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SIMULATION OF FORMING DIFFUSION ZONE AT IMPULSE ELECTRON-BEAM TREATMENT OF COATED MATERIAL

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Model of forming diffusion zone between material and coating during the process of impulse electron-beam treatment has been formulated. Algorithm of numerical problem solution on non-isothermal diffusion is developed taking into consideration the difference of spatial and temporal scales of heat conductivity and diffusion in solid substances. Dependence of diffusion zone characteristics on radiation treatment conditions is studied.

Modeling the surface thermal treatment of metals, forming the transition zone between coating and base in impulse electron-beam or laser treatment of «film-base» system there appears the necessity to solve the set of thermal conductivity and multicomponent diffusion equations together. Often they include sources and sinks of heat and mass owing to chemical transformations. Similar mathematical models allow us to add sufficiently the experimental studies due to detailed investigation of dynamic picture of diffusion zone formation.

Because of the difference in spatial and temporal scales of heat and diffusion processes in condensed phase such problems turn out to be not simple for their numerical realization as they require large calculation resources or specially developed numerical algorithms.

Available in the literature mathematical models of multicomponent diffusion transfer processes are limited for the same reason by, as a rule, isothermal approximations. The developed packets of applied programs are suitable for a sufficiently narrow range of problems.

The purpose of the present work is to construct mathematical model of diffusion zone formation in the surface layer of material in contact with coating in the process of impulse energy source treatment and to study it numerically.

1. Mathematical statement of the problem

Suppose the coating of small thickness h_1 , previously applied to the base contains ligation elements in its composition, the concentrations of which are denoted by a_i , $i=1,2$. At the initial moment of time $t=0$ ligation elements are distributed on coating uniformly, but they are absent in the base. At $t>0$ the system from the coating side is subjected to the action of pulse-periodical heat source, e.g. electron flux. We assume that the radius of electron beam is more than cross size of sample that permits us to restrict ourselves by one-dimension problem statement, which includes the heat conductivity equation and the diffusion equation of ligation elements from the coating of base thickness (fig. 1).

$$c\rho \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \lambda \frac{\partial T}{\partial x}, \quad (1)$$

$$\frac{\partial a_1}{\partial t} = -\frac{\partial J_1}{\partial x}; \quad \frac{\partial a_2}{\partial t} = -\frac{\partial J_2}{\partial x}, \quad (2)$$

where

$$J_1 = -D_{11} \frac{\partial a_1}{\partial x} - D_{12} \frac{\partial a_2}{\partial x}; \quad J_2 = -D_{21} \frac{\partial a_1}{\partial x} - D_{22} \frac{\partial a_2}{\partial x}$$

are the diffusion fluxes of legating elements, D_{ij} , are the partial diffusion coefficients; c , ρ , λ are the thermal capacity, density, and thermal conductivity of base.

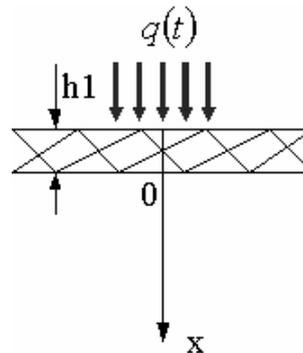


Fig. 1. Illustration on problem statement

Small thickness of filling material in comparison with that of base and that of thermal boundary layer formed in the base during treatment makes it possible not to consider the temperature and concentration distribution in this layer. Then, the boundary condition in the form of differential equation is true as a boundary condition for the temperature at $x=0$:

$$x = 0 : \lambda \frac{\partial T}{\partial x} = h_1 c_1 \rho_1 \frac{\partial T}{\partial t} + \sigma \varepsilon (T^4 - T_e^4) - q(t), \quad (3)$$

where T_e is the temperature of environment, $\sigma \varepsilon (T^4 - T_e^4)$ are the losses into the environment (vacuum), σ is the Stephan-Boltzmann constant, ε is the emittance. The kind of function $q(t)$ and initial conditions depend on the described stage of technological process. During the action of external pulse-periodical source we have

$$q(t) = q_0 f(t); \quad f(t) = \sum_{k=1}^n f_k(t),$$

where

$$f_k(t) = \begin{cases} 1, & (t_i + t_p)(k-1) \leq t < t_i + (t_i + t_p)(k-1); \\ 0, & t_i + (t_i + t_p)(k-1) \leq t < (t_i + t_p)k, \end{cases}$$

t_i is the pulse duration; t_p is the pause duration; n is the number of hits; n is the number of pulses. The power density measured in $J/(sm^2 \cdot s)$, is $q(t)$; the pulse energy is $q(t)t$.

If the source does not operate as pulses pass, then $q(t)=0$.

For concentrations at the boundary we have:

$$x = 0 : h_1 \frac{\partial a_1}{\partial t} = -J_1, \quad h_1 \frac{\partial a_2}{\partial t} = -J_2. \quad (4)$$

At infinite separation from the heated region the thermal and mass sources and sinks are absent:

$$x \rightarrow \infty : \lambda \frac{\partial T}{\partial x} = 0; \quad (5)$$

$$J_1 = 0; J_2 = 0. \quad (6)$$

Dependences of thermophysical properties on temperature are approximated by polynomials. Diffusion coefficients in common case depend on concentrations according to the power law, but on temperature is according to the Arrhenius law. Coating and base have different melting points $T_{ph,1}$, T_{ph} . Hence, to describe the property temperature dependence we have the relations:

$$D_{ij} = (D_{ij}^0 + D_{ij}^1 a_1 + D_{ij}^2 a_2 + D_{ij}^3 a_1 a_2 + D_{ij}^4 a_1^2 + D_{ij}^5 a_2^2) \times \exp\left(-\frac{E_i}{RT}\right), \quad (7)$$

$$\lambda = \lambda_0 + \lambda_1 T + \lambda_2 T^2 + \lambda_3 T^3 + \lambda_4 T^4, \quad (8)$$

$$(c\rho)_1 = L\rho_{s,1} \cdot \delta(T - T_{ph,1}) + \begin{cases} (c\rho)_{s,1}, & T < T_{ph,1}; \\ (c\rho)_{L,1}, & T \geq T_{ph,1}, \end{cases} \quad (9)$$

The relation similar to (9) is for the base. Here δ is the Dirac function.

Initial conditions for the problem (1) – (5) have the view

$$t = 0 : \left. \begin{array}{l} a_1 = a_{10}, a_2 = a_{20}, x = 0 \\ a_1 = 0, a_2 = 0, x > 0 \end{array} \right\} T = T_0, x \geq 0. \quad (10)$$

On the basis of model one need to study the influence of source parameters on the dynamics of temperature changes and redistribution of legating element concentrations.

2. Results of numerical study of model problem

The problem was solved numerically according to implicit conservative difference scheme using the sweep method. In present work a special algorithm for numerical problem solution of such type described in [1] was used. The problems on thermal conductivity and diffusion were solved by different, but consistent difference schemes. Steps in space and time were chosen from the relations of characteristic diffusion and spatial scales for heat conductivity and diffusion processes. As a characteristic spatial scale for diffusion the thickness of diffusion boundary layer forming during the time equal to one pulse duration t_i was used. For calculation of spatial scale the minimal of diffusion coefficients (calculated at the temperature close to that of base melting) was applied:

$$x_D = \sqrt{t_i D_{\min}}.$$

Spatial scale for thermal conductivity can be found similarly

$$x_T = \sqrt{\frac{t_i \lambda}{c\rho}}.$$

Then steps of difference grid in thermal conductivity and diffusion problems are connected in the following way

$$\Delta x_T / \Delta x_D = \sqrt{\lambda / (c\rho D_{\min})} = 1 / \sqrt{Le},$$

where $Le = D_{\min} / \kappa$ is the Lewis number for the given problem. Similarly one can choose the relation between steps in time. But application of implicit absolutely stable difference schemes allows us not to take care about it thoroughly.

One more problem in solving the tasks on isothermal mass transfer in condensed media consists in the following: the conditions involved for matrix of diffusion coefficients obtained for isothermal conditions and are necessary for providing a non-negativity of solution are not always met for non-isothermal problems and problems with diffusion coefficients dependent on temperature and concentration. This problem is solved in constructing numerical algorithm by using limit for model coefficients following from non-equilibrium thermodynamics [2]. Unfortunately, available in the literature experimental data for partial diffusion coefficients obtained in carrying out «isothermal» experiments fail to be used as they do not meet these conditions. Therefore, the problem of determining partial diffusion coefficients is still actual.

In calculations described below in the model problem the following parameters are used: $T_{ph1}=1710$ K, $T_{ph}=1810$ K, $\lambda=0,72-1,37 \cdot 10^{-7} T-6,00 \cdot 10^{-7} T^2+3,39 \cdot 10^{-10} T^3$, $h_i=0,05$ sm; $C_s=C_{s1}=0,68$ J/g·K, $C_L=C_{L1}=0,65$ J/g·K, $\rho_s=\rho_{s1}=7,61$ g/sm³, $\rho_L=\rho_{L1}=7,865$ g/sm³, $L_{ph}=250$ J/g, $L_{ph1}=400$ J/g, $D_{11}^0=1 \cdot 10^{-1}$ sm²/s, $D_{22}^0=1 \cdot 10^{-2}$ sm²/s, $D_{12}^0=D_{21}^0=0$, $E=140000$ J/g.

Calculations are made for two different frequencies

$$-\Gamma = \frac{1}{t_i + t_p} = 50 \text{ and } \Gamma = 10 \text{ Hz.}$$

Analysis of the results of numerical problem solution shows that distribution and maximum value of temperature and concentration in the treated sample depend on maximum power density, the number and duration of pulses as well as on source frequency.

In fig. 2, $a-c$, the temperature dependence on time in the point is presented as an example. By the end of the first pulse (fig. 2, a) of 0.005 sec duration the temperature in the coating and on the base surface at the given set of parameters does not increase 1100 K. During this time the concentration of legating elements in the coating is changing slightly (fig. 2, d). Diffusion is observed only at $T_s > 700 \dots 900$ K, which exists in the surface layer only during $\Delta t \approx 0,02$ sec. The second pulse, following the first one after the pause $t_p = 0,0015$ sec, results in higher increase in temperature that accelerates diffusion in such a way that during the second pulse and at cooling up to T_s the element concentration in the coating decreases up to $C_1^* \approx 0,0179$, but $C_2^* \approx 0,297$. Five pul-

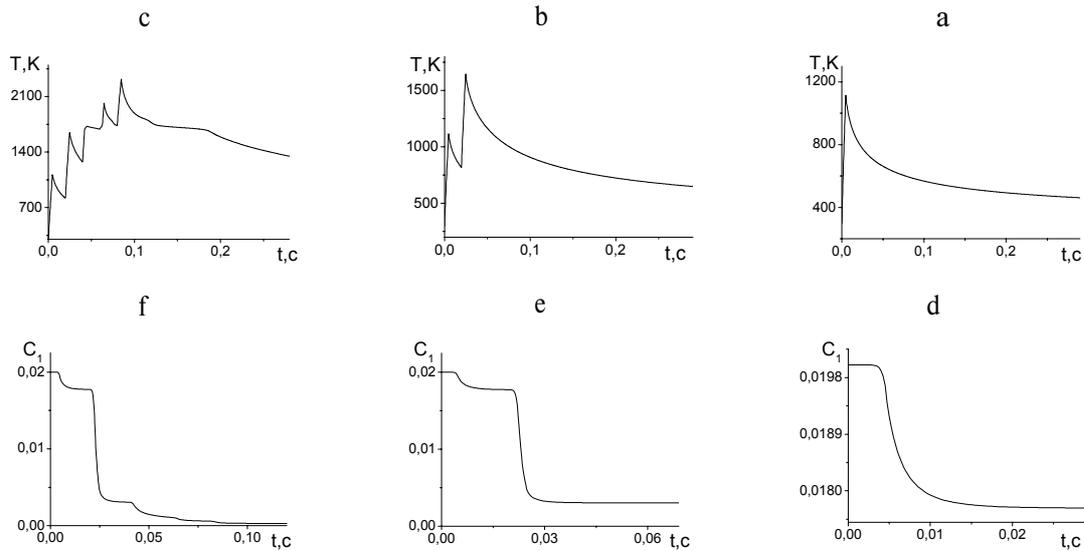


Fig. 2. Temperature and concentration dependences of ligating elements on time in the point at the following parameters: $t_i=0,005$, $t_p=0,015$ s, $q_0=6 \cdot 10^4$ W/sm²; c, d) $n=5$, б, e) $n=2$, a, f) $n=1$

ses (fig. 2, c, f) are sufficient for the concentration of the first element in the coating to decrease to nearly zero, but the second one to $C_2^* \approx 0,045$. The peaks on the temperature dependence curve on time correspond to the pulse finish; the plate does to melting (during the third pulse action and crystallisation, at cooling after 5 pulses). Concentrations of ligating elements in the coating in time follow the temperature behaviour (fig. 2, f). Spatial distribution of temperatures and concentrations in the base at different moments of time presented in fig. 3, a–c, illustrates the fact that the width of diffusion zone x_D , that can be defined by the condition

$$\min \left\{ \frac{C_i - C_i(t, x_D)}{C_{i0}} \right\} < 0,01,$$

is much less than the width of heat penetration x_T , where temperature exceeds 700...900 K during treatment.

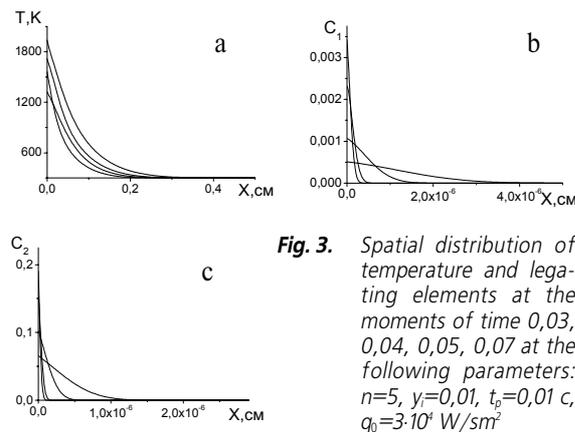


Fig. 3. Spatial distribution of temperature and legating elements at the moments of time 0,03, 0,04, 0,05, 0,07 at the following parameters: $n=5$, $y_i=0,01$, $t_p=0,01$ s, $q_0=3 \cdot 10^4$ W/sm²

Numerical values C_1^* , C_2^* in coating by the end of treatment and the values, sufficiently depend on the parameters of external impact. Such dependences are of great interest for specific schemes.

Consideration of cross diffusion flows and diffusion coefficient dependencies on concentrations shows also a sufficient influence on the process dynamics, which is not shown in the figures.

It is noted that in decreasing the source power density redistribution of concentrations takes place more smoothly, but for their essential change a large number of pulses is required.

At frequency 10 Hz the influence of source parameters on concentration distribution of legating elements in sample ($q_0=10^4$ and $t_i=0,01$, $t_p=0,09$) are practically insignificant.

3. Distribution of temperature and concentration in «copper coating – iron base» system at pulse electron-beam treatment

The model suggested was used for numerical study of concentration redistribution in «copper – iron» system, the experimental research of which by means of pulse electron stream are presented in [3]. In the experiments [3] film-base system was exposed to pulse melting by electron beam with low energy (20...30 keV) and high stream (2...3 mks, 2...10 J/sm²). The authors showed that multiple pulse melting of the given system results in increase of diffusion layer thickness by the depth sufficiently exceeding that of liquid-phase diffusion, which is essentially improves composition strength. The model (1)–(10) fully corresponds to the experimental conditions of this work.

Numerical investigation was carried out without taking into account the presence of legating elements in iron. The substance properties – base (Fe) and coating (Cu), used in calculations, were taken from [4, 5]: $T_{Cu}=1358$ K, $T_{steel}=1810$ K, $\lambda_{steel}=0,45$ W/sm K c, $h_i=0,000512$ sm; $C_{s,steel}=0,68$ J/g·K, $C_{L,steel}=0,65$ J/g·K, $\rho_{s,steel}=7,61$ g/sm³, $\rho_{L,steel}=7,8651$ g/sm³, $L_{ph,steel}=250$ J/g, $L_{phl}=20500$ J/g, $C_{s,Cu}=0,43$ J/g·K, $C_{L,Cu}=0,5$ J/g·K, $\rho_{s,Cu}=0,88$ g/sm³, $\rho_{L,Cu}=0,85$ g/sm³, $D_{H,Cu}^0=0,33$ sm²/s, $E=140000$ J/g.

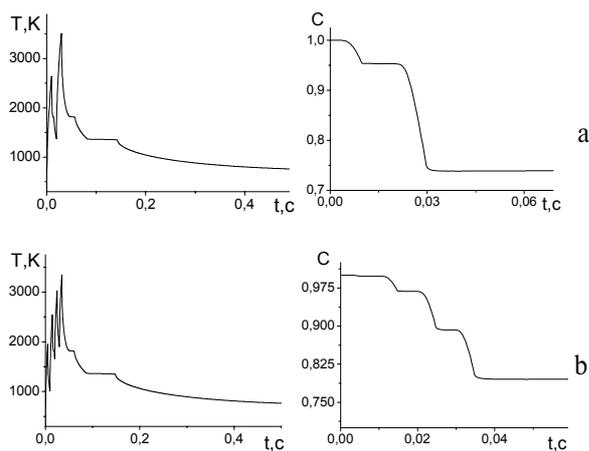


Fig. 4. Temperature and concentration dependency of legating element on time at the point at W/sm^2 : a) $n=2$, $t_i=t_p=0,01$ s, b) $n=4$, $t_i=t_p=0,005$ s

The given system is distinguished by nonlinear distribution of copper concentration, which is connected with nonlinearity of the problem and consideration of melting (fig. 4). The results depend sufficiently on the number and duration of radiation pulses even at constant energy introduced into the system in the course of treatment. Thus, for fig. 4, a, and fig. 4, b, energy density introduced into the substance is the same and equal to $6,64 \cdot 10^3$ J/sm². If this energy is introduced at two pulses, concentration in the surface layer (at the boundary of copper and iron) by the moment of system cooling up to $T=900$ K decreases to $C^* \approx 0,74$; if the same energy is introduced at 4 pulses with doubly less duration (so that full treatment time remains the same), we obtain $C^* \approx 0,79$. The width of diffusion zone formed during system treatment and cooling in the first case amounts $x_p \approx 0,0059$, but in the second one it is $x_p \approx 0,0066$ (fig. 5, a, b). Qualitative distribution of legating element concentration in the coating does not practically differ from that at varying the source parameters.

The width of heat penetration x_T and the width of diffusion zone x_D depend both on power density q_0 (i. e., on energy introduced per 1 pulse) at constancy of introduced energy (fig. 6). Treatment with equal number of pulses of different value (and of different frequency correspondingly) results in different values of heat penetration and diffusion zone. Heat penetration determined

by the specified value $T=900$ K, increases in the process of heat and melting, at cooling decreases beginning from some moment of time (fig. 6, b). Diffusion zone changes (increases) while the temperature is sufficient for diffusion, and then, it ceases to change (fig. 6, b). It is found that even insignificant increase in frequency of external influence from $\Gamma=(t_i+t_p)^{-1}$ to $\Gamma=100$ Hz results in sufficient growth of final width of diffusion zone (curves 2 and 1 in fig. 6 respectively), that is qualitative-ly consistent with the experimental data [3].

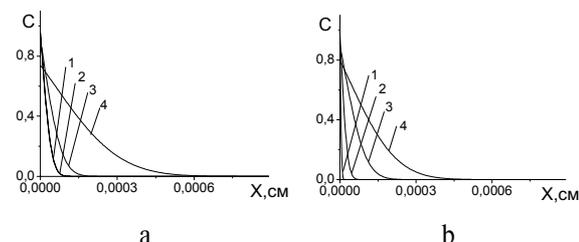


Fig. 5. Spatial distribution of Cu in Fe at different moments of time a) 1 - 0,0125; 2 - 0,025; 3 - 0,0375; 4 - 0,05 b) 1 - 0,0125; 2 - 0,025; 3 - 0,05; 4 - 0,1 with the source parameters $q_0=3,32 \cdot 10^4$ W/sm² a) $n=2$, $t_i=t_p=0,01$ s, $x_p \approx 0,0059$ b) $n=4$, $t_i=t_p=0,005$ s, $x_p \approx 0,0066$

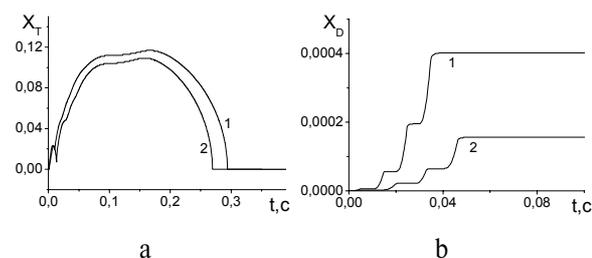


Fig. 6. Width of heat penetration a) and width of diffusion zone b) in the process of material treatment with the source parameters: 1) $q_0=3,32 \cdot 10^4$ W/sm², $t_i=t_p=0,005$ sec, $n=4$; 2) $q_0=2,5 \cdot 10^4$ W/sm², $t_i=t_p=0,00644$ sec, $n=4$

Thus, in the work the model of diffusion zone formation in the process of coating material treatment by external energy source on the basis of experimental data has been suggested. It is shown that the radiation conditions influences sufficiently the distribution of element concentration and in doing so the formation of transition zone between the coating and the base. In calculation for a concrete system the results qualitatively corresponding to the regularities observed are obtained.

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