Liepmann, H. W., and J. Laufer, "Investigation of Free Turbulent Mixing," *NACA Tech. Note No. 1257* (1947).
Lilly, G. P., "Effect of Particle Size on Particle Eddy Diffusivity," *Ind. Eng. Chem. Fundamentals*, **12**, 268 (1973).
Peskin, R. L., "Some Effect of Particle—Particle—Fluid Interactions in Two-phase Flow Systems," Ph.D. thesis, Princeton Univ., N.J. (1959).
———, "The diffusivity of Small Suspended Particles in Turbulent Fluids," National Meeting *AIChE*, Baltimore, Md. (1962).
Popper, J., N. Abuaf, and G. Hetsroni, "Velocity Measurements in a Two-Phase Turbulent Jet," *Intern. J. Multiphase Flow*, **1**, 715 (1974).
Ribeiro, M. M., and J. H. Whitelaw, "Statistical Characteristics of a Turbulent Jet," *J. Fluid Mech.*, **70**, 1 (1975).
Riley, J., and S. Corrsin, "Simulation and Computation of Dispersion in Turbulent Shear Flow," Conf. on Air Poll. Meteorology, Raleigh, N.C. (1971).
Smith, R. H., and C. T. Wang, "Contracting Cones Giving Uniform Throat," *J. Aero Sci.*, **11**, 356 (1944).
———, "An Account of Some Feature of the Phenomena in Fully Turbulent Regions," *Proc. of Iowa Thermodynamics Symposium* (1953).
Snyder, W. H., and J. L. Lumley, "Some Measurements of Particle Veocity Autocorrelation Functions in a Turbulent Flow," *J. Fluid Mech.*, **48**, 41 (1971).
Soo, S. L., and R. L. Peskin, "Statistical Distribution of Solid Phase in Two-Phase Turbulent Mortion," *Project Squid Tech. Rept. PR-80-R* (1958).
Tanaka, Z., and K. Iinoya, "Particle Size Classification by Deposition Angle in a Gas Centrifuge at Reduced Pressure," *J. Chem. Eng. Japan*, **4**, 167 (1971).
Taylor, G. I., "Diffusion by Continuous Movements," *Proc. London Math. Soc.*, **20**, Ser. 2, 196 (1921).
Tchen, C. M., "Mean Value and Correlation Problems Connected with the Motion of Small Particles in a Turbulent Fluid," Dissertation-Delft, The Hague, Martinus Nijhoff (1947).
Tollmien, W., "Rerechnung Turbulenter Ausbreitungsvorgage," *ZAMM*, **6**, 468 (1926).
Townsend, A. A., *The Structure of Turbulent Shear Flow*, Cambridge Univ. Press, Cambridge, England (1962).
Van Der Hegge Zijnen, B. G., "Measurements of the Velocity Distribution in a Plane Turbulent Jet of Air," *Appl. Sci. Res.*, **A7**, 256 (1958).
———, "Measurement of the Distribution of Heat and Matter in a Plane Turbulent Jet of Air," *ibid.*, 277 (1958).
Wygnanski, I., and H. Fiedler, "Some Measurements in the Self-Preserving Jet," *J. Fluid Mech.*, **38**, 577 (1969).

Manuscript received August 29, 1977; revision received January 3, and accepted January 5, 1978.

A Modal Approach to Dynamics of Nonlinear Processes

ROBERT J. FISHER
and
MORTON M. DENN
Department of Chemical Engineering
University of Delaware
Newark, Delaware 19711

Approximate process dynamics of certain nonlinear systems can be estimated by elementary quadratures using a modal approach. The transient response including quadratic nonlinearities is determined by the eigenvalues, eigenvectors, and adjoint eigenvectors of the linearized system equations. The only restriction is that the dominant eigenvalue of the linearized system must be widely separated from the next slowest mode. Several process models satisfy this requirement.

The method is illustrated by application to a fourth-order model of a fluidized bed. The dynamical response is in agreement with numerical solutions to the complete model equations, including estimates of finite regions of stability.

SCOPE

The field of process dynamics has been concerned almost exclusively with the response of linear systems. This restricts the analytical understanding of the response of nonlinear processes to a small region about the steady state where a linear approximation can be assumed to be valid. There are two difficulties associated with this restriction: the analyst has no a priori knowledge of the limits of applicability of the linear analysis, and information about process behavior beyond the linear region must be obtained by direct numerical simulation. Numerical simulation can be tedious for systems of high order and/or with many parameters. Numerical simulation is particularly difficult when the system time constants are widely spaced.

This paper describes a procedure by which analytical estimates of the dynamics of nonlinear processes can be obtained for systems in which the slowest time constant is widely separated from the other time constants. The procedure is an extension of the method of modal analysis and retains the multiple time scales of the full nonlinear process. The only computational information required is obtainable from the structure of the linearized system equations.

Robert J. Fisher is with Mobil Research and Development Corp., Paulsboro, N.J. 08066.

0001-1541-78-1137-0519.

CONCLUSIONS AND SIGNIFICANCE

The extension of the method of modal analysis to those nonlinear processes to which it is applicable provides a useful analytical tool for estimating the limits of linearization, regions of finite stability, and the general transient response of the process following finite perturbations. The analytical estimates of nonlinear process response have been compared quite favorably with detailed (and tedious) numerical solutions for a model of a batch catalytic fluidized bed.

The field of process dynamics has been concerned almost exclusively with the response of linear systems. This restriction is a logical consequence of the general availability of analytical tools only for systems of linear differential equations, and it leads to two related difficulties. The first is simply that the analyst has no a priori knowledge of the limits of applicability of the linear analysis. The second is that any information about process behavior beyond the linear region must be obtained by direct numerical simulation, which can be tedious for systems of high order and/or with many parameters.

There is a class of lumped parameter process models for which analytical estimates of nonlinear process dynamics can be readily obtained by extension of the method of modal analysis. The requirement for application of this method to nonlinear systems is that the first two eigenvalues of the linearized process model be widely separated. This requirement is certainly restrictive, but such systems do exist, and in these cases the nonlinear dynamics can be estimated using only information required for the linearized dynamic analysis.

NONLINEAR MODAL ANALYSIS

The general procedure for nonlinear modal analysis is described in Denn (1975) and follows from work of Eckhaus (1965) on the finite-amplitude stability of fluid mechanical systems. We consider a process whose dynamics about an equilibrium point $\mathbf{x} = \mathbf{0}$ are described by the set of differential equations

$$\frac{dx_i}{dt} = f_i(\mathbf{x}); \quad i = 1, 2, \ldots, N \tag{1}$$

In the neighborhood of $\mathbf{x} = \mathbf{0}$, we expand in a Taylor series through at least second order in $|\mathbf{x}|$ to obtain

$$\frac{dx_i}{dt} = \sum_{j=1}^{N} B_{ij}x_j + \sum_{j,k=1}^{N} C_{ijk}x_jx_k + o(|\mathbf{x}|^2) \tag{2}$$

Conventional linear process dynamics is concerned with the eigenvalues $\{\lambda_n\}$ and eigenvectors $\{\mathbf{y}_n\}$ of the matrix $\mathbf{B}$, which are defined in terms of the characteristic equation

$$\sum_{j=1}^{N} B_{ij}y_{nj} = \lambda_n y_{ni}; \quad i = 1, 2, \ldots, N; \quad n = 1, 2, \ldots, N \tag{3}$$

We assume that the eigenvalues are distinct. The eigenvectors are most conveniently normalized to unit magnitude

$$\sum_{i=1}^{N} y_{ni}y_{ni} = 1 \tag{4}$$

We also need the adjoint set of eigenvectors $\{\boldsymbol{\eta}_n\}$ defined by the characteristic equation

$$\sum_{i=1}^{N} \eta_{ni}B_{ij} = \lambda_n\eta_{nj}; \quad i = 1, 2, \ldots, N; \quad n = 1, 2, \ldots, N \tag{5}$$

The adjoint eigenvectors are orthonormal to the basic eigenvectors

$$\sum_{i=1}^{N} \eta_{ni}y_{mi} = \begin{cases} 0, & n \neq m \\ 1, & n = m \end{cases} \tag{6}$$

In modal analysis, the dynamical response is recast in terms of the temporal modes of the system. The eigenvectors $\mathbf{y}_n$ are linearly independent and form a basis in the $\mathbf{x}$ space, so the vector $\mathbf{x}$ can be expressed as a linear combination of the $\{\mathbf{y}_n\}$ as follows:

$$x_i(t) = \sum_{n=1}^{N} A_n(t)y_{ni} \tag{7}$$

The equivalence between the N components of $\mathbf{x}$ and the N modal variables A_n is established through the adjoint system by means of the orthogonality relation, Equation (6):

$$A_m(t) = \sum_{i=1}^{N} x_i(t)\eta_{mi} \tag{8}$$

Equation (2) can then be rewritten in terms of the modal variable $A_n(t)$ by substitution of Equations (3) through (8) to yield

$$\frac{dA_p}{dt} = \lambda_pA_p + \sum_{n,m=1}^{N} I_{pnm}A_nA_m + o(|\mathbf{A}|^2) \tag{9}$$

with

$$I_{pnm} = \sum_{i,j,k=1}^{N} C_{ijk}\eta_{pi}y_{nj}y_{mk} \tag{10}$$

We now make several assumptions. The first is that the eigenvalues λ_p are all real. This assumption is not critical and can be relaxed. Next, we assume that all eigenvalues are negative, so that they can be ordered $0 > \lambda_1 > \lambda_2 > \ldots$ This assumption limits consideration to steady states that are stable to infinitesimal disturbances. We then make two critical assumptions. The first is

$$|\lambda_1/\lambda_p| << 1; \quad p = 2, 3, \ldots, N \tag{11}$$

and the second is that the modal functions $A_p(t)$ are roughly ordered in magnitude according to the reciprocals of the eigenvalues. It follows from these assumptions

(Denn, 1975) that the only term which needs to be retained in the double summation in Equation (9) is the one corresponding to $n = m = 1$, which effectively uncouples the equations:

$$\frac{dA_p}{dt} = \lambda_p A_p + I_{p11} A_1^2 + o(|\mathbf{A}|^2); \quad p = 1, 2, \ldots, N \tag{12}$$

An analytical solution is then immediately obtainable:

$$A_1(t) = A_1(0)\exp(\lambda_1 t) \left\{ 1 + \frac{A_1(0) I_{111}}{\lambda_1} [1 - \exp(\lambda_1 t)] \right\}^{-1} \tag{13}$$

$$A_p(t) = A_p(0)\exp(\lambda_p t) + I_{p11} \int_0^t \exp [\lambda_p (t - \tau)] A_1^2(\tau) d\tau; \quad p = 2, 3, \ldots, N \tag{14}$$

The initial conditions $\mathbf{A}(0)$ are computed from $\mathbf{x}(0)$ by means of Equation (8), and the state variables $\mathbf{x}(t)$ are recovered from Equations (13) and (14) by means of Equation (7). It should be noted that Equations (13) and (14) explicitly retain the multiple time scales characteristic of the original process equations.

LINEARIZATION AND STABILITY LIMITS

Equations (12) to (14) provide an explicit estimate of the range of validity of a linearized dynamical analysis. The linear terms dominate the equations only as long as

$$|A_1(0)| << |\lambda_1 / I_{111}| \tag{15}$$

Since the first mode dominates the response we take $|\mathbf{A}| \approx |A_1|$ and, from Equation (7), obtain

$$|x_i(0)| << |y_{1i}| \, |\lambda_1 / I_{111}| \tag{16}$$

This condition is somewhat conservative in that it does not account for nonzero initial values of other rapidly decaying modes which will extend the range somewhat.

Equation (13) also provides an estimate of the region of stability about the steady state. All deviations from the steady state decay as long as $A_1(0) I_{111}/\lambda_1 < 1$ and grow without bound for $A_1(0) I_{111}/\lambda_1 > 1$. It then follows from Equation (8) that the system is stable for disturbances which lie within the half space

$$\frac{I_{111}}{\lambda_1} \sum_{i=1}^{N} \eta_{1i} x_i \leq 1 \tag{17}$$

Terms through third order in Equation (2) would be required to provide a good estimate of a closed region of stability; the expansion is commonly carried out to third order for problems in fluid mechanics (Denn, 1975; Eckhus, 1965).

APPLICATION: FLUIDIZED BED

The technique for estimating nonlinear dynamics can be illustrated with an example of a model of a batch catalytic fluidized bed studied by Luss and Amundson (1968). The physical assumptions are given in the original paper and are not repeated here. The system equations are

$$\frac{dP}{d\tau} = P_e - P + H_g(P_p - P) \tag{18a}$$

$$\frac{dT}{d\tau} = T_e - T + H_w(T_w - T) + H_T(T_p - T) \tag{18b}$$

$$A \frac{dP_p}{d\tau} = H_g(P - P_p) - H_g K k P_p \tag{18c}$$

$$C \frac{dT_p}{d\tau} = H_T(T - T_p) + H_T F K k P_p \tag{18d}$$

$$k = k_o \exp(-\Delta E / R T_p) \tag{18e}$$

Here P and T denote the gas partial pressure and temperature, respectively; P_p and T_p the partial pressure and temperature in the catalyst particle; and P_e and T_e the partial pressure and temperature of the entering system. The values of the dimensionless parameters used by Luss and Amundson and in the present study are given in Table 1, and the corresponding steady states are tabulated in Table 2.

Perturbation variables are defined as follows:

$$x_1 = P - P_s \qquad x_2 = T - T_s$$
$$x_3 = P_p - P_{ps} \qquad x_4 = T_p - T_{ps} \tag{19}$$

Here, the subscript s denotes the steady state value. A Taylor series expansion of Equations (18) through second order then gives

$$\frac{dx_1}{d\tau} = -(H_g + 1)x_1 + H_g x_3 \tag{20a}$$

$$\frac{dx_2}{d\tau} = -(1 + H_w + H_T)x_2 + H_T x_4 \tag{20b}$$

$$\frac{dx_3}{d\tau} = \frac{H_g}{A} x_1 - \left(\frac{H_g P_s}{A P_{ps}}\right) x_3 - \left[\frac{H_g (T_s - T_{ps}) \Delta E}{A F R T_{ps}^2}\right] x_4 - \left[\frac{H_g (T_s - T_{ps}) \Delta E (\Delta E - 2 R T_{ps})}{2 A F R^2 T_{ps}^4}\right] x_4^2 \tag{20c}$$

$$\frac{dx_4}{d\tau} = \frac{H_T}{C} x_2 + \left[\frac{H_T F (P_s - P_{ps})}{C P_{ps}}\right] x_3 + \frac{H_T}{C}\left[\frac{\Delta E (T_{ps} - T_s)}{R T_{ps}^2} - 1\right] x_4 + \left[\frac{H_T (T_{ps} - T_s) \Delta E}{C P_{ps} R T_{ps}^2}\right] x_3 x_4 + \left[\frac{H_T (T_{ps} - T_s) \Delta E (\Delta E - 2 R T_{ps})}{2 C R^2 T_{ps}^4}\right] x_4^2 \tag{20d}$$

TABLE 1. DIMENSIONLESS PARAMETERS FOR FLUIDIZED BED, FROM LUSS AND AMUNDSON (1968).

A	= 0.17143	C	= 205.7143
F	= 8 000	H_g	= 320
H_T	= 266.667	H_w	= 1.6
K	= 0.0024	Kk	= 0.0006 exp (20.7 − 15 000/T)

TABLE 2. STEADY STATE SOLUTIONS TO EQUATIONS (20) AND CORRESPONDING EIGENVALUES OF MATRIX B

	Steady state		
	1	2	3
P	0.09353	0.06705	0.00682
P_p	0.09351	0.06694	0.00653
T	690.445	758.346	912.764
T_p	690.607	759.169	915.093
λ_1	−0.006315	+0.005695	−0.008963
λ_2	−0.91165	−1.2928	−12.563
λ_3	−270.55	−270.56	−270.56
λ_4	−2187.2	−2189.5	−2258.0

TABLE 3. BASIC AND ADJOINT EIGENVECTORS FOR THE TWO STABLE STEADY STATES

	Mode (j)			
	Steady state 1			
	1	2	3	4
y_{j1}	1.3631×10^{-4}	2.6908×10^{-1}	3.5404×10^{-9}	1.690×10^{-1}
y_{j2}	-7.0368×10^{-1}	-6.5183×10^{-1}	-9.999×10^{-1}	-1.385×10^{-4}
y_{j3}	1.3673×10^{-4}	2.6915×10^{-1}	5.5816×10^{-10}	-9.8562×10^{-1}
y_{j4}	-7.1052×10^{-1}	-6.559×10^{-1}	4.814×10^{-3}	9.965×10^{-4}
η_{j1}	-2.9289	3.1738	-7.8969×10^{-3}	8.663×10^{-1}
η_{j2}	-6.7467×10^{-3}	3.4307×10^{-6}	-9.953×10^{-1}	3.168×10^{-10}
η_{j3}	-5.036×10^{-1}	5.442×10^{-1}	-2.134×10^{-4}	-8.6605×10^{-1}
η_{j4}	-1.4014	7.102×10^{-4}	9.8568×10^{-1}	-4.6887×10^{-7}
	Steady state 3			
y_{j1}	7.779×10^{-5}	1.978×10^{-2}	2.925×10^{-8}	1.597×10^{-1}
y_{j2}	-7.037×10^{-1}	-7.202×10^{-1}	-9.999×10^{-1}	-2.646×10^{-2}
y_{j3}	7.803×10^{-5}	1.907×10^{-2}	4.612×10^{-9}	-9.669×10^{-1}
y_{j4}	-7.105×10^{-1}	-6.933×10^{-1}	4.813×10^{-3}	1.973×10^{-1}
η_{j1}	$-4.248 \times 10^{+1}$	$4.377 \times 10^{+1}$	-1.652	8.598×10^{-1}
η_{j2}	-6.769×10^{-3}	2.807×10^{-5}	-9.953×10^{-1}	2.509×10^{-9}
η_{j3}	-7.305	7.232	-4.466×10^{-2}	-8.922×10^{-1}
η_{j4}	-1.406	5.559×10^{-3}	9.855×10^{-1}	-3.849×10^{-6}

The eigenvalues of the matrix **B** are given in Table 2. It is to be noted that the first and third steady states are stable, while the second is unstable. Each of the stable states satisfies the conditions on the eigenvalues, $|\lambda_1/\lambda_p| << 1$, $p \geqq 2$. The basic and adjoint eigenvectors of **B** for each of the stable steady states are recorded in Table 3. The only nonzero elements of **C** in Equations (20) are C_{344}, C_{434}, and C_{444}.

Luss and Amundson obtained numerical solutions of the full nonlinear equations for a number of initial conditions. They encountered numerical difficulties because of the extremely small step size required for numerical stability, and they ultimately adopted a procedure of setting P_p and P equal when they approached to within 1% of one another. We have solved the same cases using eigenvalues and eigenvectors of the **B** matrix and the analytical solu-

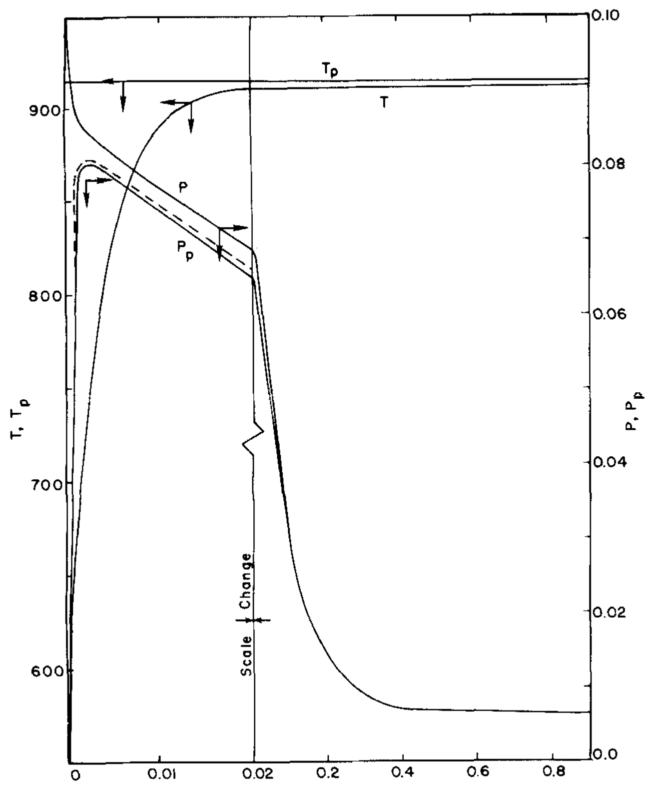

Fig. 1. Approach to the high temperature steady state. Results correspond to Figure 3 of Luss and Amundson (1968), and the dashed line is their reported numerical solution for P_p. Note the change in scale on the time axis.

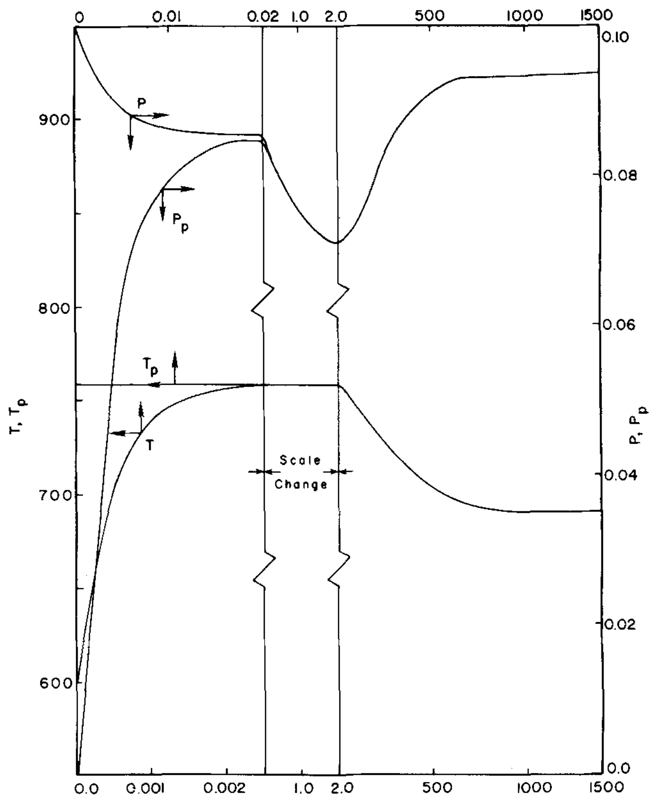

Fig. 2. Approach to the low temperature steady state. Results correspond to Figure 4 of Luss and Amundson (1968). Note the two changes in scale on the time axis and the different initial time scales for temperatures and pressures.

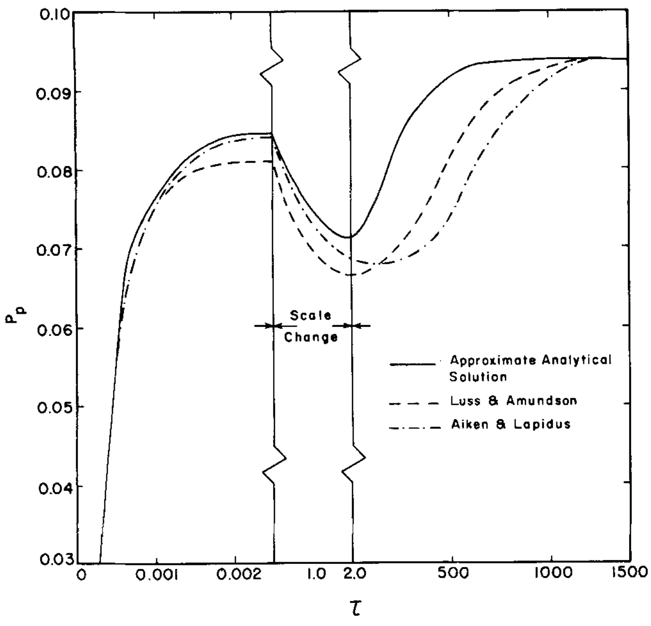

Fig. 3. Particle temperature for conditions in Figure 2; comparison of approximate analytical solution to numerical results of Luss and Amundson (1968) and Aiken and Lapidus (1974).

tions embodied in Equations (7), (13), and (14). Figure 1 shows results for a case in which the particles are initially at the temperature of the third steady state but in which there are large deviations in gas temperatures and in both partial pressures. This case corresponds to Figure 3 of Luss and Amundson, and the dashed line is their reported numerical solution for P_p. The results are almost identical, and the same is true for the other curves in the figure. Note that the multiple time scales in the process are retained in the analytical solution.

Figure 2 shows results for a case in which the particles are initially 0.1 units below the second (unstable) steady state temperature. This case corresponds to Luss and Amundson's Figure 4, and the analytical solution differs only slightly from their numerical calculation. The only deviation observable on this scale is in P_p; this is shown in Figure 3, together with a numerical solution by Aiken and Lapidus (1974) using the highly accurate Gear method for stiff systems of differential equations. The transient response in this example is quite slow, and we note that here, too, the multiple time scales have been properly accounted for by the approximate analytical solution.

The linearization limits computed from Equation (16) give essentially the same results for gas and particle properties. About steady state one, we have $\lambda_1/I_{111} = -119.8$ and $|x_1(0)|$, $|x_3(0)| << 0.016$, $|x_2(0)|$, $|x_4(0)| << 85$. About steady state three, $\lambda_1/I_{111} = 14.3$, and the linearization limits are $|x_1(0)|$, $|x_3(0)| << 0.0011$, $|x_2(0)|$, $|x_4(0)| << 10$. The calculations in Figures 1 and 2 are both initially within a region where nonlinear terms are important.

The stability estimate about the low temperature steady state from Equation (17) is

$$2.09x_1 + 0.0048x_2 + 0.36x_3 + x_4 \leqq 85.46 \qquad (21)$$

Perturbations in the gas temperature (x_2) are unimportant relative to the solid temperature (x_4) because of the rapid equilibration, and pressure perturbations are also generally unimportant because of the relative magnitudes of temperatures and pressures. This stability estimate is consistent with the behavior of the full nonlinear system, in which the upper or lower steady state is approached depending on whether the solid temperature is above or below that of the unstable middle steady state. The estimate of the allowed perturbation is a bit high, however, since the middle steady state corresponds to a value of 68.82 for the linear combination in Equation (21).

The stability estimate about the high temperature steady state is

$$30.21x_1 + 0.0048x_2 + 5.20x_3 + x_4 \geqq -10.18 \qquad (22)$$

The dominant role of the particle temperature in determining stability is again made clear, though the estimate is a very conservative one. The intermediate steady state corresponds to a value of -154.53 for the linear combination in Equation (22).

CONCLUSION

The approximate procedure described here is a powerful one for those systems whose linear responses are dominated by a single slow mode. Within the constraints imposed by the stability estimate, the method gives an accurate analytical estimate of the dynamical response, including all relevant time scales, as well as providing a self-consistent estimate of the limits of a linearized analysis and finite stability regions.

NOTATION

A = coefficient in fluidized-bed model
$A_p(t)$ = modal variable
B_{ij} = coefficient of linear terms of f_i
C = coefficient in fluidized-bed model
C_{ijk} = coefficients of quadratic terms in f_i
ΔE = activation energy
F = coefficient in fluidized-bed model
$\mathbf{f}$, f_i = rate of change of $\mathbf{x}$, x_i
H_g, H_w, H_T = coefficients in fluidized-bed model
I_{pnm} = coefficients defined by Equation (10)
K = coefficient in fluidized-bed model
k = reaction rate
k_o = preexponential factor
P = pressure
R = gas constant
T = temperature
t = time
$\mathbf{x}$, x_i = state variables
y_n, y_{ni} = eigenvectors of B_{ij}
η_{ni} = adjoint eigenvectors of B_{ij}
λ_n = eigenvalues of B_{ij}
τ = dimensionless time

Subscripts

e = entering
p = particle
s = steady state

LITERATURE CITED

Aiken, R. C., and L. Lapidus, "An Effective Numerical Integration Method for Typical Stiff Systems," *AIChE J.*, **20**, 368 (1974).
Denn, M. M., *Stability of Reaction and Transport Processes*, Prentice-Hall, Englewood Cliffs, N.J. (1975).
Eckhaus, W., *Studies in Non-Linear Stability Theory*, Springer-Verlag, New York (1965).
Luss, D., and N. R. Amundson, "Stability of Batch Catalytic Fluidized Beds," *AIChE J.*, **14**, 211 (1968).

Manuscript received April 25, 1977; revision received January 6, and accepted January 16, 1978.