NONLINEAR PROBLEMS OF HEATING THIN FILMS BY OPTICAL RADIATION

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We consider thin films deposited on a transparent substrate and heated by the absorption of optical radiation. We derive relations which are convenient for comparing calculated and experimental data.

By using optical radiation to process thin films, a number of new technological problems have been solved, and interest in the study of this problem has been increased. Processes occurring in the action of radiation on thin films have been considered in a number of papers [1-6], in which the heat-conduction problems which arise were solved by using Hankel and Laplace transforms and, as a rule, assuming that the thermophysical and optical parameters are independent of temperature. The solutions obtained in this way contain cumbersome integrals and are not very simple when inverted, which makes them inconvenient to compare with experiments [7]. In the present article the heating of thin films by optical radiation is studied by an integral method [8], which not only simplifies the solution of the problem, but also permits taking account of the temperature dependence of the thermophysical properties of the film and substrate.

We consider a boundary value heat-conduction problem in the one-dimensional approximation, valid for $r_0 \gg \sqrt{a_1 \tau}$, where r_0 is the size of the radiation spot. We assume there is ideal thermal contact between film and substrate, and that the substrate is transparent to the radiation. The system of equations and boundary conditions determining the temperature distribution in the film and substrate has the form

$$\begin{gathered}
\rho_1 c_1 \frac{\partial T_1}{\partial t} = \frac{\partial}{\partial x}\left(k_1 \frac{\partial T_1}{\partial x}\right) + \omega_1(x, t), \\
\rho_2 c_2 \frac{\partial T_2}{\partial t} = \frac{\partial}{\partial x}\left(k_2 \frac{\partial T_2}{\partial x}\right) + \omega_2(x, t), \\
k_1 \frac{\partial T_1}{\partial x}(h, t) = k_2 \frac{\partial T_2}{\partial x}(h, t), \\
T_1(h, t) = T_2(h, t), \\
\frac{\partial T_1}{\partial x}(0, t) = 0, \; T_2(\infty, t) = T_i, \\
T_1(x, 0) = T_2(x, 0) = T_i.
\end{gathered} \tag{1}$$

Subscripts 1, 2 refer to the film and substrate, respectively, and the coordinate x is measured from the front surface of the film. We set $T'_{1,2} = T_{1,2} - T_i$ everywhere, and omit primes for brevity. Since the substrate is transparent, we have for the sources $\int_0^h \omega_1 dx = Aq(t)$, $\omega_2 = 0$, $A = 1 - R - D$.

We solve system (1) by the integral heat-balance method [8], which enables us to reduce the heat-conduction problem to that of solving an ordinary differential equation. We first integrate the equation in the film with respect to x from 0 to h

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$$\int_0^h \rho_1 c_1 \frac{\partial T_1}{\partial t} dx = k_1 \frac{\partial T_1}{\partial x}(h, t) + Aq(t). \tag{2}$$

Neglecting, for simplicity, the temperature dependence of the thermophysical parameters of the film and substrate, and assuming $h < \sqrt{a_1 \tau}$, i.e., that the film is uniformly heated during the whole radiation pulse, we reduce problem (1) to the determination of the temperature distribution in the substrate:

$$\begin{gathered} \rho_2 c_2 \frac{\partial T}{\partial t} = k_2 \frac{\partial^2 T}{\partial y^2}, \\ -k_2 \frac{\partial T}{\partial y}(0, t) = Aq(t) - \rho_1 c_1 h \frac{dT_0}{dt}, \\ T(y, 0) = T(\infty, t) = 0, \; y = x - h. \end{gathered} \tag{3}$$

Here we have everywhere omitted the subscript 2 on the temperature T; $T_0 = T(0, t)$ is the film temperature, which is equal to the temperature of the substrate at the interface. We note that the condition $h < \sqrt{a_1 \tau}$, when the temperature can be taken constant over the thickness of the film and equal to the value at the substrate boundary, is usually satisfied for metallic films of thickness $h \sim 10^{-5}$ cm for $\tau > 10^{-9}$ sec. We now introduce the depth of heating of the substrate $\delta(t)$ in the standard way [8], make the substitution $T(y, t) = T_0 \varphi(\zeta)$, and integrate Eq. (3) with respect to $y = \zeta\delta$ from 0 to δ to obtain the system

$$\begin{gathered} \rho_2 c_2 B \frac{d(\delta T_0)}{dt} = -\frac{k_2 G T_0}{\delta}, \\ \rho_1 c_1 h \frac{dT_0}{dt} = \frac{k_2 G T_0}{\delta} + Aq(t), \\ B = \int_0^1 \varphi d\zeta, \quad G = \frac{d\varphi}{d\zeta}(0). \end{gathered} \tag{4}$$

We assume the absorbing power of the film varies linearly with the temperature $A = A_0 + A_1 T$, A_0, A_1 = const, and consider two cases: $A_0 > A_1 T$, $A_0 < A_1 T$.

Let $A = A_0$. The first integral of system (4) can be written in the form

$$\begin{gathered} T_0 = \frac{A_0 \gamma E(t)}{\rho_1 c_1 h(1 + \gamma)} = \frac{T_a}{1 + \gamma^{-1}}, \\ E(t) = \int_0^{t \leqslant \tau} q dt, \quad \gamma = \frac{\rho_1 c_1 h}{\rho_2 c_2 \delta B}. \end{gathered} \tag{5}$$

Equation (5) expresses the law of conservation of energy, and the parameter γ is the ratio of the volumetric heat capacities of the film and heated layer of substrate, and determines the rate of heating of the film. For the depth of heating of the substrate we obtain from (4)

$$\delta(t) = \frac{k_2 G T_0}{\rho_1 c_1 h \frac{dT_0}{dt} - A_0 q(t)}, \tag{6}$$

and the problem of determining T_0 is reduced to the integration of Abel's equation of the second kind:

$$Z \frac{dZ}{dt} = \alpha (Z + T_a)^2, \tag{7}$$

where

$$Z = T_0 - T_a; \quad \alpha = -\frac{k_2 \rho_2 c_2 B G}{(\rho_1 c_1 h)^2}.$$

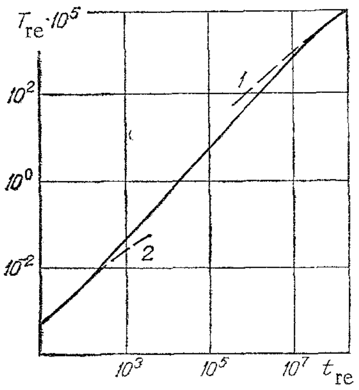

Fig. 1. Dependence of reduced temperature $T_{re} = T_0/A_0q_0$ (K·m²/W) of a copper film of thickness h = 1 μm on reduced time $t_{re} = t\alpha_1/h^2$. Dashed lines represent the limiting cases ($\gamma \gtrless 1$): 1) $T_{re} \sim t_{re}^{0.5}$; 2) $T_{re} \sim t_{re}$.

An important aspect of the application of the integral method to thermal problems is the choice of the function $\varphi(\zeta)$. Generally, in problems with surface heat sources the temperature profile is well approximated by power functions of the form $\varphi = (1 - \zeta)^m$ with $m > 1$ [8, 9]. For this class of profiles we have $B = (1 + m)^{-1}$ and $G = -m$, and hence $\frac{1}{2} < |BG| < 1$.

In the limiting cases $\gamma \gtrless 1$ it is not necessary to integrate (7), since the solution can be found from Eqs. (5) and (6). For $\gamma \gg 1$ heat transfer into the substrate is small, the contact of the film with the substrate can be considered adiabatic, and the heating of the film is determined by its volumetric heat capacity:

$$T_0 = T_a = \frac{A_0 E(t)}{\rho_1 c_1 h}. \tag{8}$$

If $\gamma \ll 1$, the rate of heating of the film is determined mainly by the amount of heat accumulated in the substrate, and we write Eqs. (5) and (6) in the form

$$T_0^2 = \frac{m+1}{m}\frac{A_0^2 q(t) E(t)}{k_2 \rho_2 c_2}. \tag{9}$$

For a constant radiation flux density $q = q_0 = \text{const}$, Eq. (9) gives $T_0 = T_a\sqrt{\alpha t}$, and by comparing this with a similar solution in [1], we find the exponent $m = \pi/(4 - \pi)$. We note that solution (9) is valid also for a strongly absorbing substrate (D = 0), and in this case corresponds to pure surface heating of the material by the energy flux.

Thus, for short radiation pulses the film can be considered thermally insulated from the substrate, and approximation (8) is used, whereas Eq. (9) applies for long pulses when steady heat transfer into the substrate is established. In intermediate cases the solution is readily found by integrating Eq. (7) numerically, e.g., by the Runge–Kutta method.

Figure 1 shows the solution of Eq. (7) for the irradiation of copper films of thickness h = 1 μm by radiation of constant intensity. For $10^{-6} \leqslant \tau \leqslant 10^{-3}$ sec the solution is determined directly by integrating Eq. (7), which in general cannot be reduced to quadratures. We note that the film temperature can be considered uniform over the thickness after a time of the order $t \sim 10h^2/\alpha_1 \sim 10^{-7}$ sec.

We now let $A = A_1 T_0$ and limit ourselves to the case $q = q_0 = \text{const}$. Clearly, the increase in absorbing power of the film with increasing temperature must lead to an acceleration of the heating rate. For adiabatic heating of the film we obtain from Eqs. (4)

$$T_0 = T_i \exp\left(\frac{A_1 q_0 t}{\rho_1 c_1 h}\right), \tag{10}$$

and for $\gamma \ll 1$ we have

$$T_0 = T_i \exp\left(\frac{A_1^2 q_0^2 t}{k_2 \rho_2 c_2}\right). \tag{11}$$

These formulas agree with the corresponding limits derived in [10].

In intermediate cases ($\gamma \sim 1$) the solution is found by solving the system of ordinary differential equations (4) for the derivatives and integrating by standard methods.

From Eqs. (8)-(11) it is easy to estimate the threshold densities of radiant energy for which the film is destroyed. If the adhesion of the film to the substrate is sufficient to maintain the liquid phase on the substrate, the film is destroyed mainly by evaporation. For poor adhestion the molten film may slide under the action of surface tension forces [1]. Destruction of the film by evaporation involves the largest expenditure of energy, and melting involves the smallest. In practice, the theshold value of the energy to destroy the film may take any value in the range $E_{\min} \leqslant E \leqslant E_{\max}$, which will be determined by the ratio of the contributions of the evaporation and sliding mechanisms.

In conclusion, we note that in solving problem (1) by the integral method, the temperature dependence of the thermophysical properties of the film and substrate can be taken into account. To do this it is necessary to make the substitution

$$\Theta_{1,2} = \int_0^{T_{1,2}} \rho_{1,2}(T)\, c_{1,2}(T)\, dT$$

in the initial equations. As a result $T_{1,2}$ in the above equations is replaced by $\Theta_{1,2}$. The film temperature T_0 is now determined from the equation $\Theta_0 = \int_0^{T_0} \rho_2 c_2 dT$.

NOTATION

T, temperature; x, Cartesian coordinate; t, time; h, film thickness; τ, duration of radiation pulse; q(t), radiation flux density; c, specific heat; ρ, density of material; k, thermal conductivity of medium; a, thermal diffusivity; A, absorptivity of film; R, reflection coefficient of film; D, transmission of film; δ, heating depth; y, ζ, new arguments; φ, auxiliary function; γ, dimensionless parameter; E, surface density of radiation energy; Θ, new variable (heat content). Subscripts: 1, film; 2, substrate; i, initial; o, boundary; a, adiabatic; re, reduced.

LITERATURE CITED

1. M. N. Libenson, "Heating and destruction of thin films by laser radiation," Fiz. Khim. Obrab. Mater., No. 2, 3-11 (1968).
2. G. L. Gurevich, "On the theory of destruction of thin films by laser radiation," Fiz. Khim. Obrab. Mater., No. 3, 5-11 (1973).
3. G. L. Gurevich and V. A. Murav'ev, "Action of laser radiation on thin films," Fiz. Khim. Obrab. Mater., No. 1, 3-8 (1973).
4. B. Ya. Lyubov and É. N. Sobol', "Quantitative analysis of the kinetics of evaporation of metallic films which have absorbed laser radiation," Fiz. Khim. Obrab. Mater., No. 6, 8-15 (1976).
5. G. R. Levinson and V. I. Smilga, "Laser processing of thin films (survey)," Kvantovaya Elektron., 3, 1637-1659 (1976).
6. G. L. Gurevich and V. A. Murav'ev, "Effect of temperature dependence of reflection coefficient on heating of thin films by laser radiation," Fiz. Khim. Obrab. Mater., No. 6, 26-29 (1973).
7. G. R. Levinson and V. I. Smilga, "Experimental study of destruction threshold of thin metallic films under laser radiation," Fiz. Khim. Obrab. Mater., No. 4, 124-128 (1971).
8. N. M. Belyaev and A. A. Ryadno, Methods of Unsteady Heat Conduction [in Russian], Vysshaya Shkola, Moscow (1978).
9. A. V. Lykov, The Theory of Heat Conduction [in Russian], Vysshaya Shkola, Moscow (1967).
10. H. S. Carslaw and J. Jaeger, Conduction of Heat in Solids, Clarendon, Oxford (1959).