An identification algorithm of model kinetic parameters of the interfacial layer growth in fiber composites

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Abstract. This paper considers the identification algorithm of parameters included in a parabolic law that is often used to predict the time dependence of the thickness of the interfacial layers in the structure of composite materials based on a metal matrix. The incubation period of the process and the speed of reaction and pressure are taken into account. The proposed algorithm of identification is based on the introduction of a minimized objective function of a special kind. The problem of identification of unknown parameters in the parabolic law is formulated in a variational form. The authors of the paper have determined the desired parameters, under which the objective function has a minimum value. It is shown that on the basis of four known experimental values of the interfacial layer thickness, corresponding to different values of temperature, pressure and the time of the interfacial layer growth, it is possible to identified four model parameters. They are the activation energy, a pre-exponential parameter, the delay time of the start of the interfacial layer formation, and the parameter determining the pressure effect on the rate of interfacial layer growth. The stability of the proposed identification algorithm is also studied.

1. Introduction
The influence of interfacial zones on physical and mechanical properties of composite materials based on a metal matrix can be significant [1, 2]. A parabolic law is often used to evaluate the interfacial layer thickness at the boundary of fibers and matrix in the metal composite materials [3-9]. This law establishes the relationship that allows determining the variation of the interfacial layer thickness depending on the exposure time of the composite sample at elevated temperature and elevated pressure. In accordance with the parabolic law, thickness of the interfacial layer \( h(t) \) is proportional to square root of the process time parameter \( \sqrt{t} \), and the proportionality coefficient is rate constant \( k \), which in its turn depends on the temperature and the pressure of the environment. To estimate the rate constant Arrhenius relationship is often used. Thus, the relationship used for estimation of the interfacial layer thickness at the boundary of fibers and the matrix is as follows:

\[
h(t) = K_0 \exp \left( -\frac{Q + \kappa P}{2RT} \right) \sqrt{t - t_0}.
\]

where \( P \) is pressure (Pa); \( Q \) is the energy of activation of the reaction zone growth process (J / mol); \( T \) is process temperature (°K); \( R = 8.314 \) is a universal gas constant (J / mol / °K); \( t \) is exposure time (sec); \( t_0 \) is an "incubation" period (sec); \( h \) is the interfacial layer thickness (m); \( K_0 \) is a pre-
exponential parameter \( \text{m/s}^{1/2} \); \( \kappa \) is a parameter determining the effect of pressure on the rate of the interfacial layer growth \((\text{m}^3/\text{mol})\).

The aim of this work is to develop an algorithm of identification of parameters, which are included in the parabolic law and characterize the process of the interfacial layer growth around the fibers in the matrix of a metal composite material. It is assumed that there are \( N \) data points, each of which is a set of four values: \( P, T, t, H \). The task is to determine unknown parameters of the model \( K_0, \tilde{Q}, \kappa, t_0 \) using \( N \) known data points.

### 2. An identification algorithm of the parabolic model parameters of the interfacial layer growth

Let us suppose that there are experimental data that are a set of \( N \) points:

\[
\begin{align*}
P_1, T_1, t_1, H_1, \\
P_2, T_2, t_2, H_2, \\
\vdots \\
P_N, T_N, t_N, H_N,
\end{align*}
\]

The problem of determination of mathematical model parameters is formulated as follows: find such model parameters for which an experimental set of points would be as little different as possible from the theoretical set of points.

Let us pre-convert the equation (1), used to estimate the interfacial layer thickness at the boundary of the fibers and the matrix, to the equivalent form:

\[
\ln(h(t)) = \beta - \tilde{Q} \frac{1}{T} - \tilde{k} \frac{P}{T} + \frac{1}{2} \ln(t-t_0),
\]

where \( \beta = \ln(K_0) \), \( \tilde{Q} = Q/(2R) \), \( \tilde{k} = k/(2R) \) are new unknown parameters of the model.

We also introduce the functions \( f_i(\beta, \tilde{Q}, \tilde{k}, t_0) \), \( (i = 1 \ldots N) \):

\[
f_i(\beta, \tilde{Q}, \tilde{k}, t_0) = \ln H_i - \ln(h(t_i)) = -\beta + \tilde{Q} \cdot \frac{1}{T_i} + \tilde{k} \cdot \frac{P_i}{T_i} + \ln\left( \frac{H_i}{\sqrt{t_i-t_0}} \right),
\]

where \( i \) is the number of experimental points, and the value of function \( h(t_i) \) is determined by (1) with the use of experimental values \( P_i, T_i, t_i \).

As the objective function we used

\[
\Phi(\beta, \tilde{Q}, \tilde{k}, t_0) = \frac{1}{K} \sum_{i=1}^{N} \left[ \ln H_i - \ln(h(t_i)) \right]^2 = \frac{1}{K} \sum_{i=1}^{N} \left[ f_i(\beta, \tilde{Q}, \tilde{k}, t_0) \right]^2.
\]

The model parameters in this approach are determined by minimizing the function \( \Phi(\beta, \tilde{Q}, \tilde{k}, t_0) \).

In case of the minimum of the objective function its partial derivatives are vanished. This leads to a system of nonlinear algebraic equations in unknown parameters \( \beta, \tilde{Q}, \tilde{k}, t_0 \). To determine the coefficients of this equation system, we will introduce the following \( N \)-dimensional vectors:

\[
\begin{align*}
V_1 &= \begin{bmatrix} -1 & \ldots & -1 \end{bmatrix}^T, \\
V_2 &= \begin{bmatrix} 1/T_1 & \ldots & 1/T_N \end{bmatrix}^T, \\
V_3 &= \begin{bmatrix} P_1/T_1 & \ldots & P_N/T_N \end{bmatrix}^T, \\
V_4 &= \begin{bmatrix} 0.5/(t_1-t_0) & \ldots & 0.5/(t_N-t_0) \end{bmatrix}^T, \\
V_5 &= \begin{bmatrix} \ln(H_1/\sqrt{t_1-t_0}) & \ldots & \ln(H_N/\sqrt{t_N-t_0}) \end{bmatrix}^T.
\end{align*}
\]

Now the system of equations can be written as:

\[
A_1 \beta + A_2 x + A_3 \cdot \tilde{k} = -D_i, \quad (i = 1 \ldots A),
\]

where

\[
\begin{align*}
A_1 &= \begin{bmatrix} 1 & \ldots & 1 \end{bmatrix}^T, \\
A_2 &= \begin{bmatrix} 1/t_1 & \ldots & 1/t_N \end{bmatrix}^T, \\
A_3 &= \begin{bmatrix} \ln(t_1-t_0) & \ldots & \ln(t_N-t_0) \end{bmatrix}^T, \\
D_i &= \begin{bmatrix} \ln(H_1/\sqrt{t_1-t_0}) & \ldots & \ln(H_N/\sqrt{t_N-t_0}) \end{bmatrix}^T.
\end{align*}
\]
where
\[ A_{ij} = \sum_{m=1}^{N} (\mathbf{V}_i)_m \cdot (\mathbf{V}_j)_m = (\mathbf{V}_i, \mathbf{V}_j), \quad D_i = (\mathbf{V}_i, \mathbf{V}_5), \quad (i, j = 1\ldots A). \] (6)

All the elements of vector \( \mathbf{D} \), and elements \( A_{11}, A_{42}, A_{43} \) of matrix \( A \) depend on the unknown quantities, that makes a system of algebraic equations (5) nonlinear. At the same time elements \( A_{ij} \) \( (i, j = 1\ldots 3) \) of matrix \( A \) do not depend on unknown variables. Considering the above-mentioned feature of the system (5), we propose the following algorithm to determine the identification parameters:

- Find the minimum value of the experimental observation time, i.e. \( t_* = \min\{t_k\}_{k=1}^N \).
- Select value \( t_0 \in [0, t_*] \).
- Consider a subsystem of the system of algebraic equations (5) with selected value \( t_0 \in [0, t_*] \), that consists of the first three equations. This subsystem is a system of linear algebraic equations for \( \beta \), \( \tilde{Q} \), \( \tilde{\kappa} \). The solution of this system allows determining these parameters (for any given value of \( t_0 \in [0, t_*] \)).

- Choosing the value of \( t_0 \) in segment \( [0, t_*] \), we strive to satisfy the last equation of the system too (5). In addition, for every \( t_0 \) parameters \( \beta \), \( \tilde{Q} \), \( \tilde{\kappa} \) should be determined again. Since the solution of the last equation of the system (5) can not belong to segment \( [0, t_*] \), instead of solving the last equation, it is recommended to minimize objective function \( \Phi(t_0) = \Phi[\beta(t_0), \tilde{Q}(t_0), \tilde{\kappa}(t_0), t_0] \) of parameter \( t_0 \) on segment \( [0, t_*] \). Here we can use any simple algorithm for minimizing a function of one variable.

To solve the problem of identification of the model parameters it is necessary to select the data points in the way that vectors \( \mathbf{V}_1, \mathbf{V}_2, \mathbf{V}_3, \mathbf{V}_4 \) are linearly independent, and the number of data points is not less than 4. It should also be noted that the increase of the number of data points helps to ensure more reliable values of the identifiable parameters of the model.

3. Results

With the proposed algorithm we performed the calculations of the model parameters for the experimental data given in [5]. In [5] it was supposed that \( \kappa \) and \( t_0 \) are equal to 0, and the data were presented for the two materials: for composite SiC/Ti2AlNb and for composite SiC/super \( \alpha_2 \). These composites reinforced with unidirectional fibers of silicon carbide of the SCS-6 brand were made with the use of two different intermetallic matrices designated as Ti2AlNb and super \( \alpha_2 \). The investigation of possible chemical reactions, occurring under the composite manufacture during the formation of the interfacial layer at the boundary of the fiber and matrix, was conducted in [5].

Table 1 shows the results of determination of the model parameters for composite SiC / Ti2AlNb (nine experimental points), and Table 2 shows the results for composite SiC / super \( \alpha_2 \) (six experimental points).

Fig. 1 and 2 show the dependence of the interphase layer thickness on the exposure time of the sample. Different figures correspond to different temperatures of the process. The points represent the experimental data, the dark line corresponds to the results of [5] and the bright line represents the results obtained using the developed algorithm. It can be seen, that the model parameters obtained by the developed algorithm correspond to the experimental data rather than the parameters specified in [5].
Table 1. Identification of model parameters (1) for composite SiC / Ti2AlNb

| T (°C) | 700 | 700 | 700 | 800 | 800 | 800 | 900 | 900 | 900 |
|--------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| t (h)  | 1000| 1500| 2000| 1000| 1500| 2000| 50  | 200 | 500 |
| h (µm) | 0.99| 1.09| 1.15| 1.93| 2.27| 2.58| 1.22| 1.95| 2.87|

The results of the identification:

|           | work [5] | developed algorithm |
|-----------|----------|---------------------|
| $K_0$ (m/s$^{1/2}$) | 14.4·10^{-6} | 6.196·10^{-6} |
| $Q$ (J/mol)   | 175709   | 154229              |

Table 2. Identification of model parameters (1) for composite SiC / super $\alpha_2$

| T (°C) | 700 | 700 | 700 | 800 | 800 | 800 |
|--------|-----|-----|-----|-----|-----|-----|
| t (h)  | 1000| 1500| 2000| 1000| 1500| 2000|
| h (µm) | 1.39| 1.44| 1.52| 2.99| 3.63| 4.04|

The results of the identification:

|           | work [5] | developed algorithm |
|-----------|----------|---------------------|
| $K_0$ (m/s$^{1/2}$) | 54377.8·10^{-6} | 3.420·10^{-6} |
| $Q$ (J/mol)   | 317664   | 139009              |

Figure 1. Time dependence of the interfacial layer thickness for the Ti2AlNb composite
a: 700°C, b: 800°C.

Figure 2. Time dependence of the interfacial layer thickness for the super $\alpha_2$ composite
a: 700°C, b: 800°C.

4. Conclusions
The algorithm which allows identifying the kinetic parameters of the parabolic law has been proposed. In order to obtain successful results of the estimations one should use the experimental data consisting of not less than 4 points. More data points will provide better results obtained during identification.
The experimental points should be selected in such a way that the introduced system of N-dimensional vectors would be linearly independent. The appropriate description of the experimental data for the intermetallic titanium-based composites has been shown.

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