# THERMOPHYSICAL RECOVERY PROCESSES DURING PULSE LASER HEATING

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We consider thermophysical processes during laser heating of an oxide with account of growth of the metallic phase at the surface.

One of the important directions in the study of thermochemical processes during the interaction of laser radiation with materials is laser and laser-plasma recovery of oxide metals, considered in [1-5], where laser heating of oxides was investigated in air or in the gas-recovery gas, particularly in a hydrogen atmosphere, leading to recovery of surface layers and formation of metallic films [3-5]. A calculation was carried out in [1] of the heating of a thermally thin oxide film on the basis of the simultaneous system of equations of thermal balance and kinetic growth of the metallic phase at the surface; laser recovery of (Cu, Co) oxide metals in a CO atmosphere was investigated experimentally, and estimates were given for calculating the kinetic constants of thermochemical reactions [2]. Results of experiments in the metals obtained by laser heating of oxides of high-melting metals in a hydrogen atmosphere are given in [3]. Laser recovery of copper from cuprous oxides in a hydrogen atmosphere was considered in [4], where it was shown that irradiation leads to instability in the surface shape of the metal formed, while the instability scale is comparable with the scale of action. The common features of the studies mentioned above are the following assumptions: we consider low-density energy fluxes and long action times, corresponding to irradiation by a continuous laser. It is noted that the thermal physics of laser recovery processes has been insufficiently investigated, particularly concerning spatial inhomogeneities (related to the inhomogeneity of the problem).

In the present study we consider the recovery features during action on the target of pulse laser radiation, corresponding to the regime of free generation (the pulse duration is $\sim$ 1 msec, the radiation wave length is 1.06 μm, and the pulse energy is $\sim$ 20 J).

During laser recovery of an oxide one of the most substantial effects is the substantial decrease in the absoption capability of the oxide–metallic film system, usually smaller by orders of magnitude than the similar variation in the amount of energy introduced into the body and passing through during its heating. This leads, in turn, to reduction in the times of heating and of thermochemical recovery reactions.

Consider the following model of the process. The oxide surface, semiinifinite and homogeneous in composition and structure, found in an atmosphere of gas-recoverer ($H_2$), is heated by radiation of a pulse laser with a Gaussian flow density distribution

$$q(r't') = q_0 \exp(-kr'^2).$$

As a result of the thermochemical reaction at the oxide surface generates a metallic phase, whose thickness increases with the flow of time. The heterogeneous growth kinetics of the metallic layer is described in the general case by the exponential law [6]

$$\frac{\partial \xi(r', t')}{\partial t'} = \frac{d_n}{\xi^n(r', t')} \exp\left[-\frac{T_n^*}{T(r' 0, t')}\right]. \quad (1)$$

For small metallic film thicknesses n = 0 (a linear law), while for an increased film thickness, when the growth process is limited by diffusion, n = 1 (parabolic law).

The low thickness of the metallic phase makes it possible to neglect its temperature gradient over depth in the statement of the thermal problem, whose mathematical formulation is:

$$\frac{1}{r'}\frac{\partial}{\partial r'} r' \frac{\partial T}{\partial r'} + \frac{\partial^2 T}{\partial z'^2} = \frac{1}{a}\frac{dT}{\partial t'},$$

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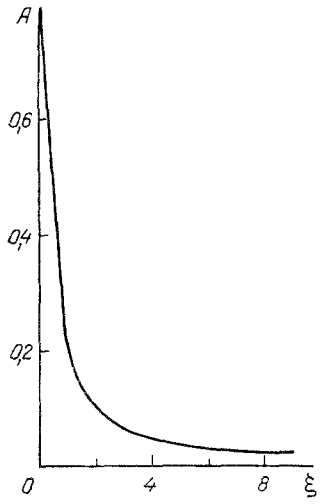


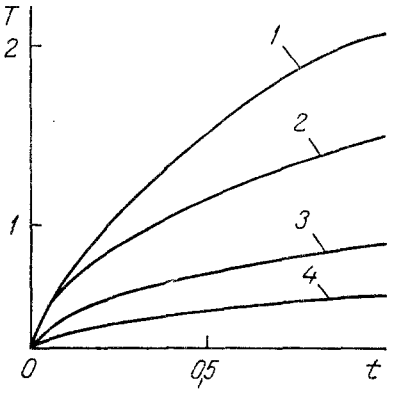




Fig. 1 Fig. 2

Fig. 1. Absorptivity of a two-layer oxide–metal film system as a function of film thickness. ξ, $10^{-8}$ m; A, dimensionless.

Fig. 2. The temperature at the center of the heating spot as a function of action times for various regimes: 1) $q_0 = 4.8 \cdot 10^9$ W/m²; 2) $3.2 \cdot 10^9$; 3) $1.6 \cdot 10^9$; 4) $0.8 \cdot 10^9$. T, $10^3$ K; t, $10^{-3}$ sec.

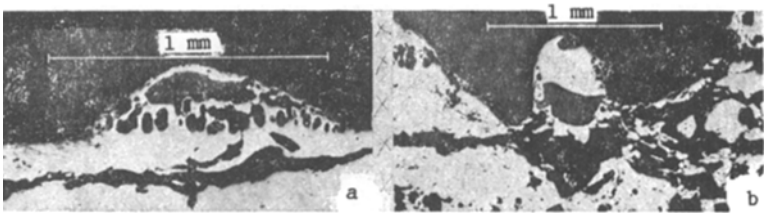



Fig. 3. Microstructure of a copper film, recovered on a $Cu_2O$ surface in hydrogen at pressure $5 \cdot 10^5$ Pa, with incident flux densities: a) $q_0 = 10^9$ w/m²; b) $2 \cdot 10^{10}$.

$$\lambda \frac{\partial T}{\partial z'}\Big|_{z'=0} = -A(\xi)\, q(r',\ t'),$$

$$\frac{\partial \xi}{\partial t'} = d_1 \exp\left(-\frac{T_1^*}{T}\right), \tag{2}$$

$$T(r',\ z',\ 0) = T(\infty,\ z',\ t') = T(r',\ \infty,\ t') = T_0,$$

and A(ξ) is the absorptivity of the two-layer metallic film–oxide system [7]:

$$A(\xi) = 1 - |r|^2, \quad r = \frac{r_{12}\exp(-2i\psi) + r_{23}}{\exp(-2i\psi) + r_{12}r_{23}} \tag{3}$$

$$r_{23} = \frac{r_{12} - r_{13}}{r_{12}r_{13} - 1}, \quad r_{12} = \frac{1 - \sqrt{\varepsilon_0}}{1 + \sqrt{\varepsilon_0}}, \quad r_{13} = \frac{1 - \sqrt{\varepsilon}}{1 + \sqrt{\varepsilon}},$$

$$\psi = \frac{2\pi\xi}{\lambda_v}\sqrt{\varepsilon_0}, \quad \sqrt{\varepsilon_0} = n_0 + i\varkappa_0, \quad \sqrt{\varepsilon} = n + i\varkappa.$$

TABLE 1. Dependence of the Recovery Zone Diameter on the Flux Density of the Incident Radiation and on the Laser Beam Diameter

| $q_0 \cdot 10^9$ W/m² | Diameter, mm | |
|---|---|---|
| | irradiation | recovery zone |
| 2,046 | 3,7 | 4,5 |
| 4,432 | 2,5 | 3,5 |
| 12,45 | 1,5 | 2,5 |

The mathematical formulation of the dependence of the absorptivity on the metallic film thickness has the same shape as for oxidation [8], but the calculations by relation (3) lead to substantially different quantitative results. Numerical analysis of relation (3) has shown fast decrease in the absorptivity of the two-layer system as a function of the thickness of the recovered layer. Figure 1 shows computational data for $A(\xi)$, showing that already for metal thicknesses $\xi \lesssim 0.01$ the quantity $A(\xi)$ is near the absorptivity value of a "pure" metal, and instead of (3) one can put in system (2) $A(\xi) \approx A_0 = \text{const}$, i.e., consider the thermal heating model of a massive body with thermophysical parameters of the oxide and with the absorptivity of the metal. A similar replacement in oxidation problems is not justified due to the smoother character of absorptivity variation, corresponding to a two-layer system of "oxides on the surface of a massive metallic body." Thus, growth of the oxide film within the limits up to 0.1 μm leads to an enchancement in absorptivity not exceeding 5% [8].

In the region considered ($q_0 \gtrsim 10^8$ W/m²) thermal losses at the surface can be neglected, and following the introduction of dimensionless parameters

$$z = \sqrt{k}\,z', \quad r = \sqrt{k}\,r', \quad t = kat' \tag{4}$$

the solution of system (2) is conveniently written in the form

$$T(r, z, t) = T_0 + T_1 \int_0^{2\sqrt{t}} \exp\left[-\left(\frac{r^2}{x^2+1} + \frac{z^2}{x^2}\right)\right] \frac{dx}{x^2+1}, \tag{5}$$

giving at the center of the heating spot

$$T(0, 0, t) = T_0 + T_1 \operatorname{arctg} 2\sqrt{t}, \quad T_1 = \frac{q_0 A_0}{\lambda \sqrt{\pi k}}. \tag{6}$$

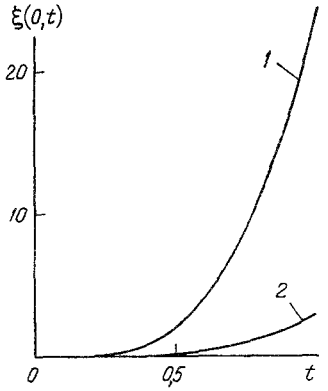
It follows from the parameters of the problem ($a \sim 10^{-7}$ m²/sec, $k \lesssim 10^7$ m$^{-2}$, $t' \lesssim 10^{-3}$ sec) that the dimensionless time satisfies $t \ll 1$, allowing to represent the temperature field distribution (5) in the form of a product of solutions of the one-dimensional heating problem by the spatial energy flow distribution [5]:

$$T(r, z, t) = T_0 + \alpha_1 \sqrt{t} \exp(-r^2) \operatorname{ierfc}\left(\frac{z}{2\sqrt{t}}\right), \tag{7}$$

where

$$\operatorname{ierfc}(x) = \int_x^\infty \operatorname{erfc}(t)\,dt, \quad \operatorname{erfc}(x) = 1 - \operatorname{erf}(x), \quad \alpha_1 = 2T_1.$$

Plots of the temperature dependences, calculated by relation (6), are shown in Fig. 2. Data analysis of the temperature dependences at the center of the heating spot indicates the existence of various regimes of laser recovery. For relatively small $q_0$ the maximum temperature achieved in the heating process does not exceed the melting temperature of the metal ($T_m Cu$ = 1356 K) (curves 3, 4), and the recovery process occurs in the solid phase. Enhancement of $q_0$ leads to melting of the metal over the oxide layer (curve 2), possessing higher melting temperature ($T_m Cu_2O$ = 1506 K), and the recovery process is already extended in the fluid phase. Oxide melting occurs with increasing $q_0$, and a special liquid metal structure is generated, fluid oxide. Further enhancement of $q_0$ leads to achievement of an expanded oxide temperature ($T_b Cu_2O$ = 2073 K) (curve 1) under the layer of the liquid metal, as a result of which a convex surface relief is formed with the appearance of cavities under the metallic layer (Fig. 3a). Cavity formation is also indicated by the effect of water evaporation processes under the metallic layer, formed at the recovery front. Further enhancement of $q_0$ leads to staggering of material from the action zone, violating the film conti-



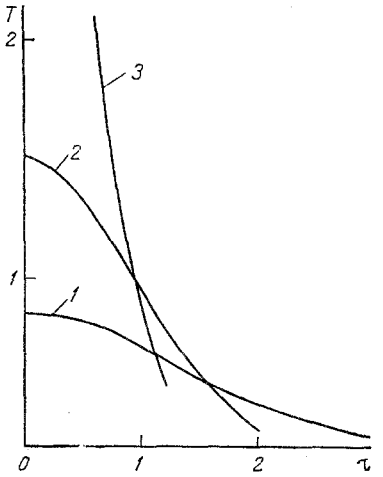




Fig. 4 Fig. 5

Fig. 4. Time dependence of a metal film thickness for various regimes of laser action: 1) $q_0 = 4.8 \cdot 10^9$ W/m$^2$: 2) $3.2 \cdot 10^9$ W/m$^2$. $\xi$, $10^{-6}$ m; t, $10^{-3}$ sec.

Fig. 5. Spatial temperature distributions at the surface for various action regimes: 1) $q_0 = 2 \cdot 10^9$ W/m$^2$; 2) $4.5 \cdot 10^9$; 3) $1.2 \cdot 10^{10}$. T, $10^3$ K; r, $10^{-3}$ m.

nuity of the melting metal due to the emergence of vapors at the surface, and to formation of characteristic craterlike surface reliefs [4]. Further increase in the flux density of the incident radiation leads to predominance of oxide destruction processes and formation of depressions in the action zone.

To estimate the thickness of the recovery layer we substitute T(r, 0, t) into (1). Taking into account that $t \ll 1$, this makes it possible to assume, with good accuracy, that $\arctan 2\sqrt{t} \approx 2\sqrt{t}$. Within the approximation given we carry out the integration (1), as a result of which we write

$$\xi(r, t) = \frac{d_1 T_1^*}{\alpha_2^2(r)} [A_1(u) - A_1(u_0)], \qquad \alpha_2(r) = \alpha_1 \exp(-r^2), \tag{8}$$

where

$$A_1(u) = \frac{T_1^*}{2u^2} - \left(\frac{T_1^*}{2} + T_0\right)\left[\frac{\exp(-u)}{u} - E_1(u)\right],$$

$$u = \frac{T_1^*}{T_0 + \alpha_2(r)\sqrt{t}}, \quad u_0 = \frac{T_1^*}{T_0}, \quad E_1(x) = \int_x^\infty \frac{\exp(-t)}{t}\, dt.$$

The time dependences of the metallic film thickness at the center of the heating spot, calculated by relation (8), are shown in Fig. 4. The nature of the substantial increase in the film thickness as a function of time is determined by the exponential temperature dependence of the film growth rate. It follows from the condition $t \ll 1$ that following the action time of a pulse the radiation temperature does not succeed in reaching a stationary value, leading to a substantial nonstationary regime of film growth of micron size. Curve 1, 2 in Fig. 4 lead to the temperature dependences 1, 2 in Fig. 2. We note the satisfactory agreement between the experimental and calculated values of the recovered oxide thickness.

The solution (5) of the thermophysical problem makes it possible to estimate the sizes and spatial structure of the recovered zone. Figure 5 shows the spatial distributions T(r, 0, t) for various $q_0$ values (curves 1-3). Taking into account $t \ll 1$, these dependences are modeled by the flux density distribution of the incident radiation. The estimates given are valid for regions in which the film thickness is sufficiently large, so as to replace the absorptivity of the two-layer system by the absorptivity of the metal.

An interesting feature of the process is the character of variation of the surface temperature of the flux density of incident radiation at distances from the center of the heating spot, corresponding to the sizes of the recovery zone (determined experimentally, see Table 1). This makes it possible to introduce the concept of effective recovery temperature $T_v$ (in the given case $T_v \approx 470$-$500$ K), i.e., to assume that for propagation of an isotherm $T_v$ at distance $r_v$ from the center of the heating spot during radiation action in the region $r < r_v$ oxide recovery occurs. Differently stated, for $T_v$ the surface temperature value is used, for which recovery occurs for a metal of such thickness ("optical"), that the system absorptivity of the system in the given region becomes equal to the absorptivity of the metal.

This makes it possible to estimate the recovery zone radius at the surface from the position of the recovery isotherm, starting from $t \ll 1$, as

$$r = \left(\frac{1}{k} \ln \frac{\alpha_1 \sqrt{t}}{\Delta T}\right)^{1/2}, \tag{9}$$

where $\Delta T = T_v - t_0$.

Substituting into (9), along with $T_v$, the oxide expansion temperature, one can thus obtain an estimation relation for crater sizes at the surface under the condition that the absorptivity value corresponds to the oxide.

We note the existence of optimal parameters of laser action for obtaining maximum thickness of continuous films of the recovered metal. For relatively low flux densities of radiation, when the surface temperature increases slowly (curves 3, 4 in Fig. 2), the film growth rate, and consequently, its achievable thickness, are low. For high flux densities of the incident radiation the temperature of the sublayer oxide reaches quickly the expansion temperature, leading to material removal from the action zone and destruction of the continuity of the metal film.

In a number of experiments one observes the existence of discontinuous films and solvent drops with sizes smaller than the zone of laser action, and widths up to 100 μm. Obviously, this is related to the motion of a melt, firstly under the action of pressure transport of the evaporating material, and, secondly, under the action of thermocapillary forces caused by the Marangoni effect, i.e., by the change in the surface tension of the melt with temperature. In the case of copper recovery from copper oxides the dependence $\sigma(T)$ has an anomalous character ($d\sigma/dT > 0$), leading to contraction of the melt toward the center of the laser action zone (see Fig. 3b).

In conclusion it must be noted that the diversity of physicochemical processes during laser recovery of oxides leads to the appearance of characteristic regimes, determining the structure and morphology of the recovered layers.

## NOTATION

$T(r, z, t)$, temperature; $\xi(r, t)$, thickness of the metal film; $r'$, $z'$, $r$, $z$, dimensional and dimensionless spatial coordinates; $t'$ and $t$, dimensional and dimensionless times; $q_0$, flux density of the incident radiation at the center of the heating spot; $k$, $\alpha$, and $\lambda$, coefficients of concentration, thermal diffusivity, and thermal conductivity; $T_0$, initial temperature; $d_n$, a constant of the recovery law; $T_n^*$, activation energy of thermochemical recovery reactions in degrees; $r_{12}$ and $r_{13}$, amplitudes of radiation reflection coefficients from a metal and an oxide; $\varepsilon_0$, $\varepsilon$, dielectric constants of the metal and the oxide; $\lambda$, wavelength of radiation; $n_0$ and $n$, refractive indices of the metal and the oxide; $a_0$, metal absorptivity; $T_m$, melting temperature; $T_b$, oxide decomposition temperature; and $\sigma(T)$, surface tension of the metal, depending on the surface temperature.

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# QUASIDYNAMIC MODELING OF HEAT-TRANSFER PROCESSES

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An analytic method is developed for describing heat-transfer dynamics, making effective use of the quasi-dynamic features of the processes. The results are given specific form for heat exchangers of two-flow, one-flow, and immersion type.

Simulative modeling of transient conditions in different technological systems is closely related to mathematical description of the dynamic changes in parameters characterizing the structural elements of such systems. It is expedient here to distinguish two classes of nonsteady processes differing in their rates: 1) fast (pulsed) processes, in which the characteristic times $\tau^*$ of parameter variation at the input to the element are commensurate with, or less then, the relaxational times $\tau_r$ of this element ($\tau^* \lesssim \tau_r$); 2) slow processes, for which $\tau^* \gg \tau_r$; henceforward, these latter processes will be referred to as quasidynamic. Often, the transition from one set of operating conditions of the system to another may be divided into two analogous stages. The quasidynamic stage is the principal component of transient processes in many complex engineering systems, which explains the increased interest in the creation of corresponding models.

Note that most traditional methods of describing the dynamics of heat-transfer processes (numerical methods, Laplacian schemes, etc.) poorly reflect, and make practically no use of, the features of slow evolution of the systems, which entails incorrect computer analysis and complicates the solution of the problem of controlling processes in real-time conditions. This leads to the need to develop high-speed analytical models taking account of the quasidynamic features of the processes which occur. The creation of such models (for the description of heat-transfer elements of cryogenic systems, in the present case) is the aim of the present work.

The basic features of the theory developed are clearly exhibited in the simplest model of dynamic heat transfer between an isothermal wall and a one-dimensional heat-carrier flow, when the evolution of the flow temperature T is described by the classical relation

$$\rho\Omega C_p \frac{\partial T}{\partial \tau} + GC_p \frac{\partial T}{\partial x} = \alpha\Pi (T_w - T).$$

Then, introducing the transport time $\tau_0 = \rho\Omega/G$ and the interaction parameter $U = \alpha\pi/(GC_p)$, the equation obtained is

$$\tau_0 \frac{\partial T}{\partial \tau} + \frac{\partial T}{\partial x} = U(T_w - T), \quad 0 < x \leqslant 1, \quad \tau > 0, \tag{1}$$

with the boundary conditions: $T(0, \tau) = T_1(\tau)$, $T(x, 0) = T_0(x)$. Assuming, for the sake of simplicity, that $\tau_0$, U, $T_W$ = const, it may readily be shown that, at time $\tau$ exceeding the transport time $\tau_0$, the output temperature $T_2(\tau) = T(1, \tau)$ is determined not by the initial distribution $T_0(x)$ but by the inlet temperature $T_1$ as a function of a delayed argument

$$T_2(\tau) = T_w + \exp(-U)[T_1(\tau - \tau_0) - T_w]. \tag{2}$$

Writing the analytical solution of Eq. (1) in the form in Eq. (2) offers the possibility of dispensing with the traditional numerical integration of Eq. (1). This form of solution removes the many problems intrinsic to the numerical method, fundamentally increases

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