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# INVESTIGATION OF MASS AND HEAT TRANSFER PROCESSES IN LASER ALLOYING OF IRON FROM PREDEPOSITED COATINGS OF REFRACTORY METALS


**A. A. Uglov, M. B. Ignat'ev, V. I. Titov, and A. P. Stepanov**



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*The laws of mass and heat transfer in laser alloying of Armco iron from predeposited coatings of molybdenum and chromium were determined experimentally.*


**Introduction.** The processes occurring during laser working of metals are characterized by strong nonuniformity, complicated hydrodynamics, high rates of cooling, and a number of other features, determining the formation of alloys with a large range of physical and chemical properties on metal surfaces. Laser surface alloying and, in particular, alloying from predeposited coatings occupies a special place in the large flow of publications on the problems of laser modification of surface [1, 2]. However most works reflect peculiar cases of the alloying of surfaces of specific brands of alloys performed in order to increase the working characteristics of the alloys. The use of different types of laser setups, treatment regimes, and materials of the base and alloying components for these purposes makes it much more difficult to systematize the experimental data. No publication gives experimental data on the development of alloying processes; the structure, the phase composition, and the distribution of the alloying component over the pool of the crystallized melt after irradiation are primarily studied. Theoretical studies of the mass and heat transfer processes primarily reduce to the construction of a model of the distribution of the alloying component by the thermocapillary mechanism [3]. Without a detailed study of the mass and heat transfer processes, determining the formation of the modified layers, it is impossible to optimize the parameters of the laser radiation and the conditions of laser treatment in the process of laser alloying from predeposited coatings. In this work, with the help of fast optical pyrometry and photoelectronic recording and metallographic and electron-microscopic studies some characteristics of the mass and heat transfer accompanying laser alloying of iron from predeposited coatings of molybdenum and chromium were determined.


A. A. Baikov Institute of Metallurgy, Academy of Sciences of the USSR, Moscow.


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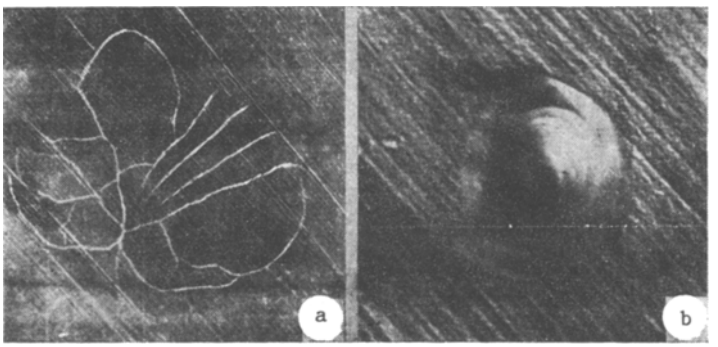


Fig. 1. Surface of chromium (a) and molybdenum (b) coatings, predeposited on Armco iron, after treatment with single pulses of laser radiation with a steep trailing edge; $\tau_p = 9 \cdot 10^{-4}$ sec, $h_{cr} = 1.5 \cdot 10^{-3}$ m, $h_{Mo} = 1.5 \cdot 10^{-3}$ m.

**Method of Study and Experimental Conditions.** Samples of Armco iron $5 \cdot 10^{-3}$ m thick and coated with molybdenum and chromium were treated with single-pulse neodymium laser radiation with a Gaussian energy distribution over the diameter of the focal spot. To follow systematically the stage of development of the processes accompanying laser alloying of iron with the coating material a special modulator cut off the target from the laser radiation. The initial pulse width was equal to $4 \cdot 10^{-3}$ sec. Thus the treatment was performed with single pulses having a steep trailing edge and the pulse width was varied with a step of $10^{-4}$ sec.

The surface temperature in the interaction zone was monitored with a fast optical pyrometer; the viewing zone was $3 \cdot 10^{-4}$ m in diameter and it was centered at the center of the treatment spot. The processes involved in the formation of the erosion laser plasma were studied with the help of fast scanning of the plasma flame through a slit diaphragm $5 \cdot 10^{-5}$ m wide, placed on the axis of the flame. The processing was performed with argon blown into the irradiation zone. The surfaces of the irradiated zones were studied with the help of a JSM-U3 scanning electron microscope. Thin transverse metallographic sections passing through the centers of the alloyed zones were studied under an NU-2E microscope.

**Discussion.** Laser heating of the coating-base system results in the appearance of stresses, associated with the difference in the coefficients of thermal expansion of the adjacent materials, in the film. For the chosen systems (Mo—Fe and Cr—Fe) the stresses in the coating in the region adjacent to the coating-base interphase boundary are tensile stresses while in the layer at the surface the stresses are compressive. At the moment when the tensile stresses exceed the adhesion strength, the coating separates from the base. It should be noted that for brittle coatings (for example, chromium) the relaxation of the tensile stresses can occur not owing to the breakdown of adhesion, but rather owing to cracking of the coating, which is in fact actually observed (Fig. 1a). In the case of treatment of molybdenum coatings cracking is not observed, and at a certain moment in time deformation of the coating in the central part of the zone of laser action occurs (Fig. 1b). The critical temperature corresponding to this zone can be determined by solving the problem of stability in the case of heating of a thin circular plate with radius R on an elastic base, described by the equation of the stress state of the coating [4] and the equation of bending of the coating [5]:

$$T_{cr} \approx 1.22h^2 (\Delta\alpha (1 + \nu_1) R^2)^{-1}. \quad (1)$$

For $R = 10^{-4}$ m and $h_{Mo} = 1.5 \cdot 10^{-5}$ m the critical temperature $T_{cr} \simeq 10^3$°C.

At the location of separation (Fig. 1b) the coating is heated adiabatically and vaporizes locally. After local destruction of the coating the molybdenum coating rolls up into a bead under the action of surface tension (Fig. 2a). As one can see from the figure, when Armco iron with a predeposited Mo coating $1.5 \cdot 10^{-5}$ m thick is irradiated the coating rolls up into a bead near the center of the heating spot, and the unmelted surface of the iron, whose temperature corresponds to the minimum of the curve 1 in Fig. 3 and is equal to 1100°C, which is in good agreement with the estimate of $T_{cr}$ presented above, is uncovered in the viewing zone of the pyrometer. After this the surface temperature passes through a second maximum, above the melting point of Armco iron. Traces of heating are present in the transverse sections, corresponding to a given pulse width, in the base material underneath the molybdenum bead formed. At the same time there are no such traces near the center of the heating spot (see Fig. 2a). This is explained by the fact that the separated Mo coating screens the surface of the substrate from the laser radiation, and its temperature is close to $T_{cr}$. In addition, the melted metal of the bead absorbs the energy from the laser pulse much more efficiently. For this

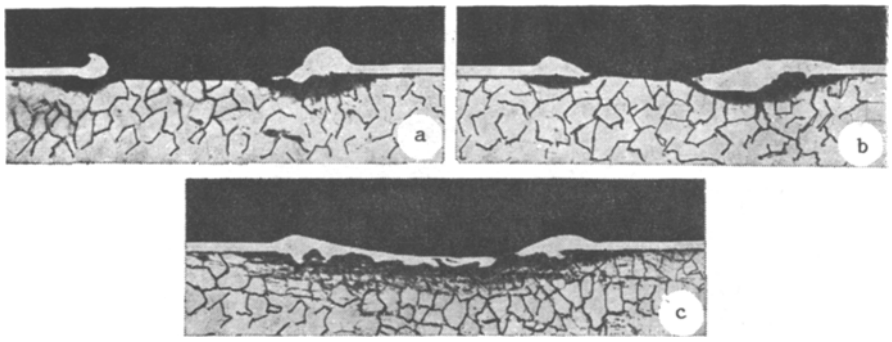


Fig. 2. Microsections of the zones of laser action on the system Mo—Fe. The duration of laser action: a, c) $1.4 \cdot 10^{-3}$ sec; b) $1.9 \cdot 10^{-3}$ sec; $h_{Mo} = 1.5 \cdot 10^{-5}$ m.

reason immediately after the material of the coating rolls up into a bead intense mass transfer of the alloying component from the ball to the center of the action zone starts (Fig. 2b).

High-speed photographic studies of the plasma flames produced by single laser pulses acting on iron coated with Mo showed that initially a microflame forms at some moment in time (Fig. 4). The position of this microflame on the time axis coincides with the first maximum on the curves 1 and 2 of the temperature-time dependence (see Fig. 3). The appearance of a microflame is explained by the fact that prior to its formation, as stated previously, local separation of the coating occurs near the maximum of the energy distribution along the diameter of the focal spot and partial vaporization of molybdenum occurs. The time interval $t_f$ from the end of the formation of the microflame to the start of intense development of an erosion plasma flame is determined by the time during which the coating rolls up into a bead ($t_{roll}$) and the heating time of the bared substrate ($t_h$):

$$t_{roll} \cong (R_1 - r_0) \frac{1}{V_{roll}} = \frac{R_1 - r_0}{\sqrt{\sigma/\rho h}} . \quad (2)$$

For a molybdenum coating with $h = 1.5 \cdot 10^{-5}$ m, $R_1 = 3 \cdot 10^{-4}$ m, assuming that $r_0 \ll R_1$, we obtain $t_{roll} = 7 \cdot 10^{-5}$ sec.

An increase in the laser power density results in a decrease of the time interval $t_f$, as indicated by the scans of the erosion flames (Fig. 4). The time interval between two maxima on the temperature oscillograms also decreases, and the value at the minimum increases (see Fig. 3). Since $V_{roll} \sim 1/\sqrt{\sigma}$ is virtually independent of the temperature the decrease in $t_f$ is associated with the decrease in $R_1$ (which is observed in the experiment), and the increase in the minimum temperature is connected with the increase in the temperature of the substrate and the possible decrease of $R_1$ to a value less than the diameter of the viewing zone. For some values of the laser power density the minimum on the temperature—time dependence degenerates (Fig. 3). This processing regime corresponds to the photoscan of the flame (Fig. 4) in which the formation of the microflame is not observed. The coating also does not roll up into a bead, and mass transfer of the alloying component starts near the center of the heating spot. However for a given power density a significant part of the alloying component evaporates and owing to the action of the recoil pressure of the vapors molybdenum and iron are spattered onto the edge of the shallow crater formed (see Fig. 2c). Thus the coating-base system melts as a single entity. It should be noted that this processing regime gives the maximum alloying depth, but it distorts the surface relief. The small peak in the curve 3 of the time dependence of the temperature (Fig. 3) is explained by the spattering of part of the melted molybdenum onto the edge of the zone of laser action. The existence of a regime in which separation of the coating is not observed can be explained as follows. Increasing the laser power density can lead to an increase in the temperature gradient at the interface of the coating and the base [6]. The magnitude of tensile stresses in the region of the coating adjacent to the Mo—Fe interphase boundary can be estimated with the help of the expression

$$\sigma_1 \sim \frac{(T_1\alpha_1 - T_2\alpha_2) E_1}{1 - \nu_1} . \quad (3)$$

Thus an increase of the temperature gradient will result in a decrease of the tensile stresses to magnitudes less than the adhesion strength of the film to the substrate. Separation of the coating does not occur. Further heating of the coating-substrate system as a single entity results in intense mass transfer of the alloying element into the base.

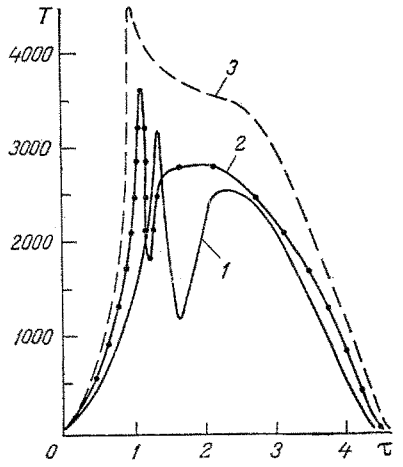


Fig. 3

Fig. 4

Fig. 3. Time dependence of the temperature near the center of the heating spot on a two-layer Mo—Fe system with different laser power density; $h_{Mo} = 1.5 \cdot 10^{-5}$ m. T, °C; $\tau$, sec·$10^{-3}$.

Fig. 4. Photographic scans of laser erosion flames with single pulse action on the two-layer system Mo—Fe; $h_{Mo} = 1.5 \cdot 10^{-5}$ m.

In the case when the coating is brittle, as already noted above, relaxation of the tensile stresses can occur owing to cracking of the coatings. Separation does not occur, thermal contact is established between the coating and the substrate, and mass transfer of chromium into the iron is observed practically along the entire surface of laser action. In contradistinction to molybdenum, the thermophysical properties of chromium are close to those of iron; the vapor pressure of chromium is higher and the vaporization temperature is lower than that of iron [7]. Taking into account the thermal resistance between the coating and the substrate, this results in significant, compared with molybdenum, losses of chromium for any regime of laser alloying of iron and, as a rule, in a lower concentration along the melt pool. In most cases the chromium coating evaporates completely near the maximum of the energy distribution along the diameter of the focal spot.

The described mechanism of interaction of the laser radiation with the chromium-iron system is also indirectly confirmed by studies of the development of a plasma flame. These studies indicate that a microflame is not formed and evaporation starts practically along the entire zone of laser action.

**Conclusions.** Thus different mechanisms of laser alloying of iron from predeposited coatings of refractory metals have been observed:

breakdown of adhesion of the coating and rolling up of the coating into a bead followed by mass transfer of the alloying element from the bead formed into the base;

heating and melting of the coating-substrate system as a single entity and mass transfer of the alloying element along the surface of the zone of laser action.

Experimental data on the time dependence of the temperature in laser processing of coating—substrate systems and the characteristics of the development of the erosion plasma flame above the surface of the zone of laser action were obtained.

## NOTATION

T, temperature; R, radius of the adhesion defect arising on heating; h, thickness of the coating; $\alpha$, coefficient of thermal expansion; $\Delta\alpha$, difference of the coefficients of thermal expansion of the coating and the substrate; E, elastic modulus; $\nu$, Poisson's ratio; $R_1$, radius of the melt zone of the coating; t, time interval; $r_0$, initial radius of the opening

formed in the coating owing to evaporation; $V_{roll}$, rate at which the melt rolls up into a bead; $\sigma$, surface tension; $\rho$, density. Indices: cr — critical; f — flame; roll — rolling up; h — heating; m — melt; 1 — coating; 2 — substrate.

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# CALCULATING KINETICS OF OXIDE FILM GROWTH DURING LASER HEATING


**A. A. Uglov, A. A. Volkov, O. G. Sagdedinov, and Yu. Yu. Krivonogov**



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*The process of oxide film growth on the surface of a copper target in air under the action of $CO_2$-laser radiation is considered.*


The processes of metal oxidation under the action of concentrated thermal fluxes are met with in various scientific and technical applications [1-3]. A wide range of studies has been dedicated to such problems, and overviews of the literature have been presented in [1, 3]. Upon heating of metal by radiation at wavelength $\lambda = 10.6$ $\mu$m in an oxidizing atmosphere the oxide film which forms upon the metal surface significantly increases absorption by the target. In a number of problems knowledge of temperature distributions and temperature gradients in the oxide film and metallic substrate is needed. Comparison of temperature values on the free oxide surface and the metal—oxide boundary is also of interest. In the model used in [2] it was proposed that the oxide film is thermally thin, while the laser radiation was in the form of periodic pulses, with the duration of the individual pulses being much shorter than the time intervals between pulses. This assumption permits replacing the time dependence of radiant flux density with the sum of delta-function pulses and assuming that the oxide film grows in the periods between individual pulses. In [4] the development of dissipative structures during metal oxidation was studied and an asymptotic law was defined for development of one-dimensional compressible structures. The oxidation process was considered only to oxide film thicknesses so small that no oscillations in the laser radiation absorption coefficient were observed.

In the present study we will consider the process of oxidation of a massive copper target by $CO_2$-laser radiation with consideration of thermal losses from the free surface of the oxide film due to convection and thermal radiation. The thermophysical parameters of the body will be considered temperature-independent. In the thermal balance condition on the oxide—air boundary the smallness of the oxide film in comparison to the depth of the thermal effect of the surface heat source was taken into account:

$$\xi(t) < \left| 2dt \exp\left(-\frac{T_d}{T_m}\right)\right|^{1/2} \ll \sqrt{a_1 t}.$$

The absorptivity of the two-layer oxide—metal system $A(\xi)$ was calculated with the well known model of [5]:

$$A(\xi) = 1 - |r|^2,\ r = \frac{r_{01}\exp(-2i\psi) + r_{12}}{\exp(-2i\psi) + r_{01}r_{12}},$$

$$r_{12} = \frac{r_{01} - r_{02}}{r_{01}r_{02} - 1},\ r_{01} = \frac{1 - \sqrt{\varepsilon}}{1 + \sqrt{\varepsilon}},\ r_{02} = \frac{1 - \sqrt{\varepsilon_0}}{1 + \sqrt{\varepsilon_0}}, \quad (1)$$


A. A. Baikov Metallurgy Institute, Academy of Sciences of the USSR, Moscow.


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