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EVAPORATION OF A SOLID INTO VACUUM

A. V. LUIKOV, T. L. PERELMAN and S. I. ANISIMOV
Heat and Mass Transfer Institute, Minsk, USSR

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Аннотация—Рассматривается вопрос о корректной постановке проблемы об испарении твердого тела в вакуум. Определены характеристики, описывающие испарение.

Показано, что при некоторых режимных параметрах скорость испарения определяется истинной кинетикой фазового превращения (твердое тело-пар). При этом имеет место волна разрежения, на фронте которой происходит перенасыщение пара. Давление пара на поверхности тела меньше насыщенного пара при данной температуре тела.

NOMENCLATURE

R,	thermal conductivity;
a,	thermal diffusivity, $a = R/c_p\rho$;
t,	time;
ω,	vapour concentration, $\omega = m.m$;
ρ,	density $= m.n$;
m,	mass of a molecule;
n,	number of molecules per unit volume, that is density of vapour molecules or concentration of molecules;
p_0,	saturation vapour pressure at T_0;
p,	vapour pressure near a solid surface;
$f(x, \vec{v})$,	distribution function;
k,	Boltzmann constant;
T,	temperature [°K];
u,	velocity of molecules;
γ,	ratio of vapour heat capacity at constant pressure to that at constant volume;
ρ,	vapour density;
λ,	mean free path of vapour molecules;
q,	specific heat flux.

1. THE FORMULATION OF A CONJUGATED PROBLEM IN EVAPORATION

For the basic quantities in evaporation of a solid, such as the evaporation rate, temperature of a gas and solid, momentum of an expanding gas, etc to be calculated, it is necessary to solve three interconnected problems: the problem of heat conduction in a solid, the hydrodynamic problem of vapour motion and the kinetic problem of boundary conditions at the evaporation front which relates the first two.

In the majority of the works devoted to the problem under consideration usually one of these problems is being solved, and instead of solving the two remaining ones assumptions insufficiently substantiated are as a rule made which appear either incorrect or correct in some limiting cases. For example, a temperature distribution within an evaporating solid is frequently found by solving the Stefan-type problem without regard for the kinetics of phase transition and the phenomena in a gaseous phase. Obviously this is valid only when the rate of a process is controlled by heat conduction, and internal vapour energy is small in comparison to the heat of evaporation. In some works [1–4] such a statement of the problem is somewhat improved by the fact that evaporation kinetics and total enthalpy jump at the interface are taken into account.

In those cases when the evaporation rate is mainly of interest the mass flow from a solid phase into a gaseous one is usually given by

$$j = \frac{p_0(T_0)}{\sqrt{(2\pi mkT)}} - \frac{p}{\sqrt{(2\pi mkT)}}, \qquad (1)$$

where $p_0(T_0)$ is the saturation vapour pressure at a temperature T_0; p and T are the pressure and temperature of the vapour near the solid surface, respectively; and the subscript "0" denotes the solid phase. In addition to the fact that equation (1) is incorrect, it is not clear, to what point in the gas the values of T and p should be referred.

In fact, the process of liquid evaporation consists not only of a vapour transfer process but also of the process of phase change (true kinetics of phase transition). Usually with liquid evaporation at atmospheric pressure the rate of kinetics of phase transition is neglected (it is considered infinitely large as compared to the vapour transfer rate). The diffusional vapour transfer rate is determined by Fick's diffusion law which for steady mass flow may be written as

$$j = D\nabla\omega = \frac{D}{\delta}(\omega_s - \omega_\infty) = \alpha_m(\omega_s - \omega_\infty) \qquad (2)$$

where α_m is the mass-transfer coefficient ($\alpha_m = D/\delta$); δ is the reduced thickness of a boundary layer; D is the vapour diffusion coefficient.

Liquid evaporation is a dynamic process; vapour molecules not only leave a liquid surface but also return to it and partially condense. The amount of liquid evaporated is the difference between the amount of the molecules which left the surface and that of molecules returned from the ambient medium.

Let the accommodation coefficient of molecules be denoted by A. It characterizes the probability of the molecule condensing on striking the surface. Then, the amount of molecules condensing on the liquid surface will be $\frac{1}{4}An\bar{u}$ where n is the number of molecules per unit volume and $\bar{u}$ is the mean thermal velocity of vapour molecules.

Assuming that the amount of molecules condensed at the surface is equal to that returned to the surface as a result of diffusion, the following equation is obtained

$$j_m = \tfrac{1}{4}Anmu = \alpha_m(\omega_\infty - \omega_s) = \tfrac{1}{4}A\omega_s\bar{u} \qquad (3)$$

where m is the mass of vapour molecules and ω is the concentration ($\omega = m\,.\,n$).

From equation (2) it follows

$$\omega_s = \frac{a_m}{\frac{1}{4}A\bar{u} + a_m}\omega_\infty, \qquad (4)$$

$$j_m = \frac{1}{[(1/\frac{1}{4}A\bar{u}) + (1/\alpha_m)]}\omega_\infty = k^*\omega_\infty \qquad (5)$$

where k^* is the proportionality factor between j_m and ω_∞ and is equal to

$$\frac{1}{k^*} = \frac{1}{\alpha_m} + \frac{1}{\frac{1}{4}A\bar{u}}. \qquad (6)$$

From equation (6) it follows that the total resistance ($1/k^*$) is equal to the sum of the diffusional resistance ($1/\alpha_m$) and the kinetic resistance ($1/\frac{1}{4}A\bar{u}$).

If the diffusional resistance is greater than the kinetic resistance ($1/\alpha_m \gg 1/\frac{1}{4}A\bar{u}$ or $\frac{1}{4}A\bar{u} \gg \alpha_m$), then from equations (4) and (5) it follows

$$\omega_s = \frac{\alpha_m}{\frac{1}{4}A\bar{u}} \ll \omega_\infty; \qquad \frac{1}{k^*} \approx \frac{1}{\alpha_m}; \qquad j_m = \alpha_m\omega_\infty \qquad (7)$$

i.e. the rate of condensation and, consequently, of evaporation is determined by the rate vapour (by diffusion) since $\alpha_m = D/\delta$. If the diffusional resistance is considerably less than the kinetic resistance ($1/\alpha_m \ll 1/\frac{1}{4}A\bar{u}$ or $\frac{1}{4}A\bar{u} \ll \alpha_m$), then we have

$$\omega_s = \omega_\infty, \qquad k^* = \tfrac{1}{4}A\bar{u} \qquad (8)$$

$$j_m = \tfrac{1}{4}A\bar{u}\omega_\infty \qquad (9)$$

i.e. the rate of condensation (evaporation) is determined by that of phase change of liquid into vapour.

Let the diffusion Nusselt number be denoted by Nu_D and the Knudsen number by Kn

$$Nu_D = \frac{\alpha_m l}{D}; \qquad Kn = \frac{\lambda}{l}; \qquad (10)$$

where l is the characteristic dimension (length of a liquid surface along a gas flow), and λ is the mean free path of vapour molecules. Then, taking into account the fact that the diffusion coefficient is proportional to the product $\bar{u}\lambda$ ($D = \Gamma \bar{u} \lambda$ where Γ is a numerical coefficient $\Gamma < 1$) it may be written

$$j_m = k^* \omega_\infty = \frac{\bar{u}\omega_\infty}{(1/\Gamma Nu_D Kn) + (1/\frac{1}{4}A)}$$

$$\approx \frac{\Gamma \bar{u} \omega_\infty}{(1/Nu_D Kn) + (1/A)} = \Gamma k^{**} \bar{u} \omega_\infty \qquad (11)$$

since, to a first approximation, it may be assumed that $\Gamma \approx \frac{1}{4}$. The total resistance $(1/k^{**})$ is equal to

$$\frac{1}{k^{**}} = \frac{1}{Nu_D Kn} + \frac{1}{A}. \qquad (12)$$

The diffusional resistance $(1/Nu_D \,.\, Kn)$ is inversely proportional to the product of the Nusselt and Knudsen numbers $(Nu_D \,.\, Kn)$.

Under ordinary conditions of liquid evaporation at atmospheric pressure the Knudsen number is small ($Kn \ll 1$), the Nusselt number being small as well. Then, the diffusional resistance $(1/N\bar{u}_D \,.\, Kn)$ is large in comparison to the kinetic one $(1/A)$, and the rate of evaporation is determined by the vapour transfer process [See equation (2)].

However, with evaporation into vacuum the Knudsen number Kn sharply increases ($Kn \sim 1$), therefore, in some evaporation regimes the diffusional resistance becomes smaller than the kinetic one ($Nu_D \,.\, Kn \gg A$), and the evaporation rate is determined by the rate of phase transition

$$j \approx \Gamma A \bar{u} \omega_s = \Gamma A \bar{u} \omega_\infty. \qquad (13)$$

Sometimes when studying evaporation under the action of an energy flux, the temperature jump at the interphase boundary is neglected, although it is clear that in such a case there should be no energy flow from one phase into another.

Since the problem on evaporation of a solid is important in practice, it is of interest to consider it in a more precise form than has been done hitherto. In the present section an attempt is made to analyse this problem. Subsequent consideration of the problem allows us to find some qualitative peculiarities of the process. which are usually omitted in the simplified consideration. The relations obtained may be further used for treating extensive experimental data on evaporation.

2. GAS DYNAMIC PROBLEM AND BOUNDARY CONDITIONS

The problem is started by considering the simplest part of the general problem on evaporation in vacuum—the vapour motion. Let it be assumed that the vapour may be considered an ideal gas of a constant adiabatic exponent γ. Possible departures from this assumption will be discussed further. Gas expansion into vacuum occurs in a rarefaction wave. Such flows are well studied, and their properties are given in detail, for example in [5, 6]. One-dimensional vapour expansion will be considered for definition purposes. Such motion is similar and represents a centered rarefaction wave. If the conditions at the surface of the solid phase are steady (or vary rather slowly), then near the surface of the solid the gas velocity should be equal to the local sound velocity [5, 6]. It is easy to understand that at the interface itself the condition under which the mass vapour velocity is equal to the velocity of sound may not be rigorously fulfilled. For this the well-known fact is sufficient to be noted that molecules moving from a solid surface have a Maxwell velocity distribution at a temperature equal to the surface temperature (molecules moving to a surface have another distribution; see below). It is possible to show that at the surface the gas velocity is not equal to that of propagation of small disturbances. In addition, it should be noted, that immediately near the surface of a solid the velocity distribution of molecules essentially differs from the equilibrium distribution, therefore, for the points near the surface, in general, there is no meaning in speaking about hydrodynamic boundary conditions which assume local equilibrium.

The equilibrium distribution is established at a distance of several mean free paths of molecules from the surface; however, as will be shown below, the parameters of this distribution essentially differ from the values applying at the surface. For these values to be defined, the kinetic equation for a "non-hydrodynamic" layer near a phase boundary has to be solved.

It should be noted that equilibrium distribution parameters may be calculated with the aid of the thirteenth-moment approximation. However, in this case such an approach is hardly justified. Indeed,' from the statement itself it is clear that the thickness of a "non-hydrodynamic" layer is several lengths of a free path, the gas state sharply changing in this region. In this case the ordinary procedure for the approximate solution of the Boltzmann equation may not be used. Such a case is similar to that of a strong shock wave (more precise, "strong shock wave of rarefaction") and for the Boltzmann equation to be solved, it is natural to use the method usually adopted for considering a structure of strong shock waves [8, 9]. This method uses a distribution function which represents the superposition of equilibrium distributions in front of and behind a shock wave with co-ordinate dependent coefficients. To substantiate such a choice of the distribution function on physical grounds one usually refers to a small width of a non-equilibrium zone, inside which are mainly molecules of equilibrium distributions in front of and behind the zone. For volumetric planes which are co-ordinate dependent to be determined, the Boltzmann equation is reduced to a differential one by the variational method or the method of moments [8, 9]. The approach described, although not mathematically rigorous, nevertheless corresponds well to the essence of the problem both in the case of a strong shock wave and in the case under consideration.

In accordance with the aforesaid, for the distribution function the following approximate expression is taken

$$f(x, \vec{v}) = \frac{1 + g(x)}{2} f_1(\vec{v}) - \frac{1 - g(x)}{2} f_2(\vec{v}) \quad (14)$$

where

$$f_1(\vec{v}) = \begin{cases} f_+(\vec{v}) & v_x > 0 \\ \beta f_-(\vec{v}) & v_x < 0 \end{cases}$$

$$f_2(\vec{v}) = f_-(\vec{v}) = n_1 \left(\frac{m}{2\pi k T_1}\right)^{\frac{3}{2}}$$

$$\exp\left(-\frac{(v_x - u_1)^2 + v_y^2 + v_z^2}{2kT_1} m\right)$$

$$f_+(\vec{v}) = n_0 \left(\frac{m}{2\pi k T_0}\right)^{\frac{3}{2}} \exp\left(-\frac{mv^2}{2kT_0}\right). \quad (15)$$

The function $g(x)$ and the parameters β, n_1, u_1, T_1 are to be determined.

For their determination there are the Boltzmann equation and the relation

$$u_1 = C(T_1) \quad (16)$$

expressing the equality between a mass gas velocity and the sound velocity at the boundary of the region where hydrodynamics is valid.† From the Boltzmann equation

$$v_x \frac{\partial f}{px} = \left(\frac{\partial f}{\partial t}\right)_{\text{collision}} \quad (17)$$

follow laws of conservation of flows of mass, momentum and energy

$$\left.\begin{aligned} &\int d\vec{v} f(x, \vec{v})\, v_x = C_1 \\ &\int d\vec{v} f(x, \vec{v})\, v_x^2 = C_2 \\ &\int d\vec{v} f(x, \vec{v})\, v_x v^2 = C_3 \end{aligned}\right\} \quad (18)$$

Relations (16) and (18) are sufficient to determine the parameters n_1, T_1, u_1 and β which completely characterize the hydrodynamic boundary conditions. Integration of equations (18) using equation (14) leads to the following system of equations

$$n_0 \cdot \sqrt{\left(\frac{kT_0}{2m\pi}\right)} = n_1 u_1 [1 + \beta \varphi_1(M)],$$

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† Relation (16) ensures steady-state conditions for the kinetic problem under consideration.

$$n_0 \frac{kT_0}{2m} = n_1 u_1^2[\tfrac{8}{5} - \beta\varphi_2(M)], \tag{19}$$

$$n_0 \left(\frac{2kT_0}{m}\right)^{\frac{3}{2}} = (\sqrt{\pi}) n_1 y_1^3 [4 + \beta\varphi_3(M)]$$

where

$$\varphi_1(M) = \left(\frac{e^{-M^2}}{M\sqrt{(\pi)}} - \operatorname{erfc} M\right),$$

$$\varphi_2(M) = \frac{1}{2}\left[\left(1 + \frac{1}{2M^2}\right)\operatorname{erfc} M - \frac{e^{-M^2}}{M\sqrt{(\pi)}}\right],$$

$$\varphi_3(M) = \frac{1}{2}\left\{\frac{[1 + (2/M^2)]}{M\sqrt{(\pi)}} e^{-M^2} - \left(1 + \frac{5}{2M^2}\right)\operatorname{erfc} M\right\}$$

$$M = u_1 \sqrt{\left(\frac{m}{2kT_1}\right)} = \sqrt{\left(\frac{\gamma}{2}\right)};$$

$$\operatorname{erfc} x = \frac{2}{\sqrt{(\pi)}} \int_x^\infty e^{-t^2}\, dt. \tag{20}$$

Boltzmann's equation (17) should be solved to find the function $g(x)$. In our case the function $g(x)$ is necessary only to assess the size of the non-equilibrium region. Owing to this fact, expressions such as $[f_0(x, \vec{v}) - f]/\tau$ may be used for the collision term. The calculations made by the variational method for such a collision term give the width of a non-equilibrium zone equal to the order of two mean free paths. Owing to this fact it is necessary in equation (20) to take $\gamma = \frac{5}{3}$ and $M = \sqrt{\frac{5}{6}}$ since at several mean free paths the internal degrees of freedom (if these exist) and the degree of condensation remain invariable.

Solving the system of equations (14)–(19) one obtains $\beta = 6{\cdot}291$, $T_1 = 0{\cdot}67\, T$ and $n_1 = 0{\cdot}31\, n_0$. Hence it is easy to calculate all the remaining hydrodynamic variables at the sound point. It is of interest to note the magnitude of the flow of molecules condensing at the surface of a solid. Assuming that the adhesion coefficient is unity (this is valid for metals), we obtain $J_-(J_+ = 0{\cdot}18$, i.e. 18 per cent of all molecules evaporated return to the surface.

The concentration of vapour molecules at the surface can also be calculated

$$n(0) = \int d\vec{v} f(0, \vec{v}) = \tfrac{1}{2}(n_0 + \beta n_1 \operatorname{erfc} M) = 0{\cdot}67\, n_0. \tag{21}$$

Obviously it is less than the concentration of saturated vapour having a temperature T_0. It is easy to show that at the sound point the concentration of vapour molecules may be higher than that of the saturated vapour. Indeed,

$$\frac{\rho_{0\,\text{sound}}}{\rho_0} = \frac{n_{0\,\text{sound}}}{n_0} \approx \exp\left[-\frac{\lambda}{T_1 k} + \frac{\lambda}{T_0 k}\right] = \exp\left[\frac{\lambda}{T_0 k}\left(-1 + \frac{T_0}{T_1}\right)\right]. \tag{22}$$

and the vapour will be saturated if $0{\cdot}31\, n_0 > n_{0\,\text{sound}}$, i.e. $T_0 < 0{\cdot}4\ \lambda/k$. This case is typical. Thus when expanding vapour into vacuum, supersaturation occurs near the surface and condensation should take place. This process will somewhat change the structure of a rarefaction wave. First of all, it ceases to be "similar" since there will appear a characteristic length determined by a finite condensation rate. Secondly, expansion (even at an infinitely large condensation rate) will already be described not by Poisson's adiabatic curve but by a "condensation" curve. The qualitative analysis of the problem of a rarefaction wave with condensation is given in [7]. If the initial supersaturation is sufficiently high, then condensation takes place as an equilibrium process and then, with a decrease in volumetric concentration of vapour molecules, there occurs freezing of the process and further expansion is described by Poisson's adiabatic curve.

3. PROBLEM ON HEAT CONDUCTION IN A SOLID

In the problem under consideration obviously there exist two limiting regimes, whose choice is determined by the conditions of heating of the solid: (1) regime in which the velocity of the evaporation front is determined by kinetics and (2) regime in which this velocity is determined by heat conduction.

Consider the first case. Its typical conditions are being created when evaporating a strongly absorbing solid under the action of radiation flow. Consider a one-dimensional steady-state problem. Let a radiation flow full on a surface ($x = 0$) of a solid $x < 0$ a part q of which is absorbed. In a co-ordinate system fixed to the phase boundary, the equations of the problem are of the form

$$\left.\begin{aligned} &+\frac{\mathrm{d}T}{\mathrm{d}z}v + a\frac{\mathrm{d}^2T}{\mathrm{d}z^2} = 0 \\ &-\varkappa\frac{\mathrm{d}T}{\mathrm{d}z}\bigg|_{z=0} = q - q' \\ &T(-\infty) = 0. \end{aligned}\right\} \qquad (23)$$

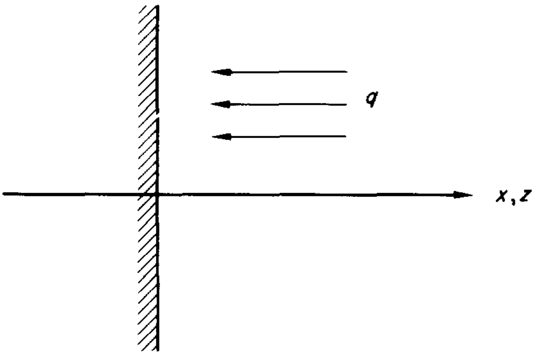

FIG. 1.

Here T is the temperature, $\varkappa$ and a are the thermal conductivity and thermal diffusivity, respectively; q' is the density of the energy flux consumed by evaporation; $z = x - vt$. For q' it is possible to write the expression

$$q' = mn_1u_1\left(\lambda + \frac{u_1^2}{2} + \frac{5}{2}\frac{k}{m}T_1 - 3\frac{k}{m}T_0\right) \qquad (24)$$

which follows from the previous paragraph of the present section (the heat capacity of a solid is $3R$ according to the law of Dulong and Petit). To transform equation (24) note that $n_1u_1 = n_Tv$ where n_T is the concentration of the solid phase, m is the mass of a molecule, v is the velocity of the evaporation front; moreover,

$$\frac{1}{2}\left(u_1^2 - \frac{5kT_1}{m}\right) = \frac{10}{3}\frac{kT_1}{m} \approx \frac{kT_0}{m}\,2.2. \qquad (25)$$

Finally

$$q' = \frac{n_0(T_0)}{1 + \beta\varphi_1(M)}\sqrt{\left(\frac{kT_0m}{2\pi}\right)}\left(\lambda - 0{\cdot}8\frac{kT_0}{m}\right) \qquad (26)$$

$$v = \frac{n_0T_0\sqrt{(kT/2\pi m)}}{n_T[1 + \beta\varphi_1(M)]}.$$

Upon integration of equation (23)

$$T = T_0\,\mathrm{e}^{-(zv/a)}. \qquad (27)$$

On satisfying the boundary condition at $z = 0$, the equation for determination of T_0 is obtained

$$1{\cdot}221\,q = n_0(T_0)\sqrt{\left(\frac{kT_0}{2\pi m}\right)}\left(\lambda + 2{\cdot}2\frac{kT_0}{m}\right). \qquad (28)$$

Upon solving this equation the velocity of the evaporation front is determined from the equation

$$v = \frac{q}{n_T(\lambda m + 2{\cdot}2\,kT_0)}. \qquad (29)$$

Consequently, the velocity of the evaporation front v is directly proportional to the specific heat flux. To compare the result obtained with the classical Stefan solution, data are presented on the rate of movement of the evaporation front obtained by solving heat conduction problems. From Stefan's solution it follows that the evaporation front propagates according to the law $\xi = \beta\sqrt{\tau}$ where β is a constant coefficient which depends on thermophysical properties of the solid [10–16]. The rate of propagation of the evaporation front will be

$$v = \frac{d\xi}{d\tau} = \tfrac{1}{2}\beta t^{-\frac{1}{2}}. \tag{30}$$

The specific heat flux will be equal to

$$q = \left(\frac{R_T C_T \rho_T}{\pi}\right)^{\frac{1}{2}} \frac{(T - T_0)\, t^{-\frac{1}{2}}}{\operatorname{erf}(\beta/2\sqrt{a_T})} \tag{31}$$

where a_T is the thermal diffusivity of the solid ($a_T = R_T/\rho_T C_T$).

From equations (30) and (31) it follows

$$v = \frac{\beta(\sqrt{\pi})\operatorname{erf}(\beta/2\sqrt{a_T})}{2\sqrt{(R_T C_T \rho_T)}\,(T_a - T_0)} \tag{32}$$

i.e. the rate of propagation of the evaporation front v is directly proportional to the specific heat flux q, as in equation (29), however, the factor of proportionality depends upon thermophysical properties of the solid (R_T, C_T, ρ_T), the evaporation front temperature T_0 and the ambient temperature T_a. Experimental data confirm relation (30) and, consequently, the relationship between v and q.

Thus, the qualitative dependence between the rate of propagation of the evaporation front and the heat flux supplied is the same, irrespective of the regime of heat sublimation. However, the mass-transfer mechanism and thermodynamic properties of a vapour on the solid surface and near it are essentially different. This explains the considerable disagreement between the experimental and predicted data for the Nusselt numbers if the latter are confirmed by the formulae of classical heat transfer between a solid surface and an ambient medium.

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Abstract—The correct formulation of the problem on evaporation of a solid into vacuum is considered. Basic evaporation characteristics are found.

It is shown that for some regime parameters the evaporation rate is determined by true kinetics of phase change (solid-vapour). There occurs a rarefaction wave, at the front of which vapour supersaturation takes place. The vapour pressure at the solid surface is less than the saturation vapour pressure at a given temperatures of the solid.

EVAPORATION D'UN SOLIDE DANS LE VIDE

Résumé—On considère la formulation correcte du problème de l'évaporation d'un solide dans le vide. On trouve les caractéristiques fondamentales de l'évaporation.

On montre que pour quelques paramètres de régime, le taux d'évaporation est déterminé par des cinétiques de conversion de phases (solide–vapeur). Une onde de raréfaction prend place au devant de laquelle existe un front de vapeur sursaturée. La pression de vapeur à la surface du solide est inférieure à celle de la vapeur saturée pour la température du solide.

VERDAMPFEN AUS DEM FESTEN AGGREGATZUSTAND IN EIN VAKUUM

Zusammenfassung—Die Arbeit behandelt die richtige Deutung des Problems der Verdampfung aus dem festen Aggregatzustand in ein Vakuum. Dabei wurden die grundlegenden Verdampfungscharakteristiken gefunden.

Es wird gezeigt, dass für einige Bereichsparameter die Verdampfungsrate durch die reine Kinetik der Phasenumwandlung bestimmt wird. Es tritt dort eine Verdünnungswell auf, an deren Front der Dampf eine Übersättigung aufweist. Der Dampfdruck an der festen Oberfläche ist kleiner als der des gesättigten Dampfes bei einer gegebenen Temperatur.