MECHANISM OF HEAT AND MASS TRANSFER IN THE VACUUM SUBLIMATION OF ICE

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Experimental data and certain theoretical results relating to the mechanism of vacuum sublimation of ice are presented. The escape of particles from the surface of the ice has been detected by high-speed photography, the velocity field has been constructed and the pressure field calculated in the vicinity of a subliming surface under vacuum conditions.

The mechanism of heat and mass transfer associated with vacuum sublimation is very complicated and so far little understood; therefore a strictly analytic (mathematical) investigation does not lead to reliable results. Many authors express contradictory views with regard both to the physical essence of the process and the choice of a method of calculating the necessary heat and mass transfer characteristics. Especially sharp discrepancies are observed in determining the coefficients of convective heat transfer, which are found from the heat and material balance equations. The question arises, why in a rarefied gas medium of low density are the coefficients of convective heat transfer, according to the results of certain authors, an order higher than the coefficients calculated on the basis of the ordinary theory of heat and mass transfer?

We set ourselves the task of finding, on the basis of special experiments, the explanation for these contradictions and discovering certain characteristics of the mechanism of heat and mass transfer in vacuum sublimation. For this purpose we conducted experiments in a vacuum apparatus – a thermobarochamber equipped with a high-speed motion-picture camera and a system for filming the sublimation process.

The thermobarochamber was a thermostated vacuum chamber permitting broad variations of the temperature ($T = 200$-$370°K$) and pressure ($p = 1 \cdot 10^{-5}$-$1 \cdot 10^{5}$ N/m^2) of the gas medium.

The experimental apparatus consisted of a vacuum chamber, a VN-4G oil fore pump, a BN-2000 booster vapor-ejector oil pump, a VA-5-4 vapor–oil diffusion unit, a four-stage Freon refrigerating machine, nitrogen trap-separators, etc.

The vacuum chamber, a cube with a volume of 0.25 m^3, was made of stainless steel. The chamber was placed in thermostated vessel with a volume of 0.5 m^3. For filming purposes the chamber was equipped with an optical-glass illuminator. The gas pressure in the chamber was measured with a thermocouple vacuum gauge. The temperature of the working body, the chamber of the working body, the chamber walls and the surrounding medium was measured with copper–constantan microthermocouples and suitable high-precision laboratory instruments. The loss of mass during sublimation was recorded on a VTK-500 laboratory balance. As the subliming body we used a sphere of ice 80 mm in diameter obtained by freezing thoroughly degassed distilled or ordinary water in a special spherical mold. The metal mold was cooled either in an ordinary refrigerator over a long period or rapidly by means of liquid nitrogen. Depending on the freezing technique, the ice was either transparent, penetrated by fine radial channels, or milky-colored without visible radial channels.

The sphere of ice with thermocouples fixed to its surface was suspended by a thin nylon thread from the beam of the laboratory balance. A light source was set up about 200 mm from the surface of the sphere. The focused beam of the lamp illuminated the surfaces of the sphere, thus acting as a source of radiant energy.

The filming experiments were conducted at constant values of the temperatures of the vacuum chamber walls and the gas medium surrounding the ice sphere at pressures $p = 40$ and 4 N/m^2.

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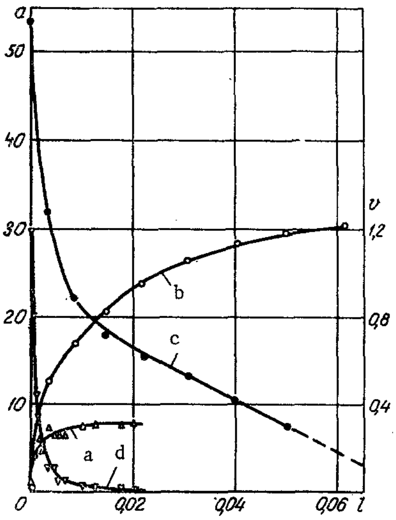

Fig. 1. Velocity (m/sec) and acceleration (m/sec^2) as functions of the distance (m) from the surface of the body for moving particles at various pressures of the surrounding medium [a and d) velocity and acceleration at p = 40 N/m^2; b and c) velocity and acceleration at p = 4 N/m^2].

All the necessary measurements were made under steady state conditions in the working chamber.

In the first series of experiments the sublimation process was photographed with a Zeitlupe-I high-speed motion-picture camera. As a result of the experiments, on the surface of the sphere we detected the presence of a "nap" of crystals that flew off into the evacuated space. In order to obtain a direct image of the trajectory of a separated particle, we photographed the process through a rotating disk with slits. At long exposures we obtained an intermittent trace of the flying particle on the film. The time interval corresponding to an individual segment of this trace was determined from the speed of the disk and the number of slits. At the same time, we photographed a reference body set up in the chamber in the plane of flight of the particles. Thus, each particle flight trajectory enabled us to construct a graph of distance versus time $l = l(\tau)$, and, using a graphic method of determining the velocity v and acceleration a, from these graphs we obtained the relations $v = v(\tau)$ and $a = a(\tau)$, i.e., from the known form of the velocity function $v = f(l, \tau)$ we determined the motion of the particle

$$v = \lim_{\Delta\tau\to 0} \frac{l_{n+1} - l_n}{\tau_{n+1} - \tau_n} = \frac{dl}{d\tau}; \quad a = \lim_{\Delta\tau\to 0} \frac{v_{n+1} - v_n}{\tau_{n+1} - \tau_n} = \frac{dv}{d\tau}. \tag{1}$$

By analyzing the photographs, we obtained a picture of the gas velocity field near the surface of the ice defined by the vector v, on the assumption that in view of its smallness the visible particle moves together with the gas. The fact that the segments of the particle trace increase in size with distance from the surface of the ice indicates the presence of acceleration.

The state of the surrounding medium (vapor–gas mixture) is described by the thermodynamic parameters $p(l, \tau)$, $T(l, \tau)$, $\rho(l, \tau)$. These parameters: pressure (p), temperature (T), and density (ρ), can be determined from the particle velocity field $v(l, \tau)$. If, as a first approximation, we assume that the particle moves together with a small volume of gas (particle motion without friction), then from the ordinary equation of hydrodynamics we can write

$$\rho \frac{dv(l, \tau)}{d\tau} = -\frac{\partial p(l, \tau)}{\partial l} - \rho(l, \tau) g \sin\vartheta, \tag{2}$$

where g is the acceleration of gravity, and ϑ the angle between the horizontal and the tangent to the flight trajectory of the particle at the point of intersection of the trajectory and the surface of the body.

If by a we denote the total acceleration ($a = dv/d\tau + g\sin\vartheta$), then Eq. (2) takes the form

$$\frac{dp}{dl} = -\rho a. \tag{3}$$

At a certain distance from the surface of the sphere the acceleration of unit volume of gas $a = 0$, i.e., the pressure gradient is also equal to zero. The thermodynamic properties of an isolated volume element of gas within the region where the acceleration $a = 0$ may be assumed equal to the properties of the surrounding medium in the vacuum chamber and can be determined using the equation of state of a perfect gas ($\rho = p/RT$).

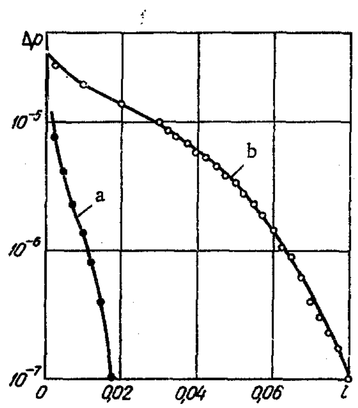

Fig. 2

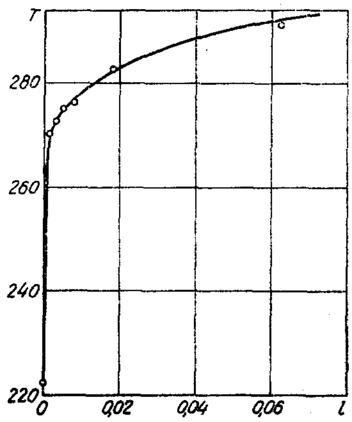

Fig. 3

Fig. 2. Pressure difference (N/m^2) as a function of distance (m) from the surface of the body at various pressures of the surrounding medium [a) at p = 40; b) 4 N/m^2].

Fig. 3. Temperature distribution (°K) near body at p = 40 N/m^2.

Thus, knowing p and T in the region where a = 0, by the method of successive approximations, on the basis of Eqs. (3) and the equations of state we determine the pressure and density field near the subliming surface.

It is clear from Fig. 1 that for pressures of about 40 N/m^2 (curves a and d) an important deformation of the velocity profile is observed at a small distance from the surface of the ice (of the order of 0.02 m). The same applies to the acceleration profile, which on the same interval falls sharply to zero, i.e., when the motion of the particle becomes uniform (acceleration a = 0).

At pressures of about 4 N/m^2 (curves b and c) the deformation of the velocity and acceleration profiles extends to a greater distance from the sublimation surface (or the order of 0.08 m), while at lower pressures of the surrounding medium we observe a much smaller drop in acceleration than at higher pressures.

If we denote by p_1 the gas pressure at the sublimation surface and by p_2 the pressure at a point in the medium where the acceleration of the particle a = 0, then it is clear from Fig. 2 that the pressure drop $\Delta p = p_1 - p_2$ depends on the pressure in the chamber, and the pressure ratio p_1/p_2, which can be related with the velocity of the gas in accordance with Bernoulli's equation

$$v^2 = \frac{2k}{k-1}\,\frac{p_1}{\rho_1}\left[1 - \left(\frac{p_2}{p_1}\right)^{\frac{k-1}{k}}\right],$$

increases as the total pressure falls.

This form of the relation $\Delta p = f(l)$ is explained by the very small change of pressure near the subliming body as compared with the pressure in the chamber (Fig. 2). The same sharp deformation of the profile is also observed in the temperature distribution near the subliming surface (Fig. 3), where the temperature gradient changes sharply at a small distance from the surface.

Values of the particle velocities determined by the photographic technique were compared with values of the rate of separation of vapor from the sublimation surface calculated from the relation $v = j_m/\rho$. The comparison revealed that the velocities determined by different methods are similar in value. Thus, for example, at p = 40 N/m^2 the velocity of the vapor leaving the surface v = 0.28 m/sec, while the particle velocity determined by photography v = 0.3 m/sec, i.e., the difference lies within the limits of experimental accuracy.

Upon analyzing the graphs of the relations (v, a, p, T) = f(l), we conclude that at the surface of the subliming body there is a layer of vapor–air mixture whose thermodynamic properties differ from the properties of the surrounding medium. It is precisely in this layer that the velocities, temperatures, and pressures change from the values at the sublimation surface to the values in the medium. The thickness of the layer δ is basically determined by the density of the medium in the vacuum chamber, where $\delta \sim 1/\rho$. In this layer, moreover, the principal resistance to convective heat and mass transfer is concentrated. This layer cannot be identified with the boundary layer at normal pressure, through which enormous volumes of vapor are transported, because of its great extent and low density.

Intense heat transfer to the sublimation surface through the rarefied layer can be realized only by electromagnetic radiation. Other methods of heat supply (convection, conduction) through a low-demsity medium are ineffective. Recent data [1] have shown that in the vacuum sublimation of ice the coefficient of convective heat transfer fluctuates between values close to unity. However, the thermal conductivity of the vapor is almost two orders less than α_c. Consequently, it may be assumed that the high sublimation rates obtained in the experiments are the result of heat transfer not by convection or conduction but by radiant energy. Under vacuum conditions the convective component can play an important part only in connection with mass transfer processes.

In the vacuum chamber very favorable conditions are created for external mass transfer, when small fluctuations of pressure, i. e., affecting the fourth decimal place, can cause mass transfer velocities of several meters per second. In the presence of an intense source of mass release (sublimation of ice in a vacuum) and in the absence of inleakage the total pressure in the chamber may be assumed equal to the saturation pressure and the chamber may be considered to contain vapor only. In this case the rate of mass transfer is determined by the vapor pressure drop. Thus, the motive force of the flow of vapor from the sublimation surface is the pressure gradient $\Delta p/l$.

In considering the heat balance constructed for the processes of sublimation of ice from the equation $q = rj_m$, it should be noted that this equation does not always satisfy the identity, since it does not take into account the phenomena of mechanical separation and entrainment of the particles. Quantitatively, this imbalance may be expressed in different proportions and will depend on the method of obtaining the ice, i. e., on its structure, etc.

On the other hand, in separating the heat flux into its individual components incorrect allowance for the radiative component leads to exaggerated values of α_c. Thus, high values of the convective component α_c may be obtained either as a result of explicitly disregarding the escape of molecular complexes or solid particles into the surrounding space or as a result of incorrectly calculating the radiative component of the heat flux. Basically, these factors alone determine the high theoretical values of α_c.

Using the equation $q = rj_m$, we assume that sublimation proceeds uniformly over the entire surface. Correspondingly, we assume that the vapor separating from the surface is uniformly distributed over the entire space. In the ideal case of uniform distribution of the energy sources around a sublimation surface having a finely crystalline structure without foreign inclusions, the sublimation process may, in fact, proceed more or less uniformly. But at the microscopic level, the sublimation process is quasi-explosive in character, with local nonuniformity of the rates of vapor release.

Subliming materials (ice, etc.) are not homogeneous but consist of many microscopic crystals and grains. Between the crystals in the body of the ice there are accumulations of gaseous inclusions, which, in the form of microvolumes, are under a certain pressure. These gaseous inclusions do not react chemically with each other or with the ice crystals. If the gaseous inclusions are contained in an absolute closed volume, they will be in hydrostatic equilibrium with the surrounding medium; however, in the case of dislocation processes leading to additional compression of the gas) or local disturbances of the continuity of the medium (leading to the formation of microcracks) the gaseous inclusions may be a source of quite intense microexplosive activity.

In the vacuum sublimation of ice the loss of mass will not be uniform over the entire surface, and as a result of the nonuniform density of gaseous inclusions and the difference in crystal orientation the losses at some points of the surface will be greater, and at others less. This leads to the formation of fine openings in the surface of the ice, which acquires a mottled rough appearance. Such a surface is capable of reducing the optical reflective properties and increasing the absorbing properties of the ice.

NOTATION

l is the distance, m;
τ is the time;

q is the specific heat flux;
j_m is the sublimation rate;
r is the latent heat of sublimation;
α_c is the coefficient of convective heat transfer;
R is the gas constant.

LITERATURE CITED

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