Nonlinear Regression Methods for Simultaneous Property Measurement

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A general procedure for the statistical estimation of thermal properties from experimental data is reviewed. The procedure, based on the techniques of nonlinear regression, includes all traditional methods of property determination and removes four specific restrictions frequently imposed on the design of property measurement experiments. This article surveys nonlinear-regression techniques and applications. A new convergence criterion for halting the iterative calculations is presented. The criterion T_{norm} is based on the Euclidean norm of the scaled normal-equation residuals, equaling zero at a minimum of the root-mean-square deviation T_{rms} and being scaled never to exceed T_{rms}.

Nomenclature

a = element of matrix, defined by Eq. (16)
$\mathbf{A}$ = matrix, defined by Eq. (17)
b = element of matrix, defined by Eq. (15)
$\mathbf{B}$ = matrix, defined by Eq. (17)
c_p = specific heat
E = experimental dependent variable
f = arbitrary function
F = sum-of-squares function, defined by Eq. (8)
g = residual of a normal equation, defined by Eq. (28)
$\mathbf{G}$ = defined following Eq. (28)
$\mathbf{I}$ = identity matrix
k = thermal conductivity
k = extinction coefficient, Eq. (4)
n = refractive index
N = scaling factor, defined by Eq. (33)
p = parameter
$\mathbf{P}$ = parameter vector
q = total number of parameters
r = total number of experimental observations
s = interpolation parameter, Eq. (21)
t = time
T = theoretical dependent variable
T_{rms} = root-mean-square difference $E - T$, defined by Eq. (9)
T_{norm} = Euclidean norm of normal-equation residuals, defined by Eq. (35)
u = defined by Eq. (4)
v = defined by Eq. (4)
x = distance
α = thermal diffusivity
δ = convergence criterion
ϵ = small, arbitrary constant
θ = angle
λ = parameter in Marquardt's algorithm
ρ = density
ρ = reflectivity, Eq. (4)

Superscripts

$-$ = quantity evaluated at the minimum value of F
n = iteration number
$*$ = transformed by Eq. (23)

Subscripts

i = index for experimental observations
j = index for parameters
k = index for parameters
$\perp$ = perpendicular-polarized component

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Introduction

ONE hypothesizes mathematical models of a physical system to understand and predict system behavior. To correlate a model to the physical phenomena, certain mathematical parameters related to the physical properties of the system must be determined experimentally. The process of "experimentally measuring a property" is simply the determination of one or more parameter values to obtain a satisfactory match between model predictions and experimental observations.

The traditional approach to property measurement is to create experimental conditions for which system behavior is described by a limited form of the model, so that each property can be measured separately. In application, these individually measured properties must be capable of predicting system behavior under more general conditions.

The digital computer has made it possible to model complex physical systems accurately. These models may describe numerous coupled phenomena which are difficult to separate when designing experiments to measure individual properties. During the past decade many restricted forms of simultaneous property measurement have been developed to provide property values for use in these complex models.[1–7] One example of such a system is the internal transfer of energy in char-forming ablative materials involving several phenomena including conduction, radiation, pyrolysis, gas flow, and deposition.

The present paper reviews a general method, based on the techniques of nonlinear regression, for simultaneously determining physical properties. The method includes all traditional methods of property determination as special cases, and removes four specific restrictions frequently imposed on the design of property measurement experiments. The authors' objective is to show the wide applicability of the nonlinear regression method and to encourage other researchers to consider the general approach when designing property measurement experiments.

Property Measurement Problem

Experiment and Model

The procedure of experimentally measuring properties consists of determining appropriate parameter values which produce the best agreement between a selected mathematical model and experimental data. In the following paragraphs

we formally describe the experimental observations and mathematical model.

In an experiment, one measures the magnitudes of one or more quantities which are designated mathematically as the dependent variables. For simplicity, we consider only one dependent variable, although one could consider an n-dimensional vector of dependent variables. The experimental dependent variable E is measured at r discrete values of the independent variables. For present purposes we restrict attention to two independent variables: distance x and time t. If one measures E at r different pairs of values of the two independent variables, the experimental data may be expressed symbolically as

$$E_i = E(x_i,t_i) \text{ for } i = 1,2, \ldots, r \tag{1}$$

The model describes a mathematical relationship between the theoretical dependent variable T and the independent variables x and t.‡ In addition, the dependent variable is also functionally dependent on certain parameters. We consider that there are q parameters, $p_1, p_2, \ldots, p_q$ which may be represented by a q-dimensional parameter vector $\mathbf{P}$. These parameters for a thermal model are related to such quantities as thermal conductivity, specific heat, reflectance, emittance, transmittance, and decomposition rate constants. Thus the theoretical dependent variable is a function not only of the two variables x and t, but also of the parameters that characterize the system. Symbolically we represent the model as

$$T = T(x,t;p_1,p_2, \ldots,p_q) = T(x,t;\mathbf{P}) \tag{2}$$

and its predictions for comparison with the experimental data E_i as

$$T_i = T(x_i,t_i;\mathbf{P}), \; i = 1,2, \ldots, r \tag{3}$$

Properties and Parameters

To pose correctly the problem of relating a model to the experiment, a distinction must be made between properties and parameters. In this section we illustrate the distinction. A parameter is a quantity to which arbitrary values are assigned; it cannot be a function of any of the variables or other parameters. A property is a characteristic quality of the system; it may be a function of any of the variables or other parameters. Consider three examples:

1) Reflectivity (perpendicular-polarized component) as defined by Fresnel's equation,[8]

$$\rho_\perp(\theta) \equiv \frac{u^2 + v^2 - 2u\cos\theta + \cos^2\theta}{u^2 + v^2 + 2u\cos\theta + \cos^2\theta} \tag{4}$$

where

$$2u^2 = [(n^2 - k^2 - \sin^2\theta)^2 + 4n^2k^2]^{1/2} + (n^2 - k^2 - \sin^2\theta)$$

$$2v^2 = [(n^2 - k^2 - \sin^2\theta)^2 + 4n^2k^2]^{1/2} - (n^2 - k^2 - \sin^2\theta)$$

The property $\rho_\perp$ is a function of the independent variable θ, the refractive index n, and the extinction coefficient k. The symbols n and k denote parameters, and their values must be determined in order to specify the value of the property $\rho_\perp$.

2) Temperature-dependent thermal conductivity,

$$k = f(T) \equiv p_0 + p_1T + p_2T^2 \ldots + p_qT^q \tag{5}$$

The property k is a function of the dependent variable T, and the $(q + 1)$ parameters, $p_0, p_1, p_2, \ldots, p_q$. Again the parameter values must be determined to specify the property value.

3) Temperature-independent thermal conductivity,

$$k \equiv p_0 \tag{6}$$

The property k is identical with the parameter p_0; this is frequently the case.

The quantities that must be evaluated to relate a mathematical model to an actual physical system are the parameters; this article might be better entitled "Nonlinear Regression Methods for Simultaneous Parameter Measurement."

Historical Development of a General Method

Conventional Restrictions

Conventional methods of parameter measurement determine the parameters from a restricted subset of the aforementioned general model and experiment. Specifically, conventional methods require the model, Eq. (2), to be of the form $T = p_1f(x,t)$ so that the parameter p_1 may be determined from the formula $p_1 = E_1/f(x_1,t_1)$. These limitations of conventional methods are summarized in the following four restrictions.

1) Boundary conditions are chosen for the general model so that the parameter vector reduces to a scalar; i.e., tests are designed to measure only one parameter at a time. This restriction requires that the experimental apparatus be carefully designed so that the experiment is described by a restricted form of the model in which the theoretical dependent variable is a function of only the one parameter. For example, a conventional experiment to measure thermal conductivity is designed with the boundary conditions controlled to produce a steady state; therefore, the experimental dependent variable, temperature, does not depend on the volumetric specific heat.

2) Only the minimum number of experimental measurements of the dependent variable is made. Thus, if only one parameter is to be determined in a given experiment, only one measurement of the dependent variable is made in excess of those measurements that are required to specify the boundary conditions for the model. In a conventional steady-state thermal-conductivity experiment, the heat flux and one temperature are measured to specify boundary conditions for the model, and a second temperature reading allows the determination of thermal conductivity. A minimum of q excess measurements must be made in an experiment in which q parameters are being determined. Conventionally, when additional measurements are taken they are replications of the minimal experiment; the parameter value is determined for each test and the average value from all tests is used as the most reliable estimate. This method of interpreting additional data is not necessarily a statistically valid procedure.

3) The boundary conditions are chosen so that a closed-form analytical expression describes the mathematical relationship between the theoretical dependent variable, the independent variables, and the parameter vector.

4) The dependent variable is a linear function of the parameters.

As discussed in the Introduction it is often not possible or convenient to design an experiment to satisfy the first restriction. If one attempts to satisfy the first restriction, one is often forced into measuring properties rather than true parameters. The first and second restrictions produce an experiment that is statistically invalid if the parameters are to be used in a model more general than the simplified form which describes the experiment. The second, third, and fourth restrictions are helpful in minimizing the required computational effort, but with high-speed digital computers these restrictions are no longer necessary.

Progress toward a General Method

Over the past decade, many investigators have presented various techniques that eliminate one or more of the aforementioned restrictions. Most of these investigators' contributions have been to present new methods of solving specific

‡ E and T denote the same dependent variable. E is the experimentally observed value; T is the theoretically predicted value.

problems, rather than to generalize their contributions to embrace both their new concepts and traditional methods. References 1–7 and 9–14 present various new methods proposed for measuring thermal conductivity, thermal diffusivity, and specific heat. The list is by no means complete, nor does it necessarily give credit to originators, but points out that an extensive number of methods have been proposed in one field alone.

Hsu[1] relaxed restrictions 1 and 4 by devising a transient method for simultaneous determination of the heat-conduction parameters $p_1 = k\rho c_p$ and $p_2 = k/\rho c_p$. In the model for his experiment, Eq. (2) had the following nonlinear closed form:

$$T = T_0 + [c_1/(1 + c_2 p_1^{1/2})] \operatorname{erfc} [(x/2)(p_2 t)^{-1/2}] \quad (7)$$

in which T_0, c_1, and c_2 are functions only of the imposed boundary and initial conditions, and erfc denotes the complementary gauss error function. From these results, the thermal conductivity k and volumetric specific heat ρc_p can be calculated.

Hsu's method illustrates one problem which clearly must be recognized: we must be careful for any given experimental design to discover the minimum number of mathematical parameters q which is required to state the mathematical model uniquely. Although we normally use three physical properties ρ, c_p, and k to describe transient conduction, only two mathematical parameters are required in the transient heat conduction equation with constant properties. We can choose these two parameters to be $p_1 = k$, $p_2 = \rho c_p$ or we could choose them to be $p_1 = k$, $p_2 = k/\rho c_p = \alpha$ or we could treat ρ as being a specified constant in the model rather than an unknown parameter and then compute the two quantities, k and c_p.

Filippov[5] recently summarized several methods that he used to measure two thermal parameters simultaneously. Filippov did not relax restriction 3, since he considered only boundary conditions for which the dependent variable could be expressed as a unique analytical function of the two parameters. He used the minimum number of measurements of the dependent variable (two) for each calculation of the parameter values, but recognized that in his transient thermal test an excess of data was available, and used this additional information as an internal cross-check of his results.

Restriction 2 can be relaxed by using the method of least squares. Thomann and Lindsjo[9] recently discussed the design of experiments for this form of data reduction; however, they used linear least-squares, which retains restrictions 3 and 4.

Nonlinear Regression

A mathematical procedure applicable for determining the optimum parameters for a mathematical model had been known for a century before the high-speed digital computer made it practical. This procedure, called nonlinear regression, nonlinear least-squares, or nonlinear estimation, was extended during the past two decades by statisticians including Box and co-workers.[15–18] A text by Draper and Smith[19] contains an excellent chapter and an extensive bibliography on nonlinear regression. The technique has been widely used in determining missile and satellite trajectories.[20] Nonlinear regression need not and should not be subjected to any of the four restrictions above; however, if these four restrictions are applied, nonlinear regression reduces to traditional parameter measurement methods.

In 1961 Beck apparently first applied this method of analysis to thermal parameter determination. Beck's first article[14] removed restrictions 2–4; he determined only one parameter, the thermal diffusivity. While recognizing the general ramifications of the method, he realized that before the method could become a practical tool, much analysis was required in learning how to design experiments that minimize the mathematical correlation between parameters. Beck's main interest[21–26] has been to design analytically an optimum experiment or experiments for the simultaneous determination of two thermal parameters.

The present authors have extended Beck's pioneering work to a charring-ablator model involving 16 thermal parameters. A recent paper[27] presents an application of the method to the simultaneous determination of six thermal parameter values; the determined values are in satisfactory agreement with conventional measurements. Other suggested applications of nonlinear regression to thermal parameter determination can be found in Refs. 28–33.

General Method of Parameter Determination

Consider a general problem in which one wishes to determine q parameters, $p_1, p_2, \ldots, p_q$, which appear in a mathematical model§ represented by Eq. (2). Consider a set of r experimental measurements (where $r \geq q$) represented by Eq. (1). Nonlinear regression is the process of determining the value of the q-dimensional parameter vector $\mathbf{P} = (p_1, p_2, \ldots, p_q)$ which will minimize the sum-of-squares function

$$F = \sum_{i=1}^{r} (E_i - T_i)^2 \quad (8)$$

where E_i and T_i are defined by Eqs. (1) and (3); thus, the procedure is to determine the value of the parameter vector $\mathbf{P}$ which will minimize the difference in a least-squares sense between theory and experiment for all available data. This value of $\mathbf{P}$ is denoted $\bar{\mathbf{P}}$ and all other quantities evaluated at the minimum value of F are marked with a bar. The sum-of-squares function is related directly to the root-mean-square (rms) difference between experimental and theoretical dependent variables:

$$T_{\text{rms}} = (F/r)^{1/2} \quad (9)$$

If a minimum of F exists, then at this minimum the gradient of F vanishes:

$$\partial F/\partial p_1 = \partial F/\partial p_2 = \ldots = \partial F/\partial p_q = 0 \quad (10)$$

or, from Eq. (8),

$$\frac{\partial F}{\partial p_k} = -2 \sum_{i=1}^{r} \left[(E_i - T_i) \frac{\partial T_i}{\partial p_k} \right] = 0 \text{ for } k = 1, \ldots, q \quad (11)$$

Equation (11) represents a set of q nonlinear equations for the q unknowns represented by the vector $\mathbf{P}$, the quantities T_i and $\partial T_i/\partial p_k$ both being functions of $\mathbf{P}$.

Newton-Raphson Iterative Method

Several methods are used for solving Eq. (11) for the parameter vector $\bar{\mathbf{P}}$. The customary Newton-Raphson procedure linearizes Eq. (11) locally by expanding T_i in a first-order Taylor series with the parameters as the independent variables in the expansion. T_i is expanded about the point T_i^n (the value of T_i at the nth interation when $\mathbf{P} = \mathbf{P}^n$) in order to obtain an improved estimation of T_i, namely T_i^{n+1}

$$\bar{T}_i \approx T_i^{n+1} \approx T_i^n + \sum_{j=1}^{q} (p_j^{n+1} - p_j^n) \left. \frac{\partial T_i}{\partial p_j} \right|_{\mathbf{P}=\mathbf{P}^n} \quad (12)$$

In this expansion, the change in parameter p_j at the nth iteration $(p_j^{n+1} - p_j^n)$ is an iteration variable for which an opti-

§ No restrictions of practical consequence are placed on the form of the model or the techniques used to solve the model. In Ref. 27, we treat a model consisting of two coupled nonlinear partial differential equations which we integrate using numerical (finite difference) techniques.

mum value is sought, and p_j^n may be regarded as a fixed constant. The parameter derivatives at $\bar{\mathbf{P}}$ in Eq. (11) are approximated by their values at the nth iteration, which is consistent with the use of a first-order Taylor series

$$(\partial T_i/\partial p_k)\big|_{\mathbf{P}=\bar{\mathbf{P}}} \approx (\partial T_i/\partial p_k)\big|_{\mathbf{P}=\mathbf{P}^n} \equiv (\partial T/\partial p_k)\big|_{(i;n)} \quad (13)$$

The so-called Normal Equations are derived from Eqs. (11–13)

$$\sum_{i=1}^{r} (E_i - T_i^n) \left.\frac{\partial T}{\partial p_k}\right|_{(i;n)} = \sum_{j=1}^{q} \left[(p_j^{n+1} - p_j^n) \sum_{i=1}^{r} \left.\frac{\partial T}{\partial p_j}\right|_{(i;n)} \left.\frac{\partial T}{\partial p_k}\right|_{(i;n)} \right] \quad (14)$$

for $k = 1, 2, \ldots, q$.

The linear form of Eq. (14) may be exhibited clearly through the use of matrix notation; by defining the elements

$$b_k^n \equiv \sum_{i=1}^{r} (E_i - T_i^n) \left.\frac{\partial T}{\partial p_k}\right|_{(i;n)} \quad (15)$$

$$a_{kj}^n = a_{jk}^n \equiv \sum_{i=1}^{r} \left.\frac{\partial T}{\partial p_j}\right|_{(i;n)} \left.\frac{\partial T}{\partial p_k}\right|_{(i;n)} \quad (16)$$

and the three matrices

$$\mathbf{B}^n \equiv \begin{bmatrix} b_1^n \\ \cdot \\ \cdot \\ \cdot \\ b_q^n \end{bmatrix}, \quad \mathbf{A}^n \equiv \begin{bmatrix} a_{11}^n & \cdots & a_{1q}^n \\ \cdot & & \cdot \\ \cdot & & \cdot \\ a_{q1}^n & \cdots & a_{qq}^n \end{bmatrix}, \quad \mathbf{P}^n \equiv \begin{bmatrix} p_1^n \\ \cdot \\ \cdot \\ p_q^n \end{bmatrix} \quad (17)$$

Equation (14) becomes

$$\mathbf{A}^n(\mathbf{P}^{n+1} - \mathbf{P}^n) = \mathbf{B}^n \quad (18)$$

or

$$\mathbf{P}^{n+1} = \mathbf{P}^n + (\mathbf{A}^n)^{-1}\mathbf{B}^n \quad (19)$$

Equation (19) is a linear equation for $\mathbf{P}^{n+1}$ which is readily solved by any of several methods once the elements of $\mathbf{A}^n$ and $\mathbf{B}^n$ have been computed.[34] The computation of these elements requires the determination of the parameter derivatives $(\partial T/\partial p_k)|_{(i;n)}$. These derivatives cannot be determined analytically unless we impose restriction 3. Since one of the prime motivations for using the general method is the ability to handle complex models solvable only by numerical techniques, we assume in the remainder of the discussion that these derivatives also must be obtained by numerical methods. The derivatives are normally approximated by finite differences;

$$\left.\frac{\partial T}{\partial p_k}\right|_{(i;n)} \approx \frac{T(x_i,t_i;p_1^n, \ldots, (1+\epsilon)p_k^n, \ldots, p_q^n) - T_i^n}{\epsilon p_k^n} \quad (20)$$

where ϵ is a small number. Thus, to solve for the elements of the matrices $\mathbf{A}^n$ and $\mathbf{B}^n$ of Eq. (19), it is necessary to solve the model represented by Eq. (2), $q + 1$ times for each iteration: first at $\mathbf{P}^n$ and then varying in succession each parameter by a small amount.

A second method for determining the derivatives is the method of parametric differentiation discussed by Chapman and Kirk.[35] They apply the method to a model which is an nth-order nonlinear ordinary differential equation. For this model, the method requires at each iteration the numerical solution of the nth-order nonlinear ordinary differential equation and q independent nth-order linear ordinary differential equations with variable coefficients. Blizzard and Jirka[28] attempted to apply parametric differentiation to a model which is a nonlinear partial differential equation; however, they incorrectly differentiated the model.[29] To the authors' knowledge, the method has not been correctly applied to partial differential equations; if it were, it would require at each iteration the numerical solution of the nonlinear partial differential equation and q independent linear partial differential equations with variable coefficients. Thus, the finite difference and the parametric differentiation methods require a comparable number of calculations.

Alternate Iterative Methods

The standard iterative procedure outlined above becomes impractical at large values of q unless steps are taken to insure rapid convergence. There are three steps that should be taken to speed convergence to $\bar{\mathbf{P}}$ and to minimize computer time.

1) The experiments should be carefully designed to reduce correlation between parameters. The reader is referred to the work of Box,[15–18] Beck,[21–26] and Pfahl.[31]

2) The initial estimate of the parameter vector P should be carefully chosen. Kitrell et al.[36] discuss several procedures.

3) The basic Newton-Raphson method of solving the nonlinear equations may be modified. The following paragraphs outline two different techniques.

Booth and Peterson[37] and Hartley[38] have presented interpolative methods for speeding convergence. The interpolative methods determine the point where the sum-of-squares function F is a minimum along the line represented by

$$\mathbf{P}^n + s(\mathbf{P}^{n+1} - \mathbf{P}^n), \text{ where } 0 < s < \infty \quad (21)$$

If the true minimum of F is not near the line defined by Eq. (21), the method is of little value; we have found that the method is of value for $q \leq 2$.

Marquardt[39] has presented an algorithm which is widely used. The method reduces F at each iteration unless $\mathbf{P}^n$ is a stationary point. The matrices $\mathbf{A}^n$, $\mathbf{B}^n$, and $\mathbf{P}^n$ of Eq. (17) are scaled as follows:

$$\mathbf{B}^{*n} \equiv \begin{bmatrix} b_1^{*n} \\ \cdot \\ \cdot \\ \cdot \\ b_q^{*n} \end{bmatrix}, \quad \mathbf{A}^{*n} \equiv \begin{bmatrix} 1 & \cdots & a_{1q}^{*n} \\ \cdot & & \cdot \\ \cdot & & \cdot \\ a_{q1}^{*n} & \cdots & 1 \end{bmatrix}, \quad \mathbf{P}^{*n} \equiv \begin{bmatrix} p_1^{*n} \\ \cdot \\ \cdot \\ p_q^{*n} \end{bmatrix} \quad (22)$$

where

$$b_k^{*n} \equiv \frac{b_k^n}{(a_{kk}^n)^{1/2}}, \quad a_{kj}^{*n} \equiv a_{jk}^{*n} \equiv \frac{a_{kj}^n}{(a_{kk}^n a_{jj}^n)^{1/2}}, \quad \text{and } p_k^{*n} \equiv (a_{kk}^n)^{1/2} p_k^n \quad (23)$$

The scaled $\mathbf{A}^n$ matrix $\mathbf{A}^{*n}$ is a symmetric positive definite matrix with diagonal elements of unity. It is known as the matrix of simple correlation coefficients.[39] For a perfectly designed experiment, $\mathbf{A}^{*n}$ should approach the identity matrix,

$$\mathbf{I} \equiv \begin{bmatrix} 1 & 0 & \cdots & 0 \\ 0 & 1 & & \cdot \\ \cdot & & & \cdot \\ \cdot & & & \cdot \\ 0 & 0 & \cdots & 1 \end{bmatrix}$$

In terms of the above scaled matrices, Eq. (18) becomes

$$\mathbf{A}^{*n}(\mathbf{P}^{*n+1} - \mathbf{P}^{*n}) = \mathbf{B}^{*n} \quad (24)$$

Marquardt's algorithm modifies this equation to

$$(\mathbf{A}^{*n} + \lambda^n \mathbf{I})(\mathbf{P}^{*n+1} - \mathbf{P}^{*n}) = \mathbf{B}^{*n} \quad (25)$$

where λ^n is a scalar constant which may be changed at each iteration. If $\lambda^n = 0$, Eq. (25) reduces to the Newton-Raphson method. If $\lambda^n \to \infty$, the direction of change in $\mathbf{P}$ approaches that of the steepest-descent method; however, in this case $||\mathbf{P}^{n+1} - \mathbf{P}^n|| \to 0$ and convergence may be very

slow. Marquardt[39] provides rules for the selection of λ^n so that F is reduced at every iteration.

Quasi-Newton Methods

With the aforementioned methods of solution, a large portion of the computing time must be devoted at each iteration to calculating the parameter derivatives by finite differences. Many quasi-Newton methods have been proposed which do not require the calculation of all of the derivatives at each iteration.[40-45] Zeleznik[46] has recently presented a unified derivation of the quasi-Newton methods. Vitale and Taylor[47] compared the Booth-Peterson-Hartley method[37,38] with the Davidon-Fletcher-Powell method[40,41]; they concluded that the choice of method depends on the model. The authors have used the simplest variant of the quasi-Newton methods and have found it extremely helpful as q becomes large, provided $\mathbf{P}^n$ is in the vicinity of $\mathbf{P}$ so that the neglected terms of the Taylor series expansion are small. This variant assumes that the matrix $\mathbf{A}^{*n}$, and consequently $(\mathbf{A}^{*n})^{-1}$, does not change from iteration to iteration. The inverse matrix is calculated on the first iteration; on succeeding iterations only $\mathbf{B}^{*n}$ is calculated to determine $\mathbf{P}^{*n+1}$. After several iterations the inverse matrix must be recalculated.

Convergence Criteria

As one carries out the foregoing iterative procedure and hopefully approaches a solution to the set of equations, there are three questions to be asked of the latest estimate, $\mathbf{P}^n$, to the vector $\bar{\mathbf{P}}$: 1) Is the sequence $\mathbf{P}^n$ approaching a value of $\mathbf{P}$ for which F is a minimum? 2) How closely do the model predictions and experimental data agree for the set of parameters $\mathbf{P}^n$? 3) How good an estimate of $\bar{\mathbf{P}}$ is $\mathbf{P}^n$? We consider these three questions in order.

The first question is difficult to answer analytically, if the derivatives must be determined by numerical means. Therefore, it is customary to assume that if $\mathbf{P}^n$ is a reasonable answer which, for arbitrarily selected surrounding points, is a local minimum, then $\mathbf{P}^n$ is an estimate of a value of $\bar{\mathbf{P}}$ for which F is a minimum rather than some other form of stationary point.

The second question is answered by computing the value of T_{rms} for $\mathbf{P} = \mathbf{P}^n$ since T_{rms} is a direct measure of the agreement between the model and the experiment. Our goal is to have T_{rms} approach zero; however, it will never reach zero since there is never perfect agreement between the numerical model and the experiment. The three sources of this disagreement are experimental error, imperfections in the model, and numerical approximations in the finite difference calculations.

The third question becomes important when the successive values of T_{rms} do not approach zero. One needs to have a method of determining whether the reason for large values of T_{rms} is that the model does not fully describe the relevant physical processes, or that the iterative procedures have not approached a solution to Eq. (11). The following discussion develops a new measure of convergence, T_{norm}, to answer this third question. There are certain properties that T_{norm} should have. 1) It should equal zero for $\mathbf{P}^n = \bar{\mathbf{P}}$. 2) It should have the same units as T_{rms}. 3) It should be scaled for convenient interpretation; a logical choice being that it never exceed T_{rms}.

In functional form the q equations represented by Eq. (11) may be rewritten as

$$f_k(\mathbf{P}) \equiv \sum_{i=1}^{r} \left[(E_i - T_i) \frac{\partial T_i}{\partial p_k} \right] = 0 \text{ for } k = 1, \ldots, q \quad (26)$$

These equations will be satisfied only when $\mathbf{P} = \bar{\mathbf{P}}$. The solution to the equations will not be changed if each equation is multiplied by an arbitrary, nonzero, constant, N_k, so that the units of each equation will be (units of T) instead of (units of T^2/units of p_k). Therefore, we may write Eq. (26) as

$$N_k f_k(\mathbf{P}) = 0 \text{ for } k = 1, \ldots, q \quad (27)$$

Again this equation will be satisfied only when $\mathbf{P} = \bar{\mathbf{P}}$. At the nth iteration when $\mathbf{P} = \mathbf{P}^n$, the equality in general will not be true and instead

$$N_k f_k(\mathbf{P}) = g_k(\mathbf{P}) \text{ for } k = 1, \ldots, q \quad (28)$$

The functions g_k can be considered to be elements of a vector, $\mathbf{G}(\mathbf{P}) \equiv [g_1(\mathbf{P}), g_2(\mathbf{P}), \ldots, g_q(\mathbf{P})]$, in q-dimensional space. The norm of the vector $\mathbf{G}(\mathbf{P})$ can be used as a measure of the degree to which $\mathbf{P}$ satisfies Eq. (27), since only when the norm of $\mathbf{G}(\mathbf{P})$ vanishes will $\mathbf{P} = \bar{\mathbf{P}}$. Therefore, we define

$$T_{\text{norm}} \equiv ||\mathbf{G}(\mathbf{P})|| \equiv \left(\sum_{k=1}^{q} g_k^2 \right)^{1/2} \equiv \left(\sum_{k=1}^{q} N_k^2 f_k^2 \right)^{1/2} \quad (29)$$

From the definition of f_k in Eq. (26),

$$T_{\text{norm}} = \left(\sum_{k=1}^{q} \left\{ N_k^2 \left[\sum_{i=1}^{r} (E_i - T_i) \frac{\partial T_i}{\partial p_k} \right]^2 \right\} \right)^{1/2} \quad (30)$$

To satisfy requirement 3, we apply Cauchy's inequality[48]:

$$\left[\sum_{i=1}^{r} a_i b_i \right]^2 \leq \sum_{i=1}^{r} a_i^2 \sum_{j=1}^{r} b_j^2 \quad (31)$$

with $a_i = E_i - T_i$ and $b_j = \partial T_j / \partial p_k$ to obtain the inequality:

$$T_{\text{norm}} \leq \left(\sum_{i=1}^{r} (E_i - T_i)^2 \sum_{k=1}^{q} \left\{ N_k^2 \sum_{j=1}^{r} \left(\frac{\partial T_j}{\partial p_k} \right)^2 \right\} \right)^{1/2} \quad (32)$$

Therefore, if we choose the arbitrary constants N_k so that

$$N_k^2 = \left[1 \Big/ rq \sum_{j=1}^{r} \left(\frac{\partial T_j}{\partial p_k} \right)^2 \right] \quad (33)$$

then we obtain the desired result that

$$T_{\text{norm}} \leq \left\{ \sum_{i=1}^{r} (E_i - T_i)^2 \Big/ r \right\}^{1/2} \equiv T_{\text{rms}} \quad (34)$$

Thus, substituting Eq. (33) into Eq. (30) completely specifies T_{norm};

$$T_{\text{norm}} \equiv \left(\frac{1}{rq} \sum_{k=1}^{q} \left[\left\{ \sum_{i=1}^{r} (E_i - T_i) \frac{\partial T_i}{\partial p_k} \right\}^2 \Big/ \sum_{j=1}^{r} \left(\frac{\partial T_j}{\partial p_k} \right)^2 \right] \right)^{1/2} \quad (35)$$

Equation (35) is evaluated at $\mathbf{P} = \mathbf{P}^n$ to obtain T_{norm^n}. This definition of T_{norm} meets the three requirements which were specified for a measure of the degree to which a given vector $\mathbf{P}$ satisfies Eq. (26).

A geometrical interpretation of T_{norm} may be derived; substitution from Eqs. (11) and (16) into Eq. (35) yields

$$T_{\text{norm}} \equiv \frac{1}{2} \left[\frac{1}{rq} \sum_{k=1}^{q} \left(\frac{\partial F}{(a_{kk})^{1/2} \partial p_k} \right)^2 \right]^{1/2} \quad (36)$$

Substitution of the definition of $p_k{}^{*n}$ from Eq. (23) yields

$$T_{\text{norm}} \equiv \frac{1}{2} (rq)^{-1/2} \left[\sum_{k=1}^{q} \left(\frac{\partial F}{\partial p_k{}^*} \right)^2 \right]^{1/2} \quad (37)$$

The term in brackets in Eq. (37) is the norm of the gradient of F in the scaled parameter space; thus

$$T_{\text{norm}} \equiv \tfrac{1}{2} (rq)^{-1/2} ||\text{grad}^* F|| \quad (38)$$

It is important to note that the gradient of F is not invariant under the transformation of Eq. (23).[39] Other criteria of convergence have been presented based on norms of various transformed gradients of F; however, in the authors' experience,

this choice is preferable since the scaling relative to T_{rms} permits rapid interpretation of the progress of the iterations.

All criteria for ending the iterative calculations have certain shortcomings; the appropriate criterion depends on the experiment, the model, and the technique used to solve the nonlinear equations. For the ablation problems considered by the authors, not only the parameter values, but the model validity was unknown; for this type of problem, we recommend the use of $T_{norm} \leq \delta_1$ and $T_{norm}/T_{rms} \leq \delta_2 < 1$ as criteria of convergence. If $T_{norm} \to 0$, but T_{rms} remains large, one then has an indication that he should question either the validity of his model or the accuracy of his experimental data. Other criteria have been suggested by Booth and Peterson.[37] Beck[23] recommended the criterion $|(p_k{}^{n+1} - p_k{}^n)/p_k{}^n| \leq \delta_3$, for $k = 1, 2, \ldots, q$.

Computer Programs

It is apparent that the magnitude of the computations for large values of q, for extensive sets of experimental data, or for complex models necessitates the use of a high-speed digital computer. The authors have incorporated the basic nonlinear regression calculations as well as the variations presented in Ref. 29 into a digital computer program written in the Fortran language; a mathematical model describing a charring ablator[27,49] forms a subroutine of the program. Programs by Booth and Peterson[37] and Marquardt[50] are available. Meeter[51] has written a general program which accepts the model as a subroutine.

Model Identification

Frequently, in addition to determining parameter values, one must identify the appropriate model; identification is an important problem when modeling complex physical systems. This problem of "finding out how a system works" has been described by Box and Hill[52] in an excellent article. The choice of model or competing models must precede the determination of parameters; however, the adequacy of the model cannot be decided until the parameter values are determined. Thus, model identification and parameter determination are inseparable.

Beck[53] recently investigated these problems for heat conduction with temperature-dependent thermal conductivity described by Eq. (5). He concluded that to minimize computation and to aid in model identification, one should first estimate the dominant parameter, p_0, by assuming the other lesser parameters $p_1, p_2, \ldots, p_q$ to be zero. If this temperature-independent model, Eq. (6), proves inadequate, then one should consider the linear term and determine p_0 and p_1. Since p_0 should not change significantly in the new model, and since p_1 should be small, one iteration might suffice to determine both parameters.

The present authors[30] developed through practical experience a procedure for determining the thermal diffusivity of a charring ablator, which anticipated the results of Beck's analysis. The temperature-independent diffusivity was first calculated; if this model did not produce adequate agreement with the experiment, a linear temperature-dependent diffusivity model was used. The new calculations were started at the previously determined constant value.

Concluding Remarks

Simultaneous parameter measurement by means of nonlinear regression is a general approach which embraces all traditional techniques. Recent developments have made the general method a practical alternative to individual parameter measurement in areas of thermophysics where coupled energy transfer and storage processes cannot be isolated conveniently. These developments include analytical studies on the design of optimal experiments for simultaneous measurement of two parameters, new iterative techniques for rapidly solving nonlinear equations, and a convergence criterion introduced in this paper.

This paper has been presented in the hope of stimulating other researchers to adopt the method. The authors have demonstrated the successful application of the method to the simultaneous determination of six thermal parameters describing a charring ablator.[27] There are many other problems where this general method should prove useful, such as the elastic-plastic regime in solids, multistep chemical reactions, radiant transfer through an absorbing medium, and visco-elastic flow or turbulent flow of fluids.

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