VAPORIZATION OF MATTER THROUGH INTERACTION WITH INTENSE ENERGY FLUXES

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UDC 532.529.5/6

Surface and volume mechanisms for the evaporation of matter through interaction with intense radiation fluxes are discussed. Calculations are performed which assume the existence of both fluctuation and steady-state bubbles in a material and which include superheating of the material ahead of the inward-travelling vaporization wave. The dependence of the incident energy flux density on the average thickness of the energy deposition zone for which the transition from surface to volume vaporization occurs is obtained for aluminum, copper, and lead.

1. There are two different points of view with respect to the vaporization of material through the incidence of high radiation fluxes, for example, laser radiation on a metal. A number of authors [1-3] assume the material is vaporized from the surface while others [4-6] suppose that volume vaporization in the heated zone dominates. Under conditions resembling the experimental conditions in [6], volume vaporization certainly is the predominant mechanism. The point is the heated layer was thick in those experiments and the temperature of the material was high (close to critical). With expansion of the material surface tension is reduced and heterophase fluctuations are considerably favored.

In the general case, unfortunately, quantitative evaluations of the limits of one or the other vaporization mechanism is made difficult by the absence of reliable information about a whole series of factors, about the rate of bubble growth in liquid metals, for example, etc. Experimental data [1, 3], as correctly noted in [3], does not yet permit one to answer this question unambiguously since the differences in the parameters measured in these experiments are small for different assumptions about the vaporization mechanism. An attempt is made below to evaluate the region of dominance of one or the other vaporization mechanism.

2. In the interaction of radiation in the optical range at comparatively low flux densities, even with opaque materials, the vapor layer which is formed is transparent (at least, for small thicknesses of such a layer) [1-3]. Because of the vaporization, radiation has a chance to penetrate into deeper layers of the material, to heat them, and to give rise to vaporization, etc. – a vaporization wave is created. Radiation absorption and phase transition occur in a rather thin layer, and the discussion can be confined to the plane problem Since the main portion of the energy is consumed in vaporization in this mode, one can estimate the wave velocity and the mass consumption $\dot m$ from the following relation:

$$\dot m = \rho_0 D = q^\circ_r (1 - K_r)/Q \tag{2.1}$$

where ρ_0 is the density of the condensed solid, D is the velocity of the vaporization wave front, q_r° is the constant radiation flux density incident on the solid, K_r is the average reflection coefficient for this radiation, and Q is the heat of vaporization per gram of material. In the derivation of Eq. (2.1), it was assumed the temperatures of the cold material fed into vaporization wave can be neglected in comparizon with phase transition temperature T_V. The enthalpy of the condensed absolutely cold material is assumed

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Moscow. Translated from Zhurnal Prikladnoi Mekhaniki i Tekhnichekoi Fiziki, No. 5, pp. 49-57, September-October, 1972. Original article submitted April 20, 1972.

to be $(-Q)$, and the enthalpy of the gaseous absolutely cold material is assumed to be zero. In addition, for simplicity, the enthalpy h of the vapor and its kinetic energy $\frac{1}{2}u^2$, where u is mass velocity, are neglected in comparison with Q.

Equation (2.1) for $q_r^0(1-K_r) = 5$ MW/cm^2 and $Q = 10$ kJ/g (Al) gives $\rho_0 D = 5 \cdot 10^2$ g/cm^2-sec and $D = 2$ m/sec. Thus the vaporization wave moves relative to the material ahead of it at an essentially subsonic velocity. Such a quasitationary propagation mode, of course, is established only for a certain time after the initiation of vaporization. We do not consider here those properties which may be associated with nonstationary states of motion of the vaporization wave. We limit the discussion to the quasistationary problem.

Let a constant radiation flux q_r^0 incident on a solid decrease exponentially with depth as given by

$$q_r = q_r^0 (1 - K_r) \exp[-(x - x_v)/l_0] \tag{2.2}$$

where q_r is the radiation flux density at the depth x, x_v is the rightward-moving vaporization boundary, and l_0 is the characteristic thickness of the energy deposition zone in the solid. We assume for simplicity that reflection occurs at the surface of the solid.

A similar vaporization wave can also be propagated through a material through the interaction with it of other forms of energy; for example, this can occur through energy deposition from Joule heating caused by the passage of an electric current created by the penetration of a powerful magnetic field into the material [7]. A sharp decrease of conductivity in vapors leads to their "transparency" to a magnetic field. The boundary of the region of energy deposition is displaced along with the material being vaporized. The depth of penetration of the field in copper is $3 \cdot 10^{-2}$ cm at a time of the order of 1 μsec. This depth is considerably greater than the depth of penetration in metals for radiation in the optical range. In other cases (for example, through interaction with an intense electron beam), propagation of a vaporization zone is also possible although it is nonstationary and screened by the vapor layer. In this case, calculations relative to the stationary problem may be used for an evaluation with the understanding q_r^0 means the energy flux density penetrating to the vaporization zone. Finally, and in the case of optical radiation also, the quantity l_0 may markedly exceed the characteristic depth in metals; as an example, consider the semitransparent materials used in the experiment [6].

The energy flux density is determined through ordinary thermal conductivity by the relation

$$q_T = -K\partial T/\partial x \tag{2.3}$$

where K is the coefficient of thermal conductivity and T is the temperature of the solid. The energy equation takes the form

$$\rho_0 c_p \frac{\partial T}{\partial t} + \frac{\partial q}{\partial x} = 0 \qquad (q = q_T + q_r) \tag{2.4}$$

where c_p is the heat capacity at constant pressure, t is time, and q is the total thermal flux density.

We shall seek a solution of Eq. (2.4) in the form $T = T(x-Dt)$. Using the boundary condition $T \to 0$ when $q \to 0$, we obtain from Eq. (2.4)

$$q = \rho_0 c_p D T \tag{2.5}$$

Note that the radiation flux density q_r° at the boundary of the solid including partial reflection must satisfy the condition

$$q_r^\circ (1 - K_r) = \rho_0 D (c_p T_0 + Q) \tag{2.6}$$

From Eqs. (2.2), (2.3), (2.5), and (2.6), we obtain

$$-\frac{K}{q_r^\circ(1-K_r)} \frac{\partial T}{\partial x} + \exp[-(x-x_v)/l_0] = \frac{T}{T + Q/c_p} \tag{2.7}$$

We reduce Eq. (2.7) to dimensionless form by introducing $T^* = T/T_0$ and $x^* = x/l_0$:

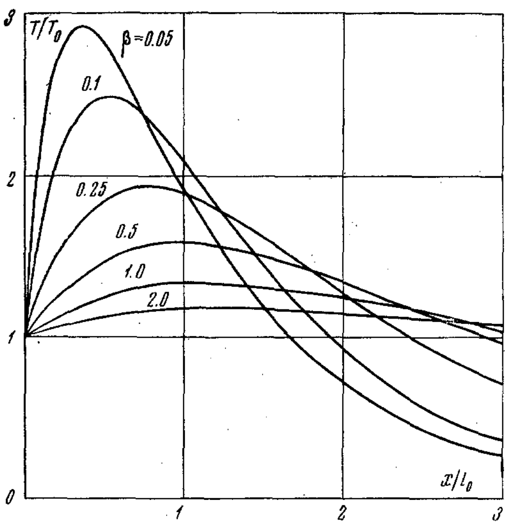

Fig. 1

$$-\beta\frac{\partial T^*}{\partial x^*}+\exp(-x^*+x_v{}^*)=T^*\alpha,\quad \alpha=\frac{1}{1+Q/c_pT} \qquad (2.8)$$

$$\beta=\frac{KT_0}{q_r{}^\circ(1-K_r)\,l_0}$$

The solution of Eq. (2.8) has the form

$$T^*(x^*)=\frac{1}{\alpha+\beta}\exp\left(-\frac{\alpha x^*}{\beta}\right)\left[\exp\left(x^*\left(\frac{\alpha}{\beta}-1\right)\right)-1+\alpha-\beta\right] \qquad (2.9)$$

This equation indicates that there is always some superheating of the condensed solid ($T/T_0 > 1$) ahead of the inwardly travelling vaporization wave because of thermal conductivity.

The physical significance of this is that vaporization of material through surface vaporization occurs because of the influx of heat by ordinary thermal conductivity from some region where the radiation energy is deposited (vaporization occurs because of the supply of heat from within). Thus the distribution of temperature with depth in units of l_0 is shown in Fig. 1 for $\alpha = 0.25$.

It is clear that superheating increases as the value of the parameter β decreases, or as the value of q_r°, which appears in β, increases, for example.

The dependence of the maximum superheating T_{max}/T_0 on the parameter δ_T/l_0 is shown in Fig. 2 where δ_T is the width of the zone of heating through thermal conductivity. The quantity δ_T and the residence time t_T of particles of the condensed solid in the heating zone can be estimated from the obvious relations

$$\delta_T=\sqrt{at_T},\qquad t_T=\delta_T/D,\qquad \delta_T=a/D$$

Here, a is the thermal diffusivity of the material ($a^2 = K/c_p\rho_0$).

We draw attention to the fact the thickness δ of the heated layer and also the particle residence time in the heated layer increase as the radiation flux density $q_r{}^\circ$ and the velocity D decrease. While $\delta_T = 10^{-3}$-10^{-4} cm and $t_T = 10^{-1}$-10^{-2} μsec for a velocity D = 10 m/sec (for $q_r{}^\circ(1-K_r) = 30$ MW/cm^2) and for a thermal diffusivity of 0.1-1 cm^2/sec, the thickness δ_T increases to 10^{-2}-10^{-3} cm and the residence time to 10-100 μsec for a velocity of 1 m/sec, i.e., for a flux density $q_r{}^\circ(1-K_r) = 3$ MW/cm^2.

It is clear from Fig. 2 that for that for l_0 = const, the maximum superheating of the material increases when there is a reduction in the coefficient of thermal conductivity and consequently in δ_T. The residence time $t_{T,q}$ for particles in the region of the vaporization wave is defined as the ratio between the width $\delta_{T,q}$ of the heated zone, i.e., the larger of the quantities l_0 and δ_T, and the velocity D of the vaporization wave. This velocity, is turn, can be determined from Eq. (2.6), given q_r°.

We consider the heating of the liquid (we neglect the heat of fusion in comparison with the heat of vaporization) at a constant pressure equal to the phase transition pressure $p_v(T_v)$. Calculations were made for aluminum, copper, and lead. The required physical constants were taken from the appropriate handbooks, for example, the $p_v(T_v)$ relationship was represented in the form

$$\lg p_v=A-B/T_v$$

where A and B are constants, and was extrapolated to the critical point which for lack of other information was defined as $p_c = \rho_0 c_0{}^2/27$ [6] where c_0 is the velocity of sound for the material in the normal state. The value of the critical pressure p_c for lead was taken from [8]. In the boiling point range at pressures from normal to critical, values of the parameter α for Al, Cu, and Pb vary within the following limits, respectively: 0.18-0.56, 0.18-0.57, and 0.26-0.39.

The value of the reflection coefficient K_r was taken to be 0.52, 0.50, and 0.66 respectively for Al, Cu, and Pb.

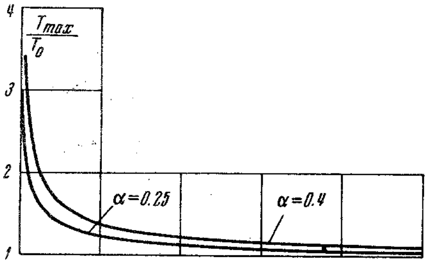

Fig. 2

The value of the coefficient of thermal conductivity K was taken from [9, 10] with K decreasing abruptly by approximately a factor of 2 in the transition through the fusion temperature.

The pressure p_V was determined from the relation

$$p_v/q_r^\circ = C \tag{2.10}$$

where C is a constant in the stationary case. Equation (2.10) can be considered as an interpolation of the experimental data. It agrees with practically all theoretical calculations (based on any of the theories). The quantity C was taken to be 4, 3, and 18 dyn-sec/J for Al, Cu, and Pb, respectively.

By increasing the liquid temperature $T = T_V + \Delta T$, where ΔT is the absolute superheating of the material, and by maintaining a constant pressure $p = p_V$, the pressure inside vapor bubbles will be increased. The vapor pressure is only a function of the temperature as was shown in [11].

Under these conditions, a bubble of radius

$$r_c = 2\sigma / \Delta p \qquad (\Delta p = p(T) - p_v(T_v)) \tag{2.11}$$

will be in unstable equilibrium with the liquid. Here, r_c is the critical radius at which a bubble continues to grow spontaneously.

3. On the basis of the theory of heterophase fluctuations [11, 12], one can estimate the rate of formation I of supercritical bubbles (nuclei) per second and per cubic centimeter of liquid:

$$I = N_0 \sqrt{\frac{2\sigma}{\pi m_p M}} \exp\left(-\frac{\omega}{kT}\right) \exp\left(-\frac{4\pi r_c^2 \sigma}{3kT}\right) \tag{3.1}$$

Here, N_0 is the number of molecules per cm^3, σ is the coefficient of surface tension, m_p is the proton mass, M is molecular weight, ω is the heat of vaporization per molecule, k is the Boltzmann constant, and $T = T_V + \Delta T$ is the temperature of the material. Following [13], the quantities σ and ω were taken in the form

$$\sigma = \sigma_0 \left(\frac{T_c - T}{T_c - T_v}\right), \qquad \omega = \omega_0 \left(\frac{T_c - T}{T_c - T_v}\right) \tag{3.2}$$

where σ_0 and ω_0 correspond to the boiling point values at normal pressure.

An actual liquid has solid particle impurities, inhomogeneities, etc. and also a certain steady-state size distribution of bubbles per cm^3, $N_S(r)$. We shall attempt to determine the effect on vaporization of the difference between an actual liquid (in this case, a liquid metal) and an ideally pure liquid. M. G. Sirotyuk [14] measured the relation $N_S^\circ(r)$ for the number of bubbles per cm^3 of distilled water having a radius greater than r. Between the functions $N_S(r)$ and $N_S^\circ(r)$ there is obviously the following relation:

$$N_s^\circ(r) = \int_r^\infty N_s(r)\,dr$$

The experimental relations $N_S^\circ(r)$ and $N_S(r)$ can be approximated within an order of magnitude by the functions

$$N_s^\circ(r) = \frac{10^{-10}}{2r^2}, \quad N_s(r) = \frac{10^{-10}}{r^3} \tag{3.3}$$

where the value of the constant appearing in Eqs. (3.3) plays a minor role.

According to the theory of Ya. I. Frenkel' [11], dependence of the number of bubbles per cm^3 in the neighborhood of the boiling point, $N_f(r)$, is of a probabilistic nature:

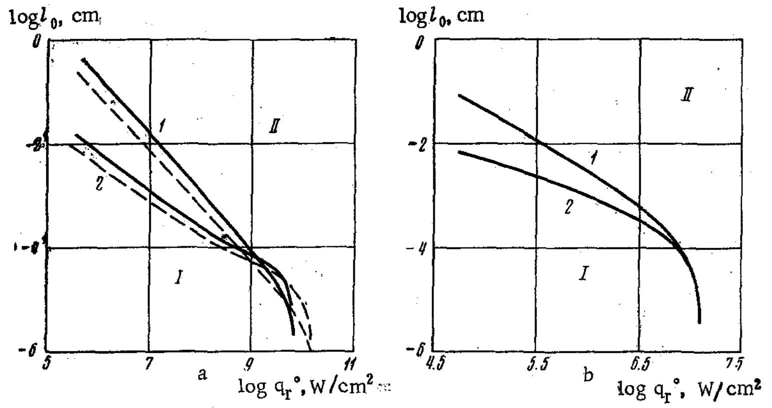

Fig. 3

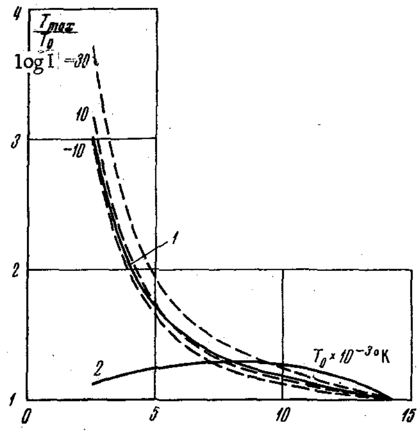

Fig. 4

$$N_f(r) = N_0 \exp(-4\pi\sigma r^2/3kT) \tag{3.4}$$

Unfortunately, the functions $N_s°(r)$ and $N_s(r)$ have not been determined for metals, and we therefore assume Eqs. (3.3) are also valid for the molten metals mentioned above. Note that the air saturation of water in the experiments [14] was 0.025 cm^3/ml, and this may be even greater for metals, for example, 3.2 cm^3/ml in cast iron [15].

We define a condition for the comparability of surface and volume vaporization by means of the equality of the area s_0 on which the radiation is incident and the total bubble area s_b, made up of the area s_1 of bubbles growing from already existing bubbles and of the area s_2 of bubbles formed, during the residence time t in the heating wave:

$$s_b = s_1(t) + s_2(t) = \int\limits_{r_c(t)}^{\infty} N(r, t)\, s(t)\, dr + \int\limits_0^t \int\limits_{r_c(t)}^{\infty} I(r, t)\, dr\, s(t)\, dt$$
$$s(t) = 4\pi R^2(t), \quad R(t) = r_c(t) + \int\limits_0^t v(t)\, dt \tag{3.5}$$

Here, s(t) and R(t) are the area and radius of a bubble, $r_c(T(t))$, $\Delta p(T(t))$, $\sigma(T(t))$, and T(t) are taken from Eqs. (2.11), (3.2), and (2.9), the equality x = Dt is used, N(r, t) is taken from Eqs. (3.3) or (3.4) depending on the kind of bubble, and I(r, t) is determined from Eq. (3.1) in the case of fluctuations.

The quantity v(t) appearing in Eq. (3.5) is the rate of radial growth of a bubble. In first approximation and neglecting viscosity effects, v can be set equal to the maximum Rayleigh velocity

$$v(t) = \sqrt{{}^2/_3 \Delta p(T(t)) / \rho_0} \tag{3.6}$$

Note that in Eq. (3.5) we do not consider boiling at centers of vapor formation – depressions in the surface of heat-emitting walls which are filled with gas or vapor – since we are considering a plane case without the presence of a wall or of hydrophobic particles.

As an estimate, one can assume the bubble area s_f in the case of fluctuation theory will be

$$s_f = 4\pi (v t_{T,q})^2 I\, t_{T,q} \delta_{T,q} \tag{3.7}$$

Here we make the following assumptions relative to Eq. (3.5):

$$T(t) = T_{\max}, \quad r_c = 0, \quad v(t) = v(T_{\max}), \quad I = I(T_{\max}), \quad t = t_{T,q}$$

and neglect the term corresponding to the growth of already existing bubbles.

On the other hand, the second term in Eq. (3.5) goes to zero when only steady-state bubbles are present, and for the same assumptions their area s_S is

$$s_s = 4\pi (vt_{T,q})^2 N_s \delta_{T,q} \quad (N_s = N_s(r_c)) \tag{3.8}$$

Satisfaction of the condition for equality of areas mentioned above makes it possible to determine the dependence of the incident flux density q_r° on the characteristic thickness l_0 of the energy deposition zone. The resultant curves obtained respectively for Al, Cu (dashed curve), and Pb are shown in Figs. 3a and 3b.

In Fig. 3a and subsequent figures, curves denoting fluctuation bubbles are marked with the number 1 and those related to steady-state bubbles are marked 2.

For both assumptions (there are only fluctuation bubbles or there are only steady-state bubbles), it is clear that the l_0 for which the transition from surface boiling (region I) to volume boiling (region II) occurs decreases as q_r° increases. Note that for steady-state bubbles this relation falls below that for fluctuation bubbles over a considerable range of q_r°. This is evidence that the values of l_0 for which the transition from surface to volume boiling occurs for given values of q_r° are reduced when steady-state bubbles are present.

It is obvious that under the natural assumption both types of bubbles are present a generalized relation $q_r^\circ(l_0)$ will correspond to the lower portion of the curves at a given value of q_r°; i.e., for example, in aluminum fluctuation bubbles will play the leading role in the transition from surface to volume vaporization when $l_0 < 6.0 \cdot 10^{-5}$ cm, steady-state bubbles when $6.0 \cdot 10^{-5}$ cm $\leq l_0 \leq 1.2 \cdot 10^{-2}$ cm, once again fluctuation bubbles when $1.2 \cdot 10^{-2}$ cm $< l_0 \leq 5.0 \cdot 10^{-1}$ cm, and volume vaporization occurs in any case when $l_0 > 5.0 \cdot 10^{-1}$ cm.

As an illustration, we point out that in the interaction of aluminum with laser radiation having a free path $l_0 = 10^{-5}$ cm, it is found that the transition from surface to volume vaporization occurs with fluctuation bubbles upon exceeding $q_r^\circ = 5 \cdot 10^9$ W/cm^2 and upon exceeding $q_r^\circ = 6.0 \cdot 10^9$ when it is assumed steady-state bubbles are present [14]. When both types of bubbles are present, the transition between the two vaporization mechanisms will be accomplished at the lower value $q_r^\circ = 5.0 \cdot 10^9$ W/cm^2, i.e., fluctuation bubbles will be the determining factor in this case. Note that the q_r° scale of values in Fig. 3a and b corresponds to surface temperature values obtained from Eq. (2.10) and the $p_V(T_V)$ relation. Thus $q_r^\circ = 5 \cdot 10^5$ W/cm^2 corresponds to the initiation of boiling in Al at p = 1 atm ($T_V = 2621$°K) and $q_r^\circ = 7 \cdot 10^9$ W/cm^2 to the critical point ($T_C = 14{,}200$°K).

Maximum values of the superheating (T_{max}/T_0) obtained for the condition $s_0 = s_b$ according to fluctuation theory and under the assumption steady-state bubbles are present are shown in Fig. 4 as a function of the surface temperature T_0 for Al. It is clear that these curves are very different, particularly for flux densities equal to, or somewhat greater than, the radiation flux density producing vaporization. The indicated values are $T_{max}/T_0 = 3.0$ and 1.1, respectively, from fluctuation theory and under the assumption steady-state bubbles are present. As q_r° increases, these curves approach each other and even cross. As indicated above, this is evidence of a shift in the dominant type of bubble in the transition from surface to volume vaporization. As T_0 increases to T_C, the maximum superheating T_{max}/T_0 ends to unity because $\sigma \to 0$ and $\omega \to 0$. Assuming the existence of both types of bubbles, the generalized superheating curve will be represented by the lower portions of the curves in Fig. 4.

Figure 4 shows superheating values T/T_0 as a function of T_0, calculated from fluctuation theory [11, 12], which are sufficient to produce critical bubble formation rates I equal to 10^{30}, 10^{10}, and 10^{-10} sec^{-1}-cm^{-3} in molten Al under steady-state conditions. For example, it is clear that a small change in T/T_0 leads to an order-of-magnitude change in I. This fact was used in the evaluation of s_b.

Thus the presence of steady-state bubbles in many ways facilitates the transition from surface to volume vaporization as shown by calculations, limited by the scope of the assumptions, of the interaction between semitransparent materials and laser radiation (with the thickness of the energy deposition zone much greater than the average range of optical radiation in metals, $l_0 \sim 10^{-4}$-10^{-5} cm). However, for typical laser values of l_0 for metals, surface vaporization should occur first followed by volume vaporization as q_r° increases with the transition at $5 \cdot 10^9$, $3 \cdot 10^9$, and 10^7 W/cm^2 for Al, Cu, and Pb, respectively; in this case, the presence of steady-state bubbles obviously has little effect on the boundary between the predominant vaporization mechanisms.

The author thanks I. V. Nemchinov for suggesting the topic and for valuable discussions.

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