RELAXATION PHENOMENA IN NONSTATIONARY EVAPORATION OF LIQUID

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Based on the phenomenological theory – the thermodynamics of irreversible processes – an analysis of nonstationary process of liquid evaporation is carried out. It is shown that a sharp change in the heat-flux magnitude causes the occurrence of processes of oscillating relaxation of vapor generation in the system.

Introduction. The processes of evaporation and condensation are very important in such fields of contemporary engineering as vapor-generating systems and contact heat exchangers in industrial power equipment, vacuum metallurgy, production of superpure substances and samples with special properties, in microelectronics and different chemical technologies, in systems of supply of internal combustion engines, cryogenic rocket engines, and also in different technological processes of the pharmaceutical industry. Especially important, from the point of view of exploitation safety, are the processes of evaporation in engineering calculations of transient regimes of vapor-generating systems of high-power electric stations.

At the present time, the problem of a stationary evaporation-condensation process has been rather well studied. Among the earlier works in this area are the investigations carried out by Herz and Knudsen [1, 2], who suggested the well-known Herz-Knudsen relation for the rate of evaporation under stationary conditions:

$$J = \beta \, \frac{p_{\rm s} - p_{\infty}}{\sqrt{2\pi m k T}} \,,$$

where β is the empirical coefficient of evaporation that characterizes the ratio of the number of molecules that were reflected by the phase interface to the number of molecules incident on this surface and p_s is the pressure of the saturated vapors of a liquid at the temperature *T*. From the point of view of molecular-kinetic theory, the rate of evaporation of a condensed phase with a temperature much smaller than the binding energy of atoms is determined by two processes: separation of atoms from a surface due to thermal motion and their return as a result of collision with the atoms of a gas phase. Thus, the Herz-Knudsen relation represents a difference of two one-sided Maxwellian fluxes of particles. A detailed review of earlier works on the phenomena of evaporation and condensation can be found in [3, 4].

Further investigations of the processes of evaporation and condensation were carried out on the basis of the Boltzmann kinetic equation and its models at small transfer velocities. In [5, 6], the first kinetic theory of evaporation was probably constructed for the first time with account for the change in the function of gasphase distribution due to macroscopic motion of the medium. At small degrees of the nonequilibrium state, for the resultant flux of substance J = nu there is a small (in comparison with the mean thermal velocity of molecules) macroscopic velocity of vapor motion u in the direction of the normal to the phase interface; n is the concentration of molecules per unit volume. In [7–9], a more detailed kinetic analysis of the processes of

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evaporation and condensation is made, in which the use of a two-flow function in simulation of the process makes it possible to take into account both the macroscopic motion of the vapor phase and the discontinuous character of the distribution function near the phase interface. The Boltzmann kinetic equation is solved by the method of moments in a stationary formulation of the problem. In [10], similar methods are used for the analysis and numerical solution of one-dimensional stationary problems that simulate intense stationary processes of evaporation and condensation. As a result of the solution of the gas-dynamics equations for the region exterior to the Knudsen layer, the approximation of the function of vapor distribution on the outer boundary of the Knudsen layer was obtained.

In [11], the process of recondensation at a small temperature difference was considered. The distribution function was obtained as a result of approximate solution of the Boltzmann stationary equation. The dependence of the distribution function on the vapor velocity was taken into account in a zero approximation. In [12], the problem of vapor recondensation was considered from the standpoint of the contemporary kinetic theory by solving the Boltzmann equation with correct formulation of the problem. Conditions for the occurrence of an inverse temperature profile on the phase interface for a continuous regime were established. At the present time, the development of methods of solving the Boltzmann kinetic equation has made it possible to carry out a detailed investigation of the problem of stationary evaporation or condensation. A review of publications on the contemporary state of the problem can be found, for instance, in [13].

The processes of nonstationary evaporation or condensation, when the magnitude of the molecular flux of an evaporating substance is a function of time, have been hardly studied at all up to now. In this case, in accordance with the Le Chatelier–Brown principle, a rapid change in the magnitude of the flux of an evaporating substance must lead to such changes in a system that could result in compensation of this change [14, 15].

The effect of the dynamics of a change in the magnitude of the flux of the evaporating substance on the rate of evaporation can be obtained in hydrodynamic approximation. As is seen from the Herz-Knudsen formula, the magnitude of pressure over the surface of the Knudsen layer is not constant and depends on the hydrodynamic conditions of vapor flow and must be found from the Navier–Stokes equations:

$$\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} = -\frac{1}{\rho} \frac{\partial p}{\partial x} + v \frac{\partial^2 u}{\partial x^2}.$$
(1)

Having integrated (1) over x and used the theorem of the mean, we obtain an expression for the pressure over the surface of the Knudsen layer:

$$p(t) = p_{\infty} - \rho l_{\rm ef} \frac{\partial u}{\partial t} - \frac{\rho}{2} u^2 + \mu \frac{\partial u}{\partial x}, \qquad (2)$$

where $l_{\rm ef}$ is the characteristic macroscopic scale of the process.

Substituting expression (2) into the Herz-Knudsen equation finally yields

$$J(t) = \frac{p_{\rm s} - p_{\infty} - \rho l_{\rm ef} \frac{\partial u}{\partial t} - \frac{\rho}{2} u^2 + \mu \frac{\partial u}{\partial x}}{\sqrt{2\pi m k T}}.$$
(3)

It should be noted that the term of the form $\rho l_{ef} \frac{\partial u}{\partial t}$ in Eq. (3) determines the inertia properties of the vapor flow, $\frac{\rho}{2}u^2$ is the dependence of the vapor-generation rate on the kinetic energy of the flow, and $\mu \frac{\partial u}{\partial x}$ repre-

sents the effect of viscous friction, which in turn depends on the specific configuration of the system of vapor removal, the presence of sections of contraction or expansion of the channel, curvilinear sections, steps, etc.

The problem of nonstationary evaporation of a condensed substance into vacuum was solved numerically in [16] on the basis of the Boltzmann homogeneous model equation. Also analyzed there was the process of transition from a free molecular regime of vapor expansion in the initial stage of evaporation to the regime of motion of a solid medium. As a result of numerical solution of the kinetic equation, the time dependences for the temperature, evaporating substance flux density, and vapor density were obtained as a result of numerical solution of the kinetic equation. Of interest is the wavy character of the change in temperature due to relaxation phenomena in establishment of the reverse molecular flux. Collisions of atoms in a gas phase lead to the incipience of the reverse molecular flux and to a decrease of the "pure" evaporation rate. As shown in [16], the period during which the reverse flow is stabilized is equal to about 20 times that of the free path, and it can be conventionally regarded as the time of the development of the hydrodynamic regime of vapor motion, even though actually such a regime is not attained inside the Knudsen layer. It was also shown in [16] that pulse heating can create conditions under which the hydrodynamic regime of evaporation does not exist at all.

Thus, a detailed investigation of the dependence of the evaporation rate on the thermodynamic parameters of the system in the case of a nonstationary process is of considerable scientific interest. The aim of the present work is a theoretical and experimental investigation of the process of nonstationary evaporation in a liquid-saturated vapor system in the case of a rapid change of one or several thermodynamic parameters of the system.

Phenomenological Description of Substance Evaporation in the Thermodynamics of Irreversible Processes. In the thermodynamics of irreversible processes, it is customary to use linear phenomenological laws [14, 15] which in the neighborhood of equilibrium can be written in the form

$$J_i = \sum_j L_{ij} \frac{\partial \phi}{\partial \alpha_j}, \tag{4}$$

where $J_i = \partial \alpha_i / \partial t$ is the generalized velocity and $\partial \phi / \partial \alpha_j = X_j$ is the generalized force. Here, the phenomenological coefficients are interrelated via the Onsager reciprocity relation $L_{ik} = L_{ki}$. In the case of two phases characterized by parameters α_1 and α_2 that are interrelated via the relations

$$\alpha = \frac{\alpha_1}{\alpha_1 + \alpha_2}, \quad 1 - \alpha = \frac{\alpha_2}{\alpha_1 + \alpha_2}, \quad \alpha_1 + \alpha_2 = 1,$$

the thermodynamic potential of the system is written as

$$\phi = \phi_1 \alpha + \phi_2 \left(1 - \alpha \right) \,. \tag{5}$$

Equations (4) and (5) yield

$$J = L \left(\phi_1 - \phi_2 \right) \,. \tag{6}$$

For the process of evaporation, expression (6) represents the Herz-Knudsen formula for the evaporation rate. Assuming L to be equal to $\beta v/4$, we obtain

$$J = \beta \frac{\nu}{4} \left(n_{\rm s} - n \right) \,, \tag{7}$$

where $v = \sqrt{8kT/\pi m}$.

Expression (4) represents a series expansion of the functional dependence between the flux and generalized thermodynamic force, in which only the first order is taken into account. In [17], it is shown that for highly intense transient processes in the expression for a generalized flow one has to take into account a larger number of expansion terms:

$$J_{i} = L_{i}^{\tau} \frac{\partial J_{i}}{\partial t} = \sum_{k} \left(L_{ik} X_{k} + L_{ik}^{\tau} \frac{\partial X_{k}}{\partial t} \right).$$
(8)

Equation (8) can also be obtained on the basis of the Le Chatelier–Brown principle, according to which a sharp change in the magnitude of the flux in a system must induce the appearance of a gradient of the corresponding thermodynamic parameter that hinders this change:

$$\frac{dJ_i}{dt} = -\sum_j L_{ij}^{**} \left(\frac{\partial \phi}{\partial \alpha_j}\right)_{\text{ind}}.$$
(9)

Combining expressions (4) and (9), we obtain Eq. (8).

Thus, for a two-phase system with highly intense transient processes expression (6) becomes

$$J = L_J \frac{dJ}{dt} + L (\phi_1 - \phi_2) + L_{\phi} \frac{d}{dt} (\phi_1 - \phi_2) .$$
⁽¹⁰⁾

For nonstationary evaporation, expression (7), with (8) and (10) taken into account, is represented as

$$J = \tau_{\rm v} \frac{\partial J}{\partial t} + \beta \frac{v}{4} (n_{\rm s} - n) + \tau_{\rm p} \beta \frac{v}{4} \frac{\partial (n_{\rm s} - n)}{\partial t}$$
(11)

or, assuming that vapor obeys the perfect gas state law and using an explicit expression for the arithmetic mean velocity of molecules and also omitting the last term in Eq. (11), for the vapor flux we obtain

$$J = \tau_{\rm v} \frac{\partial J}{\partial t} + \frac{\beta}{\sqrt{2\pi m k T}} \left(p_{\rm s} - p \right) \,. \tag{12}$$

where τ_v is the characteristic time of relaxation of the vapor mass flux.

The first and third terms in Eq. (11) reflect the inertia properties of a molecular flux of the evaporating substance, which according to the Le Chatelier–Brown principle hinders an instantaneous change in the flux with change in the thermodynamic parameters of the system. It should be noted that a similar expression for the rate of vapor generation under nonstationary conditions can be obtained from hydrodynamic considerations on substitution into Eq. (3) of the relation J(t) = nu, which connects the rate of vapor generation J(t)with hydrodynamic velocity of vapor u:

$$J(t) = \frac{p_{\rm s} - p_{\infty} - \frac{\rho l_{\rm ef}}{n} \frac{\partial J}{\partial t} - \frac{\rho}{2n} J^2 + \frac{\mu}{n} \frac{\partial J}{\partial x}}{\sqrt{2\pi m k T}}$$
(13)

or

$$J(t) = \beta V(n_{\rm s} - n) + \frac{\partial J}{\partial t} \frac{\beta l_{\rm ef}}{\nu}.$$
(14)

From Eq. (14) it is seen that $\tau_v = \beta l_{ef}/v$.

Dynamics of Pressure Change in a Closed Volume in Nonstationary Evaporation. The character of pressure change in evaporation in a closed volume when the magnitude of this pressure is smaller than the pressure of saturated vapors at a prescribed temperature seems to have been investigated for the first time in [18, 19]. The relationship between an increase in pressure and evaporation rate can be obtained assuming that the vapor pressure in a volume is leveled out instantaneously (i.e., it is the same throughout the entire volume). The concentration of vapor molecules in the volume in evaporation is a function of time and can be presented in the form

$$n(t) = n_0 + S \int_0^t J dt .$$
 (15)

Assuming the system to be thermostatted, differentiating the equation of state of a perfect gas with respect to time, and taking into account Eq. (7), we obtain

$$\frac{dp}{dt} = \frac{\beta S}{V} \left(p_{\rm s} - p \right) \left(\frac{kT}{2\pi m} \right)^{1/2}.$$
(16)

In [18], the integral representation of (16) was used:

$$\ln (p_{\rm s} - p) = -\frac{\beta S}{V} \left(\frac{kT}{2\pi m}\right)^{1/2} t + \text{const} .$$
⁽¹⁷⁾

Relations (16) and (17) were checked experimentally; they hold with a sufficient accuracy for slow processes when the evaporating-substance flux density J changes smoothly and monotonically:

$$\left|\tau_{v}\frac{\partial J}{\partial t}\right| << 1.$$
⁽¹⁸⁾

But when the rate of change in the evaporating substance density is rather great, the system of thermodynamic equations that describe the process must take into account the inertia term in expression (12).

We consider the process of liquid evaporation due to constant supply of heat to the system. The amount of vapor formed per unit time from a unit surface can be calculated in this case from the energy conservation equation:

$$Jm = \frac{P_Q V}{hS} \,, \tag{19}$$

where P_0 is the specific volumetric power of heat generation in the system.

The last term in Eq. (12) describes the change in the rate of evaporation due to the relaxation of local pressure and, therefore, it can be neglected, since the speed of this process is close to the speed of sound. Assuming that the vapor obeys the equation of state of a perfect gas and using Eq. (12), it is possible to obtain the equation that describes the dynamics of a change in pressure in the system in nonstationary evaporation:

$$\frac{\tau_{\rm v}V}{SkT}\frac{d^2p}{dt^2} + \frac{V}{SkT}\frac{dp}{dt} + \frac{\beta}{\sqrt{2\pi mkT}}\left(p_{\rm s} - p\right) = 0.$$
⁽²⁰⁾



Fig. 1. Change in the pressure (a) and dimensionless value of additional vapor generation (b) as functions of time on instantaneous cessation of heat supply to the system and drop in the pressure.

Solution of Eq. (20) will be obtained in the form

$$p_{\rm s} - p = p_{\rm m} \exp\left(-\frac{t}{2\tau_{\rm v}}\right) \cos\left(\left[\frac{\beta SkT}{2V\tau_{\rm v}\sqrt{2\pi mkT}}\right]^{1/2}t\right),\tag{21}$$

where the quantity $p_{\rm m}$ will be determined from Eq. (19):

$$p_{\rm m} = \frac{P_Q V \sqrt{2\pi m kT}}{\beta h Sm}$$

Equation (21) describes a change in pressure on instantaneous cessation of the heat supply to the system. The characteristic graph of the dependence of $p_s - p$ on time t is presented in Fig. 1a.

The amount of vapor formed in the system from the instant of cessation of the heat supply is calculated from the equation

$$\Delta G(t) = \frac{P_Q V}{hSm} \exp\left(-\frac{t}{2\tau_v}\right) \cos\left(\left[\frac{\beta SkT}{V\tau_v \sqrt{2\pi mkT}}\right]^{1/2} t\right).$$
(22)

Using the quantity

$$\tau_{\rm r} = \frac{V}{\beta S v}$$
,

as one other characteristic scale of time measurement and the quantity

$$\Delta G_{\rm r} = \frac{P_Q V}{hSm} \, \tau_{\rm r} \, ,$$

as the scale of measurement of the amount of vapor, we can write Eq. (22) in a dimensionless form as

$$\Delta G(t) = \frac{1}{\tau_{\rm r}} \exp\left(-\frac{t}{2\tau_{\rm v}}\right) \cos\left(\left[\frac{1}{4\tau_{\rm v}\tau_{\rm r}}\right]^{1/2} t\right). \tag{23}$$

In our opinion, the quantity τ_r is associated with the relaxation of mass transfer that involves condensation of the supersaturated vapor.



Fig. 2. Graph of the excess amount of vapor to be accumulated in the system from the time of cessation of the heat supply in comparison with the process of slow vapor generation.

Fig. 3. Dimensionless additional vapor generation vs. the phenomenological parameter τ_v/τ_r that characterizes the inertia properties of the system.

The time dependence of the dimensionless quantity of additional vapor generation caused by the instantaneous cessation of heat supply to the system is presented in Fig. 1b (for comparison the dashed curve represents the graph of the dimensionless quantity of additional vapor generation calculated from the Herz-Knudsen relation [18]). The excessive amount of vapor which will be accumulated in the system from the instant of cessation of the heat supply in comparison with the process of slow vapor generation described by the Herz-Knudsen relation can be calculated from the formula (see Fig. 2)

$$\Delta G_{\rm G-K}(t) = \frac{1}{\tau_{\rm r}} \int_{0}^{t} \left(\exp\left(-\frac{t}{2\tau_{\rm v}}\right) \cos\left(\left[\frac{1}{4\tau_{\rm v}\tau_{\rm r}}\right]^{1/2}t\right) - \exp\left(-\frac{t}{4\tau_{\rm r}}\right) \right) dt$$

The magnitude of additional vapor generation can be obtained by integrating Eq. (23) over the time from zero to infinity:

$$\Delta G^* = 2 \left(\frac{\tau_{\rm v}}{\tau_{\rm r}} \right)^{3/2} / \left(1 + \frac{\tau_{\rm v}}{\tau_{\rm r}} \right). \tag{24}$$

As is seen from Eq. (24), the value of the additional vapor generation is a function of the inertia parameter τ_v . The dependence of the dimensionless value of the additional vapor generation caused by instantaneous cessation of the heat supply to the system on the dimensionless number τ_v/τ_r , which characterizes the inertia properties of the process, is presented in Fig. 3.

Experimental Methods and Equipment. As the object of investigation in this experiment we used a hermetically sealed cell equipped with optical windows, branch pipes for vapor evacuation, and a heating element. The vapor pressure in the cell was measured by a piezoceramic probe connected to a charge-sensitive amplifier and an oscillograph. The cell had several copper-constantan thermocouples imbedded for measuring the liquid temperature near the phase interface and the vapor temperature. The liquid in the cell could be heated either by a heater or as a result of volumetric evolvement of Joulean heat in electrolysis. On the experimental rig described, two types of experiments were conducted to investigate the relaxation of the thermodynamic parameters of the vapor in the cell: (1) with a sharp reduction of vapor pressure over the phase-change surface and (2) with a rapid change in the liquid temperature as a result of pulse heating. The experiments were run mainly on degassed distilled water.



Fig. 4. Oscillogram of pressure relaxation in a thermostatted volume on a sharp decrease in the vapor pressure over the liquid surface as a result of rapid evacuation of the cell.

Results of Measurements and Discussion. Figure 4 presents the characteristic graph of pressure relaxation in the cell on sudden reduction of vapor pressure over the liquid surface as a result of rapid evacuation of the cell. The graph shows that the process is of an autooscillatory character.

The experimental graph in Fig. 3 can be approximated with a certain degree of error by relation (21). Though the pressure oscillations observed are of an explicitly anharmonic nature, the graph at least allows us to find that

$$\left[\frac{\beta SkT}{V\tau_{\rm v}\sqrt{2\pi mkT}}\right]^{1/2} = 2\pi \; ; \; \frac{1}{2\tau_{\rm r}} = 5 \; , \tag{25}$$

whence $\tau_v \sim 0.1$ and $\tau_r \sim 0.01$. The anharmonic nature of the pressure oscillations is due to the different mechanisms of heat exchange in the system: (1) during increase in the vapor generation flux, heat is absorbed in phase transition due to liquid cooling; (2) during decrease in the vapor generation flux, the vapor is partially condensed and heat is evolved in the microparticles of the condensate and subsequently is transferred to the gas phase. At the same time, the quantity τ_v for water in the case of surface evaporation can be evaluated from the molecular-kinetic theory. If we consider the density of the flux of vapor molecules directly near the liquid surface as a difference of two molecular fluxes (evolved by the liquid and absorbed by the phase interface from the surrounding vapor) and use, for calculation of the density of the flux of molecules, the nonstationary distribution function obtained in the 13-moment approximation of the solution of the Boltzmann kinetic equation, the expressions for τ_v and τ_r can be represented as [20]

$$\tau_{\rm v} = \frac{2\beta m\delta}{(2-\beta)\sqrt{2\pi mkT}}, \quad \tau_{\rm r} = \frac{V}{\beta S}\sqrt{\frac{\pi m}{8kT}},$$

where δ is the thickness of the Knudsen layer over the phase interface. From the physical viewpoint, the presence of inertia properties in the process of vapor generation is associated with the additional expenditure of energy in rapid passage of molecules through the Knudsen layer.

Evaluation for distilled water under normal conditions yields $\tau_{\nu} \sim 0.005$ and $\tau_r \sim 0.01.$

The excess of the experimental value over the theoretical one indicates that the great contribution to the inertia parameter τ_v is due to the nonstationary macroscopic processes of vapor motion.

Conclusions. The application of the method of irreversible thermodynamics to the phenomenological analysis of nonstationary evaporation and also the results of experimental investigation of pressure relaxation

in a thermostatted volume on sharp change in the flux of vapor generation points to the substantial effect of the inertia properties of a thermodynamic system on the dynamics of the development of a stationary process. In calculation of nonstationary conditions of operation of industrial fuel-power engineering equipment, it is necessary to take into account such phenomena as additional vapor generation and inductive vapor drop on a sudden change in the vapor-generation flux.

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NOTATION

p, pressure; T, temperature; R, universal gas constant; J, specific density of the molecular vapor flux; u, flow velocity; v, arithmetic mean velocity of molecules; m, mass of one molecule; n, density number of molecules; α , generalized coordinate; ϕ , thermodynamic potential; v, kinematic viscosity; ρ , vapor density; S, surface area of evaporation; V, volume of the system; h, specific heat of vapor generation; τ , characteristic time of relaxation; μ , dynamic viscosity; t, time; L, phenomenological coefficients; k, Boltzmann constant. Subscripts: ∞ , initial parameters of the system; s, parameters at the saturation of liquid; i, j, k, current subscripts; r, relaxation of the thermodynamic system; v, relaxation of the process of vapor generation; m, maximum value; Q, heat release in a thermodynamic system; p, relaxation of pressure inhomogeneity over the space.

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