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ESTIMATION OF KINETIC PARAMETERS OF COMPOSITE MATERIALS DURING THE CURE PROCESS USING THE COMBINED WAVELET REGULARIZATION METHOD

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The limitation of the experimental methods in thermophysical characterization of composite materials leads to an increased use of inverse parameter estimation techniques. However, in some situations the convergence of the inverse algorithm is impossible due to the correlation of the involved parameters and the existing noises in measurement data. Several different approaches have been used to tackle this problem. In this article, a new approach is utilized to solve it. This new technique combines the wavelet denoising and Levenberg–Marquardt regularization method. In order to examine this technique, a highly ill-posed problem is considered as a test case, that is, the estimation of the composite kinetic parameters during the cure process.

Keywords: *Wavelet denoising, parameter estimation, composite kinetic parameter, Levenberg–Marquardt.*

Introduction. Inverse methods are commonly used for thermophysical parameter estimation problems. Beck [1] estimated the thermal conductivity simultaneously with the volumetric heat capacity of nickel from one-dimensional transient temperature measurements. Scott and Beck [2] determined these thermal properties for carbon/epoxy composites as functions of temperature and fiber orientation. They also developed a methodology for estimating these two properties in the same composite materials during curing [3]. The researches done by Jurkowski et al. [4] and Garnier et al. [5] showed that small sensitivity coefficients or the unbalance of the sensitivity matrix results in the instability of the estimation procedure. This particular remark correlates with the fact that both the Gauss and modified Box–Kanemasu methods [6] have been found to show that the resulting instabilities cause the divergence of the method when used with models that contain correlated or nearly correlated thermal properties.

Several different approaches have been used to consider this problem. One approach is to modify the experimental design. For example, Loh and Beck [7] were able to estimate simultaneously both the thermal conductivity and volumetric heat capacity of an anisotropic carbon/epoxy composite through the use of nine thermocouples embedded at various locations within a sample. Correlation may still take place but the use of multiple sensors alleviated the problem. Nevertheless, modifications of the experimental design, such as the use of internal sensors, are not always feasible, especially when nondestructive testing is required. In addition, the use of embedded thermocouples can be a source of important bias.

Another approach is applying regularization methods, which can be employed in two ways. Although these methods introduce a bias into the estimation, they significantly stabilize the solution. One type of regularization known as the "Tikhonov regularization" adds a penalty term to the objective function. Additional comments on this type of regularization can be found in the book by Woodbury [8, chapter 2]. The Levenberg–Marquardt (LM) method regularizes the Gauss one when this procedure is applied. The iterative regularization is another type of regularization method. The main idea of this method is to stop the iterative procedure sufficiently closely but not very near to the final optimum point (Ozicik and Orlande, [9]).

It should be noted that the regularization techniques manipulate the original noisy data and use them "as are" for estimating the unknowns. Some authors have proposed to preprocess the data obtained from the sensors before the application of the inverse heat conduction problems (IHCP). This approach consists of different digital filtering methods. Al-Khalidy [10] used digital filter formulation to smooth noisy sensor data in parabolic and hyperbolic inverse

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heat conduction. In this method, each data value is replaced by a combination of itself and a number of adjacent data. Beck [11] proposed a prefiltering formula that is much simpler than that used by Al-Khalidy where every data are replaced by $y_m = (Y_{m-1} + 2Y_m + Y_{m+1})/4$ (here m is the index of time step). Kalman [12] introduced a filtering technique referred to as Kalman filtering. This technique has been used by some authors to solve IHCPs. Ji and Jang [13] applied the Kalman filter to a one-dimensional problem with errors in the measured temperatures by comparing the calculated heat transfer rate with that obtained experimentally. Tuan et al. [14] used the Kalman filter and a real-time least-squares algorithm to solve a two-dimensional inverse heat conduction problem.

In this article, a noise reduction technique based on wavelet transform is used to modify the sensor data before they are used by the IHCP methods. Wavelets as an alternative approach for data analysis have been utilized successfully for signal separation and noise reduction [15]. In this approach, the discrete wavelet transform (DWT) is used to decompose the signal acquired from each sensor in a multiresolution filter bank structure. The wavelet denoising algorithm is then applied for noise reduction that modifies the wavelet coefficients derived during wavelet decomposition. The reconstruction phase is applied to the modified coefficients in order to recover the noise reduced signal. The denoised signal is then considered as an input to the inverse algorithm. Here, we do not explain the wavelet denoising algorithm, and readers who are not acquainted with this subject are referred to our previous paper [16].

The parameter estimation problem defined in what follows is solved by the LM method using sensor data modified by the wavelet method. This inverse problem concerns the estimation of the kinetic parameters of the composite materials during the cure process.

Problem Description. One major step of manufacturing a composite material that consists of a thermosetting resin matrix is using elevated temperature and pressure to cure the material. A comprehensive thermal modeling of such a cure process is rather complicated. For simplification, we assume that the convective heat-transfer effect by resin flow is negligible, and the resin as well as the fiber are at the same temperature at any point in time. With these assumptions, for a one-dimensional heat conduction the governing equations can be expressed as follows [17]:

$$\rho C_p \frac{\partial T}{\partial t} = k \frac{\partial^2 T}{\partial x^2} + \rho H_{\mathrm{r}} \frac{d\alpha}{dt} ,$$

$$\frac{d\alpha}{dt} = F(\alpha, T) , \tag{1}$$

where $d\alpha/dt$ is the cure rate and the function $F(\alpha, T)$ represents the kinetic characterization and determines the cure rate. In this work, $F(\alpha, T)$ is modeled as suggested in [18, 19]:

$$F(\alpha, T) = K(T)\, \alpha^m (1 - \alpha)^n , \tag{2}$$

where m and n describe the order of the curing mechanism and K obeys the Arrhenius law

$$K = A \exp\left[-\frac{E}{RT}\right] . \tag{3}$$

The initial and boundary conditions for Eqs. (1) are as follows:

$$T(x, 0) = T_0 , \quad \alpha(x, 0) = 0 \quad \text{as} \quad -L \le x \le L ;$$

$$T(-L, t) = T_{\mathrm{c}} , \quad T(L, t) = T_{\mathrm{c}} \quad \text{as} \quad 0 \le t \le t_{\mathrm{f}} . \tag{4}$$

In the inverse problem considered in this study, the kinetic parameters m, n, A, and E are unknown and are estimated from measured temperatures of several sensors during the cure process. Before the application of the IHCP algorithm, for estimation of the kinetic parameters the measured temperatures are denoised.

A function of the sum of square errors that should be minimized is defined here, as is commonly used in the inverse methods:

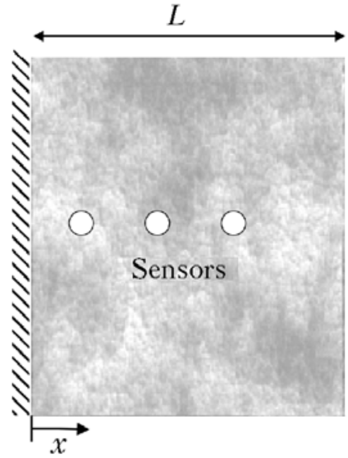

Fig. 1. Geometry of the problem.

$$S = \sum_{j=1}^{N} \sum_{i=1}^{M} (T_{ji} - Y_{ji})^2 , \tag{5}$$

where Y_{ji} are the measured temperatures and T_{ji} are the calculated ones at the measurement locations with the use of the proposed model (Eqs. (1)) and S is a function of the unknown kinetic parameters m, n, A, and E.

The Levenberg–Marquardt method is utilized in this study in order to minimize the objective function S. This method is an iterative technique that finds the unknown parameters for which the minimum point of the function S is reached. Details of the LM method have been described completely in [9, chapter 2]. Due to the nonlinearity of this inverse problem, the sensitivity coefficients with respect to each of the unknowns in each iteration of LM method should be recalculated as

$$X_i = \frac{T(\beta_i + \varepsilon\beta_i) - T(\beta_i)}{\varepsilon\beta_i} + o(\varepsilon\beta_i) , \tag{6}$$

where β stands for any of the parameters m, n, A, and E and ε is a small number of the order of 10^{-5} [20].

Results and Discussion. Consider the system shown in Fig. 1 which is made of carbon fiber and epoxy matrix with the following thermal and kinetic properties [21]: ρ = 1520 kg/m^3, C_p = 0.942 kJ/(kg·K), k = 4.457·10^{-4} kW/(m·K), A = 80.8·10^8 sec^{-1}, E = 78.61 kJ/mol, m = 0.673, n = 1.774, H_r = 120 kJ/kg.

We consider here that the length of the slab is equal to 2 cm, and the experiment time to 2000 sec. The initial temperature of the slab was assumed to be 300 K and the boundary condition temperature 400 K. Of particular interest, before starting any estimation procedure, is the careful analysis of both the degree of correlation of the unknown parameters and the condition number of the sensitivity matrix. These quantities are important, since the small magnitudes of the sensitivity coefficients and near-linear dependence between the coefficients are the limiting factors for the stability and thus for the convergence of gradient-based estimation procedures. In addition, computation of the condition number of the Fisher information matrix X^TX can allow assessment of any ill-conditioning characteristic of the estimation problem [23].

The condition number of the Fisher information matrix X^TX can be defined as

$$cond = \frac{|\lambda_1|}{|\lambda_{np}|} , \tag{7}$$

where λ_1 and $\lambda_{\rm np}$ are respectively the largest and smallest eigenvalues of the X^TX matrix (which has a rank n_p). The condition number for actual parameters is equal to 1.02·10^{17}, which shows a large difference between the highest and lowest sensitivity coefficient magnitudes and linear dependence between the parameter sensitivity coefficients.

Table 1. Correlation Matrix

Parameters	A	E	m	n
A	1.0000	0.9847	–0.1159	0.4182
E	0.9847	1.0000	–0.2747	0.2785
m	–0.1159	–0.2747	1.0000	0.4962
n	0.4182	0.2785	0.4962	1.0000

Table 2. Estimated Parameters for Errorless, Noisy, and Denoised Data

Quantities	$A \cdot 10^{-8}$, sec^{-1}	δA, %	E, kJ/mol	δE, %	m	δm, %	n	δn, %
Errorless	74.67	7.59	78.36	0.3	0.673	–0.06	1.77	0.23
Noisy	12.25	84.9	69.02	12.2	0.997	–48.225	0.436	75.44
Denoised	68.32	15.44	78.41	0.25	0.641	4.65	1.736	2.11

The correlation matrix for this problem, which can be determined on the basis of the procedure described by Walpole and Myers [22], is presented in Table 1. It is the correlation matrix for the real values of kinetic parameters. These values indicate that there is a linear dependence between the parameters A and E in the vicinity of original values. The two following conclusions can be drawn from the matrix: 1) there is a possibility that the estimated values of A and E differ from the real values, but both yield the same result; 2) this matrix can be computed during the estimation process and, when any its component approximates unity, the procedure is stopped, because in such cases more iterations will not cause any noticeable changes in the solutions. The the above discussion shows that the convergence and solution of the parameter estimation will not be easily obtained.

An initial condition is required for starting the estimation. Since the kinetic parameters have wide ranges of values, one cannot choose a single suitable value as the initial condition that yields good results for all composite materials. However, the following proposed values can be considered as primary initial conditions: $A = 10^{10}$ sec^{-1}, E = 1 kJ/mol, m =1, n = 1.

For testing the proposed algorithm, noisy data have to be used; therefore, the data used in this study include: 1) errorless data; 2) data containing errors having a Gaussian distribution with a standard deviation of 0.01% of the maximum temperature; 3) data obtained by denoising (using the wavelet algorithm) of the second set data. Here, three sensors are used for temperature measurement, which is the minimum number of sensors to gain acceptable errorless results. The sensor distances from x = 0 are taken as 0.25, 0.40, and 0.55 cm with the reading interval of 2 sec.

The LM parameter estimation method was carried out with the use of FORTRAN software. First, the estimation results for the non-noisy data are presented. The stopping criterion for this case was

$$S_n < \varepsilon_1 , \tag{8}$$

where ε_1 is an arbitrary small number.

Table 2 includes the values of the parameters and percentage deviations from the real parameters for all the three cases considered. The α-dependences of the estimated and real cure rates are presented in Fig. 2.

The results verify the validity of our parameter estimation procedure and suitability of the initial conditions (see Fig. 2a). At the next stage, the kinetic parameters are estimated on the basis of the same initial conditions with the use of the noisy data. Here, the stopping criterion is also determined by Eq. (8), where $\varepsilon_1 = \sigma^2 MN$. As is shown in Fig. 2c and Table 2, the results obtained for this case are not acceptable. One may reach a more acceptable final solution utilizing a trial and error process in choosing initial conditions. However, this technique is not desirable because it requires significant efforts and is time-consuming. Thus we propose a preprocessing manner as follows: Before using the sensor data in the inverse algorithm, they should first be denoised by the wavelet denosing algorithm, and then the denosied data are to be inserted into the inverse algorithm as an input.

In this study, the wave-menu of the MATLAB toolbox is used for the wavelet denoising calculations. The use of this toolbox for denoising is a simple technique and does not represent a computational load. In our data analysis, we used a conservative thresholding criteria referred to as "penalize high" and hard thresholding in MATLAB. We used coif2 for analyzing wavelets in a multiresolution structure with eleven levels for decomposition.

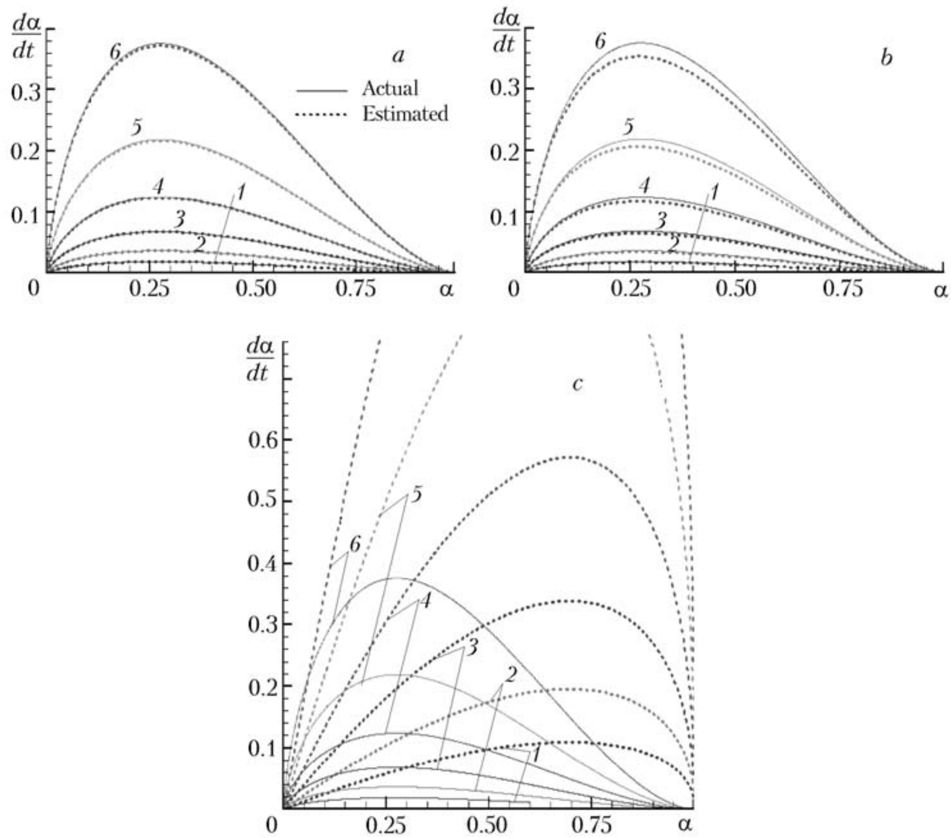

Fig. 2. Simulation of isothermal cure rate based on actual parameters (solid curves) and those from errorless (a), denoised (b), and noisy (c) data (dashed curves): 1) T = 100; 2) 110; 3) 120; 4) 130; 5) 140; 6) 150°C. $d\alpha/dt$, min^{-1}.

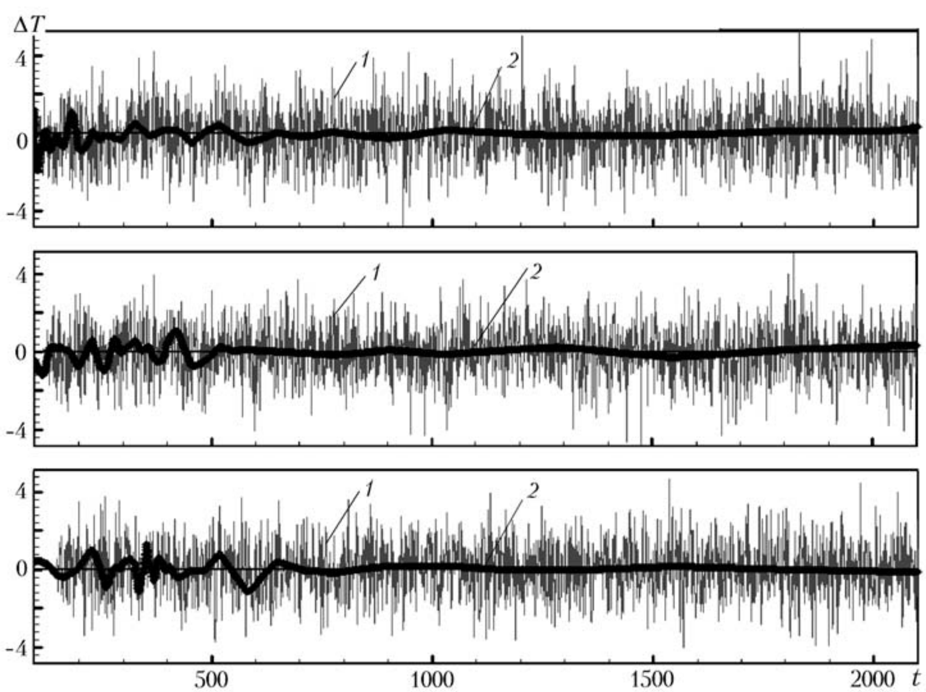

Fig. 3. Difference between noisy and errorless data (1) and that between denoised and errorless data (2) for three sensors. ΔT, K; t, sec.

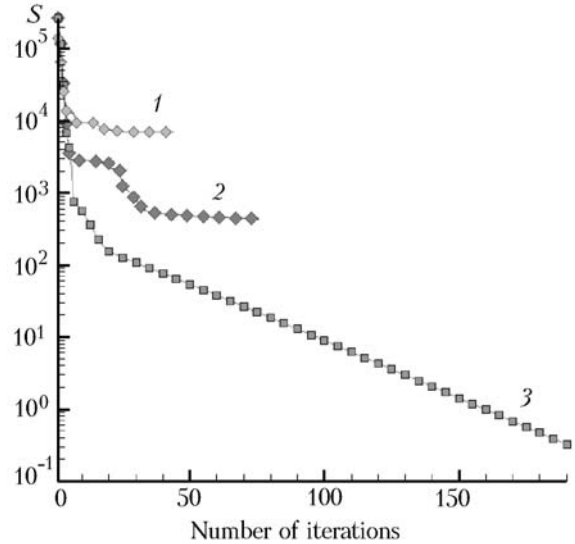

Fig. 4. Reduction history of the objective function for various cases: noisy (1), denoised (2), and non-noisy (3) data.

Both the isothermal cure rate with the use of the parameters obtained from the denoised data and the actual parameters are shown in Fig. 2b. The results (including ones presented in Table 2) indicate that the denoising of data yields an acceptable and accurate solution. In this case, the same initial conditions of non-noisy case were used, and the stopping criterion for this case was

$$|S_{n+1} - S_n| < \varepsilon_2 . \tag{9}$$

The difference ΔT between the noisy and errorless data of three sensors is compared with that between the denoised and errorless data in Fig. 3. Figure 4 enables one to compare the reduction of the objective function with respect to the number of iterations for three cases studied.

Conclusions. Noise reduction in temperature measurement by wavelets was combined with the traditional Levenberg–Marquardt method in order to estimate kinetic parameters in a cure process. This technique has a considerable advantage in comparison with the traditional Levenberg–Marquardt method. In the case where the Levenberg–Marquardt algorithm fails to reach a reasonable solution, the applied method yields a very precise solution with the same number of sensors.

NOTATION

A, kinetic parameter, sec^{-1}; C_p, specific heat, J/(kg·K); E, activation energy, kJ/mol; H_r, total reaction heat, kJ/kg; k, thermal conductivity, W/(m·K); K, kinetic rate constant, sec^{-1}; L, slab thickness, m; m and n, kinetic exponents; M, total number of time steps; N, number of sensors; R, gas constant, kJ/(mol·K); S, objective function; t, time, sec; t_f, final time, sec; T, temperature, K; T_0, initial temperature, K; T_c, boundary condition temperature, K; ΔT, temperature difference, K; X, sensitivity matrix; x, coordinate, m; Y, dimensionless measured temperatures; α, degree of cure; β, true parameter vector; ε, ε_1, ε_2, small numbers; λ, matrix eigenvalue; ρ, density, kg/m^3; σ, standard deviation of the measured temperature errors.

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