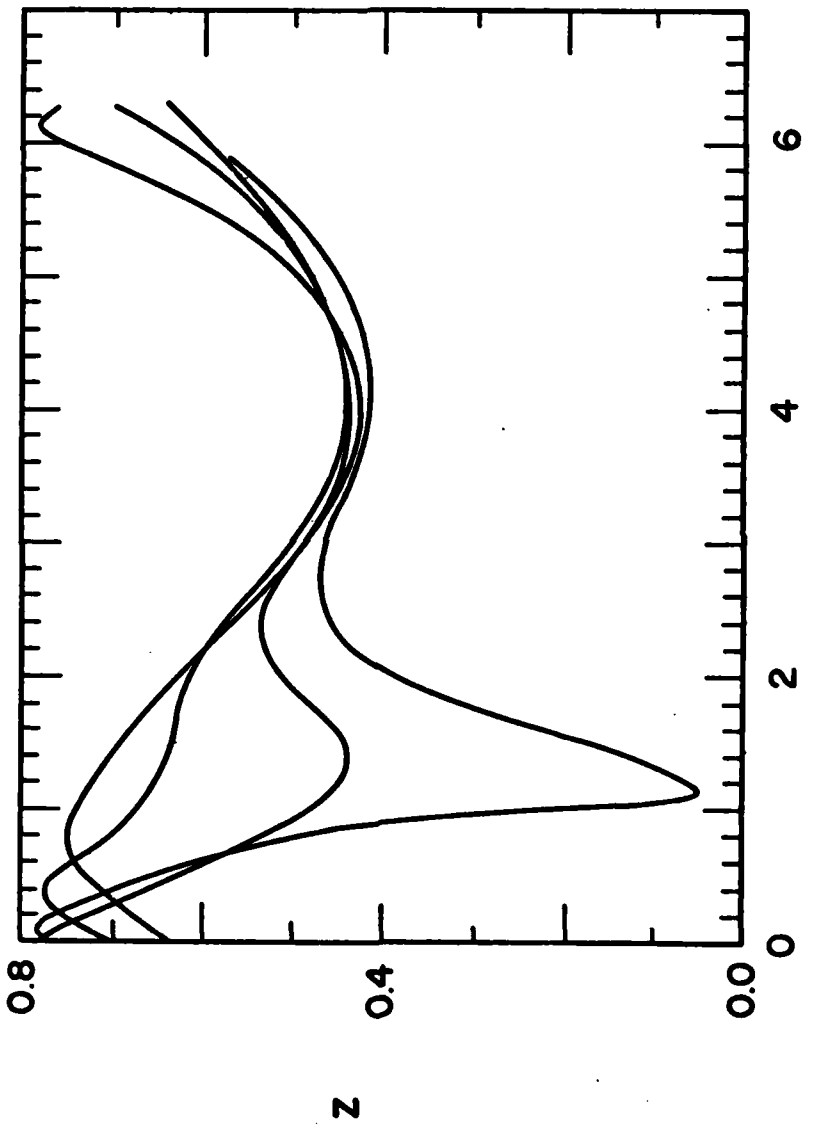

Figure 4. Concentration of B vs. Time

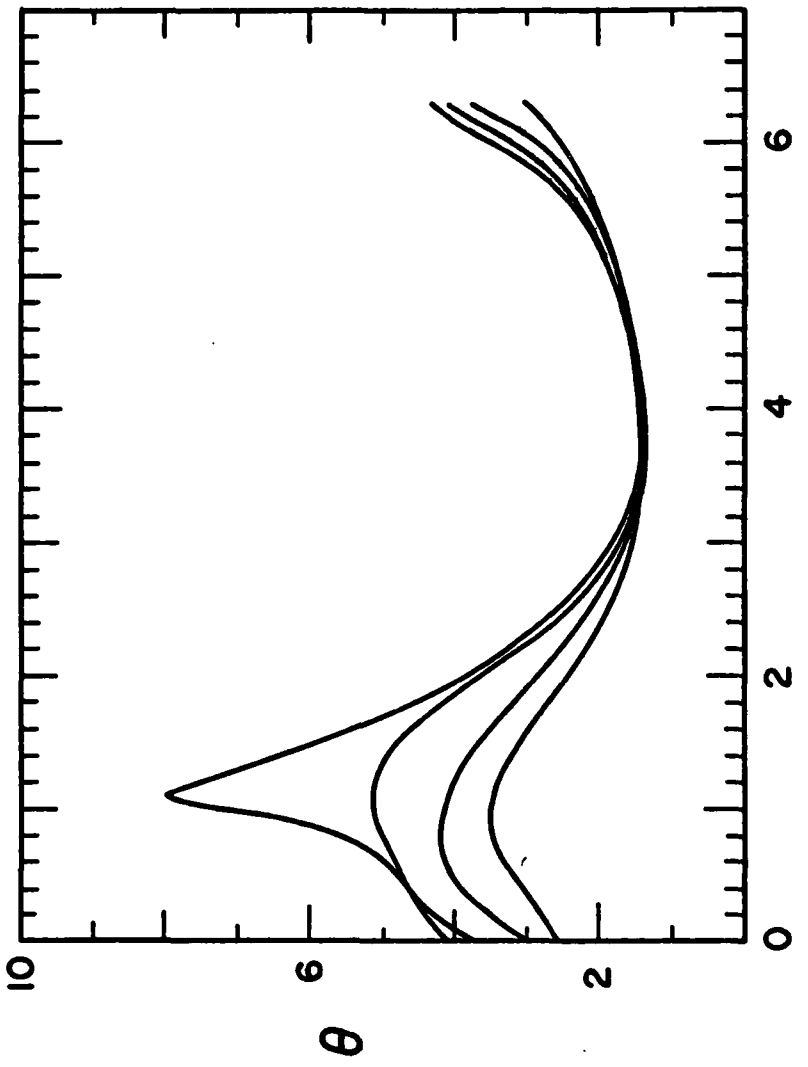

Figure 5. Temperature vs. Time

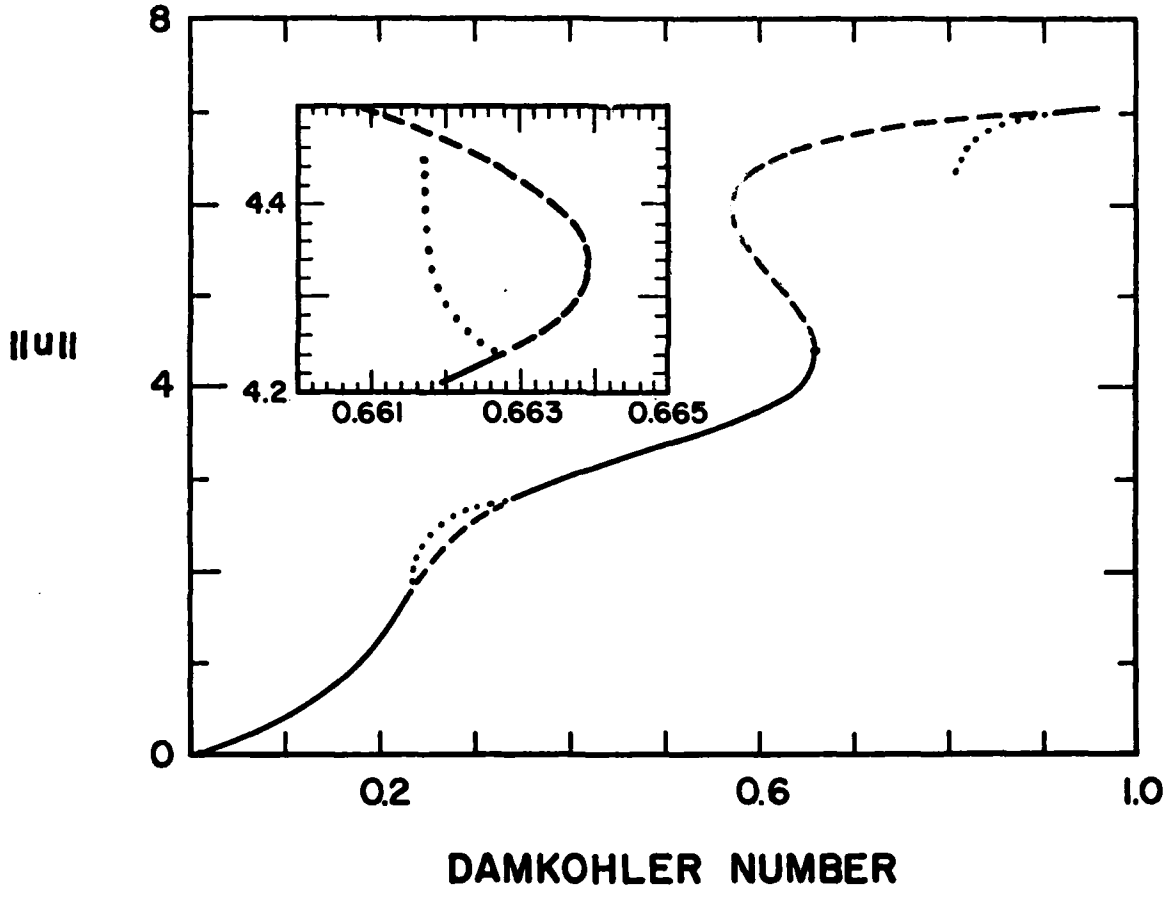

Figure 6. Response Diagram with Three Periodic Branches
(B = 8.0, α = 1.4, β = 2.0, σ = .04)

increases as one moves out along the branch and becomes infinite as the branch terminates. The next figure illustrates the increasing amplitude of the limit cycles as the orbits apparently approach a separatrix in a complete phase plane. (We have not traced all the branches to completion but simply illustrate the utility of the numerics in such computations. For example, the period of solutions at the third Hopf point is 13.81 which increases to 175.64 at the last point shown on this branch.) The occurance of "short" branches which bifurcate very near a steady state limit point and contain solutions whose period approach infinity are found quite often in this problem. A recent theoretical analysis of this particular type of bifurcation phenomenon is presented by Keener [42].

The example in Figure 8 shows a steady state branch containing a region of three solutions and also three Hopf bifurcation points. Periodic reactor operation is possible for a limited region of D between the second and third Hopf points and for an extremely narrow range of D between the first bifurcation point and the turning point contained on the first periodic branch.

The results in Figures 9 and 10 are similar with the first diagram exhibiting five steady states and two branches of oscillatory solutions which do not connect. (The steady state branch which crosses the raised x-axis in Figure 9 continues down to a limit point at $D = 0.062$ and $\|u\| = 1.25$ and then turns back to the origin.) The response curve in the next figure again contains two separate periodic branches as the steady state branch is stretched into a 1-3-1-3-1 pattern. The most interesting result in this case is the direction of the periodic branches toward the steady limit points. The periodic branches (presumably stable) appear to become nearly tangent to the limit points insuring the existence of a stable solution for all values of D. As in all other examples of this type of branch, the period of the oscillation becomes increasing large as we move away from the bifurcation point. Analytical results concerning the above phenomenon have recently been obtained by Golubitsky and Langford [43, 44].

The last figure shows a 1-3-1 steady state curve with two periodic branches. The second branch in this example bifurcates to the right but then sharply turns back to the left at $D = 0.136$ creating of course possible jumps between oscillatory states and high

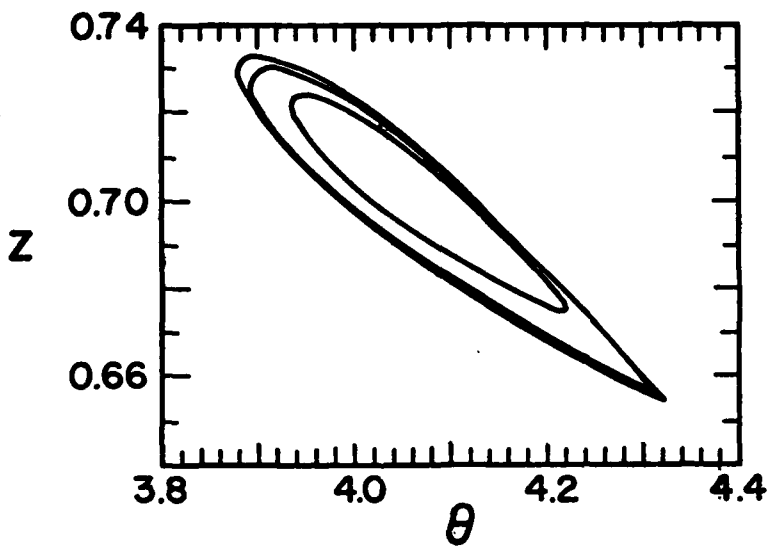

Figure 7. Phase Plot at Points Along the Upper Branch from Figure 6

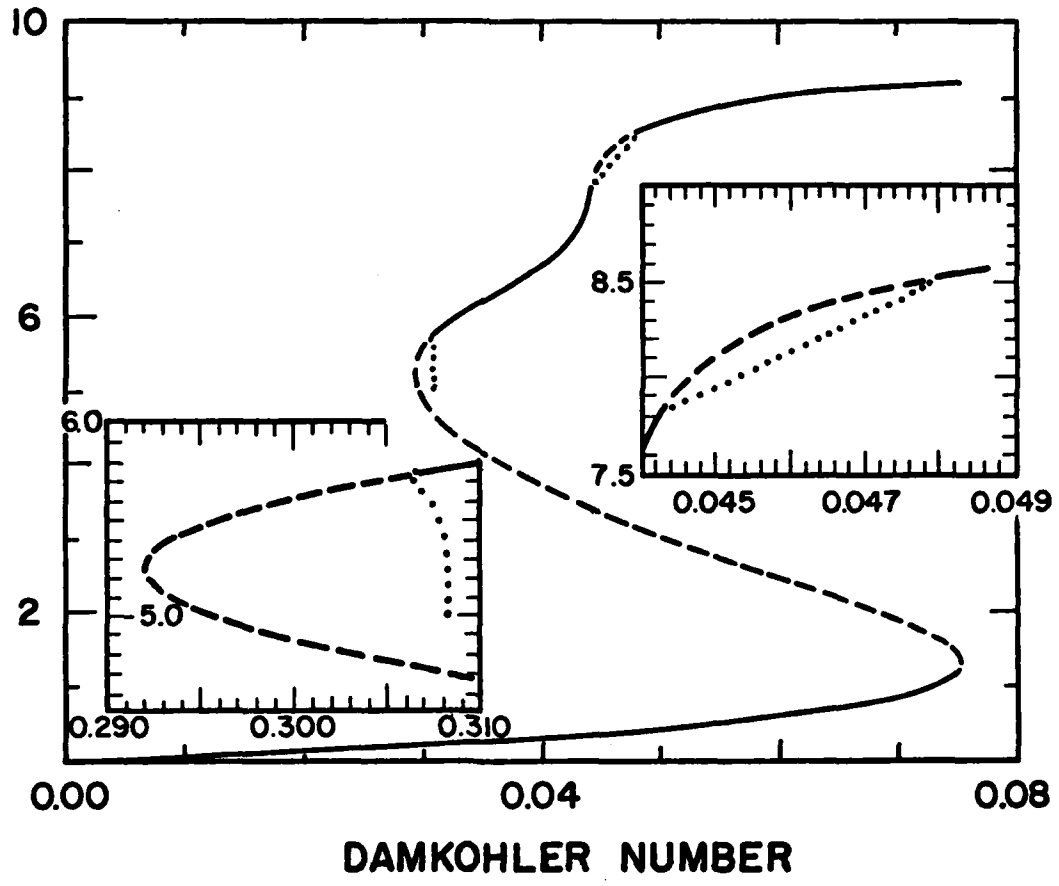

Figure 8. Response Diagram with Parameters (B = 12.0, α = 0.6, β = 1.0, σ = .01)

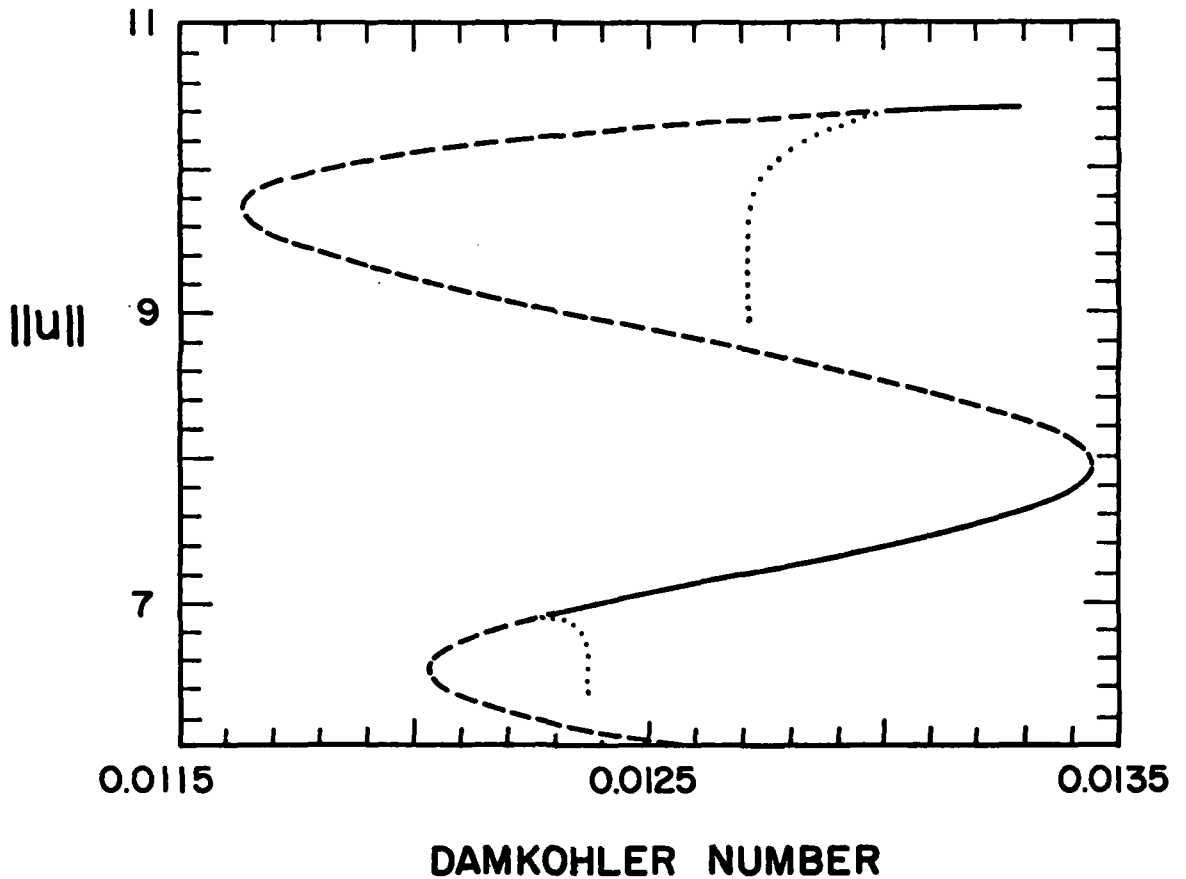

Figure 9. Response Diagram Exhibiting Five Steady States and Two Periodic Branches ($B = 14.0$, $\alpha = 0.6$, $\beta = 1.0$, $\sigma = .01$)

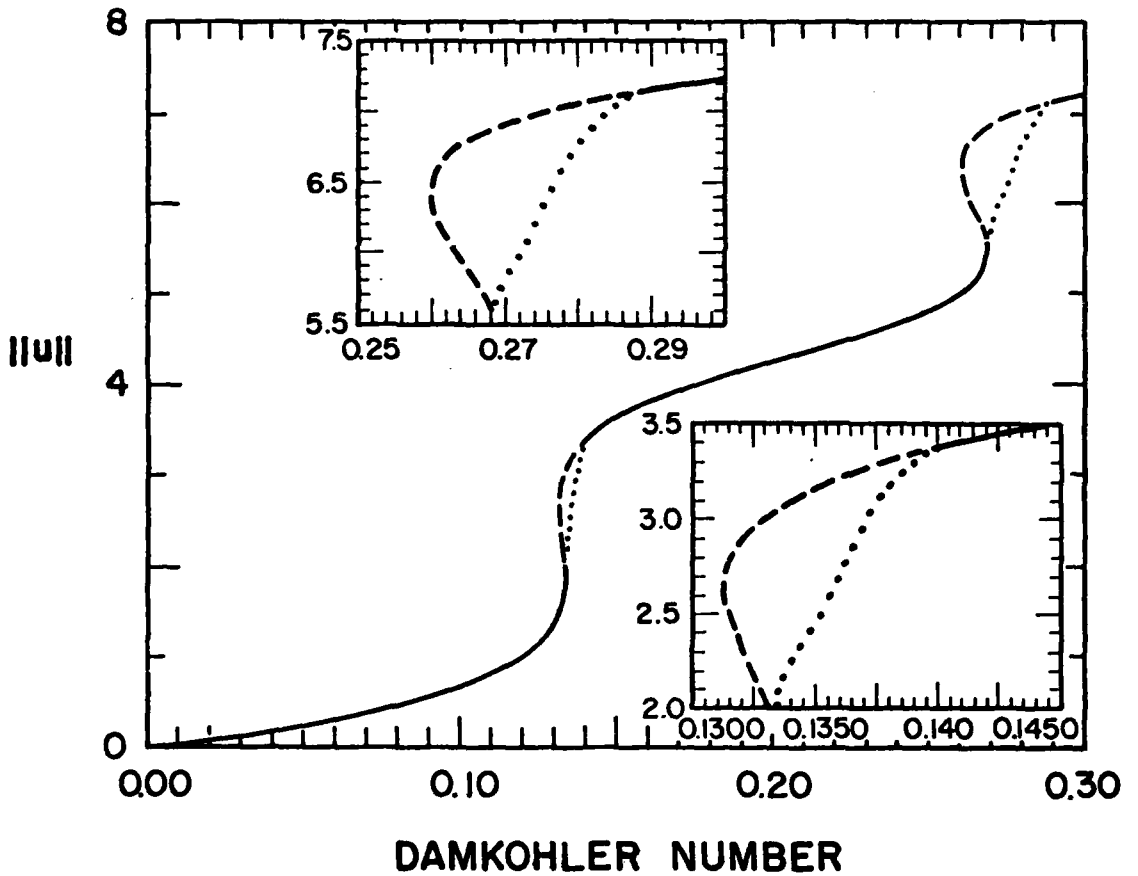

Figure 10. Response Diagram Illustrating 1-3-1-3-1 Multiplicity with Two Periodic Solutions ($B = 8.0$, $\alpha = 1.0$, $\beta = 1.0$, $\sigma = .01$)

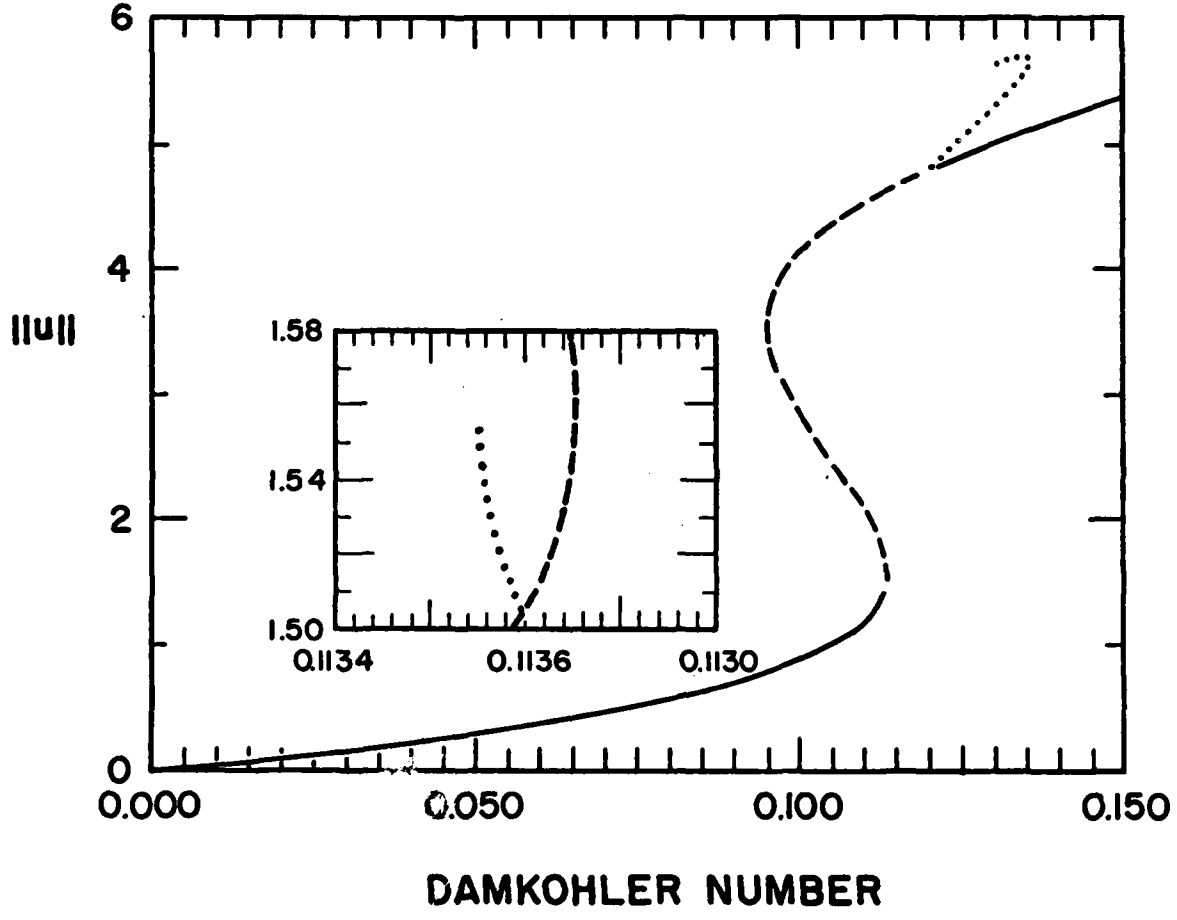

Figure 11. Response Diagram for Parameters B = 15, α = .05, β = 2.4, σ = .03. (Note Turning Point on Periodic Solution)

temperature (or conversion) steady states. While the existence of turning points on solution branches is often seen in the CSTR case, they have not been reported earlier on branches which do not join.

CONCLUSION

We present a simple continuation technique for branches of periodic solutions. This technique is easy to numerically implement and allows computation to proceed past turning points as well as along unstable branches. Without essential change the procedure can be used to switch solution branches at the Hopf bifurcation points. The techniques were applied to the CSTR with consecutive $A \rightarrow B \rightarrow C$ reactions. The computational results reveal several types of dynamic behavior not reported in earlier studies of this reactor or the simpler $A \rightarrow B$ case.

It is to be emphasized that the numerical methods presented here are completely general for nonlinear autonomous systems of differential equations. Extension of the method to more general operator equations than those discussed in this paper is not difficult, in principle. We believe these techniques can find wide application in the bifurcation analysis of mathematical model- in several fields of research.

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We present a continuation technique for branches of periodic solutions which can be applied to autonomous systems of nonlinear differential equations. Coupling the technique with Hopf bifurcation expansions enables one to compute entire periodic solution branches including those with turning points and unstable solutions. We apply these methods to a continuously

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ABSTRACT (cont.)

stirred tank reactor with consecutive $A \rightarrow B \rightarrow C$ reactions. Our computations reveal dynamic phenomena not seen in previous reactor studies. The results include response diagrams exhibiting stable and unstable periodic branches that contain multiple turning points. The discovery of these points indicates the reactor may jump from a steady state to a periodic orbit or may jump from one periodic orbit to another.


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