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INVERSE PROBLEM SOLUTION FOR DIFFUSION AND OXIDATION PROCESSES IN MATERIALS FOR GAS TURBINE BLADES

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ABSTRACT

A model of high temperature gas corrosion of alloys for gas turbine blades have been proposed. The model describes the diffusion of alloying element and oxidant in surface of material and kinetics of corrosion scale formation.

The main aim of this paper is inverse problem solution to estimate four parameters of diffusion and oxidation in the mathematical model using the data of short time laboratory testing. To determine the unknown parameters the procedure of iterative identification, based on Gauss-Newton's technique, was applied.

The results of inverse test problems solution both for exact and inexact input concentration measurement values of alloying elements during short time laboratory experiments are given in the paper.

Keywords: modeling, inverse problem, diffusion, oxidation.

NOMENCLATURE

A -sensitivity matrix
A^T -matrix transposed to matrix A
C -concentration
D -diffusion coefficient
E -unite matrix
F -root mean square criterion of solution quality
k -uptaking coefficient
P -vector of desired parameters
ΔP -increment of vector of desired parameters
W -mass source
x -coordinate
Z -sensitivity coefficient of concentration C to desired parameter P
α -regularization parameter
β -mass transfer coefficient
ε -coefficient of proportionality
ρ -random number
τ -time

Indexes

C -tied alloying element
T -total alloying element
m -total number of space-time points of concentration measurement
M -model
Me - alloying element
n -total number of desired parameters
Ox -oxidant
E -experimental
i -number of diffusant, number of desired parameter
j -number of space-time point of concentration measurement
l -iteration number
1 -oxidant
2 -alloying element

INTRODUCTION

Problems of high-temperature diffusion and oxidation in alloys are important for engineering and industry. Diffusion and oxidation models are necessary for life time prediction and optimization of the protective coatings for gas turbine blades for long time operation.

The mathematical model for diffusion of scale forming elements, the diffusion of oxidants from the gas medium and the

kinetics of oxide scale formation are developed by Krukovsky and Kartavova (1996) and described by systems diffusion equations of alloying elements and oxidant with oxidation processes taking into consideration by mass source term.

To make corrosion and diffusion predictions for a long time by means of mathematical model seems to be possible only if rather exact model's coefficients are on hand. It is possible on the base of effective modern methods of parameters estimation in diffusion and oxidation models using experimental data.

The parametrical sensitivity analysis of the calculated characteristics at the measurement points (concentration distribution and oxidation depth of alloys treated at high temperature) to variation in the desired parameters (coefficients of mass transfer, diffusion, etc.) is very useful for investigation of desired parameters sensitivity as the first step.

The physical and mathematical models under investigation as well as estimate four parameters of diffusion and oxidation in the model using the data of short time laboratory testing are considered below.

PHYSICAL MODEL

The physical model of scale formation at the external metal boundary was deduced in the following way.

The scale forming alloying elements diffuse to the boundary metal from the alloy. The oxidant absorbed from the gas medium diffuses in the reverse direction. The oxidant combines with alloying elements to form an oxide scale

$$aMe + bOx \rightarrow Me_aOx_b \qquad (1)$$

The typical spatial concentration distribution of one scale forming element in oxidation areas is shown in Fig.1.

In the oxidation areas the concentration of the alloying element rises with a decreased in the concentration of the alloying element dissolved in the solid solution of the alloy adjacent to the oxide area . The redistribution of alloying elements in the surface layer is determined by scale formation. The diffusion mobility of the scale forming element cannot be considered without knowledge of the element form: whether the element is in a metal phase state or is combined with an oxidant

Proceeding from that the following assumption was made: only elements not tied up in the oxide are diffusing. To quantify experimental data by element distribution in the oxide and in the solid solution in the surface layer of the oxidized alloy, the concentration curve of the alloying element (curve 3) is separated into two : curve 1 - uncombined (not tied up in oxide) alloying element in a solid, curve 2 - combined (tied up in oxide) alloying element. It is possible to separate the surface layer into the following main areas (Fig. 1) : compact oxide area $x_0<x<x_1$; inner area of oxidation $x_1<x<x_2$; depletion area $x_2<x<x_3$. The compact oxide area $x_0<x<x_1$ contains a compact oxide film. The concentration of the combined alloying element is a maximum ($C=C_{max}$) in this area. The area $x_2<x<x_3$ contains a mixture of oxides and denuded solid solution of the alloy . x_0 is the exterior boundary of scale . By convention, x_2, the boundary of inner oxidation, means the maximum depth when oxides occur (are observed). According to it, inner (unsteady in depth) oxidation in all layer volume takes place in such area. The external boundary concentration (x x1) takes on a maximum value, and the inner ones ($x=x_2$) take on minimum (for example, 5% of maximum) values. The oxidant concentration takes on the minimum in such an area. Below this there is diffusion area of dealloying by the base alloying element without oxides $x_2<x<x_3$. By convention, the dealloying area $x_2<x<x_3$ means a zone where the alloying element concentration ranges from minimum to closely maximum values (for example, 95% of maximum values) on the boundary $x=x_3$. It is necessary to note hat for alloys containing a number of alloying elements, the areas of oxidation and dealloying may differ and overlap one another.

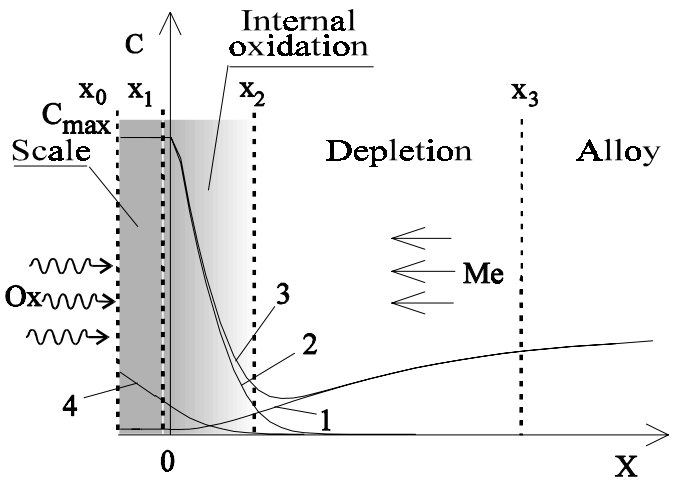

Figure 1. Quantitative concentration distribution of alloying element Me and oxidant Ox in the one layer. Curve 1 - uncombined alloying element which is not tied up in the scale; 2 - combined alloying element; 3 - total concentration of alloying element, 4 - oxidant.

It should be noted that during oxidation the volume of the alloy surface layer is not constant. Heat-resistant alloys form a dense film such that the volume of the oxide should exceed that the metal needed for oxide formation. The increase in the surface volume must be taken into account. The considered physical processes are unsteady: the surface boundary x_0 moves outwards with an increase in the volume, the boundaries x_1, x_2, x_3 and are move inwards the alloy.

MATHEMATICAL MODEL

The diffusion process of untied alloying elements (i=2 - Me) or oxidant (i=1 - Ox) towards the area of the formation of

scale products within the calculation domain $0<x<\infty$ can be described by the following set of equations:

$$\frac{\partial C_i}{\partial \tau} = \frac{\partial}{\partial x}\left[D_i \frac{\partial C_i}{\partial x}\right] - W_i, \qquad (1)$$

$$\tau > 0, \quad x_0 < x < \infty, \qquad x_0 = x_0(\tau)$$

$$C_i = C_i(x,\tau),\ D_i = D_i(x, C_i),$$

$$W_i = W_i(C_1, C_2)$$

initial condition:

$$C_i(x,0) = C_i(x), \quad x_0(0) = 0, \qquad (2)$$

boundary conditions:

$$D_i \frac{\partial C_i(x_0,\tau)}{\partial x} = \beta_i \cdot [C_i(x_0,\tau) - C_{i\infty}]$$

$$\frac{\partial C_i(\infty,\tau)}{\partial x} = 0,. \qquad (3)$$

For an oxidant $\beta_1 \neq 0$, for alloying element $\beta_2=0$.

The volume mass source of alloying element or oxidant is described by the function:

$$W_i = \begin{cases} k_i \cdot C_1 \cdot C_2, & C_2^C < C_{2\max}^C \\ 0, & C_2^C \geq C_{2\max}^C \end{cases}, \qquad (4)$$

where coefficients k_1 and k_2 are in the ratio of mass sources of alloying element and oxygen going for the formation of oxide layer, C_2^C is the concentration of the tied element for coordinate x can be calculated by the equation:

$$C_2^C(x,\tau) = \int_0^\tau W_2(x,\tau)\, d\tau, \qquad (5)$$

and total concentration of alloying element or oxidant:

$$C_i^T(x,\tau) = C_i^C(x,\tau) + C_i(x,\tau), \qquad (6)$$

where C_i is untied element or oxidant concentration.

Outer boundary x_0 moves according with oxide layer formation and mismatch of oxide and metal substrate densities:

$$x_0(\tau) = x_0(0) - \int_{x_0}^{x_\infty} C_2(x,0)dx + \int_{x_0}^{x_\infty} C_2(x,\tau)dx - \\ -(1+k_s)\cdot \frac{\rho_{All}}{\rho_{Oxd}} \cdot \int_{x_0}^{x_\infty} C_2^C(x,\tau)dx \qquad (7)$$

The diffusion and oxidation mathematical model is integrated by numerical finite differences methods using the implicitly approximating formulation. It permits one to take into consideration the most general peculiarities of the problem formulation: - the multilayer, the relationships between coefficients and coordinate, time and concentration. During each time step it is multiple solved the system of equations for oxidant, then the system of equations for alloying element. The implicitly connection between these system solution was carried by equation for mass sours (4) and was realized by means of iteration algorithm.

To make oxidation and diffusion predictions for a long time by means of mathematical model (1)-(7) seems to be possible only if rather exact coefficients are on hand.

The aims of this paper are to carry out the parametrical sensitivity analysis of the model and to determine parameters of this model by means of inverse problem solution using the data of computational experiment for both accurate and disturbed concentration distributions as well as of laboratory testing.

METHOD OF PARAMETRICAL IDENTIFICATION

There are a number of model parameters that can be defined only by means of an identification using experimental concentration distribution of alloying element in sample's surface layer on holding in the oxidation atmosphere at high temperatures.

The diffusion coefficients of alloying element D_{Me} and oxidant D_{Ox}, the mass transfer coefficient β_{Ox} of oxidant from gas medium and mass uptaking coefficient k_{Ox} of oxidant in the model (1)-(7) can be referred to a number of such parameters.

As experimental data for inverse problem solutions, the data of concentration distribution of alloying element in alloy surface layer at various time moments is used. The inverse problem statement reduces to an identification of four above mentioned parameters so that the following condition satisfies:

$$F = \left\{ \sum_{j=1}^{m} \left[C_{jM}(P) - C_{jE}\right]^2 / m \right\}^{0,5} \approx \delta \qquad (8)$$

where $C_{j,iM}$ and $C_{j,iE}$ are total alloying element concentration at jth point of space and time computed and experimental data (Fig.2); m is the number of space and time points at which the alloying element concentration measurements were made; P is vector of desired parameters including D_{Me}, D_{Ox}, β_{Ox}, k_{Ox}; δ is the root mean square error of concentration measurement making up around 0.8% for real measurements.

The identification of model parameters had been run by the technique described by Krukovsky (1996). In according with it technique to find the vector of the desired P parameters an iterative algorithm based on the Gauss-Newton is applied:

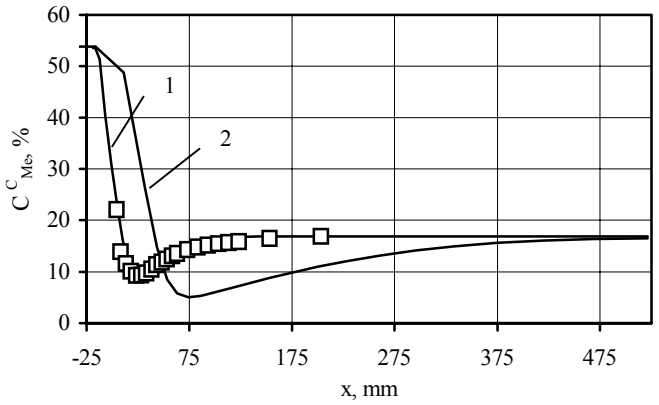

Figure 2. Relationship between total alloying element (Cr) concentration and coordinate at the 1000 h (Curve 1), T=900°C, - experiment (made by Dr. A. Rybnikov, Polzunov Central Boiler and Turbine Institute, St. Petersburg, Russia) and 20000 h (Curve 2).

$$P^{l+1} = P^{l} + \Delta P^{l} \ , \qquad (9)$$

where l is the number of iteration (l = 0,1,2,...), ΔP^{l} is an increment in the vector of the desired P parameters on the l+1th iteration. The ΔP^{l} increment is determined by solution of the following system of liner algebraic equations (SLAE)

$$\begin{cases} Z^{l}_{1,1}\Delta P^{l}_{1} + \cdots + Z^{l}_{1,i}\Delta P^{l}_{i} + \cdots + Z^{l}_{1,n}\Delta P^{l}_{n} = C^{l}_{1M} - C_{1E} \\ \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \\ Z^{l}_{j,1}\Delta P^{l}_{1} + \cdots + Z^{l}_{j,i}\Delta P^{l}_{i} + \cdots + Z^{l}_{j,n}\Delta P^{l}_{n} = C^{l}_{jM} - C_{jE} \\ \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \\ Z^{l}_{m,1}\Delta P^{l}_{1} + \cdots + Z^{l}_{m,i}\Delta P^{l}_{i} + \cdots + Z^{l}_{m,n}\Delta P^{l}_{n} = C^{l}_{mM} - C_{mE} \end{cases} ,(10)$$

where $Z^{l}_{j,i}$ is the sensitivity (derivative) function of the concentration at the jth space and time point of measurement $C_{j,E}$. (j=1,2,...,m) to variation in the **i**th parameter P^{l}_{i} (i=1,2,...,n) of P vector, $C^{l}_{j,M}$ are values of the model concentrations at the points corresponding to the experimental $C_{j,E}$. Values $C^{l}_{j,M}$ are computed by direct oxidation and diffusion problem solution with known (given at the l=0) parameters at the previous iteration l . Values $C_{j,M}$ are computed at the every iteration at the same space points the values $C_{j,E}$ were measured or assigned.

Redefined set of line algebraic equations is solved by the least-square technique. If express the matrix of the sensitivity coefficients $Z_{i,j}$ in term **A**, and right-hand side in term **B**, the SLAE solution with m×n dimensionality is achieved by following SLAE solution with n×n dimensionality:

$$\mathbf{A}^{\mathbf{T}} A \cdot \Delta P = \mathbf{A}^{\mathbf{T}} B \ , \qquad (11)$$

where A^T is the matrix transposed to the matrix A.

The iteration processes (9)-(10) results in the minimization of the quadratic criterion of solution quality (8).

The exit out of iteration process is made after the fulfilled condition:

$$\max_{i}\left|\Delta P^{l}_{i} / P^{l}_{i}\right| \cdot 100 \le 0{,}5 \ . \qquad (12)$$

For stable inverse problem solutions the following complex of regularization procedures is proposed:

1. Account of lower and upper bounds on desired inverse problem solution (desired parameters).

2. Iterative regularization, stop of iteration procedure if the condition (12) satisfies.

3. Tikhonov's (1987) classical regularization using in the SLAE solving stage. According with it method instead of SLAE (10) the following SLAE is solved:

$$\left(\mathbf{A}^{\mathbf{T}} A + \alpha \cdot E\right) \cdot \Delta P = \mathbf{A}^{\mathbf{T}} B \ , \qquad (13)$$

where E is unity matrix, α is regularization parameter selecting by various ways described by Tikhonov (1987). SLAE (13) is similar with Levenberg-Marquardt method. In our case α was not varied during the iterative procedure and was chosen by means of so called test inverse problem solution with known values of measurement errors.

INVERSE PROBLEM SOLUTION TESTING

In practical point of view the parametrical analysis of concentration $C_{j,iM}$ distribution sensitivity to desired parameters of mathematical model (diffusion, mass transfer and uptaking coefficients) is in great interest. As coefficient of sensitivity of concentration to ith desired model parameter the value Z_k is assumed:

$$Z_i = \frac{\Phi\left(P^0_i + \delta P_i\right)}{\delta P_i / P^0_i} , \qquad (14)$$

$$\Phi\left(P^0_i + \delta P_i\right) = \\ = \left\{ \sum_{j=1}^{m} \left[C_{jM}\left(P^0_1, ..., P^0_i + \delta P_i, ..., P^0_n\right) - C_{jM}\left(P^0\right) \right]^2 / m \right\}^{0,5} , \qquad (15)$$

$$\delta P_i = 0.1 \cdot P^0_i \qquad (16)$$

where P^0_i is initial values of desired parameters' vector, δP_i is the disturbance of ith parameter.

Table I. Initial, accurate, and estimated parameters.

Para-me-ter	Accurate	Initial	Estimated					
			at the accurate concentration			at the disturbed concentration		
			τ_1=500h.	τ_2=1000h	$\tau_1+\tau_2$	τ_1=500h.	τ_2=1000h	$\tau+\tau_2$
D_{me}	$3.0\cdot10^{-16}$	$3.0\cdot10^{-15}$	$3.44\cdot10^{-16}$	$3.53\cdot10^{-16}$	$3.27\cdot10^{-16}$	$3.13\cdot10^{-16}$	$3.21\cdot10^{-16}$	$3.03\cdot10^{-16}$
D_{ox}	$1.0\cdot10^{-16}$	$1.0\cdot10^{-15}$	$7.81\cdot10^{-17}$	$7.82\cdot10^{-17}$	$8.57\cdot10^{-17}$	$8.60\cdot10^{-17}$	$8.67\cdot10^{-17}$	$8.99\cdot10^{-17}$
k_{Ox}	$3.0\cdot10^{-7}$	$3.0\cdot10^{-8}$	$2.34\cdot10^{-7}$	$2.22\cdot10^{-7}$	$2.55\cdot10^{-7}$	$2.44\cdot10^{-7}$	$2.27\cdot10^{-7}$	$2.21\cdot10^{-7}$
β_{Ox}	$1.0\cdot10^{-10}$	$1.0\cdot10^{-11}$	$1.18\cdot10^{-10}$	$1.16\cdot10^{-10}$	$1.21\cdot10^{-10}$	$1.29\cdot10^{-10}$	$1.15\cdot10^{-10}$	$1.28\cdot10^{-10}$

For mathematical model sensitivity evaluation, the parametrical sensitivity analysis has been carried concerning four model parameters which will be sought by oxidation and diffusion inverse problem solution for one oxide-forming element redistribution in Ni-based alloy EI607A (Ni-16.9Cr-1.0Al-2.3Ti). Diffusion redistribution and oxidation computations were run with magnitudes of accurate coefficients given in Table I for time moments 500 and 1000 h.

As accurate model concentrations the computed chromium distribution (Table II) of total alloying element is assumed. Modifying one of four parameters in accordance with expression (9) the total element distribution $C_{j,iM}$ is computed. Sensitivity coefficient for D_{Me} calculated by (14) is formed 1.5. The sensitivity coefficients for another parameters are given in Table III.

As sensitivity analysis results show the most affectable on inverse problem solution parameters are chromium and oxygen diffusion coefficients, that is mean at the identification namely these two parameters will be searched best of all. Nevertheless the coefficients of mass uptaking and mass transfer affect on solution too, but its influences are visibly less, the β_{Ox} in 3 times smaller then D_{Me} and k_{Ox} in 2 times smaller then D_{Me} corresponding.

Let us proceed to consider the next stage of the proposed systematic formulation referred to as the estimation of model parameters. To well estimate the model parameters the test inverse problem solution is previously needed. For inverse problem solution testing the familiar scheme was used:

1. Direct oxidation and diffusion problem solution with given input parameters , that is a computational experiment to calculate the chromium distribution is refereed to as “precision” data at two time points 500 h (j=1) and 1000 h (j=2) and at the space points given in Table II.

2. Inverse oxidation and diffusion problem solution with accurate data (Table II) and comparison estimated model parameters with given one for direct problem (stage 1).

3. Inverse oxidation and diffusion problem solution with disturbed data modeling real errors of concentrate measurements. Comparison results with given parameters.

Inverse problem solution test has been carried with initial coefficients given in Table I, D_{Me} and D_{Ox}, greater then accurate ones on an order, and, β_{Ox} and k_{Ox} lower then accurate ones on an order.

Table II. Accurate and disturbed concentration of chromium on coordinate.

Coordinate x, μm	Accurate		Disturbed		Coordinate x, μm	Accurate		Disturbed	
	τ_1=500 h	τ_2=1000h	τ_1=500h	τ_2=1000h		τ_1=500 h	τ_2=1000h	τ_1=500h	τ_2=1000 h
5.42007	17.5951	20.9496	18.2652	21.7474	83.839	17.0733	16.2167	16.7881	15.9457
9.93464	14.0771	16.8658	13.6272	16.3267	101.133	16.6207	16.2697	16.8752	16.5189
14.8238	10.9868	11.9755	11.1788	12.1848	120.666	16.6932	16.5772	16.8957	16.7785
20.3727	10.2973	9.3822	10.7287	9.7754	142.568	16.8866	16.8564	16.8994	16.8693
26.9201	11.7739	9.312	11.6775	9.2357	166.972	17.3002	17.294	16.8999	16.8938
34.795	13.5017	10.3629	13.2398	10.162	194.012	16.6673	16.6665	16.8995	16.8991
44.2626	14.6597	11.7092	14.7621	11.7910	223.824	16.5667	16.5668	16.9006	16.9001
55.5098	15.9574	13.569	15.8796	13.5029	256.542	17.1596	17.1589	16.8995	16.8993
68.6668	16.1412	14.6035	16.5143	14.9411					

Table III. Sensitivity Z_i coefficients' magnitudes for model parameters being subject of identification.

D_{Me}	k_{Ox}	β_{Ox}	D_{Ox}
1.5	0.74	0.54	1.68

Simultaneous searching of four parameters is carried with accurate concentration distribution (Table II) using information from one (t=500 h) and two calculation time points (t=500 h and 1000 h). The computational estimation of desired parameters is run with initial parameters (Table II) by identification technique (1)-(7) described above and using the accurate concentration data about obtained at the 500 h as experimental information C_{jE}. To obtain the stable solution the regularization parameter α in (13) was taken as $1\cdot10^{-5}$. Root mean square criterion F variation with number of iteration is shown in Fig. 3.

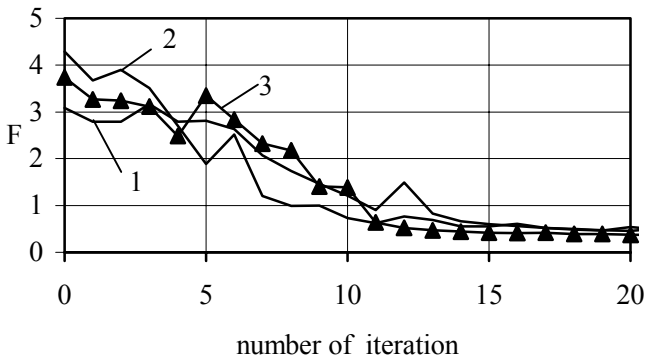

Figure 3. Relationship between root mean square quality solution criterion of inverse problem and number of iteration for various experimental time moments: 1 - one experimental time moment at the 500 h; 2 - one experimental time moment at the 1000 h; 3 - two experimental time moments at the 500 h and 1000 h.

In much the same line the computational estimations of desired parameters were run using the data about concentration at 1000 h (curves 2) and at 500 h and 1000 h simultaneously as experimental information. The use of experimental information about chromium distribution at the two exposures is quite evidently efficient since the solution runs more stable and in addition the iteration regularization comes into action (calculation stop at the 34 iteration on fulfilling the condition (12)).

For real error modeling the random measurement errors have been put on the concentration distribution by according to the following expression:

$$C_{j\ni} = C_{jM}^{T}\left(1+\varepsilon\cdot\rho_j\right), \qquad (16)$$

where ρ_j is random number given by the normal low within the range ±3 with mathematical expectation 0 and variance 1. The disturbance range of C equal to ε =5%. Disturbed concentration of chromium given in Table II.

The searching at the same four initial parameters with disturbed concentration was run using two time moments information with the same initial desired parameters (Table I).

Thus it was shown the possibility of simultaneous estimation of four parameters of mathematical model using the information about element distribution with error at one and two time moments.

Results of practical mass transfer inverse problem solution are given in Fig. 2. Coefficients $D_{Me}=3.05\cdot10^{-16}$ m^2/s, $D_{Ox}=1.12\cdot10^{-16}$ m^2/s, $\beta_{Ox}=1.03\cdot10^{-10}$ m/s, $k_{Ox}=3.04\cdot10^{-7}$ 1/s were determined using experimental concentration distribution obtained after experimental exposure of samples made of EI893 alloy during 1000 h at 900°C. Curve 1 is model distribution at the same time, curve 2 is computed prediction.

So, proposed technique can be applied for identification of mass transfer parameters as function of concentration at various temperature with final purpose of model use for long time prediction of concentration distribution and oxidation of alloying element.

CONCLUSIONS

1. The inverse problem solution technique and parameters' identification results are considered in the paper.

2. The possibility of identification of four model coefficients D_{Me}, D_{Ox}, β_{Ox}, k_{Ox} using experimental information of one time experimental exposure as well as of two time moments.

3. The computational stability of proposed inverse problem solution were investigated. The use of experimental information about chromium distribution at two exposure the solution runs more stable and accurate.

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