will be needed. What attributes of the controlled system these indices should reflect is not clear. Here it is imperative to reflect on the lessons of the past, for they have shown repeatedly that the primitive and superficial statements of control objectives must be transformed into criteria that have fundamental meaning within the ultimate framework of the theory. For example, the gain-bandwidth product and the relative orientation of eigenvectors are both far removed from simple statements of minimal process excursions, but both bear centrally and directly on accomplishing the latter. There is more than a suspicion that the work of genius is needed here, for without it the control configuration problem will likely remain in a primitive, hazily stated, and wholly unmanageable form.

Indeed, the same may be said for all aspects of the chemical process control problem. If not genius, then perception and the courage to tackle problems of an unfamiliar and unfriendly character. The insidious trend of the past decade to seek mere translations of the control techniques arising in other fields has left the chemical engineering profession destitute of incisive investigation and substantial resolution of its own unique problems. Instead only the elementary ideas should have been *borne across* the chasm separating spacecraft control from chemical process control and the seeds allowed to germinate in the virgin but unexplored valleys of the latter. That has not happened, but it must happen before practitioners can reasonably be expected to use the results of chemical process control theory. And it must be made to happen by those with experience in process engineering; there are few others who can perceive the problems and goals clearly and realistically. In fact, the chemical engineer is viewed by others to be in an extremely enviable position owing both to the wealth of control problems in his domain and his knowledge of processes. It would not be realistic to say, however, that he will be able to solve his problems singlehandedly; they are much too difficult. But if he can recognize those problems and respond with an imaginative attack and an inventive and pioneering spirit, there shall be some hope of narrowing the gap. The gap is present indeed, but contrary to the views of many, it is the theoretician who must close it.

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THE AUTHOR

The author is a pragmatic New Englander who has been trying to teach idealistic Californians about process control and dynamics for the last decade. He writes that his handful of publications on these topics, while not all of a theoretical genre, is in no way excluded from the set of gap-makers criticized in this essay.

A New Method of Parameter Estimation in Linear Systems

A time domain method is given for estimating the matrix or related parameters in linear systems with constant coefficients and real eigenvalues. The method consists of a one-dimensional search for the local minima of a scalar function $\mu(\lambda)$, which provide the eigenvalues of the system matrix and the matrix itself when observable. Applications are given to the determination of a transfer function and the estimation of the rate matrix of a monomolecular reaction system. Questions of accuracy, number, and type of measurements required are discussed.

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SCOPE

Process identification and parameter estimation constitute an important part of theoretical and applied work in the areas of process modeling and control, chemical kinetics, and transport phenomena. The present paper is concerned with parameter estimation in linear systems of ordinary differential equations with constant coefficients

and real eigenvalues, that is, nonoscillatory. The term linear system refers to linearity with respect to the state variables, whereas the parameters almost always enter in a nonlinear way. Several methods of nonlinear parameter estimation exist, but when the number of parameters is large they become subject to problems of slow convergence or divergence, or convergence to an irrelevant extremum, caused by the complicated and unpredictable nature of the response surface. The estimation of the matrix of a system of n equations involves n^2 unknowns, which even for $n = 3$ is a rather prohibitive task for straightforward nonlinear estimation.

In view of these difficulties of multiparameter estimation, special methods have been developed exploiting the structure of linear systems to reduce the estimation problem to smaller more manageable units. Such methods are classified to frequency response and time domain methods. Frequency response methods have been used primarily in process control and although they provide accurate results they are limited by the need of complicated and expensive experiments. Time domain methods, experimentally simpler but somewhat less accurate, are widely used in process control, kinetics, and other areas. Many of these methods have been summarized in a recent review article by Nieman et al. (1971).

The present paper presents a time-domain method whereby the multiparameter estimation problem is reduced to a one-dimensional search for the eigenvalues of the system matrix. Following the determination of the eigenvalues, other parameters of the system such as rate constants and diffusion coefficients are easily determined. In addition to computational simplicity, the method aims at suggesting type and amount of measurements required for meaningful estimation. The method is limited to systems with real eigenvalues; therefore, it cannot be applied to oscillatory systems.

CONCLUSIONS AND SIGNIFICANCE

A new method is given for estimating the parameters of a linear system with constant coefficients and real eigenvalues. The method is based on finding linear combinations of the measurements which contain a single exponential in time so that the eigenvalues of the system matrix can be determined independently of each other. This decomposition is carried out by constructing a positive definite matrix $Q(\lambda)$ from the measurements. The smallest eigenvalue $\mu(\lambda)$ of $Q(\lambda)$ attains local minima at $\lambda = \lambda_1, \ldots, \lambda = \lambda_n$ which are precisely the eigenvalues of the system matrix. From $\lambda_1, \ldots, \lambda_n$ and some additional information obtained, the system matrix and other desired parameters can be determined. The problem is thus reduced to a one-dimensional search for the minima of $\mu(\lambda)$, thereby avoiding the pitfalls of multiparameter estimation. In addition, the reduction to one dimension provides a visual comprehension of the response surface and the effects thereon of number and type of measurements.

Although confidence intervals are not derived a qualitative analysis of the accuracy of estimation is presented. It is found that the accuracy depends critically on the relative magnitude of the error free value $\mu^*(\lambda)$ of $\mu(\lambda)$ and the residual error R. Estimation is basically possible only when $R < \mu^*(\lambda)$. Any factor that increases $\mu^*(\lambda)$ relative to R will improve the accuracy of estimation. For fixed error level, the accuracy of the estimation is dramatically improved when the measurements include all states of the system. Equally important is the utilization of data from at least n initial states.

An application is given to the determination of a process transfer function. The results are very satisfactory although they are subject to an intrinsic limitation of the problem. This is the difficulty of determining large negative eigenvalues, especially when the measurements involve only the first component of the state vector. A second application is made to the determination of the rate matrix in a monomolecular reaction system. The results obtained are very good even at high levels of error. It is expected that the method is also applicable to the estimation of diffusivities and other parameters in systems governed by the diffusion equation by restricting attention to the first few eigenvalues of the system. One such application has been given elsewhere (Gavalas and Seinfeld, 1972).

The main advantage of the method presented is the independent determination of each eigenvalue and eigenvector, which avoids problems of divergence or convergence to spurious extrema. The estimation, essentially reduced to the evaluation of a scalar function $\mu(\lambda)$, is easily programmable on the computer. The method requires a relatively more accurate measurement of the initial state, which may be difficult in some systems. Its main limitation is that it requires measurements at precisely controlled times. This control of time is easily arranged in process control and transport experiments but is difficult in kinetics. The method is somewhat related to that of Wei and Prater (1962) and a comparison of the two is given in the last section.

GENERAL THEORY

The theory will be restricted to linear systems with constant coefficients and with real and distinct eigenvalues. The governing differential equations and the measurements will be given as

$$\dot{x} = Ax \tag{1}$$

$$y = Hx + \epsilon \tag{2}$$

where x, y, ϵ are the state vector, measurement vector, and random error vector of dimensions n, r, r, $(r \leq n)$, respectively. The constant matrices A and H have dimensions nxn and rxn, respectively. No forcing term has been included in Equation (1). The testing is carried out by bringing the system to some initial state x_0 at $t = 0$ and measuring its subsequent output as it freely evolves towards equilibrium at times $t_1 = 0, t_2, \ldots, t_N$. We shall consider m initial states $x_0(\alpha)$, $\alpha = 1, \ldots, m$ which span the space, that is, the nxm matrix X_0 defined by

$$(X_0)_{ij} = x_{0i}(j) \tag{3}$$

has rank n, which implies $m \geq n$. The solution of Equation (1) at time t_k, corresponding to the initial state $x_0(\alpha)$ will be denoted by $x(k, \alpha)$. Similarly, $y(k, \alpha)$, $\epsilon(k, \alpha)$ will denote the corresponding values of y and ϵ. Thus, the totality of measurements will be given by

$$y(k,\alpha) = Hx(k,\alpha) + \epsilon(k,\alpha)$$
$$\alpha = 1,\ldots,m\ ;\ k = 1,\ldots,N \tag{4}$$

In most cases we deal with stable systems so that we may designate by $-\lambda_1,\ldots,-\lambda_n$ the eigenvalues of the matrix A, where $\lambda_i \geqq 0$. The corresponding eigenvectors $v^{(1)},\ldots,v^{(n)}$ are linearly independent and span the state-space; therefore the solution $x(t)$ corresponding to an initial state $x_0(\alpha)$ can be written as

$$x(t,\alpha) = T\,e^{-\Lambda t}\,T^{-1}\,x_0(\alpha) \tag{5}$$

where T has columns the eigenvectors of A,

$$T = (v^{(1)}|v^{(2)}|\ldots v^{(n)}) \tag{6}$$

and

$$\Lambda = \begin{bmatrix} \lambda_1 \ldots 0 \\ \ldots\ldots \\ 0 \ldots \lambda_n \end{bmatrix} \tag{7}$$

The measurement y is now

$$y(k,\alpha) = HTe^{-\Lambda t_k}\,T^{-1}\,x_0(\alpha) + \epsilon(k,\alpha) \tag{8}$$

which can also be written in the more compact form

$$Y(k) = \mathrm{HT}e^{-\Lambda t_k}\,T^{-1}\,X_0 + E(k) \tag{9}$$

where the matrices X_0, $Y(k)$, $E(k)$ are defined by

$$X_{0i\alpha} = x_{0i}(\alpha) \quad i = 1,\ldots,n;\quad \alpha = 1,\ldots,m \tag{10}$$

$$Y_{i\alpha}(k) = y_i(k,\alpha) \quad i = 1,\ldots,r;\quad \alpha = 1,\ldots,m \tag{11}$$

$$E_{i\alpha}(k) = \epsilon_i(k,\alpha) \quad i = 1,\ldots,r;\quad \alpha = 1,\ldots,m \tag{12}$$

In order to proceed in the simplest possible fashion we shall first examine the case $m = n$ and then present the necessary modifications for the case $m > n$. When $m = n$, X_0 is a nonsingular nxn matrix; therefore, the matrix

$$Z = X_0^{-1}T \tag{13}$$

exists. By post-multiplying Equation (9) by Z there is obtained

$$Y(k)Z = HTe^{-\Lambda t_k} + E(k)Z \tag{14}$$

If $z(1),\ldots,z(n)$ are the columns of Z we can write Equation (14) as

$$\sum_{\alpha=1}^{n} y_i(k,\alpha)z_\alpha(j) = (HT)_{ij}\,e^{-\lambda_j t_k} + \sum_{\alpha=1}^{n} \epsilon_i(k,\alpha)z_\alpha(j)$$
$$i = 1,\ldots,r\ ;\ j = 1,\ldots,n \tag{15}$$

This equation states the existence of multipliers $z_1,\ldots,z_n$ such that the vector $z_1y(t,1) + \ldots z_ny(t,n)$ contains only a single exponential in time. In particular, when $t = 0$, that is, $k = 1$, Equation (15) becomes

$$\sum_{\alpha=1}^{n} y_i(1,\alpha)z_\alpha(j) = (HT)_{ij} + \sum_{\alpha=1}^{n} \epsilon_i(1,\alpha)z_\alpha(j) \tag{16}$$

Upon elimination of $(HT)_{ij}$ between Equations (15) and (16) there is obtained

$$\sum_{\alpha=1}^{n} z_\alpha(j)\,[y_i(k,\alpha) - y_i(1,\alpha)e^{-\lambda_j t_k}]$$
$$= \sum_{\alpha=1}^{n} z_\alpha(j)\,[\epsilon_i(k,\alpha) - \epsilon_i(1,\alpha)e^{-\lambda_j t_k}] \tag{17}$$
$$i = 1,\ldots,r\ ;\ j = 1,\ldots,n$$

Equation (17) suggests the formulation of a sum of error squares

$$J = \sum_{k=1}^{N}\sum_{i=1}^{r}\left\{\sum_{\alpha=1}^{n} z_\alpha[y_i(k,\alpha) - y_i(1,\alpha)\,e^{-\lambda t_k}]\right\}^2 \tag{18}$$

The problem now is to minimize J with respect to z and λ simultaneously, under the normalization constraint $z_1^2 + \ldots + z_n^2 = 1$. We will shortly see that J has n local minima and hence there are n solutions $(\lambda_j, z(j))$, $j = 1,\ldots,n$. Equation (18) places special emphasis on the first measurement $y(1,\alpha)$. This slight disadvantage can be turned to an advantage in kinetic and other systems where the state can be measured more accurately initially than in subsequent times.

The quantity J can be written as a quadratic form

$$J = z^T Q(\lambda) z \tag{19}$$

where

$$Q_{ij}(\lambda) = \sum_{k=1}^{N}\sum_{l=1}^{n}\Big\{[y_l(k,i) - y_l(1,i)e^{-\lambda t_k}]$$
$$[y_l(k,j) - y_l(1,j)e^{-\lambda t_k}]\Big\} \tag{20}$$
$$i,j = 1,\ldots,n$$

In the absence of measurement error, the matrix $Q(\lambda)$ is positive definite for all λ except $\lambda = \lambda_1,\ldots,\lambda_n$ for which it becomes positive semidefinite. For any given λ, the minimum $\mu(\lambda)$ of J under the constraint $z_1^2 + \ldots +$

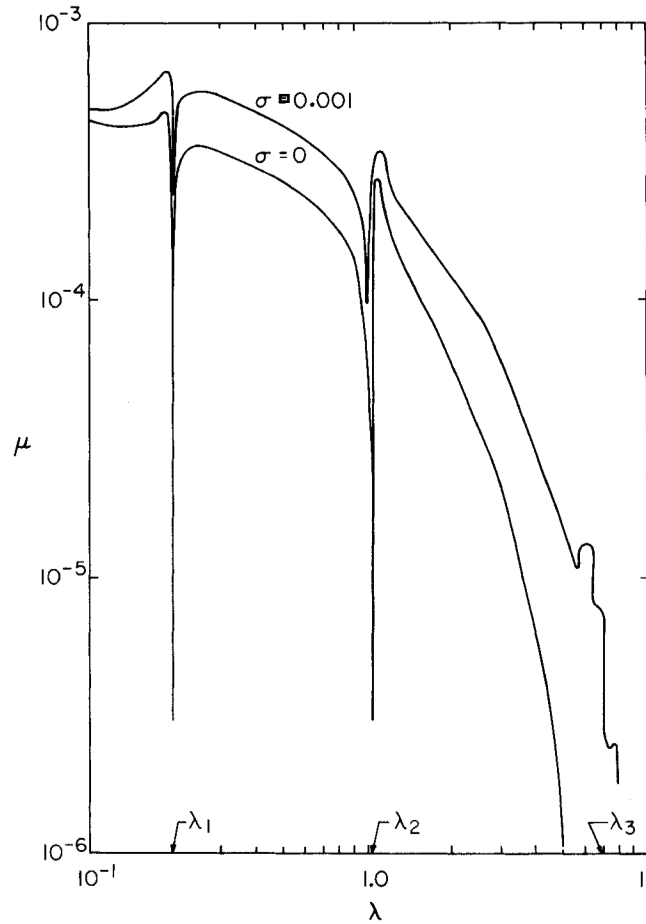


Fig. 1. The function $\mu(\lambda)$ of Example 1.

$z_n^2 = 1$, it attained at some vector $z = z(\lambda)$. It is well known that $\mu(\lambda)$ is the smallest eigenvalue of $Q(\lambda)$ and $z(\lambda)$ the corresponding eigenvector. Let us consider the behavior of the function $\mu(\lambda)$. In the absence of measurement error, $\mu(\lambda)$ attains its minimum value, zero, at the points $\lambda_1, \ldots, \lambda_n$. The vectors $z(\lambda_1), \ldots, z(\lambda_n)$ are precisely the columns of the matrix Z of Equation (13). In the presence of measurement error, $\mu(\lambda)$ attains local minima at points $\hat{\lambda}_1, \ldots, \hat{\lambda}_n$ which approximate $\lambda_1, \ldots, \lambda_n$ provided the error is sufficiently small. The minima $\mu(\hat{\lambda}_1), \ldots, \mu(\hat{\lambda}_n)$ are positive and proportional to the error variance σ^2.

The problem has been reduced to a simple one-dimensional search. The function $\mu(\lambda)$ is plotted and the points of local minima $\hat{\lambda}_1, \ldots, \hat{\lambda}_n$ and the corresponding vectors $z(\hat{\lambda}_1), \ldots, z(\hat{\lambda}_n)$ are recorded. Denoting by $\hat{Z}$ the corresponding matrix we can go back to Equation (13) and see what information can be obtained about the matrix T. The initial state matrix X_0 in general is not observable. What is observable is the matrix Y_0 so that multiplying Equation (13) by HX_0 we obtain an estimate of the matrix HT

$$H\hat{T} = Y_0\hat{Z} \tag{21}$$

Since H is $r \times n$, $r < n$, this relationship does not completely specify $\hat{T}$. This is as far as the estimation can go. Although $\lambda_1, \ldots, \lambda_n$ have been estimated, the matrix T and hence the matrix A cannot be completely estimated. The reason is that there are infinitely many pairs of X_0 and T that would give the same set of observations $Y(k)$.

In certain systems, for example in chemical kinetics, the initial states $x_0(\alpha)$ may be known even though subsequent measurements involve only some of the components of the state vector. In such cases T is uniquely estimated as $\hat{T} = X_0\hat{Z}$ and A is estimated as

$$\hat{A} = -\hat{T}\hat{\Lambda}\hat{T}^{-1} \tag{22}$$

In other systems as that of example 1, the matrix T can be readily determined once the eigenvalues $\lambda_1, \ldots, \lambda_n$ are found.

Let us now investigate the case of redundant observations $m > n$. We can still find a matrix Z such that

$$X_0Z = T \tag{23}$$

However, since X_0 is $n \times m$, the matrix Z is not unique and cannot be expressed as in Equation (13). The development leading to the definition of the quadratic form (19) is still valid. However, the matrix $Q(\lambda)$ has somewhat different properties. In the absence of measurement error, $Q(\lambda)$ has zero as an eigenvalue of multiplicity $m - n$ for each λ, except for $\lambda_1, \ldots, \lambda_n$ where zero is an eigenvalue of multiplicity $m - n + 1$. In the presence of measurement error, $Q(\lambda)$ is positive definite, but possesses $m - n$ small eigenvalues of order σ^2. This suggests that we order the eigenvalues of $Q(\lambda)$ as follows

$$\underset{1}{\mu(\lambda)} < \underset{2}{\mu(\lambda)} < \ldots < \underset{m-n+1}{\mu(\lambda)} < \ldots < \underset{m}{\mu(\lambda)} \tag{24}$$

and define

$$\mu(\lambda) = \underset{m-n+1}{\mu(\lambda)} \tag{25}$$

The local minima $\hat{\lambda}_1, \ldots, \hat{\lambda}_n$ of the quantity $\mu(\lambda)$ provide the desired estimates of the eigenvalues $\lambda_1, \ldots, \lambda_n$. The values $\mu(\hat{\lambda}_j)$ are of the same order of magnitude as $\underset{i}{\mu}(\hat{\lambda}_j)$, $i = 1, \ldots, m - n$.

SPECIAL CASE: ALL STATES MEASURED

This case allows an alternative and simpler approach and also allows the complete determination of the matrix A. By setting $H = I$, Equation (8) becomes

$$y(k, \alpha) = Te^{-\Lambda t_k}T^{-1}x_0(\alpha) + \epsilon(k, \alpha) \tag{26}$$

which upon multiplication with T^{-1} gives

$$T^{-1}y(k, \alpha) = e^{-\Lambda t_k}T^{-1}x_0(\alpha) + T^{-1}\epsilon(k, \alpha) \tag{27}$$

If $\gamma(1), \ldots, \gamma(n)$ are the rows of the matrix T^{-1} then

$$\sum_{l=1}^{n} \gamma_l(j)y_l(k, \alpha) = e^{-\lambda_j t_k}(T^{-1}x_0(\alpha))_j + \sum_{l=1}^{n} \gamma_l(j)\epsilon_l(k, \alpha) \tag{28}$$

$$k = 1, \ldots, N; \quad \alpha = 1, \ldots, m$$

which implies the existence of multipliers $\gamma_1, \ldots, \gamma_n$ such that the quantity $\gamma_1y_1(k, \alpha) + \ldots + \gamma_ny_n(k, \alpha)$ contains only a single exponential, the same for all α. Equation (28) becomes for $k = 1$ $(t = 0)$:

$$\sum_{l=1}^{n} \gamma_l(j)y_l(1, \alpha) = (T^{-1}x_0(\alpha))_j + \sum_{l=1}^{n} \gamma_l(j)\epsilon_l(1, \alpha) \tag{29}$$

By eliminating $(T^{-1}x_0(\alpha))_j$ between Equations (28) and (29), there is obtained

$$\sum_{l=1}^{n} \gamma_l(j)[y_l(k, \alpha) - y_l(1, \alpha)e^{-\lambda_j t_k}] = \sum_{l=1}^{n} \gamma_l(j)[\epsilon_l(k, \alpha) - \epsilon_l(1, \alpha)e^{-\lambda_j t_k}] \tag{30}$$

$$k = 1, \ldots, N; \quad \alpha = 1, \ldots, m$$

This last equation suggests the formulation of the sum of error squares

$$J_1 = \sum_{k=1}^{N} \sum_{\alpha=1}^{m} \left\{ \sum_{l=1}^{n} \gamma_l[y_l(k, \alpha) - y_l(1, \alpha)e^{-\lambda_j t_k}] \right\}^2 \tag{31}$$

This can also be written as a quadratic form

$$J_1 = \gamma^T W(\lambda)\gamma \tag{32}$$

where

$$W_{ij}(\lambda) = \sum_{k=1}^{N} \sum_{\alpha=1}^{m} \{[y_i(k, \alpha) - y_i(1, \alpha)e^{-\lambda t_k}][y_j(k, \alpha) - y_j(1, \alpha)e^{-\lambda t_k}]\} \tag{33}$$

This quadratic form is positive semidefinite in the absence of error and positive definite in the presence of error. The problem has been reduced to one of minimizing J_1 with respect to λ and γ simultaneously, under the constraint $\gamma_1^2 + \ldots + \gamma_n^2 = 1$. For any given λ we let $\nu(\lambda)$ be the minimum of J_1 attained at some vector $\gamma(\lambda)$. Again, $\nu(\lambda)$ and $\gamma(\lambda)$ is the smallest eigenvalue and the corresponding eigenvector of $W(\lambda)$. For sufficiently low measurement

error, $\nu(\lambda)$ attains local minima at $\hat{\lambda}_1, \ldots, \hat{\lambda}_n$ which provide the required estimates of the eigenvalues of the matrix A. The corresponding vectors $\gamma(\hat{\lambda}_1), \ldots, \gamma(\hat{\lambda}_n)$ are the rows of a matrix T^{-1} which can be used to obtain an estimate of the system matrix A as

$$\hat{A} = -\hat{T}\hat{\Lambda}\hat{T}^{-1} \tag{34}$$

Notice that the present treatment does not require $m \geqq n$. In fact, m could be one, but the accuracy of estimation is drastically reduced when $m < n$.

RELIABILITY OF ESTIMATES AND COMPUTATIONAL CONSIDERATIONS

For any given value of λ, the term $\exp(-\lambda t_k)$ decreases and eventually the quantities $y_l(k, i)$, $y(1, i)\exp(-\lambda t_k)$ become smaller than the standard deviation σ of the measurement error. Such measurements obviously cannot be useful to the estimation of an eigenvalue of magnitude λ. It is therefore suggested that for a given λ the summation over k in Equation (20) is carried out only as far as t_k satisfies

$$\sigma \leqq y_M e^{-\lambda t_k} \tag{35}$$

where

$$y_M = \max_{i,\alpha} |y_i(1, \alpha)| \tag{36}$$

As a consequence, the estimation of the negative eigenvalues with the larger absolute value will be less accurate because the number of useful measurements will be smaller.

The computation of the eigenvalues of a matrix, even a symmetric one such as Q or W, entails considerable numerical error. To reduce this error it is suggested to use one of the more efficient methods such as the Householder method (Noble, 1969) along with double precision arithmetic. Even so, the smaller eigenvalues will be calculated less accurately than the larger ones. This property is relevant because the eigenvalues of $Q(\lambda)$ happen to be widely separated in magnitude, for example, the eigenvalues of a typical 3×3 Q matrix are $\mu_1 = 6.70 \times 10^{-3}$, $\mu_2 = 1.42 \times 10^{-2}$, $\mu_3 = 3.13 \times 10^{1}$. It is rather unfortunate then that the eigenvalue of interest $\mu = \mu_1$ is also the one computed with the least accuracy. In the examples considered, this property has not caused any difficulty, but it would be of interest to seek computational methods which determine the smallest eigenvalue with accuracy. Another method for locating the estimates $\hat{\lambda}_j$, especially in the case $m = n$, is to seek the local minima of $\det[Q(\lambda)]$ rather than the minima of $\mu(\lambda)$.

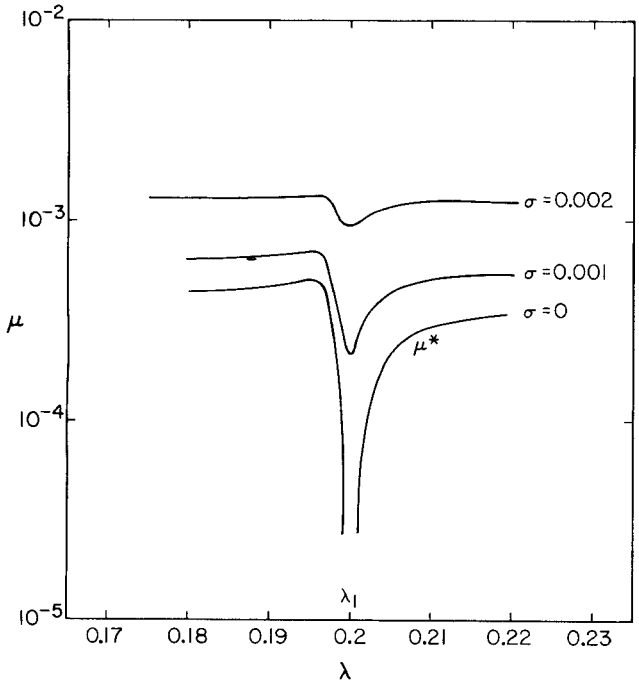


Fig. 2. The function $\mu(\lambda)$ of Example 1 in the neighborhood of λ_1.

With regard to the question of reliability, it is difficult to derive confidence intervals for the estimates λ_j. However, two basic reliability problems can be treated in a qualitative and elementary fashion. One is how to distinguish whether a local minimum in the curve $\mu(\lambda)$ is significant or represents a spurious random fluctuation. The other is whether the data in a given region of λ are accurate enough to reveal a true minimum in that region. Both problems can be discussed by comparing the behavior of the functions $\mu^*(\lambda)$, $\mu(\lambda)$. Figure 2 compares $\mu^*(\lambda)$ and $\mu(\lambda)$ in the neighborhood of λ_1 for two values of the error variance. At each point λ, the quantity $\mu(\lambda)$ is made up of two contributions. One arises from the deviation $\lambda - \lambda_1$ and its magnitude is given by the height $\mu^*(\lambda)$ of the error free curve. The other contribution is the residual measurement error whose mean value can be computed from Equations (17) and (18) after neglecting the terms $\epsilon_i(1, \alpha)\exp(-\lambda t_k)$ as

$$R \cong \sigma^2 \sum_{k=1}^{N} \sum_{i=1}^{r} \sum_{\alpha=1}^{m} z_\alpha^2 = rN\sigma^2 \tag{37}$$

The two contributions $\mu^*(\lambda)$ and R are not additive. There is interaction such that $\mu(\lambda) \leqq \mu^*(\lambda) + R$ and the minimum of $\mu(\lambda)$ shifts to a point $\hat{\lambda}_1 \neq \lambda_1$. At that minimum, the contribution $\mu^*(\lambda)$ roughly vanishes and $\mu(\hat{\lambda}_1) \cong R$. The minimum value $\mu(\hat{\lambda}_1)$ should be substantially lower than neighboring values of $\mu(\lambda)$ to be distinguishable. Hence in the neighborhood of a minimum $\hat{\lambda}_1$, $\mu(\lambda)$ should be substantially higher than R, for example 1.5 R or more, which, in turn, requires that $\mu^*(\lambda)$ should be of the order of R or higher. To summarize, a section of the curve $\mu(\lambda)$ can be used to detect a minimum only if its magnitude is about 1.5 R or higher. A local minimum of $\mu(\lambda)$ is in turn significant only if it is approximately equal to R. The quality of estimation basically depends on the relative magnitude of $\mu^*(\lambda)$ and R. Any factor that increases $\mu^*(\lambda)$ for fixed R would also improve the accuracy of estimation.

DETERMINATION OF A PROCESS TRANSFER FUNCTION

In process control, the dynamics of a linear system is often formulated in terms of its transfer function. Consider, for example, the transfer function of a one-input one-output third-order system with a pure time delay,

$$G(s) = \frac{Ke^{-s\tau_d}}{(\tau_1 s + 1)(\tau_2 s + 1)(\tau_3 s + 1)} \tag{38}$$

The time constants τ_1, τ_2, τ_3, the gain K, and the time delay τ_d are traditionally determined by frequency response or time domain testing. The latter testing usually involves a step or pulse input and subsequent interpretation of the data by one of several methods. The earliest is the graphi-

cal method of Oldenburg and Sartorius (Ramo and Woodridge, 1961). Limited to second-order systems, it requires data accurate enough for graphical differentiation and cannot simultaneously utilize data resulting from different test inputs. A straightforward nonlinear least squares estimation, on the other hand, suffers from the difficulties of multiparameter estimation. Other more efficient methods have been developed and are reviewed by Nieman et al. (1971).

To apply our method, measurements are required as the system moves freely from some initial state to equilibrium. For a third-order system at least three linearly independent initial states should be used. These can be produced by various forcing inputs such as combinations of finite pulses, ramps, etc. At time $t = 0$ the input is interrupted and the system evolves freely to equilibrium, governed by the equation

$$a\dddot{x} + b\ddot{x} + d\dot{x} + x = 0 \tag{39}$$

where $a = \tau_1\tau_2\tau_3$, $b = \tau_1\tau_2 + \tau_2\tau_3 + \tau_3\tau_1$, $c = \tau_1 + \tau_2 + \tau_3$. If we set $x_1 = x$, $x_2 = \dot{x}$, $x_3 = \ddot{x}$, Equation (39) can be written in the standard form of Equation (1),

$$\begin{aligned} \dot{x}_1 &= x_2 \\ \dot{x}_2 &= x_3 \\ \dot{x}_3 &= -\frac{1}{a}x_1 - \frac{c}{a}x_2 - \frac{b}{a}x_3 \end{aligned} \tag{40}$$

The measurements must incorporate the time delay

$$y(t) = x_1(t - \tau_d) + \epsilon(t) \tag{41}$$

The measurement matrix H is simply (1, 0, 0) and for $t_k > \tau_d$ Equation (15) gives

$$\sum_{\alpha=1}^{3} y(k,\alpha)z_\alpha(j) = T_{1j}\, e^{\lambda_j\tau_d}\, e^{-\lambda_j t_k} + \sum_{\alpha=1}^{3} \epsilon(k,\alpha)z_\alpha(j) \tag{42}$$

which can be evaluated at some $t_{k0} > \tau_d$ to give

$$\sum_{\alpha=1}^{3} y(k_0,\alpha)z_\alpha(j) = T_{1j}e^{\lambda_j\tau_d}\, e^{-\lambda_j t_{k_0}} + \sum_{\alpha=1}^{n} \epsilon(k_0,\alpha)z_\alpha(j) \tag{43}$$

Eliminating the quantity $T_{1j}e^{\lambda_j\tau_d}$ between (42) and (43) there is obtained

$$\sum_{\alpha=1}^{3} z_\alpha(j)[y(k,\alpha)e^{-\lambda_j t_{k_0}} - y(k_0,\alpha)e^{-\lambda_j t_k}] = \sum_{\alpha=1}^{3} z_\alpha(j)[\epsilon(k,\alpha)e^{-\lambda_j t_{k_0}} - \epsilon(k_0,\alpha)e^{-\lambda_j t_k}] \tag{44}$$

The matrix Q must now be defined as

$$Q_{ij}(\lambda) = \sum_{k=1}^{N} \{[y(k,i)e^{-\lambda t_{k_0}} - y(k_0,i)e^{-\lambda t_k}][y(k,j)e^{-\lambda t_{k_0}} - y(k_0,j)e^{-\lambda t_k}]\} \tag{45}$$

Thus, the presence of a pure time delay requires only a slight modification of the method. The time delay τ_d and the gain K cannot be determined from relaxation type experiments. They require a forcing type experiment such as a step input. After the determination of the eigenvalues, however, the determination of K and τ_d from a step input experiment is straightforward.

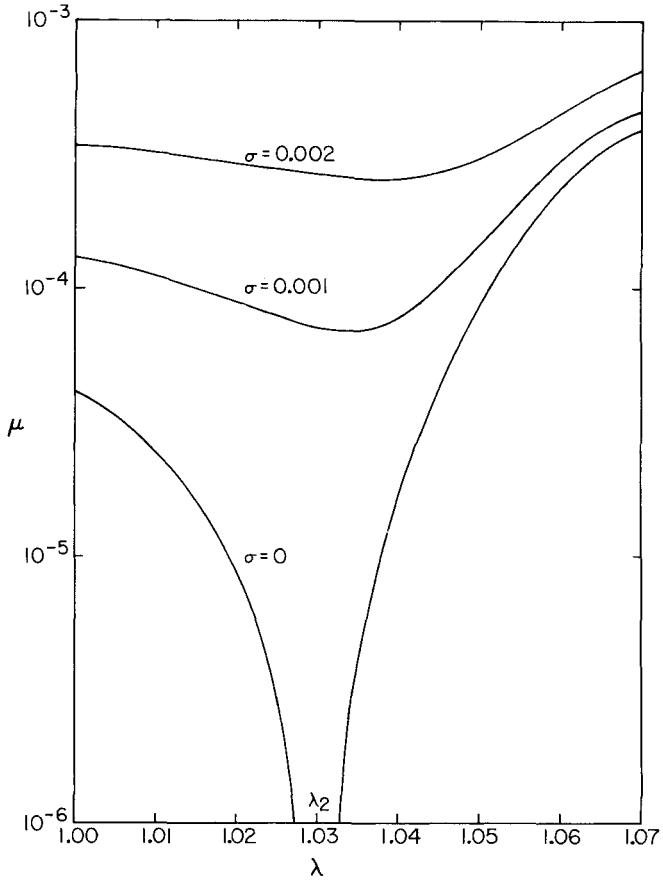


Fig. 3. The function $\mu(\lambda)$ of Example 1 in the neighborhood of λ_2.

We shall now present the results of a simulation study. The eigenvalues of the system matrix $-\lambda_i = -1/\tau_i$ have been assigned the values $\lambda_1 = 0.2$, $\lambda_2 = 1.03$, $\lambda_3 = 6.9$. The system (40) was integrated in the range $0 \leq t \leq 20$ for the initial states

$$X_0 = \begin{bmatrix} 1 & 1 & 0 & 0.5 & 0 & 1 \\ 0 & -1 & 1 & -1.5 & 1 & 0 \\ 0 & 2 & -1 & 0.5 & 1 & -1.5 \end{bmatrix} \tag{46}$$

Results were recorded at intervals of 0.1 and the measurements were generated by adding normally distributed random error $n(0, \sigma^2)$ of mean zero and variance σ^2,

$$y(k,\alpha) = x_1(k,\alpha) + n(0,\sigma^2; k,\alpha) \tag{47}$$
$$\alpha = 1, \ldots, 4; \quad k = 1, \ldots, N = 201$$

The results presented in Figures 1, 2, and 3 were obtained with $m = 3$, that is, with the first three columns of the initial state matrix X_0. As shown in Figures 2 and 3, the first two eigenvalues λ_1, λ_2 are estimated extremely well for $\sigma \leq 0.002$. Actually, very good results ($\hat{\lambda}_1 = 0.198$, $\hat{\lambda}_2 = 1.054$) were obtained with an error level as high as $\sigma = 0.01$.

The estimation of the third eigenvalue λ_3 has been found very difficult. Figure 1 shows the gross features of the function $\mu(\lambda)$ for $\sigma = 0$ and $\sigma = 0.001$. The curve $\mu(\lambda)$ has sharp minima at $\lambda_1 = 0.002$, $\lambda_2 = 1.03$ but its behavior becomes erratic at larger values of λ. For $\lambda > 5$, $\mu^*(\lambda) << R$ and the curve $\mu(\lambda)$ does not contain useful information. The local minima at $\lambda = 5.6$, $\lambda = 7.5$ are

insignificant and the true minimum at $\lambda = 6.9$ has disappeared. This difficulty in determining large λ_j is inherent in the system and can be explained as follows: for $\lambda > 5$, t_k must be less than 1.4 to satisfy Equation (35). The summation of Equation (20) is thus limited to less than fourteen terms with the result of relatively low accuracy in the estimation of any $\lambda_j > 5$. There is a more important reason, however, which is related to the type of measurements used. The eigenvector of the matrix of system (40) corresponding to an eigenvalue λ_j is $(1, -\lambda_j, \lambda_j^2)$. The transformation T is therefore given by

$$T = \begin{bmatrix} 1 & 1 & 1 \\ -\lambda_1 & -\lambda_2 & -\lambda_3 \\ \lambda_1^2 & \lambda_2^2 & \lambda_3^2 \end{bmatrix}$$

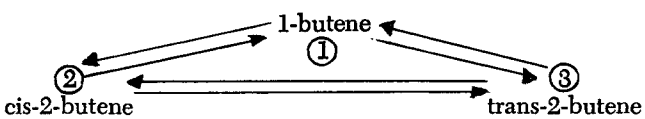
According to Equation (5), the coefficients of $e^{-\lambda_j t_k}$ in $x_1(k, \alpha)$, $x_2(k, \alpha)$, $x_3(k, \alpha)$ are $(T^{-1}x_0)_j$, $-\lambda_j(T^{-1}x_0)_j$, $\lambda_j^2(T^{-1}x_0)_j$, respectively. For large λ_j the exponential $e^{-\lambda_j t_k}$ makes very little contribution in the measured component x_1, whence the low value of $\mu^*(\lambda)$ for large λ. The only way of improving the estimation in the region of large λ is to lower σ, by improving the accuracy of measurements or by the use of duplicate measurements.

The possibility of improving the estimation by using all six columns of the matrix X_0 of Equation (46) was also examined. This increase in the number of initial states used resulted in estimates of λ_1, λ_2 very close to the ones obtained with $m = 3$. The larger eigenvalue λ_3 was again impossible to estimate.

ESTIMATION IN MONOMOLECULAR REACTION SYSTEMS

The method presented in the previous sections bears some similarity to the well-known method of Wei and Prater (1962) in that both decompose the problem to the independent estimation of each eigenvector and its corresponding eigenvalue. In the method of Wei and Prater this decomposition is achieved by a judicious sequence of experiments; in our method it is achieved purely by computation. The method of Wei and Prater is of special interest in kinetics because it does not require accurate control of the times of composition measurements, which is especially difficult in flow reactors. It does require, however, a large number of measurements and these must include all components of the state vector. As a consequence it has not been applied outside the domain of chemical kinetics. Our method requires accurate control of the measurement times, which is often experimentally difficult in kinetics. A less serious limitation is the need of a relatively accurate measurement of the initial state. In kinetics at least this presents no difficulty. The method is broadly applicable to areas such as process control and transport processes where the time of measurements can be controlled without difficulty. In these areas, as well as in kinetics, it has the advantage that the amount of data required is relatively small and the measurements need not include all components of the state vector.

In this section we shall give an application to a monomolecular reaction system, assuming accurate control of time. For this purpose we shall use one of Wei's and Prater's example, the isomerization of butene by means of the reaction network



The mole fractions x_1, x_2, x_3 satisfy a linear system

$$\dot{x} = Ax \tag{48}$$

where the matrix A is known to have the eigenvalue zero corresponding to an equilibrium vector x^*

$$Ax^* = 0 \tag{49}$$

It will be assumed that measurements are made on all three components

$$y_i = x_i + \epsilon_i \qquad i = 1, 2, 3 \tag{50}$$

The data given by Wei and Prater (1962) are time-free; therefore, we have used simulated data computed with the matrix

$$A = \begin{bmatrix} -0.7245 & 0.2381 & 0.0515 \\ 0.5327 & -0.5273 & 0.1736 \\ 0.1918 & 0.2892 & -0.2251 \end{bmatrix} \tag{51}$$

and the initial state matrix

$$X_0 = \begin{bmatrix} 1 & 0 & 0 & 0.5 \\ 0 & 1 & 0 & 0.5 \\ 0 & 0 & 1 & 0 \end{bmatrix} \tag{52}$$

The data were generated in the range $0 \leq t \leq 6$ at intervals of 0.4 and were corrupted by Gaussian random error with mean zero and variance σ^2.

For systems like the present which possess a nonzero equilibrium state, the estimation procedure must be slightly modified. Due to the presence of measurement error, the first local minimum of $\mu(\lambda)$ or $\nu(\lambda)$ will be attained at a point $\lambda \neq 0$, slightly positive or negative. In the first case all the eigenvalues of the estimated matrix will be negative, predicting zero as an equilibrium state. In the second case the estimated matrix will possess a positive eigenvalue, predicting unstable behavior. This

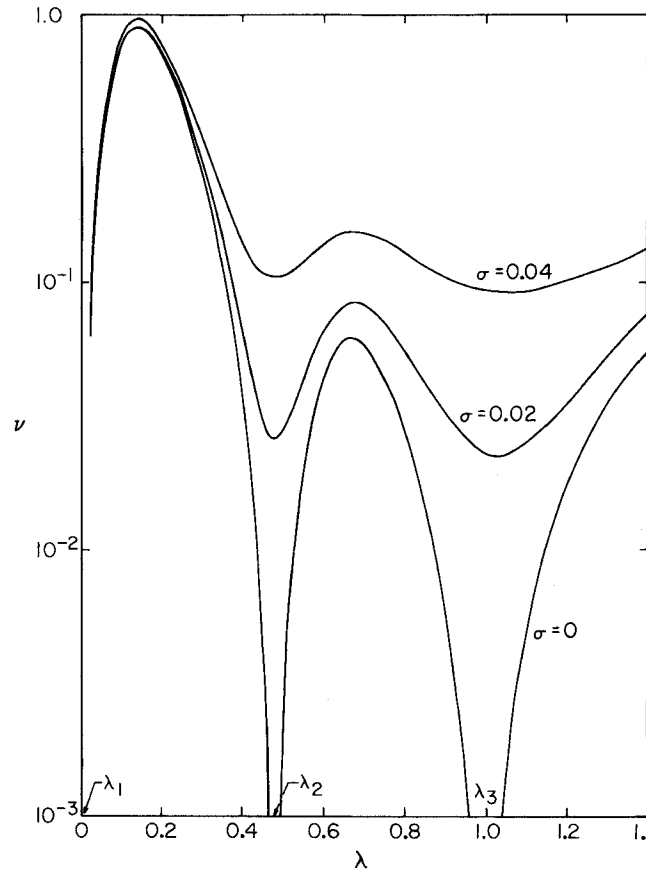


Fig. 4. The function $\nu(\lambda)$ of Example 2 for $m = 4$.

TABLE 1. RESULTS OF ESTIMATION FOR SYSTEM OF EXAMPLE 2

$\sigma = 0$ True values			$\sigma = 0.02$ Estimates			$\sigma = 0.04$ Estimates		
λ_1	λ_2	λ_3	$\hat{\lambda}_1$	$\hat{\lambda}_2$	$\hat{\lambda}_3$	$\hat{\lambda}_1$	$\hat{\lambda}_2$	$\hat{\lambda}_3$
0.000	0.4769	1.000	0.000	0.4775	1.028	0.000	0.4800	1.052
$\gamma(\lambda_1)$	$\gamma(\lambda_2)$	$\gamma(\lambda_3)$	$\gamma(\hat{\lambda}_1)$	$\gamma(\hat{\lambda}_2)$	$\gamma(\hat{\lambda}_3)$	$\gamma(\hat{\lambda}_1)$	$\gamma(\hat{\lambda}_2)$	$\gamma(\hat{\lambda}_3)$
0.5773	0.7070	0.8802	0.5789	0.6982	0.9010	0.5802	0.6954	0.9171
0.5773	0.5063	−0.4723	0.5829	0.5141	−0.4339	0.5881	0.5150	−0.3970
0.5773	−0.4938	0.0474	0.5702	−0.4982	0.0384	0.5635	−0.5012	−0.0370
	A			$\hat{A}$			$\hat{A}$	
−0.7245	0.2381	0.0515	−0.7615	0.2344	0.0497	−0.7864	0.2419	0.0888
0.5327	−0.5273	0.1736	0.5702	−0.5157	0.1740	0.5962	−0.5205	0.1281
0.1918	0.2892	−0.2251	0.1903	0.2891	−0.2283	0.1875	0.2941	−0.2251

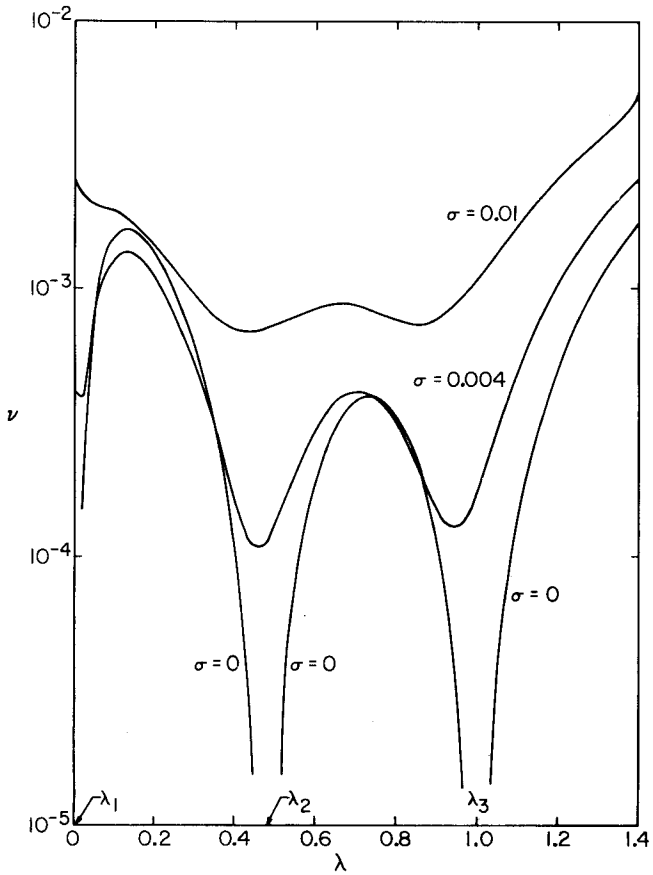


Fig. 5. The function $\nu(\lambda)$ of Example 2 for $m = 1$.

alteration of the stability characteristics can be easily corrected whenever, as in the present example, the existence of a nonzero equilibrium state is a priori known. The eigenvalue $\lambda_1 = 0$ is automatically accepted and the vector $z(0)$ or $\gamma(0)$ which minimizes $z^TQ(0)z$ or $\gamma^TW(0)\gamma$ respectively is computed. These quantities must be used in place of the quantities corresponding to the first local minimum of $\mu(\lambda)$ or $\nu(\lambda)$. The remaining eigenvalues are estimated using the general procedure. In this way the estimated matrix $\hat{A}$ is guaranteed to possess the zero eigenvalue.

As all three states x_1, x_2, x_3 were measured, the method of the second section was used. The results appear on Figure 4 and Table 1. Even at very high error levels, for example, $\sigma = 0.02$ corresponds to an average error of 12%, the estimation results are satisfactory. The eigenvectors and the matrix A are estimated less accurately than the eigenvalues. This is a common property of computational methods for determining the eigenvalues and eigenvectors of matrices. The larger in absolute value eigenvalues are subject to higher estimation error because their contribution to the measurements diminishes more rapidly. Predictably, the measurement of all three states makes it possible to carry out the estimation in the presence of much larger error than in the previous example.

The results presented thus far were obtained using all four columns of the matrix X_0 of Equation (52). No significant change in the estimates was obtained when the number of initial states was reduced to three. When only one initial state was used the estimation was seriously affected. Figure 5, which was computed with the first column of X_0, shows that in order to obtain results comparable with those of Figure 4, the error level has to be reduced by a factor of about ten. Already at $\sigma = 0.01$ the minimum at $\lambda = 0$ disappears and the other two minima are considerably displaced.

NOTATION

A = system matrix
E = error matrix
$G(s)$ = transfer function
H = measurement matrix
I = identity matrix
J, J_1 = criterion for minimization, sum of error squares
m = number of initial states, number of experiments
N = number of measurement times
n = number of components of the state vector
$Q(\lambda)$ = a matrix defined by Equation (20)
R = residual error
r = number of components of measurement vector
$v^{(j)}$ = the jth eigenvector of A
T = a matrix defined by Equation (6)
t = time
X_0 = initial state matrix
$W(\lambda)$ = a matrix defined by Equation (33)
x = state vector
x_0 = initial state vector
Y_0 = initial measurements matrix

Y = measurement matrix
Z = matrix defined by Equation (23)
$z(j)$ = the jth column of Z

Greek Symbols

α = index characterizing the initial state
$\gamma(j)$ = the jth row of T^{-1}
ϵ = measurement error vector
λ_j = the jth eigenvalue of A
$\mu_j(\lambda)$ = the jth eigenvalue of $Q(\lambda)$
$\mu(\lambda)$ = the smallest eigenvalue of $Q(\lambda)$, or as defined by Equation (25)
$\nu(\lambda)$ = the smallest eigenvalue of $W(\lambda)$
σ = the error standard deviation
τ_1, τ_2, τ_3 = time constants
τ_d = time delay

Special Symbols

$\wedge$ = indicates estimates
o = indicates initial conditions
T = as superscript indicates matrix transpose

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Flow Rate-Pressure Gradient Measurements in Periodically Nonuniform Capillary Tubes

Flow rate-pressure gradient measurements have been performed on 15 different test capillary tubes, each of which consisted of short, alternating segments of two different diameters. The Reynolds number range covered extended from 2 to 700, based on the conditions in the narrower capillary segment. Darcy's law was found valid up to about Re = 30-50, whereas the Forchheimer equation described the data over the entire range of Re covered. The Forchheimer equation has been obtained from a nondimensional form of the volume averaged momentum equation by using the averaging theorem due to Slattery and to Whitaker. The parameters α and β have obtained precise hydrodynamic definitions. The experimental data have been treated in terms of the Forchheimer equation: the parameters α and β have been calculated using various definitions for the area of flow. Dimensionless permeability, equal to the ratio of measured-to-Poiseuille permeability has been found to be a minimum-type function of small-to-large capillary diameter ratio. Dimensionless inertial parameter β^* has been found to be a maximum-type function of the capillary diameter ratio, if the calculation was based on the area of flow equal to the cross sectional area of the narrower capillary segment, in every case. The maximum occurred at the same value of the capillary diameter ratio as the minimum for the dimensionless permeability.

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SCOPE

It has been recognized for a long time that the flow channels (pores) in porous media contain constrictions and expansions, resulting in conical flow (for example, Scheidegger, 1957; Dullien and Batra, 1970) in the channels. The various expressions used to calculate permeabilities, however, usually neglect any effect conical flow may have on the permeability. Therefore, it seemed interesting to determine the magnitude of this effect experimentally. A highly simplified model of the flow channels in porous media has been chosen for this purpose, the permeability of which, excluding the effects of conical flow, could be calculated easily by the Hagen-Poiseuille equation. The measured permeability could then be compared with the predicted value, resulting in an estimate of the magnitude of excess viscous dissipation due to the existence of convergent-divergent flow in the capillaries. The model used